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Finite expansion of the inverse matrix in the polarization propagator method

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Abstract. An alternative theoretical approach to the polarization propagator based on a new finite expansion of a finite-dimensional matrix is presented. The general equations for such an expansion are derived and the validity conditions stated. This method is used to accomplish an approximate scheme for the self-energy of the particle–hole propagator within the superoperator formalism. Within this scheme each contribution includes corrections to infinite order in electronic interaction and so describes collective effects in a natural way. Individual contributions can be interpreted as describing the propagation of the interaction through a particular subset of electronic excitations. Comparison with other known approximation levels, such as the random-phase approximation, is also analyzed.

Key words: Polarization propagator – Particle–hole propagator – Self-energy – Inverse matrix

1 Introduction

Propagator methods have been widely used in atomic and molecular physics for studying electronic transitions (neutral excitations, ionization and electron attachment processes, etc.) and molecular response properties. Different kinds of propagators appear in the literature according to the nature of the physical property under study, such as the electron propagator (EP), the particle–hole (p-h) propagator, the polarization propagator (PP), etc. [1–12]. Propagator methods appear as strikingly attractive, since excitation energies and transition moments can be evaluated without calculation of the excited states and energies of the system under consideration.

Several approximation schemes for propagators have been developed. Among those which may be based on a diagrammatic expansion the random-phase approximation (RPA) for the PP [1], the outer valence Green function for the EP [6] and the algebraic diagrammatic construction for the EP [13] or for the PP [14] should be mentioned. Other methods are the so-called algebraic ones, such as the equation-of-motion method [15] and the equivalent superoperator formulation of the propagator via inner projection techniques [2, 5, 7–10, 12, 16–18].

Using the superoperator formalism, truncating the operator manifold used in the inner projection and with a suitable choice of the ground-state reference state, a perturbative expansion of the PP is achieved, where the order concept is defined by the fluctuation potential, i.e., the electronic repulsion minus the Fock potential [7, 18]. Analogous expressions hold for the EP [2, 8, 9, 16, 17]. It should be mentioned that this perturbative expansion of the propagator can also be represented diagrammatically [9, 18]. To a given order, the perturbative scheme of the PP takes into account collective effects of the system, represented by the infinite sum of certain classes of diagrams [19].

The second-order PP approach (SOPPA) has been succesfully used to calculate excitation energies [20, 21] and response properties (Ref. [12] and references therein [22, 23]). However, in certain low convergent systems, results at the SOPPA level were unsatisfactory, and improvements in the reference ground state were necessary to obtain reliable results (Ref. [24] and references therein). It is known that in such systems, collective effects play an important role [7, 18, 19]. For a given order n, the PP includes collective effects in a selective way, since only those infinite sums of diagrams which begin at order n or lower will appear in the n-order approach to the PP.

In this article a new method called the finite expansion of the inverse matrix (FEIM) is presented and applied to the propagator expression within the superoperator formalism. The underlying idea of the FEIM method is to express each element of the inverse of an N-dimensional matrix, $\mathbf{X}$, as a finite sum of terms, where each term in the expansion is characterized by a definite subset of matrix indices $K = \{i_1 < i_2 < \cdots < i_n\}_{1 \le n \le N}$ (i_j refers to a general index) and represents the sum of all terms that in a conventional perturbative series of $\mathbf{X}^{-1}$ involve matrix

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elements $\mathbf{X}_{mn}$ with $m, n \in K$. Since the number of indices subsets, K, is finite (for a finite-dimension matrix), the number of terms in the FEIM remains finite. A related method called "succesive inversion" was proposed to calculate the inverse matrix [25–27].

The FEIM method is used in the analysis of the associated self-energy of the p-h propagator, where each individual contribution to the finite expansion can be considered as describing the propagation of the interaction through the elements of the particular subset, K. It should be noted that for the p-h propagator, matrix indices refer to electronic excitations from occupied to virtual molecular orbitals. Each contribution includes corrections to infinite order in the electronic interaction, i.e., involves a sum of diagrams to infinite order, and so it may be asserted that each term of the expansion takes into account collective effects in a natural way.

For a finite operator manifold and an approximate reference state, the approach to the self-energy within the FEIM method is readily obtained by including in the finite expansion only certain terms which may be selected according to the type of interaction terms and the transmission paths one wishes to include in the calculation, and not according to a given perturbative order in the electronic interaction.

This method offers the feature of selecting terms according to physical criteria, and so those which are suspected of being negligible may in a first attempt, be omitted and others may be included, regardless of the perturbative order at which they first appear. One may suppose at first glance that the higher the order at which a particular term in the expansion first appears, the less influence it should have; however, when dealing with infinite series of diagrams, the perturbative order at which they begin cannot be considered as a definite criterion. If results obtained with a particular selection of terms should be improved, new terms may be simply added to the existing ones in a straighforward way.

