Equivalence theorems between the solutions of the fourth-order modified contracted Schrödinger equation and those of the Schrödinger equation

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Four basic theorems involving the correlation matrices are demonstrated here. One of these theorems establishes, as a necessary and sufficient condition, that the solutions of the fourth-order modified contracted Schrödinger equation correspond uniquely to those of the Schrödinger equation. The complete equivalence between these two equations is demonstrated. Another equation equivalent to the Schrödinger equation but involving only the correlation matrices is also obtained as a consequence of the second theorem.

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I. INTRODUCTION

The convenience of replacing the search for the N-electron wave function by that of the second-order reduced density matrix (2-RDM) was pointed out long ago by the great pioneers in this field: Husimi [1], Löwdin [2], Mayer [3], McWeeny [4] Ayres [5], and Coulson [6]. It is not possible to refer here to the many valuable contributions that followed these early works. However, the interested reader may find a reliable account in a series of books and reviews [7–13] and, in particular, in the recent book by Coleman and Yukalov [6].

In 1987, Valdemoro applied the RDM contracting mapping to the matrix representation in the N-electron space of the Schrödinger equation [14] and obtained the family of contracted Schrödinger equations (CSE), which can be shown to be equivalent to the integrodifferential equations originally proposed by Cho [15] in 1962 and then reported independently by Cohen and Frishberg [16,17] and by Nakatsuji [18] in 1976. What hindered the use of these equations is that they are indeterminate [19]. An approximate algorithm for building a 2-RDM in terms of the 1-RDM was reported by Valdemoro [20] and subsequently the procedure was extended in order to approximate higher-order RDMs in terms of the lower-order ones [21,22]. The use of these approximations permitted the removal of the 2-CSE indeterminacy, thus allowing its iterative solution [23]. This started a new line of research that not only shed light on the RDM's theory but is also established a new approach to study accurately the electronic structure of fermion systems without having recourse to the N-electron wave function [24–36].

Alcoba and Valdemoro [37] have recently reported and implemented a new family of equations: the modified contracted Schrödinger equations (MCSE), which have significant properties. The most important of this family of equations is the 4-MCSE, which transforms the search for the N-electron eigenfunction into the search for the solution of a fourth-order self-contained equation. The relevance of this approach lies in the fact that no approximation is, in principle, needed. The price that must be paid is that the size of the problem augments. A new feature of the MCSEs is that the correlation matrices play a central role in them. This is the reason why this paper aims to state and prove four theorems involving the correlation matrices. The first three theorems shed light upon the narrow link existing between the CSEs, the Schrödinger equation, and the correlation vanishing terms recently reported [36,37]. The interrelation between the CSE, the Schrödinger equation, and the cancellation of the high-order correlation effects lies at the root of the MCSE because it shows that it is possible to construct a self-contained equation that could be equivalent to the Schrödinger equation. The fourth theorem proves that when the 4-MCSE, in its self-contained form, is satisfied by a set of 1-, 2-, 3-, and 4-RDMs, which must be N representable [38,39], then these matrices and the ensuing energy correspond to eigenstates of the system and eigenvalue, considered respectively. The converse is also proved. This one-to-one correspondence between the 4-MCSE and the Schrödinger equation shows that the 4-MCSE *is not an artifice* but a relevant equation in many-body theory.

The notation and basic background are described in the following section, where some new related formulas are also given. In Sec. III, the problem is stated and solved by establishing and proving the above-mentioned theorems. A brief discussion about some of the implications that these theorems have when applied in practice is given in the concluding section.

II. BASIC DEFINITIONS

In what follows, we consider that the system under study has a fixed and well-defined number of particles, N. We will also consider that the one-electron space is spanned by a finite basis set of $2\times K$ orthonormal spin orbitals. Under these conditions a p-RDM corresponding to an N-electron state Ψ may be defined in second quantization formalism as

$$ {}^{p}D_{i_1 i_2 \ldots i_p ; j_1 j_2 \ldots j_p} = \frac{1}{p!} \langle \Psi | b^{\dagger}_{i_1} b^{\dagger}_{i_2} \ldots b^{\dagger}_{i_p} b_{j_p} \ldots b_{j_2} b_{j_1} | \Psi \rangle, \tag{1} $$

where $b^{\dagger}$ and b are the fermion creator and annihilator operators, respectively.

In this formalism the many-body Hamiltonian may be written as

$$ \hat{H} = \sum_{k,l,r,s} {}^{0}H_{rs;kl}\, b^{\dagger}_{r} b^{\dagger}_{s} b_{l} b_{k}, \tag{2} $$

where

$$ {}^0H_{rs;kl} = \frac{1}{2}\left(\frac{\varepsilon_{rk}\delta_{sl}+\varepsilon_{sl}\delta_{rk}}{N-1} + \langle rs|kl\rangle\right), \tag{3}$$

where ε groups the one-electron integrals and $\langle rs|kl\rangle$ are the two-electron integrals in the Condon and Shortley notation.

The representation of this Hamiltonian in the N-electron space yields the Hamiltonian matrix, $\underline{\mathcal{H}}$, whose diagonalization provides the full configuration interaction (FCI) eigenstates. The matrix representation of the Schrödinger equation in this space takes the form

$$ \underline{\mathcal{H}}\ \underline{\mathcal{D}}^{\Psi} = E_{\Psi}\underline{\mathcal{D}}^{\Psi} = \underline{\mathcal{D}}^{\Psi}\underline{\mathcal{H}}, \tag{4}$$

where Ψ is the considered eigenstate of the Hamiltonian and $\hat{\mathcal{D}}^{\Psi} = |\Psi\rangle\langle\Psi|$ is the N-electron density operator.

The fact that $\underline{\mathcal{D}}^{\Psi}$ commutes with $\underline{\mathcal{H}}$ will not be explicitly taken into account in what follows. Thus only the equation derived from the left part of Eq. (4) will be considered. It should, therefore, be kept in mind that the relations derived in a similar way from the right part of Eq. (4) are equally true.

The application of the matrix contracting mapping into the two-electron space [14,40,41] to both sides of relation (4) (left side equation), leads to

$$ \langle\Psi|\hat{H}b_i^{\dagger}b_j^{\dagger}b_q b_p|\Psi\rangle = E\langle\Psi|b_i^{\dagger}b_j^{\dagger}b_q b_p|\Psi\rangle = E2!\,{}^2D_{ij;pq}. \tag{5}$$

Replacing the explicit form of $\hat{H}$ into Eq. (5) and transforming the left-hand side (lhs) of the equation into its normal form one obtains the 2-CSE, which may be written as

$$ \begin{aligned} 2!E\,{}^2D_{ij;pq} &= 2!2({}^0H\,{}^2D)_{ij;pq} + 3!2\sum_{k,l,s}({}^0H_{js;kl}\,{}^3D_{pqs;ikl} \\ &\quad + {}^0H_{si;kl}\,{}^3D_{pqs;ljk}) + 4!\sum_{k,l,r,s}{}^0H_{rs;kl}\,{}^4D_{pqrs;ijkl} \\ &\equiv {}^2\mathcal{M}_{ij;pq}. \end{aligned} \tag{6}$$

In what follows the matrix ${}^p\underline{\mathcal{M}}$ will denote the matrix representation of that side of the p-CSE that involves the ${}^0H$ elements.