In Sect. 2 the general equations for a finite expansion of the inverse of a Hermitian matrix are derived. In Sect. 3 a comparison of the FEIM method with the perturbative series of the inverse is presented and a diagrammatic interpretation of the FEIM is outlined. Section 4 is devoted to the FEIM analysis of the p-h self-energy and finally, in Sect. 5 a short summary is given.

2 Decomposition of the inverse matrix as a finite sum of terms

Let us consider a Hermitian matrix $\mathbf{X}$ and its inverse $\mathbf{Y}$, both of finite dimension N, partitioned as

$$\mathbf{X} = \begin{pmatrix} \mathbf{T} & \mathbf{M} \\ \mathbf{M}^\dagger & \mathbf{W} \end{pmatrix} , \tag{1}$$

$$\mathbf{Y} = \begin{pmatrix} \mathbf{R} & \mathbf{N} \\ \mathbf{N}^\dagger & \mathbf{Z} \end{pmatrix} , \tag{2}$$

where $\dim(\mathbf{T}) = \dim(\mathbf{R})$.

In what follows, matrix indices may label either individual matrix elements or whole submatrices. The partitioning of matrices $\mathbf{X}$ and $\mathbf{Y}$ is chosen depending on whether one seeks an expression for the diagonal element or block (ii) of $\mathbf{X}^{-1}$ or for the nondiagonal one (ij). In the former case, $\mathbf{T}$ and $\mathbf{R}$ are characterized by index i and in the latter by indices i and j. Elements with these particular choices of indices can always be brought to the upper left position of the matrix by redefinition of the indices.

We intend to prove that element (ij) of $\mathbf{Y}$ [for the (ii) element just take $j = i$] may be written in the form of a finite expansion as

$$\mathbf{Y}_{ij} \equiv \mathbf{R}_{ij} = \mathbf{F}^{ij} + \sum_{\substack{k \\ k \neq i,j}}^{N} \mathbf{F}^{ij}(k) + \sum_{\substack{k<l \\ k,l \neq i,j}}^{N} \mathbf{F}^{ij}(k,l) + \sum_{\substack{k<l<m \\ k,l,m \neq i,j}}^{N} \mathbf{F}^{ij}(k,l,m) + \cdots + \mathbf{F}^{ij}(k,l,\ldots,N) \ . \tag{3}$$

In Eq. (3) $\mathbf{F}^{ij}$ represents the contribution to element (ij) of $\mathbf{X}^{-1}$ arising from elements of matrix $\mathbf{X}$ which involve only indices i and j, i.e., $\mathbf{X}_{ii}$, $\mathbf{X}_{ij}$, $\mathbf{X}_{ji}$ and $\mathbf{X}_{jj}$. The term $\mathbf{F}^{ij}(k)$ collects the contribution of elements with indices i, j and k, where k should be different from the other indices, since if $k = i$ or $k = j$, this contribution has already been taken into account in $\mathbf{F}^{ij}$. The term $\mathbf{F}^{ij}(k,l)$ collects the contributions of elements with indices i, j, k and l, and so on. Indices in each term $\mathbf{F}^{ij}(k,l,\ldots)$ are always different, since contributions with repeated indices were included in previous $\mathbf{F}^{ij}(\ldots)$ which involve a smaller number of indices.

A natural way to interpret this expansion would be to consider that $\mathbf{F}^{ij}$ represents some kind of direct interaction between i and j; $\mathbf{F}^{ij}(k)$ represents the same type of interaction, but transmitted through k, and so on. We stress the fact that $\mathbf{F}^{ij}(k)$ collects all interaction terms between i and j where k is involved. $\mathbf{F}^{ij}(k,l)$ represents the interaction transmitted through k and l, and so on.

Using the partitioning technique [27] $\mathbf{R}$ may be written as (indices i and j are omitted when no confusion may arise)

$$\mathbf{R} = [\mathbf{T} - \mathbf{M}\mathbf{W}^{-1}\mathbf{M}^\dagger]^{-1} = \left[\mathbf{I} + \mathbf{T}^{-1}\mathbf{M}\mathbf{W}^{-1}\mathbf{M}^\dagger + (\mathbf{T}^{-1}\mathbf{M}\mathbf{W}^{-1}\mathbf{M}^\dagger)^2 + \cdots\right]\mathbf{T}^{-1} \equiv \mathbf{J}\mathbf{T}^{-1} \ . \tag{4}$$

It is assumed that the inverse matrix of $\mathbf{T}$ exists. From the comments after Eq. (2), this means that $\det(\mathbf{X}_{ii}) \neq 0$ (when indices label individual matrix elements, this is simply equivalent to $\mathbf{X}_{ii} \neq 0$) and that the matrix formed by $\mathbf{X}_{ii}$, $\mathbf{X}_{jj}$, $\mathbf{X}_{ij}$ and $\mathbf{X}_{ji}$ also has an inverse.

Since $\dim(\mathbf{W}) < N$, it can be considered as an inductive hypothesis that elements $(\mathbf{W}^{-1})_{kl}$ have the same type of expansion as those of Eq. (3); moreover, the only difference in the expansions of $(\mathbf{X}^{-1})_{kl}$ and $(\mathbf{W}^{-1})_{kl}$ is that in the latter case terms involving indices i or j do not appear.

The product $\mathbf{M}\mathbf{W}^{-1}\mathbf{M}^\dagger$ can thus be expressed as

$$\mathbf{M}\mathbf{W}^{-1}\mathbf{M}^\dagger = \sum_{\substack{k \\ k \neq i,j}} \mathbf{K}^{(ij)}(k) + \sum_{\substack{k<l \\ k,l \neq i,j}} \mathbf{K}^{(ij)}(k,l) + \cdots \ . \tag{5}$$

Matrices $\mathbf{K}^{(ij)}$ only depend on indices i and j through matrices $\mathbf{M}$. If the diagonal element (ii) of the inverse were sought, the $\mathbf{G}^{(ii)}$ would be complex numbers; for the non-diagonal (ij) ones, they would be 2×2 matrices and when indices represent submatrices they will be matrices of higher dimension.

Using the FEIM methzod for $\mathbf{W}$, $\mathbf{M}\mathbf{W}^{-1}\mathbf{M}^{\dagger}$ may be written as

$$\begin{aligned}(\mathbf{M}\mathbf{W}^{-1}\mathbf{M}^{\dagger})_{pq} &= \sum_{k,l} \mathbf{M}_{pk}(\mathbf{W}^{-1})_{kl}\mathbf{M}^{\dagger}_{lq} \\ &= \sum_{k,l} \mathbf{M}_{pk}\Bigg[\mathbf{F}^{kl} + \sum_{\substack{m \\ m\neq k,l}} \mathbf{F}^{kl}(m) \\ &\quad + \sum_{\substack{m<n \\ m,n\neq k,l}} \mathbf{F}^{kl}(m,n) + \cdots\Bigg]\mathbf{M}^{\dagger}_{lq} \ , \end{aligned} \tag{6}$$

where $p, q \in \{i, j\}$.

The order of factors is preserved in order to include the case when $\mathbf{F}^{kl}(\cdots)$ are true matrices, which occurs when indices label submatrices. Grouping together terms with $k = l$ and with $k \neq l$, Eq. (6) can be expressed as

$$\begin{aligned}(\mathbf{M}\mathbf{W}^{-1}\mathbf{M}^{\dagger})_{pq} &= \sum_{k} \mathbf{M}_{pk}\Bigg[\mathbf{F}^{kk} + \sum_{\substack{m \\ m\neq k}} \mathbf{F}^{kk}(m) \\ &\quad + \sum_{\substack{m<n \\ m,n\neq k}} \mathbf{F}^{kk}(m,n) + \cdots\Bigg]\mathbf{M}^{\dagger}_{kq} \\ &\quad + \sum_{k\neq l} \mathbf{M}_{pk}\Bigg[\mathbf{F}^{kl} + \sum_{\substack{m \\ m\neq k,l}} \mathbf{F}^{kl}(m) \\ &\quad + \sum_{\substack{m<n \\ m,n\neq k,l}} \mathbf{F}^{kl}(m,n) + \cdots\Bigg]\mathbf{M}^{\dagger}_{lq} \ , \end{aligned} \tag{7}$$

and collecting together terms according to an increasing number of different sum indices, the product $\mathbf{M}\mathbf{W}^{-1}\mathbf{M}^{\dagger}$ can be finally expressed as

$$\begin{aligned}&(\mathbf{M}\mathbf{W}^{-1}\mathbf{M}^{\dagger})_{pq} \\ &= \sum_{k} \mathbf{M}_{pk}\mathbf{F}^{kk}\mathbf{M}^{\dagger}_{kq} + \sum_{k<l}\Big[\mathbf{M}_{pk}\mathbf{F}^{kk}(l)\mathbf{M}^{\dagger}_{kq} \\ &\quad + \mathbf{M}_{pk}\mathbf{F}^{kl}\mathbf{M}^{\dagger}_{lq} + \mathbf{M}_{pl}\mathbf{F}^{ll}(k)\mathbf{M}^{\dagger}_{lq} + \mathbf{M}_{pl}\mathbf{F}^{lk}\mathbf{M}^{\dagger}_{kq}\Big] \\ &\quad + \sum_{k<l<m}\Big[\mathbf{M}_{pk}\mathbf{F}^{kk}(l,m)\mathbf{M}^{\dagger}_{kq} + \mathbf{M}_{pl}\mathbf{F}^{ll}(k,m)\mathbf{M}^{\dagger}_{lq} \\ &\quad + \mathbf{M}_{pm}\mathbf{F}^{mm}(k,l)\mathbf{M}^{\dagger}_{mq} + \mathbf{M}_{pk}\mathbf{F}^{kl}(m)\mathbf{M}^{\dagger}_{lq} \\ &\quad + \mathbf{M}_{pl}\mathbf{F}^{lk}(m)\mathbf{M}^{\dagger}_{kq} + \mathbf{M}_{pk}\mathbf{F}^{km}(l)\mathbf{M}^{\dagger}_{mq} \\ &\quad + \mathbf{M}_{pl}\mathbf{F}^{lm}(k)\mathbf{M}^{\dagger}_{mq} + \mathbf{M}_{pm}\mathbf{F}^{mk}(l)\mathbf{M}^{\dagger}_{kq} \\ &\quad + \mathbf{M}_{pm}\mathbf{F}^{ml}(k)\mathbf{M}^{\dagger}_{lq}\Big] + \cdots \ . \end{aligned} \tag{8}$$