This equation, whose main variable is the 2-RDM, also depends on the 3- and 4-RDMs, which is the cause of its indeterminacy. As reported by Colmenero and Valdemoro [23] in 1994, the interdependence that—at least apparently—exists among the 2-, 3-, ..., p-CSEs may be decoupled by constructing satisfactory approximations of the 3- and 4-RDMs in terms of the 2- and 1-RDMs. The algorithms used [21,22] were an extension of those proposed by Valdemoro in 1992 for approximating the 2-RDM in terms of the 1-RDM. Since then, several new improvements have been implemented in order to enhance the accuracy of the algorithms [24,25,29–31,42].

A. Decomposition of the RDMs and correlation matrices structure

The anticommuting rules satisfied by the fermion operators, together with the resolution of the identity operator, render possible the decomposition of a p-RDM element into a sum of terms involving lower-order RDM elements and terms describing pure q-body correlation effects [26–30]. Let us consider now the decomposition of the 2-RDM that provides the simplest example.

$$ \begin{aligned} 2!\,{}^2D_{ij;ml} &= \langle\Psi|b_i^{\dagger}b_j^{\dagger}b_l b_m|\Psi\rangle = -{}^1D_{i;l}\delta_{j;m} \\ &\quad + \langle\Psi|b_i^{\dagger}b_m b_j^{\dagger}b_l|\Psi\rangle, \end{aligned} \tag{7}$$

and let us write the resolution of the identity, $\hat{I}$, as

$$ \hat{I} = |\Psi\rangle\langle\Psi| + \sum_{\Psi'\neq\Psi}|\Psi'\rangle\langle\Psi'| \equiv \hat{P} + \hat{Q}, \tag{8}$$

where $|\Psi\rangle$ represents the eigenstate of the Hamiltonian being studied, $|\Psi'\rangle$ represent the remaining part of the spectrum and the projector $\hat{P}$ and its orthogonal complement $\hat{Q}$ are defined as

$$ \hat{P} \equiv |\Psi\rangle\langle\Psi|, \tag{9}$$

$$ \hat{Q} \equiv \sum_{\Psi'\neq\Psi}|\Psi'\rangle\langle\Psi'|. \tag{10}$$

When inserting $\hat{I}$ in the last term of Eq. (7) between b_m and $b_j^{\dagger}$ one has

$$ 2!\,{}^2D_{ij;ml} = {}^1D_{i;m}\,{}^1D_{j;l} - {}^1D_{i;l}\delta_{j;m} + {}^2C_{ij;ml}, \tag{11}$$

where

$$ {}^2C_{ij;ml} = \langle\Psi|b_i^{\dagger}b_m\hat{Q}b_j^{\dagger}b_l|\Psi\rangle \equiv \sum_{\Psi'\neq\Psi}{}^1D_{i;m}^{\Psi\Psi'}\,{}^1D_{j;l}^{\Psi'\Psi}. \tag{12}$$

The matrices ${}^1D^{\Psi\Psi'}$ are first-order transition reduced density matrices. The ${}^2\underline{C}$ matrices can be interpreted as describing the simultaneous virtual excitations and deexcitations of two electrons of the system.

Recalling the basic relation obtained by taking the expectation value of the anticommutator of two fermion operators:

$$ {}^1\delta_{i;j} = {}^1D_{i;j} + {}^1\overline{D}_{i;j}, \tag{13}$$

where the first-order hole reduced density matrix (1-HRDM) is defined as

$$ {}^1\overline{D}_{i;j} = \langle\Psi|b_j b_i^{\dagger}|\Psi\rangle, \tag{14}$$

we can rewrite the decomposition of the 2-RDM as follows:

$$ \begin{aligned} 2!\,{}^2D_{ij;ml} &= {}^1D_{i;m}\,{}^1D_{j;l} - {}^1D_{i;l}\,{}^1D_{j;m} - {}^1D_{i;l}\,{}^1\overline{D}_{j;m} \\ &\quad + {}^2C_{ij;ml}. \end{aligned} \tag{15}$$

While the first two terms of the right-hand side (rhs) of Eq. (15) describe the uncorrelated portion of the 2-RDM [26,28,32], the last two terms of Eq. (15) are the analytical expression of the second-order cumulant [31–35,43–45] that describes the correlated portion of this RDM [26,27,32]. Thus, Eq. (15) shows that the cumulant can be decomposed in two terms: one term that is a product of two one-particle density matrices describing the correlation effects through the 1-HRDM and one term expressed as an element of the ${}^2\underline{C}$ matrix that cannot be decomposed in terms of lower-order density matrices—which is why the ${}^2\underline{C}$ matrices have been called pure two-body correlation matrices [27–30,36].

It should be mentioned here that the choice and the order in which the fermion operators are anticommuted in Eq. (7) is not unique. Thus, one may as well have

$$2!\,{}^2D_{ij;ml} = -{}^1D_{i;l}\,{}^1D_{j;m} + {}^1D_{i;m}\delta_{j;l} - {}^2C_{ij;lm}. \tag{16}$$

There are still two other possible equivalent expressions that correspond to the permutation of the creator indices and to the joint permutation of the creator and the annihilator indices. It should perhaps be mentioned here that in order to have an antisymmetric ${}^2C$ matrix one must consider the matrix

$${}^2\mathcal{C}^A_{ij;ml} = \frac{{}^2C_{ij;ml} - {}^2C_{ij;lm} - {}^2C_{ji;ml} + {}^2C_{ji;lm}}{4}, \tag{17}$$

in which case the 2-RDM takes the form

$$2!\,{}^2D = {}^1D \wedge {}^1D + {}^1D \wedge {}^1I + {}^2\mathcal{C}^A, \tag{18}$$

where ${}^1I$ is the unit matrix of dimensions $(2\times K)\times(2\times K)$, and the wedge symbol represents de Grassman product [46]. In the demonstrations that center our attention here it is not necessary to consider antisymmetrized matrix forms. Thus, the theoretical implications and the practical implementation of this property in the 4-MCSE will be reported elsewhere [47].

The decomposition of the 3- and 4-RDMs may be carried out in a similar way as in the 2-RDM case [29]. These decomposition formulas are given in the Appendix. We will just mention now that the decomposition of the 3- and 4-RDMs generate a set of different pure three-body and pure four-body correlation matrices. Thus, besides the different matrices arising as the result of the permutation of their indices, other structural varieties occur in the higher-order matrices. Consequently, the pure three-body correlation matrices are

$${}^{(3;2,1)}C_{ijk;mlr} = 2! \sum_{\Psi' \neq \Psi} {}^2D^{\Psi\Psi'}_{ij;ml}\,{}^1D^{\Psi'\Psi}_{k;r}, \tag{19}$$

$${}^{(3;1,1,1)}C_{ijk;mlr} = \sum_{\Psi',\Psi'' \neq \Psi} {}^1D^{\Psi\Psi'}_{i;m}\,{}^1D^{\Psi'\Psi''}_{j;l}\,{}^1D^{\Psi''\Psi}_{k;r}, \tag{20}$$

and the ${}^{(3;1,2)}C_{ijk;mlr}$, whose formula may be easily be inferred.

The only pure four-body correlation matrices that will be used in what follows are

$${}^{(4;2,2)}C_{ijkl;mnrs} = 2!\,2! \sum_{\Psi' \neq \Psi} {}^2D^{\Psi\Psi'}_{ij;mn}\,{}^2D^{\Psi'\Psi}_{kl;rs}, \tag{21}$$

$${}^{(4;2,1,1)}C_{ijkl;mlrs} = 2! \sum_{\Psi',\Psi'' \neq \Psi} {}^2D^{\Psi\Psi'}_{ij;mn}\,{}^1D^{\Psi'\Psi''}_{k;r}\,{}^1D^{\Psi''\Psi}_{l;s}, \tag{22}$$

but, as in the three-order case, all four-order combinations of 1-, 2-, and 3-transition RDMs are possible.

B. The pure *p*-body correlation matrices

The interesting physical-mathematical properties of the pure *p*-body correlation matrices deserve by themselves a separate study [48]. Nevertheless, we will describe here three types of properties that are needed in the following developments.

1. Interrelation among correlation matrices

Let us first consider the manner in which two different C matrices of the same order are interrelated. As an example of how to proceed, let us consider the ${}^{(4;2,1,1)}\underline{C}$ and the ${}^{(4;2,2)}\underline{C}$ matrices.

$$\begin{aligned}
{}^{(4;2,2)}C_{ijkl;mnrs} &= 2!\,2! \sum_{\Psi' \neq \Psi} {}^2D^{\Psi\Psi'}_{ij;mn}\,{}^2D^{\Psi'\Psi}_{kl;rs} \\
&= \langle\Psi|b_i^\dagger b_j^\dagger b_n b_m \hat{Q} b_k^\dagger b_l^\dagger b_s b_r|\Psi\rangle \\
&\quad - \langle\Psi|b_i^\dagger b_j^\dagger b_n b_m \hat{Q}\{b_k^\dagger b_s \delta_{lr} - b_k^\dagger b_r b_l^\dagger b_s\}|\Psi\rangle \\
&= -{}^{(3;2,1)}C_{ijk;mns}\delta_{lr} \\
&\quad + \langle\Psi|b_i^\dagger b_j^\dagger b_n b_m \hat{Q}\{b_k^\dagger b_r|\Psi\rangle\langle\Psi|b_l^\dagger b_s \\
&\quad + b_k^\dagger b_r \hat{Q} b_l^\dagger b_s\}|\Psi\rangle \\
&= -{}^{(3;2,1)}C_{ijk;mns}\delta_{lr} + {}^{(3;2,1)}C_{ijk;mnr}\,{}^1D_{ls} \\
&\quad + {}^{(4;2,1,1)}C_{ijkl;mnrs}.
\end{aligned}$$

Thus,

$$\begin{aligned}
&{}^{(4;2,2)}C_{ijkl;mnrs} - {}^{(4;2,1,1)}C_{ijkl;mnrs} \\
&\quad = {}^{(3;2,1)}C_{ijk;mnr}\,{}^1D_{ls} - {}^{(3;2,1)}C_{ijk;mns}\delta_{lr}.
\end{aligned} \tag{23}$$

The reasoning followed here is general and, therefore, sets the way in which to handle these matrices.

2. The vertical contraction of these correlation matrices

Let us now consider a different kind of interrelation arising from what will be called *vertical contraction*. As usual, the sum is carried out over a common creator and annihilator index but what is specific to this kind of contraction is that this common index occupies the same place in the creator and annihilator labels, respectively. In order to render the reasoning more transparent we will consider concrete cases, but the procedure and the results are general.

(1) The indices to be contracted are the last ones (or the first ones) in the label. There are two possible cases:

Case (a):

$$\sum_l {}^{(4;2,1,1)}C_{ijkl;mnrl} = \sum_l \langle\Psi|b_i^\dagger b_j^\dagger b_n b_m \hat{Q} b_k^\dagger b_r \hat{Q} b_l^\dagger b_l|\Psi\rangle = \langle\Psi|b_i^\dagger b_j^\dagger b_n b_m \hat{Q} b_k^\dagger b_r \hat{Q}\hat{N}|\Psi\rangle = 0. \tag{24}$$

Note that in this case the string of operators $\hat{Q}\hat{N}$ acting on $|\Psi\rangle$ vanish.

Case (b):

$$\sum_l {}^{(4;2,2)}C_{ijkl;mnrl} = \langle\Psi|b_i^\dagger b_j^\dagger b_n b_m \hat{Q} b_k^\dagger \hat{N} b_r|\Psi\rangle = (N-1)\,{}^{(3;2,1)}C_{ijk;mnr}. \tag{25}$$

(2) The indices to be contracted are interior.

$$\sum_k {}^{(4;2,1,1)}C_{ijkl;mnks} = \sum_k \langle\Psi|b_i^\dagger b_j^\dagger b_n b_m \hat{Q} b_k^\dagger b_k \hat{Q} b_l^\dagger b_s|\Psi\rangle = \langle\Psi|b_i^\dagger b_j^\dagger b_n b_m \hat{Q}\hat{N}\hat{Q} b_l^\dagger b_s|\Psi\rangle = N\,{}^{(3;2,1)}C_{ijl;mns}. \tag{26}$$

Note that in this case the string of operators $\hat{Q}\hat{N}\hat{Q}$ becomes $N\hat{Q}$.

3. The vanishing products of the Hamiltonian and the correlation matrices

A recently reported property of the correlation matrices [36], which plays a central role in our developments, derives from the basic relation

$$\langle\Psi|\hat{H}\hat{Q}\hat{\Theta}|\Psi\rangle = 0, \tag{27}$$

where $\hat{\Theta}$ may be any operator. The reason is that the projectors $\hat{Q}$ and $\hat{P}$ are complementary to each other and, as we have mentioned, the Ψ is assumed to be an eigenstate of the Hamiltonian.

As an example, let us consider that $\hat{\Theta}$ is a two-body density operator. Then, Eq. (27) becomes

$$0 = \langle\Psi|\hat{H}\hat{Q} b_i^\dagger b_j^\dagger b_l b_k|\Psi\rangle = \sum_{pqrs} {}^0H_{pq;rs}\,{}^{(4;2,2)}C_{pqij;rskl} = {}^{(4;2,2)}0_{ij;kl}. \tag{28}$$

This type of vanishing terms are described as ${}^{(4;2,2)}0_{ij;kl}$ where only the indices that do not enter into the sum appear explicitly in the symbol.

As has been mentioned in Ref. [36], this property causes the cancellation of the high-order correlation effects through the action of the Hamiltonian and explains why, when the Hamiltonian only has two-body operators, there is a one-to-one correspondence between the 2-RDM and the wave function corresponding to an eigenstate of the Hamiltonian [49].

III. EQUIVALENCE THEOREMS BETWEEN THE SOLUTIONS OF THE FOURTH-ORDER MODIFIED CONTRACTED SCHRÖDINGER EQUATION AND THOSE OF THE SCHRÖDINGER EQUATION

A. Presenting the problem

As will be shown here, the *vanishing terms* play a determinant role in the 4-MCSE theory. In fact, there are several equivalent forms of generating the 4-MCSE, which only differ on the kind of vanishing terms appearing in the derivation. To understand the physical meaning and the interrelations existing among the different vanishing terms is not of only theoretical interest. Thus, the question of whether one must take these terms explicitly into account when solving iteratively the 4-MCSE is of the utmost practical relevance. It will also be shown that an equivalence exists between the 4-MCSE and the Schrödinger equation.

In order to present the problem we will first derive the 4-MCSE in a different way from that followed in Ref. [37] and then we will recall the general lines of the previous derivation.