By comparing Eqs. (5) and (8), the first $\mathbf{K}^{(ij)}(\cdots)$ matrices can be identified as

$$\mathbf{K}^{(ij)}_{pq}(k) = \mathbf{M}_{pk}\mathbf{F}^{kk}\mathbf{M}^{\dagger}_{kq} \ , \tag{9}$$

$$\begin{aligned}\mathbf{K}^{(ij)}_{pq}(k,l) &= \mathbf{M}_{pk}\mathbf{F}^{kk}(l)\mathbf{M}^{\dagger}_{kq} + \mathbf{M}_{pk}\mathbf{F}^{kl}\mathbf{M}^{\dagger}_{lq} \\ &\quad + \mathbf{M}_{pl}\mathbf{F}^{ll}(k)\mathbf{M}^{\dagger}_{lq} + \mathbf{M}_{pl}\mathbf{F}^{lk}\mathbf{M}^{\dagger}_{kq} \ , \end{aligned} \tag{10}$$

$$\begin{aligned}\mathbf{K}^{(ij)}_{pq}(k,l,m) &= \mathbf{M}_{pk}\mathbf{F}^{kk}(l,m)\mathbf{M}^{\dagger}_{kq} + \mathbf{M}_{pl}\mathbf{F}^{ll}(k,m)\mathbf{M}^{\dagger}_{lq} \\ &\quad + \mathbf{M}_{pm}\mathbf{F}^{mm}(k,l)\mathbf{M}^{\dagger}_{mq} + \mathbf{M}_{pk}\mathbf{F}^{kl}(m)\mathbf{M}^{\dagger}_{lq} \\ &\quad + \mathbf{M}_{pl}\mathbf{F}^{lk}(m)\mathbf{M}^{\dagger}_{kq} + \mathbf{M}_{pk}\mathbf{F}^{km}(l)\mathbf{M}^{\dagger}_{mq} \\ &\quad + \mathbf{M}_{pl}\mathbf{F}^{lm}(k)\mathbf{M}^{\dagger}_{mq} + \mathbf{M}_{pm}\mathbf{F}^{mk}(l)\mathbf{M}^{\dagger}_{kq} \\ &\quad + \mathbf{M}_{pm}\mathbf{F}^{ml}(k)\mathbf{M}^{\dagger}_{lq} \ . \end{aligned} \tag{11}$$

The general term may be expressed as

$$\begin{aligned}&\mathbf{K}^{(ij)}_{pq}(k_1, k_2, \ldots, k_n) \\ &= \sum_{l_1=1}^{n} \mathbf{M}_{pk_{l_1}}\mathbf{F}^{k_{l_1}k_{l_1}}(k_{l_2}, \ldots, k_{l_n})\mathbf{M}^{\dagger}_{k_{l_1}q} \\ &\quad + \sum_{\substack{l_1,l_2=1 \\ l_1\neq l_2}} \mathbf{M}_{pk_{l_1}}\mathbf{F}^{k_{l_1}k_{l_2}}(k_{l_3}, \ldots, k_{l_n})\mathbf{M}^{\dagger}_{k_{l_2}q} \ , \end{aligned} \tag{12}$$

where $p, q \in \{i, j\}$, $k_1 < k_2 < \cdots < k_n$ and the subset $(k_{l_2}, \ldots, k_{l_n})$ is the ordered set $(k_1, k_2, \ldots, k_n)$, where k_{l_1} is missing [an analogous interpretation holds for $(k_{l_3}, \ldots, k_{l_n})$].