The initial point for generating the 4-MCSE is similar to that described in Sec. II B. Thus, starting with the expression for the 4-CSE,

$$4!E\,{}^4D_{ijkl;pqrs} = \langle\Psi|\hat{H} b_i^\dagger b_j^\dagger b_k^\dagger b_l^\dagger b_s b_r b_q b_p|\Psi\rangle \equiv {}^4\mathcal{M}_{ijkl;pqrs}, \tag{29}$$

one then proceeds to modify the order of the fermion operators with the ultimate aim of obtaining operator strings of the $b^\dagger b^\dagger bbb^\dagger b^\dagger bbb^\dagger b^\dagger bb$ type in mind. Then the unit operator (8) is inserted between bb and $b^\dagger b^\dagger$. Also, when the lhs of Eq. (5) appears it will be replaced by ${}^2\underline{\mathcal{M}}$. In order to increase the readability of the paper and since no difficult operations are involved, the intermediate steps are reported in the Appendix. The resulting equation is

$$\begin{aligned}4!E\,{}^4D_{ijkl;pqrs}=&(\delta_{ql}\delta_{kp}-\delta_{pl}\delta_{kq})\,{}^2\mathcal{M}_{ij;rs}+(\delta_{ql}\delta_{ks}-\delta_{kq}\,{}^1D_{l;s})\,{}^2\mathcal{M}_{ij;pr}\\&+(\delta_{lr}\delta_{kq}-\delta_{ql}\,{}^1D_{k;r})\,{}^2\mathcal{M}_{ij;ps}-(\delta_{lp}\delta_{ks}-\delta_{kp}\,{}^1D_{l;s})\\&\times{}^2\mathcal{M}_{ij;qr}-(\delta_{kp}\delta_{lr}-\delta_{lp}\,{}^1D_{k;r})\,{}^2\mathcal{M}_{ij;qs}+2!\,{}^2\mathcal{M}_{ij;pq}\,{}^2D_{kl;rs}-\delta_{ql}(E\,{}^{(3;2,1)}C_{ijk;psr}+{}^{(5;2,2,1)}0_{ijk;psr})\\&-\delta_{kq}(E\,{}^{(3;2,1)}C_{ijl;prs}+{}^{(5;2,2,1)}0_{ijl;prs})+\delta_{lp}(E\,{}^{(3;2,1)}C_{ijk;qsr}+{}^{(5;2,2,1)}0_{ijk;qsr})+\delta_{kp}(E\,{}^{(3;2,1)}C_{ijl;qrs}\\&+{}^{(5;2,2,1)}0_{ijl;qrs})+E\,{}^{(4;2,2)}C_{ijkl;pqrs}+{}^{(6;2,2,2)}0_{ijkl;pqrs}\\=&\,{}^4\mathcal{M}_{ijkl;pqrs}.\end{aligned}\tag{30}$$

When one assumes that the Ψ are eigenstates of the Hamiltonian, the vanishing terms ${}^{(6;2,2,2)}\underline{0}$ and ${}^{(5;2,2,1)}\underline{0}$ disappear from the equation yielding [37],

$$\begin{aligned}E\,{}^4D_{ijkl;pqrs}=&(\delta_{ql}\delta_{kp}-\delta_{pl}\delta_{kq})\,{}^2\mathcal{M}_{ij;rs}\\&+(\delta_{ql}\delta_{ks}-\delta_{kq}\,{}^1D_{l;s})\,{}^2\mathcal{M}_{ij;pr}\\&+(\delta_{lr}\delta_{kq}-\delta_{ql}\,{}^1D_{k;r})\,{}^2\mathcal{M}_{ij;ps}\\&-(\delta_{lp}\delta_{ks}-\delta_{kp}\,{}^1D_{l;s})\,{}^2\mathcal{M}_{ij;qr}\\&-(\delta_{kp}\delta_{lr}-\delta_{lp}\,{}^1D_{k;r})\,{}^2\mathcal{M}_{ij;qs}\\&+{}^2\mathcal{M}_{ij;pq}\,{}^2D_{kl;rs}-\delta_{ql}E\,{}^{(3;2,1)}C_{ijk;psr}\\&-\delta_{kq}E\,{}^{(3;2,1)}C_{ijl;prs}+\delta_{lp}E\,{}^{(3;2,1)}C_{ijk;qsr}\\&+\delta_{kp}E\,{}^{(3;2,1)}C_{ijl;qrs}+E\,{}^{(4;2,2)}C_{ijkl;pqrs}.\end{aligned}\tag{31}$$

This equation is self-contained, since the highest-order RDM involved is the 4-RDM from which all the other matrices appearing in the equation may be derived either through contraction or by applying the decomposition techniques described previously.

Note that the lhs of Eq. (29) arising directly from the contraction of the $E_\Psi\underline{D}^\Psi$ member of the Schrödinger equation is left untouched. Now, in our previous derivation [37] the reasoning followed was to some extent different. Let us insert the unit operator into Eq. (29) as follows:

$$\begin{aligned}4!E\,{}^4D_{ijkl;pqrs}=&\langle\Psi|\hat{H}\hat{I}b_i^\dagger b_j^\dagger b_k^\dagger b_l^\dagger b_s b_r b_q b_p|\Psi\rangle\\\equiv&\,4!E\,{}^4D_{ijkl;pqrs}+{}^{(6;2,4)}0_{ijkl;pqrs}.\end{aligned}\tag{32}$$

Then the rhs of this equation could be set equal to the rhs of Eq. (30) and the high-order vanishing terms would disappear leaving just the ${}^{(4,2,2)}\underline{0}$. When the vanishing terms are set equal to zero both derivations yield the same final result. However, while in the derivation reported here we are keeping the structure obtained by contraction from the Schrödinger equation, where the energy corresponds to an eigenvalue, in the other derivation the energy is just an expectation value. But if the vanishing terms would have to be evaluated explicitly in order to solve the equation—that is, if Eq. (30) must be considered instead of Eq. (31)—then the equation would not be self-contained. On the other hand, the equation derived in Ref. [37], where only the ${}^{(4;2,2)}\underline{0}$ appeared, would still be easy to handle.

Another important question is whether satisfying the 4-MCSE, as in the 2-CSE case, is a necessary and sufficient condition so that a set of N-representable RDMs corresponds to an eigenstate of the Schrödinger equation and the energy simultaneously obtained coincides with the eigenvalue.

B. The theorems

Theorem 1. Let us assume that the pure four-body correlation matrix ${}^{(4;2,2)}\underline{C}$ is derived from the decomposition of an N-representable $\underline{4}$-RDM, then

$${}^{(4;2,2)}\underline{0}=\underline{0},\tag{33}$$

will be satisfied by this four-body correlation matrix *if and only if* the density matrix, preimage of the 4-RDM, satisfies the Schrödinger equation, Eq. (4).

Proof. (a) Let us assume that the Schrödinger equation, Eq. (4), is satisfied for Ψ. It then follows that

$$\begin{aligned}0=\langle\Psi|\hat{H}\hat{Q}b_i^\dagger b_j^\dagger b_l b_k|\Psi\rangle=&\sum_{pqrs}{}^0H_{pq;rs}\,{}^{(4;2,2)}C_{pqij;rskl}\\=&\,{}^{(4;2,2)}0_{ij;kl},\end{aligned}\tag{34}$$

for all i,j,k,l.

(b) The second part of the theorem is proved as follows. Through the use of the resolution of the identity it is easy to see that ${}^{(4;2,2)}\underline{0}$ can be rewriten as

$${}^{(4;2,2)}\underline{0}=E2!\,{}^2\underline{D}-{}^2\underline{\mathcal{M}}.\tag{35}$$

Now, by hypothesis

$${}^{(4;2,2)}\underline{0}=\underline{0},\tag{36}$$

which implies

$$E2!\,{}^2\underline{D}-{}^2\underline{\mathcal{M}}=\underline{0},\tag{37}$$

that is, the 2-CSE is satisfied. By the Nakatsuji's theorem [18], whose second quantization equivalent was given by Mazziotti [31], it follows that the Schrödinger equation is satisfied. The proof is thus completed.

Theorem 2. Let us assume that the pure four-body correlation matrix ${}^{(4;2,1,1)}\underline{C}$ is derived from the decomposition of an N-representable $\underline{4}$-RDM, then

$$^{(4;2,1,1)}\underline{0}=\underline{0}, \tag{38}$$

will be satisfied by this four-body correlation matrix *if and only if* the density matrix, preimage of the 4-RDM, satisfies the Schrödinger equation, Eq. (4).

Proof. (a) Let us assume that the Schrödinger equation, Eq. (4), is satisfied for Ψ. It then follows that

$$0=\langle\Psi|\hat{H}\hat{Q}b_i^\dagger b_k\hat{Q}b_j^\dagger b_l|\Psi\rangle=\sum_{pqrs}{}^0H_{pq;rs}{}^{(4;2,1,1)}C_{pqij;rskl}$$
$$={}^{(4;2,1,1)}0_{ij;kl}, \tag{39}$$

for all i,j,k,l.

(b) The second part of the proof is as follows. We first use the fact that the fourth-order correlation matrix $^{(4;2,1,1)}\underline{C}$ can be contracted to the third-order one $^{(3;2,1)}\underline{C}$, as has been previuosly shown in Sec. II B 2. Thus, our hypothesis

$$^{(4;2,1,1)}\underline{0}=\underline{0}, \tag{40}$$

implies that

$$^{(3,2,1)}\underline{0}=\underline{0}. \tag{41}$$

Now, making use of relation (23) and the resolution of the identity, it follows that

$$E2!\,{}^2D_{pq;rs}-{}^2\mathcal{M}_{pq;rs}={}^{(4;2,2)}0_{pq;rs}=-\delta_{rq}{}^{(3;2,1)}0_{p;s}$$
$$+{}^{(3;2,1)}0_{p;r}{}^1D_{q;s}+{}^{(4;2,1,1)}0_{pq;rs},$$

for all p,q,r,s. Since the rhs of this relation vanishes, so does the lhs, that is, the 2-CSE is satisfied. By the Nakatsuji's theorem, it follows that the Schrödinger equation is satisfied. The proof is thus completed.

Attention should be called to a relevant implication of Eq. (38). Thus, in a similar way that expression (33) is equivalent to demanding that the 2-CSE is satisfied, the equation

$$^{(4;2,1,1)}\underline{0}=\underline{0}, \tag{42}$$

implies that the equation

$$E\,{}^2C_{ij;pq}=\sum_{k,l,r}{}^0H_{kl;ri}{}^{(3;2,1)}C_{klj;rpq}$$
$$-\sum_{k,l,s}{}^0H_{kl;is}{}^{(3;2,1)}C_{klj;spq}$$
$$+\sum_{k,l,r,s}{}^0H_{kl;rs}{}^{(4;3,1)}C_{klij;rspq}, \tag{43}$$

involving the correlation matrices must also be satisfied and also that this last equation is equivalent to the Schrödinger equation. This reasoning can be easily extended through the use of the following theorem.

Theorem 3. Let us assume that the correlation matrix $^{(p;2,x,y,\ldots)}\underline{C}$ is obtained from the decomposition of an N-representable p-RDM, then the following relation

$$^{(p;2,x,y,\ldots)}\underline{0}=\underline{0}, \tag{44}$$

with $p>4$ and $x+y+\cdots=p-2$ will be satisfied by the corresponding p-correlation matrix *if and only if* the density matrix, preimage of the p-RDM, satisfies the Schrödinger equation, Eq. (4).

Proof. (a) Let us assume that the Schrödinger equation, Eq. (4), is satisfied for Ψ. It then follows that

$$0=\langle\Psi|\hat{H}\hat{Q}b_{i_1}^\dagger\ldots b_{i_x}^\dagger b_{t_x}\ldots b_{t_1}\hat{Q}b_{j_1}^\dagger\ldots b_{j_y}^\dagger b_{v_y}\ldots b_{v_1}\hat{Q}\ldots|\Psi\rangle$$
$$=\sum_{pqrs}{}^0H_{pq;rs}{}^{(p;2,x,y,\ldots)}C_{pqi_1\ldots i_xj_1\ldots j_y\ldots;rst_1\ldots t_xv_1\ldots v_y\ldots}$$
$$={}^{(p;2,x,y,\ldots)}0_{i_1\ldots i_xj_1\ldots j_y\ldots;t_1\ldots t_xv_1\ldots v_y\ldots}. \tag{45}$$

for all $i_1,\ldots,i_x,j_1,\ldots,j_y,\ldots,t_1,\ldots,t_x,v_1,\ldots,v_y,\ldots$.

(b) The second part of the proof is as follows. We first consider the particular case when $y,\ldots=0$. Recalling the rules given in Sec. II B 2, it can be shown that the vanishing term of the form $^{(p;2,x)}\underline{0}$ can be contracted to $^{(4;2,2)}\underline{0}$. Thus, Eq. (44) implies

$$^{(4;2,2)}\underline{0}=\underline{0}. \tag{46}$$

In all the other cases it can be shown that the vanishing term of the form $^{(p;2,x,y,\ldots)}\underline{0}$ can be contracted to $^{(4;2,1,1)}\underline{0}$. Thus, Eq. (44) implies

$$^{(4;2,1,1)}\underline{0}=\underline{0}. \tag{47}$$

The proof is completed by following the same reasoning as in the two previous theorems (second part of the proofs).

Theorem 4. Assuming that the correlation matrices $^{(3;2,1)}\underline{C}$ and $^{(4;2,1,1)}\underline{C}$ are obtained from the decomposition of a set of N-representable 3- and 4-RDM, then the 4-MCSE, Eq. (31), will be satisfied by this set of RDMs *if and only if* the density matrix, preimage of the 3- and 4-RDM, satisfies the Schrödinger equation, Eq. (4).

Proof. (a) It is easy to see that if the Schrödinger equation Eq. (4) is satisfied, then, by contraction, the equation

$$4!E\,{}^4\underline{D}={}^4\underline{\mathcal{M}},$$

must hold; and, therefore, the equivalent equation Eq. (30) must also hold. Another consequence of Eq. (4) is that Eq. (27) is also satisfied and, therefore, the vanishing terms appearing in the rhs of this equation vanish. Thus, the 4-MCSE Eq. (31) holds.

(b) We prove the converse. Contracting Eq. (31) over the last two indices, we obtain

$$2!E\,{}^2\underline{D}={}^2\underline{\mathcal{M}},$$

and this last matrix equation (2-CSE) implies that the Schrödinger equation is satisfied. The proof is thus completed.

The main consequence of this theorem is that the equivalence between the 4-MCSE and the Schrödinger equation is established.

The contraction of the 4-MCSE, which is not too obvious, is described in detail in the Appendix due to its length, although it does not involve any difficult operation.