According to Eq. (4), matrix $\mathbf{J}$ can be expressed as

$$\mathbf{J} = \sum_{n=0}^{\infty}\left[\mathbf{T}^{-1}\mathbf{M}\mathbf{W}^{-1}\mathbf{M}^{\dagger}\right]^{n} \ . \tag{13}$$

Using Eq. (5) and the multinomial expansion, $\mathbf{J}$ may be written as

$$\begin{aligned}\mathbf{J} &= \sum_{n=0}^{\infty}\left[\mathbf{T}^{-1}\left(\sum_{k}\mathbf{K}^{(ij)}(k) + \sum_{k<l}\mathbf{K}^{(ij)}(k,l) + \cdots\right)\right]^{n} \\ &= \sum_{n=0}^{\infty}\sum_{\substack{\{n_k\}\{n_{kl}\} \\ \{n_{klm}\}\cdots \\ \sum n_{kl\ldots}=n}}\Bigg(\frac{n!}{n_1!n_2!\ldots}\prod_{k}\left[\mathbf{T}^{-1}\mathbf{K}^{(ij)}(k)\right]^{n_k} \\ &\quad \times \prod_{k<l}\left[\mathbf{T}^{-1}\mathbf{K}^{(ij)}(k,l)\right]^{n_{kl}} \\ &\quad \times \prod_{k<l<m}\left[\mathbf{T}^{-1}\mathbf{K}^{(ij)}(k,l,m)\right]^{n_{klm}}\ldots\Bigg) \\ &= \mathbf{J}_0 + \sum_{k}\mathbf{J}(k) + \sum_{k<l}\mathbf{J}(k,l) + \cdots \ . \end{aligned} \tag{14}$$

It is expedient to analyze the expansion of $\mathbf{J}$ in Eq. (14) according to the number of indices $k, l, m, \ldots$ involved in the sums of its right-hand side (rhs). The term $\mathbf{J}_0$ corresponds to the case $n = 0$ in the multinomial expansion, and thus

$$\mathbf{J}_0 = \mathbf{1} \ . \tag{15}$$

Using this result in Eq. (4) and by comparing with Eq. (3), the first terms in the FEIM can be identified as

$$\mathbf{F}^{ii} = \mathbf{T}^{-1} \equiv (\mathbf{X}_{ii})^{-1} \ , \tag{16}$$

$$\mathbf{F}^{ij} = (\mathbf{T}^{-1})_{ij} \equiv \left[\begin{pmatrix} \mathbf{X}_{ii} & \mathbf{X}_{ij} \\ \mathbf{X}_{ji} & \mathbf{X}_{jj} \end{pmatrix}^{-1} \right]_{ij} \ . \tag{17}$$

In deriving Eqs. (16) and (17) different partitions of $\mathbf{X}$ were taken in each case, according to what was said after Eq. (2).

The next term to be considered corresponds to the case where all but one of the n_i are zero, which will be noted n_p. According to Eq. (14) the associated contribution takes the form

$$\begin{aligned} \mathbf{J}(p) &= \sum_{n=1}^{\infty} \left[\mathbf{T}^{-1} \mathbf{K}^{(ij)}(p) \right]^n = \sum_{n=0}^{\infty} \left[\mathbf{T}^{-1} \mathbf{K}^{(ij)}(p) \right]^n - \mathbf{1} \\ &= \left[\mathbf{1} - \mathbf{T}^{-1} \mathbf{K}^{(ij)}(p) \right]^{-1} - \mathbf{1} \ . \end{aligned} \tag{18}$$

With contributions of Eqs. (15) and (18), the matrix $\mathbf{R}$ may be written, following Eq. (4) as

$$\begin{aligned} \mathbf{R} &= \left[\mathbf{1} + \sum_{p}^{N} \left[(\mathbf{1} - \mathbf{T}^{-1} \mathbf{K}^{(ij)}(p))^{-1} - \mathbf{1} \right] \right] \mathbf{T}^{-1} \\ &= \mathbf{T}^{-1} + \sum_{p}^{N} \left[(\mathbf{T} - \mathbf{K}^{(ij)}(p))^{-1} - \mathbf{T}^{-1} \right] \ . \end{aligned} \tag{19}$$

By comparing Eqs. (3) and (19) and taking into account the expressions of Eqs. (16) and (17), one obtains

$$\mathbf{F}^{ij}(k) = \left[[\mathbf{T} - \mathbf{K}^{(ij)}(k)]^{-1} - \mathbf{T}^{-1} \right]_{ij} \ . \tag{20}$$

In the usual case where indices refer to single matrix elements, the term $\mathbf{F}^{ii}(k)$ is expressed as

$$\begin{aligned} \mathbf{F}^{ii}(k) &= \frac{\mathbf{M}_{ik}\mathbf{M}_{ki}}{\mathbf{T}_{ii}(\mathbf{T}_{ii}\mathbf{T}_{kk} - \mathbf{M}_{ik}\mathbf{M}_{ki})} \\ &\equiv \frac{|\mathbf{X}_{ik}|^2}{\mathbf{X}_{ii}(\mathbf{X}_{ii}\mathbf{X}_{kk} - |\mathbf{X}_{ik}|^2)} \ , \end{aligned} \tag{21}$$

where Eq. (9) has been used.