IV. CONCLUSION

When discussing in Ref. [37] the questions that remained open in the iterative solution of the 4-MCSE, there was a particularly intriguing one: What was the role played by the vanishing terms? Should one try to solve the 4-MCSE form where the vanishing terms appeared explicitly? Or, alternatively: Should one impose them to be zero, thus removing them from the equation, and keeping them only as convergence tests? We think the answer is clear, in view of the results just reported. Thus, if one wishes to obtain a set of RDM's and the energy that would correspond to the Schrödinger equation solutions, one must solve a form of the 4-MCSE such as Eq. (31), where the vanishing terms do not explicitly appear. On the other hand, evaluating the ${}^{(4;2,2)}\underline{0}$ matrix is the best convergence test, since when this matrix is null we may be sure that all the p-CSE for $p\geqslant 2$ are satisfied and hence so is the Schrödinger equation. In practice, of course, we can only aim at values of the matrix ${}^{(4;2,2)}\underline{0}$ close to zero, since the matrices involved will probably be only approximately N-representable. In fact, the error in the evaluated ${}^{(4;2,2)}0$ matrix is a reliable and severe measure of the calculation error.

Thus, the results just reported guarantee that any convergent method applied on the 4-MCSE will converge to those sets of RDM's corresponding to the FCI solutions. Therefore, the method cannot converge to other N-representable sets—as in the case of the 1-CSE that presents at least two N-representable solutions, those corresponding to the FCI and Hartree-Fock solutions, and which makes, in principle, the equation useless to perform calculations [27].

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APPENDIX

1. Decomposition formulas of the 3- and 4-RDM

It has been shown that the 3- and 4-RDM may be decomposed in terms of the pure three- and four-body correlation matrices as [29]

$$\begin{aligned}3!\,{}^3D_{ikm;jln}=&-2!\,{}^2D_{ik;jn}\delta_{ml}+2!\,{}^2D_{ik;ln}\delta_{jm}\\&+2!\,{}^2D_{ik;jl}\,{}^1D_{m;n}+{}^{(3;2,1)}C_{ikm;jln},\end{aligned}\tag{A1}$$

and

$$\begin{aligned}4!\,{}^4D_{ijkl;pqrs}=&\,2!\,{}^2D_{ij;rs}(\delta_{qk}\delta_{pl}-\delta_{pk}\delta_{ql})+3!\,{}^3D_{ijl;qrs}\delta_{kp}+3!\,{}^3D_{ijk;prs}\delta_{lq}-3!\,{}^3D_{ijl;prs}\delta_{k;q}-3!\,{}^3D_{ijk;qrs}\delta_{l;p}\\&+2!\,{}^2D_{ij;pq}2!\,{}^2D_{kl;rs}+{}^{(4;2,2)}C_{ijkl;pqrs}.\end{aligned}\tag{A2}$$

2. Details of the 4-MCSE derivation

We rearrange the operators in the term $\langle\Psi|\hat{H}b_i^\dagger b_j^\dagger b_k^\dagger b_l^\dagger b_s b_r b_q b_p|\Psi\rangle$, hereafter called A as described in the text. The different steps are

$$\begin{aligned}A=&\,\delta_{ql}\langle\Psi|\hat{H}b_i^\dagger b_j^\dagger b_k^\dagger b_p b_s b_r|\Psi\rangle-\langle\Psi|\hat{H}b_i^\dagger b_j^\dagger b_k^\dagger b_q b_l^\dagger b_p b_s b_r|\Psi\rangle\\=&\,\delta_{ql}\delta_{kp}\langle\Psi|\hat{H}b_i^\dagger b_j^\dagger b_s b_r|\Psi\rangle-\delta_{ql}\langle\Psi|\hat{H}b_i^\dagger b_j^\dagger b_p b_k^\dagger b_s b_r|\Psi\rangle-\langle\Psi|\hat{H}b_i^\dagger b_j^\dagger b_k^\dagger b_q b_l^\dagger b_p b_s b_r|\Psi\rangle.\end{aligned}$$

Recalling the definition of ${}^2\underline{\mathcal{M}}$, Eq. (6),

$$\begin{aligned}A=&\,\delta_{ql}\delta_{kp}\,{}^2\mathcal{M}_{ij;rs}-\delta_{ql}\langle\Psi|\hat{H}b_i^\dagger b_j^\dagger b_p b_k^\dagger b_s b_r|\Psi\rangle-\delta_{kq}\langle\Psi|\hat{H}b_i^\dagger b_j^\dagger b_l^\dagger b_p b_s b_r|\Psi\rangle+\langle\Psi|\hat{H}b_i^\dagger b_j^\dagger b_q b_k^\dagger b_l^\dagger b_p b_s b_r|\Psi\rangle\\=&\,(\delta_{ql}\delta_{kp}-\delta_{pl}\delta_{kq})\,{}^2\mathcal{M}_{ij;rs}+\delta_{ql}\delta_{ks}\,{}^2\mathcal{M}_{ij;pr}+\delta_{ql}\langle\Psi|\hat{H}b_i^\dagger b_j^\dagger b_p b_s b_k^\dagger b_r|\Psi\rangle+\delta_{kq}\langle\Psi|\hat{H}b_i^\dagger b_j^\dagger b_p b_l^\dagger b_s b_r|\Psi\rangle\\&+\delta_{lp}\langle\Psi|\hat{H}b_i^\dagger b_j^\dagger b_q b_k^\dagger b_s b_r|\Psi\rangle-\langle\Psi|\hat{H}b_i^\dagger b_j^\dagger b_q b_k^\dagger b_p b_l^\dagger b_s b_r|\Psi\rangle.\end{aligned}\tag{A3}$$

Inserting the unit operator $\hat{I}$ into the third term of Eq. (A3),

$$A=(\delta_{ql}\delta_{kp}-\delta_{pl}\delta_{kq})\,{}^2\mathcal{M}_{ij;rs}+\delta_{ql}\delta_{ks}\,{}^2\mathcal{M}_{ij;pr}+\delta_{ql}\langle\Psi|\hat{H}b_i^\dagger b_j^\dagger b_p b_s \hat{I} b_k^\dagger b_r|\Psi\rangle+\delta_{kq}\langle\Psi|\hat{H}b_i^\dagger b_j^\dagger b_p b_l^\dagger b_s b_r|\Psi\rangle$$
$$+\delta_{lp}\langle\Psi|\hat{H}b_i^\dagger b_j^\dagger b_q b_k^\dagger b_s b_r|\Psi\rangle-\langle\Psi|\hat{H}b_i^\dagger b_j^\dagger b_q b_k^\dagger b_p b_l^\dagger b_s b_r|\Psi\rangle$$

and using the resolution of the identity Eq. (8), we have

$$A=(\delta_{ql}\delta_{kp}-\delta_{pl}\delta_{kq})\,{}^2\mathcal{M}_{ij;rs}+\delta_{ql}\delta_{ks}\,{}^2\mathcal{M}_{ij;pr}+\delta_{ql}\langle\Psi|\hat{H}b_i^\dagger b_j^\dagger b_p b_s \hat{Q} b_k^\dagger b_r|\Psi\rangle-\delta_{ql}\,{}^1D_{k;r}\,{}^2\mathcal{M}_{ij;ps}$$
$$+\delta_{kq}\langle\Psi|\hat{H}b_i^\dagger b_j^\dagger b_p b_l^\dagger b_s b_r|\Psi\rangle+\delta_{lp}\langle\Psi|\hat{H}b_i^\dagger b_j^\dagger b_q b_k^\dagger b_s b_r|\Psi\rangle-\langle\Psi|\hat{H}b_i^\dagger b_j^\dagger b_q b_k^\dagger b_p b_l^\dagger b_s b_r|\Psi\rangle.$$

Carrying out similar operations at different places yields

$$\begin{aligned}
A=&(\delta_{ql}\delta_{kp}-\delta_{pl}\delta_{kq})\,{}^2\mathcal{M}_{ij;rs}+\delta_{ql}\delta_{ks}\,{}^2\mathcal{M}_{ij;pr}-\delta_{lp}\delta_{ks}\,{}^2\mathcal{M}_{ij;qr}+(\delta_{lr}\delta_{kq}-\delta_{ql}\,{}^1D_{k;r})\,{}^2\mathcal{M}_{ij;ps}+\delta_{lp}\langle\Psi|\hat{H}b_i^\dagger b_j^\dagger b_s b_q \hat{I} b_k^\dagger b_r|\Psi\rangle\\
&-\delta_{kp}\langle\Psi|\hat{H}b_i^\dagger b_j^\dagger b_q b_l^\dagger b_s b_r|\Psi\rangle+\delta_{ql}\langle\Psi|\hat{H}\hat{I}b_i^\dagger b_j^\dagger b_p b_s \hat{Q} b_k^\dagger b_r|\Psi\rangle-\delta_{kq}\langle\Psi|\hat{H}b_i^\dagger b_j^\dagger b_r b_p \hat{I} b_l^\dagger b_s|\Psi\rangle\\
&+\langle\Psi|\hat{H}b_i^\dagger b_j^\dagger b_q b_p \hat{I} b_k^\dagger b_l^\dagger b_s b_r|\Psi\rangle\\
=&(\delta_{ql}\delta_{kp}-\delta_{pl}\delta_{kq})\,{}^2\mathcal{M}_{ij;rs}+(\delta_{ql}\delta_{ks}-\delta_{kq}\,{}^1D_{l;s})\,{}^2\mathcal{M}_{ij;pr}-\delta_{lp}\delta_{ks}\,{}^2\mathcal{M}_{ij;qr}+(\delta_{lr}\delta_{kq}-\delta_{ql}\,{}^1D_{k;r})\,{}^2\mathcal{M}_{ij;ps}\\
&+\delta_{lp}\langle\Psi|\hat{H}b_i^\dagger b_j^\dagger b_s b_q \hat{I} b_k^\dagger b_r|\Psi\rangle-\delta_{kp}\delta_{lr}\,{}^2\mathcal{M}_{ij;qs}-\delta_{kp}\langle\Psi|\hat{H}b_i^\dagger b_j^\dagger b_q b_r \hat{I} b_l^\dagger b_s|\Psi\rangle\\
&-\delta_{ql}(E\,{}^{(3;2,1)}C_{ijk;psr}+{}^{(5;2,2,1)}0_{ijk;psr}),
\end{aligned}$$

where we have used the definitions Eqs. (19) and (27). The same operations are now used several times

$$\begin{aligned}
A=&(\delta_{ql}\delta_{kp}-\delta_{pl}\delta_{kq})\,{}^2\mathcal{M}_{ij;rs}+\delta_{ql}\delta_{ks}\,{}^2\mathcal{M}_{ij;pr}-\delta_{lp}\delta_{ks}\,{}^2\mathcal{M}_{ij;qr}+(\delta_{lr}\delta_{kq}-\delta_{ql}\,{}^1D_{k;r})\,{}^2\mathcal{M}_{ij;ps}+\delta_{lp}\langle\Psi|\hat{H}b_i^\dagger b_j^\dagger b_s b_q \hat{I} b_k^\dagger b_r|\Psi\rangle\\
&-\delta_{kp}\langle\Psi|\hat{H}b_i^\dagger b_j^\dagger b_q b_l^\dagger b_s b_r|\Psi\rangle+\delta_{ql}\langle\Psi|\hat{H}\hat{I}b_i^\dagger b_j^\dagger b_p b_s \hat{Q} b_k^\dagger b_r|\Psi\rangle-\delta_{kq}\langle\Psi|\hat{H}b_i^\dagger b_j^\dagger b_r b_p \hat{I} b_l^\dagger b_s|\Psi\rangle\\
&+\langle\Psi|\hat{H}b_i^\dagger b_j^\dagger b_q b_p \hat{I} b_k^\dagger b_l^\dagger b_s b_r|\Psi\rangle\\
=&(\delta_{ql}\delta_{kp}-\delta_{pl}\delta_{kq})\,{}^2\mathcal{M}_{ij;rs}+(\delta_{ql}\delta_{ks}-\delta_{kq}\,{}^1D_{l;s})\,{}^2\mathcal{M}_{ij;pr}-\delta_{lp}\delta_{ks}\,{}^2\mathcal{M}_{ij;qr}+(\delta_{lr}\delta_{kq}-\delta_{ql}\,{}^1D_{k;r})\,{}^2\mathcal{M}_{ij;ps}\\
&+\delta_{lp}\langle\Psi|\hat{H}b_i^\dagger b_j^\dagger b_s b_q \hat{I} b_k^\dagger b_r|\Psi\rangle-\delta_{kp}\delta_{lr}\,{}^2\mathcal{M}_{ij;qs}-\delta_{kp}\langle\Psi|\hat{H}b_i^\dagger b_j^\dagger b_q b_r \hat{I} b_l^\dagger b_s|\Psi\rangle-\delta_{ql}(E\,{}^{(3;2,1)}C_{ijk;psr}+{}^{(5;2,2,1)}0_{ijk;psr})\\
&-\delta_{kq}\langle\Psi|\hat{H}\hat{I}b_i^\dagger b_j^\dagger b_r b_p \hat{Q} b_l^\dagger b_s|\Psi\rangle+\langle\Psi|\hat{H}\hat{I}b_i^\dagger b_j^\dagger b_q b_p \hat{Q} b_k^\dagger b_l^\dagger b_s b_r|\Psi\rangle+2!\;{}^2\mathcal{M}_{ij;pq}\,{}^2D_{kl;rs}\\
=&(\delta_{ql}\delta_{kp}-\delta_{pl}\delta_{kq})\,{}^2\mathcal{M}_{ij;rs}+(\delta_{ql}\delta_{ks}-\delta_{kq}\,{}^1D_{l;s})\,{}^2\mathcal{M}_{ij;pr}-(\delta_{lp}\delta_{ks}-\delta_{kp}\,{}^1D_{l;s})\,{}^2\mathcal{M}_{ij;qr}+(\delta_{lr}\delta_{kq}-\delta_{ql}\,{}^1D_{k;r})\,{}^2\mathcal{M}_{ij;ps}\\
&-(\delta_{kp}\delta_{lr}-\delta_{lp}\,{}^1D_{k;r})\,{}^2\mathcal{M}_{ij;qs}+\delta_{lp}\langle\Psi|\hat{H}\hat{I}b_i^\dagger b_j^\dagger b_s b_q \hat{Q} b_k^\dagger b_r|\Psi\rangle-\delta_{kp}\langle\Psi|\hat{H}\hat{I}b_i^\dagger b_j^\dagger b_q b_r \hat{Q} b_l^\dagger b_s|\Psi\rangle-\delta_{ql}(E\,{}^{(3;2,1)}C_{ijk;psr}\\
&+{}^{(5;2,2,1)}0_{ijk;psr})-\delta_{kq}(E\,{}^{(3;2,1)}C_{ijl;prs}+{}^{(5;2,2,1)}0_{ijl;prs})+E\,{}^{(4;2,2)}C_{ijkl;pqrs}+{}^{(6;2,2,2)}0_{ijkl;pqrs}+2!\;{}^2\mathcal{M}_{ij;pq}\,{}^2D_{kl;rs}\\
=&(\delta_{ql}\delta_{kp}-\delta_{pl}\delta_{kq})\,{}^2\mathcal{M}_{ij;rs}+(\delta_{ql}\delta_{ks}-\delta_{kq}\,{}^1D_{l;s})\,{}^2\mathcal{M}_{ij;pr}+(\delta_{lr}\delta_{kq}-\delta_{ql}\,{}^1D_{k;r})\,{}^2\mathcal{M}_{ij;ps}-(\delta_{lp}\delta_{ks}-\delta_{kp}\,{}^1D_{l;s})\,{}^2\mathcal{M}_{ij;qr}\\
&-(\delta_{kp}\delta_{lr}-\delta_{lp}\,{}^1D_{k;r})\,{}^2\mathcal{M}_{ij;qs}-\delta_{ql}(E\,{}^{(3;2,1)}C_{ijk;psr}+{}^{(5;2,2,1)}0_{ijk;psr})-\delta_{kq}(E\,{}^{(3;2,1)}C_{ijl;prs}+{}^{(5;2,2,1)}0_{ijl;prs})\\
&+\delta_{lp}(E\,{}^{(3;2,1)}C_{ijk;qsr}+{}^{(5;2,2,1)}0_{ijk;qsr})+\delta_{kp}(E\,{}^{(3;2,1)}C_{ijl;qrs}+{}^{(5;2,2,1)}0_{ijl;qrs})+2!\;{}^2\mathcal{M}_{ij;pq}\,{}^2D_{kl;rs}\\
&+E\,{}^{(4;2,2)}C_{ijkl;pqrs}+{}^{(6;2,2,2)}0_{ijkl;pqrs},
\end{aligned}$$