It is important to note that if the nondiagonal part of matrix $\mathbf{X}$ defines a certain perturbative order, $\mathbf{F}^{ii}(k)$ represents a correction to the diagonal element of the inverse which begins in the second order but which includes in addition terms summed up to infinite order.

An analogous expression for $\mathbf{F}^{ij}(k)$ can be obtained from Eqs. (9) and (20) in the form

$$\begin{aligned} \mathbf{F}^{ij}(k) = \Bigg[& \begin{pmatrix} \mathbf{T}_{ii} - \frac{\mathbf{M}_{ik}\mathbf{M}_{ki}}{\mathbf{T}_{kk}} & \mathbf{T}_{ij} - \frac{\mathbf{M}_{ik}\mathbf{M}_{kj}}{\mathbf{T}_{kk}} \\ \mathbf{T}^{*}_{ij} - \frac{\mathbf{M}^{*}_{ik}\mathbf{M}^{*}_{kj}}{\mathbf{T}_{kk}} & \mathbf{T}_{jj} - \frac{\mathbf{M}_{jk}\mathbf{M}_{kj}}{\mathbf{T}_{kk}} \end{pmatrix}^{-1} \\ & - \begin{pmatrix} \mathbf{T}_{ii} & \mathbf{T}_{ij} \\ \mathbf{T}^{*}_{ij} & \mathbf{T}_{jj} \end{pmatrix}^{-1} \Bigg]_{ij} \ . \end{aligned} \tag{22}$$

The next terms to be considered are those which involve two different indices, k and l. The corresponding term in Eq. (14) is

$$\begin{aligned} \mathbf{J}(k,l) = \sum_{n=1}^{\infty} & \sum_{\substack{n_k n_l n_{kl} \\ n_k + n_l + n_{kl} = n \\ n_k, n_l \neq n}} \frac{n!}{n_k! n_l! n_{kl}!} \left[\mathbf{T}^{-1} \mathbf{K}^{(ij)}(k) \right]^{n_k} \\ & \times \left[\mathbf{T}^{-1} \mathbf{K}^{(ij)}(l) \right]^{n_l} \left[\mathbf{T}^{-1} \mathbf{K}^{(ij)}(k,l) \right]^{n_{kl}} \ . \end{aligned} \tag{23}$$

Terms with $n_k = n$ and $n_l = n$ are excluded, since they have already been taken into account in terms $\mathbf{J}(k)$ and $\mathbf{J}(l)$, respectively.

By adding and subtracting terms with $n = 0$, $n_k = n$ and $n_l = n$, the sum of Eq. (23) can be written as

$$\begin{aligned} \mathbf{J}(k,l) = & \sum_{n=0}^{\infty} \sum_{\substack{n_k n_l n_{kl} \\ n_k + n_l + n_{kl} = n}} \frac{n!}{n_k! n_l! n_{kl}!} \left[\mathbf{T}^{-1} \mathbf{K}^{(ij)}(k) \right]^{n_k} \\ & \times \left[\mathbf{T}^{-1} \mathbf{K}^{(ij)}(l) \right]^{n_l} \left[\mathbf{T}^{-1} \mathbf{K}^{(ij)}(k,l) \right]^{n_{kl}} - \mathbf{1} \\ & - \sum_{n=1}^{\infty} \left[\mathbf{T}^{-1} \mathbf{K}^{(ij)}(k) \right]^{n} - \sum_{n=1}^{\infty} \left[\mathbf{T}^{-1} \mathbf{K}^{(ij)}(l) \right]^{n} \ . \end{aligned} \tag{24}$$

The second sum of the first term on the rhs of Eq. (24) is the multinomial expansion of $[\mathbf{T}^{-1}(\mathbf{K}^{(ij)}(k) + \mathbf{K}^{(ij)}\ (l) + \mathbf{K}^{(ij)}(k,l)]^n$ and according to Eqs. (18) and (20), the last two terms in Eq. (24) correspond to

$$\sum_{n=1}^{\infty} \left[\mathbf{T}^{-1} \mathbf{K}^{(ij)}(k) \right]^{n} = \mathbf{F}^{ij}(k)\mathbf{T} \ , \tag{25}$$

with an analogous expression for l.

In this way, $\mathbf{J}(k,l)$ may be expressed in a condensed form as

$$\begin{aligned} \mathbf{J}(k,l) = & \Big[\mathbf{1} - \mathbf{T}^{-1} \mathbf{K}^{(ij)}(k) - \mathbf{T}^{-1} \mathbf{K}^{(ij)}(l) \\ & - \mathbf{T}^{-1} \mathbf{K}^{(ij)}(k,l) \Big]^{-1} - \mathbf{1} \ - \mathbf{F}^{ij}(k)\mathbf{T} - \mathbf{F}^{ij}(l)\mathbf{T} \ . \end{aligned} \tag{26}$$

Finally, from Eqs. (3) and (4), the term $\mathbf{F}^{ij}(k,l)$ may be obtained as

$$\begin{aligned} \mathbf{F}^{ij}(k,l) &= \mathbf{J}(k,l)\mathbf{T}^{-1} \\ &= \Big[[\mathbf{T} - \mathbf{K}^{(ij)}(k) - \mathbf{K}^{(ij)}(l) - \mathbf{K}^{(ij)}(k,l)]^{-1} \\ &\quad - \mathbf{T}^{-1} \Big]_{ij} - \mathbf{F}^{ij}(k) - \mathbf{F}^{ij}(l) \ . \end{aligned} \tag{27}$$

It can be inferred that $\mathbf{F}^{ij}(k,l)$ is expressed in terms of the results of previous calculations.

Proceeding in an analogous way, the general term can be readily obtained as

$$\begin{aligned} & \mathbf{F}^{ij}(k_1, k_2, \ldots, k_n) \\ &= \Bigg[\Bigg(\mathbf{T} - \sum_{l=1}^{n} \mathbf{K}^{(ij)}(k_l) - \sum_{l<m}^{n} \mathbf{K}^{(ij)}(k_l, k_m) - \cdots \\ &\quad - \sum_{\substack{k_{l_1} < \cdots < k_{l_{n-1}} \\ \in \{k_1, k_2, \ldots, k_n\}}} \mathbf{K}^{(ij)}(k_{l_1}, \ldots, l_{l_n}) \\ &\quad - \mathbf{K}^{(ij)}(k_1, k_2, \ldots, k_n) \Bigg)^{-1} - \mathbf{T}^{-1} \Bigg]_{ij} \\ &\quad - \sum_{l=1}^{n} \mathbf{F}^{ij}(k_l) - \sum_{l<m}^{n} \mathbf{F}^{ij}(k_l, k_m) - \cdots \end{aligned}$$

$$- \sum_{\substack{k_{l_1} < \cdots < k_{l_{n-1}} \\ \in \{k_1, k_2, \ldots, k_n\}}} \mathbf{F}^{ij}(k_{l_1}, \ldots, k_{l_{n-1}}) \ , \tag{28}$$

where $k_1 < k_2 < \ldots < k_n$.

3 Comparison of the FEIM method with the perturbative expansion of the inverse

For a better understanding of the FEIM method, let us consider a perturbative expansion of $\mathbf{X}^{-1}$ in the form

$$\mathbf{X}^{-1} = [\mathbf{X}_0 + \mathbf{X}_1]^{-1} = \mathbf{X}_0^{-1} - \mathbf{X}_0^{-1}\mathbf{X}_1\mathbf{X}_0^{-1} + \mathbf{X}_0^{-1}\mathbf{X}_1\mathbf{X}_0^{-1}\mathbf{X}_1\mathbf{X}_0^{-1} - \cdots \ , \tag{29}$$

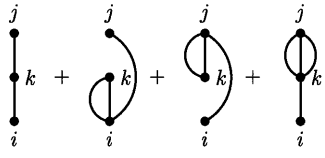
where $\mathbf{X}_0$ is a diagonal matrix and $\mathbf{X}_1$ can be regarded as a perturbation.

The basic idea of the FEIM is to group in a single term, $\mathbf{F}^{ij}(k_1, k_2, \ldots, k_m)$, all the contributions to element (ij) of $\mathbf{X}^{-1}$ that, on the rhs of Eq. (29), involve products of matrix elements $\mathbf{X}_{mn}$ with $m, n \in \{i, k_1, k_2, \ldots, k_m, j\}$.

This can also be seen in a pictorial way. By choosing the partition in Eq. (29) such that $(\mathbf{X}_1)_{kk} = 0$ and by representing by dots the diagonal terms $(\mathbf{X}_0)_{kk}$ and by segments joining those dots the terms $(\mathbf{X}_1)_{kl}$, $\mathbf{F}^{ij}(k_1, k_2, \ldots, k_m)$ may be represented by a sum of infinite diagrams constructed following these rules:

- Draw $m + 2$ dots aligned labeled $\{i, k_1, k_2, \cdots, k_m, j\}$.
- Beginning with i and ending with j (or vice versa), join all dots with lines. Each dot must be joined at least once.
- There are no closed loops, since $(\mathbf{X}_1)_{kk} = 0$.

The term $\mathbf{F}^{ij}(k_1, k_2, \ldots, k_m)$ is obtained by summing all the diagrams drawn joining the $m + 2$ dots in all possible manners, following the rules given previously. The first diagrams of the $\mathbf{F}^{ij}(k)$ term look like

$\mathbf{F}^{ij}(k) =$  $+ \cdots .$ (30)

The number of lines is the number of $(\mathbf{X}_1)_{mn}$ terms that appears in each diagram, i.e., the order in the interaction represented by $\mathbf{X}_1$. Of course, if we chose the partitioning of $\mathbf{X}$ in Eq. (29) by removing the constraint $(\mathbf{X}_1)_{ii} = 0$, closed loops should also be included.