where we also have used the definition Eq. (21). It then follows that

$$\begin{aligned}
A=&(\delta_{ql}\delta_{kp}-\delta_{pl}\delta_{kq})\,{}^2\mathcal{M}_{ij;rs}+(\delta_{ql}\delta_{ks}-\delta_{kq}\,{}^1D_{l;s})\,{}^2\mathcal{M}_{ij;pr}+(\delta_{lr}\delta_{kq}-\delta_{ql}\,{}^1D_{k;r})\,{}^2\mathcal{M}_{ij;ps}-(\delta_{lp}\delta_{ks}-\delta_{kp}\,{}^1D_{l;s})\,{}^2\mathcal{M}_{ij;qr}\\
&-(\delta_{kp}\delta_{lr}-\delta_{lp}\,{}^1D_{k;r})\,{}^2\mathcal{M}_{ij;qs}-\delta_{ql}(E\,{}^{(3;2,1)}C_{ijk;psr}+{}^{(5;2,2,1)}0_{ijk;psr})-\delta_{kq}(E\,{}^{(3;2,1)}C_{ijl;prs}+{}^{(5;2,2,1)}0_{ijl;prs})\\
&+\delta_{lp}(E\,{}^{(3;2,1)}C_{ijk;qsr}+{}^{(5;2,2,1)}0_{ijk;qsr})+\delta_{kp}(E\,{}^{(3;2,1)}C_{ijl;qrs}+{}^{(5;2,2,1)}0_{ijl;qrs})+2!\;{}^2\mathcal{M}_{ij;pq}\,{}^2D_{kl;rs}\\
&+E\,{}^{(4;2,2)}C_{ijkl;pqrs}+{}^{(6;2,2,2)}0_{ijkl;pqrs},
\end{aligned}$$

which is the rhs of Eq. (30).

3. Details of the 4-MCSE contraction

The equation to be contracted is

$$4!E\,^4D_{ijkl;pqrs}=(\delta_{ql}\delta_{kp}-\delta_{pl}\delta_{kq})\,^2\mathcal{M}_{ij;rs}+(\delta_{ql}\delta_{ks}-\delta_{kq}\,^1D_{l;s})\,^2\mathcal{M}_{ij;pr}+(\delta_{lr}\delta_{kq}-\delta_{ql}\,^1D_{k;r})\,^2\mathcal{M}_{ij;ps}-(\delta_{lp}\delta_{ks}-\delta_{kp}\,^1D_{l;s})$$
$$\times^2\mathcal{M}_{ij;qr}-(\delta_{kp}\delta_{lr}-\delta_{lp}\,^1D_{k;r})\,^2\mathcal{M}_{ij;qs}+2!\,^2\mathcal{M}_{ij;pq}\,^2D_{kl;rs}-\delta_{ql}E\,^{(3;2,1)}C_{ijk;psr}-\delta_{kq}E\,^{(3;2,1)}C_{ijl;prs}$$
$$+\delta_{lp}E\,^{(3;2,1)}C_{ijk;qsr}+\delta_{kp}E\,^{(3;2,1)}C_{ijl;qrs}+E\,^{(4;2,2)}C_{ijkl;pqrs}.$$

In order to contract this equation we impose $l=s$ and add over this common index,

$$\begin{aligned}3!(N-3)E\,^3D_{ijk;pqr}=&(N-3)(-\delta_{kq}\,^2\mathcal{M}_{ij;pr}+\delta_{kp}\,^2\mathcal{M}_{ij;qr}\\&+^1D_{k;r}\,^2\mathcal{M}_{ij;pq})-2E\,^{(3;2,1)}C_{ijk;pqr}\\&+\sum_l\delta_{kp}E\,^{(3;2,1)}C_{ijl;qrs}\\&-\sum_l\delta_{kq}E\,^{(3;2,1)}C_{ijl;prs}\\&+\sum_l E\,^{(4;2,2)}C_{ijkl;pqrs},\end{aligned}\tag{A4}$$

where we have implicitly assumed that the RDMs and TRDMs are antisymmetric matrices. From the relations derived in Sec. II B2, it follows that

$$\sum_l\,^{(3;2,1)}C_{ijl;prl}=0,$$

$$\sum_l\,^{(3;2,1)}C_{ijl;qrl}=0,$$

and

$$\sum_l\,^{(4;2,2)}C_{ijkl;pqrl}=(N-1)^{(3;2,1)}C_{ijk;pqr},$$

which allows us to rewrite Eq. (A4) as follows:

$$\begin{aligned}3!(N-3)E\,^3D_{ijk;pqr}=&(N-3)(-\delta_{kq}\,^2\mathcal{M}_{ij;pr}+\delta_{kp}\,^2\mathcal{M}_{ij;qr}\\&+^1D_{k;r}\,^2\mathcal{M}_{ij;pq}+E\,^{(3;2,1)}C_{ijk;pqr}).\end{aligned}$$

It must be noted that this last equation is the 3-MCSE multiplied by the factor $(N-3)$ [37].

In order to carry out a second contraction we impose now $k=r$ and add over this common index. Proceeding in a similar way as before, one finally has

$$2!(N-3)(N-2)E\,^2D_{ij;pq}=(N-3)(N-2)\,^2\mathcal{M}_{ij;pq},$$

which is the result used in Sec. III B.

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