On modifying the elements with index k_1 in matrix $\mathbf{X}$, $\mathbf{R}_{ij}$ will change; however, in the expansion of Eq. (3) only the terms $\mathbf{F}^{ij}(\cdots)$ involving index k_1 will be altered. Thus, the FEIM provides a quantitative measure of the influence of elements of the original matrix on its inverse.

4 The FEIM method and the self-energy of the p-h propagator

4.1 The PP

The Fourier transform of the double-time Green function (propagator) of two operators A and B may be written in the superoperator formalism [16, 17] as

$$\langle\langle \mathsf{A}; \mathsf{B} \rangle\rangle_E = (\mathsf{A}^\dagger | (E\hat{\mathsf{I}} - \hat{\mathsf{H}})^{-1} \mathsf{B}) \ , \tag{31}$$

where $\langle\langle \mathsf{A}; \mathsf{B} \rangle\rangle_E$ is the propagator in the energy domain and $\hat{\mathsf{H}}$ and $\hat{\mathsf{I}}$ refer to the superoperators Hamiltonian and identity, respectively, the action of which on an arbitrary element of the operators space $\{\mathsf{A}_j\}$ is defined as [16, 17]

$$\hat{\mathsf{H}}\mathsf{A}_j = [\mathsf{H}, \mathsf{A}_j]_- \ , \tag{32}$$

$$\hat{\mathsf{I}}\mathsf{A}_j = \mathsf{A}_j \ . \tag{33}$$

A binary product between operators is introduced by means of the mean value

$$(\mathsf{X}|\mathsf{Y}) = \langle \psi_0^N | [\mathsf{X}^\dagger, \mathsf{Y}] | \psi_0^N \rangle \ , \tag{34}$$

where $|\psi_0^N\rangle$ is the exact reference ground state of the system of N particles.

Following the Møller–Plesset partitioning, the Hamiltonian of the system may be expressed in terms of a zero-order (Fock operator) and a first-order part, as follows:

$$\mathsf{H} = \mathsf{F} + \mathsf{V} \ , \tag{35}$$

where

$$\mathsf{F} = \sum_i \varepsilon_i \mathsf{a}_i^\dagger \mathsf{a}_i \ , \tag{36}$$

$$\mathsf{V} = \frac{1}{4} \sum_{ijkl} \langle ij||kl\rangle \mathsf{a}_i^\dagger \mathsf{a}_j^\dagger \mathsf{a}_l \mathsf{a}_k - \sum_{ij\alpha} \langle i\alpha||j\alpha\rangle \mathsf{a}_i^\dagger \mathsf{a}_j \ . \tag{37}$$

In what follows, indices α, β, γ, ... (m, n, p, ...) denote occupied (virtual) Hartree–Fock spin-orbitals, and i, j, k, ... refer to unrestricted indices.

In fact, Eq. (31) may be written in a more useful form via the inner projection technique [28] as

$$\langle\langle \mathsf{A}; \mathsf{B} \rangle\rangle_E = (\mathsf{A}^\dagger | \tilde{\mathbf{h}}) (\mathbf{h} | E\hat{\mathsf{I}} - \hat{H} | \tilde{\mathbf{h}})^{-1} (\mathbf{h} | \mathsf{B}) \ , \tag{38}$$

where the superoperator inverse is avoided in favor of an inverse matrix and $\mathbf{h}$ stands for a complete operator basis [29]. When A and B are number-conserving one-electron operators, $\langle\langle \mathsf{A}; \mathsf{B} \rangle\rangle_E$ is called the PP, in which case $\mathbf{h}$ may be chosen as [29]

$$\{\mathbf{h}\} = \{\mathbf{h}_2, \mathbf{h}_4, \ldots\} \ , \tag{39}$$

where $\mathbf{h}_{2N}$ includes both the Nparticle-Nhole excitation (Np-Nh) and Nhole-Nparticle (Nh-Np) deexcitation manifolds, defined as

$$\{\mathbf{h}_2\} = \{\mathbf{q}^\dagger, \mathbf{q}\} = \{\mathsf{a}_m^\dagger \mathsf{a}_\alpha, \mathsf{a}_\alpha^\dagger \mathsf{a}_m\} \ , \tag{40}$$

$$\{\mathbf{h}_4\} = \{\mathbf{q}^\dagger \mathbf{q}^\dagger, \mathbf{q}\mathbf{q}\} = \{\mathsf{a}_m^\dagger \mathsf{a}_\alpha \mathsf{a}_n^\dagger \mathsf{a}_\beta, \mathsf{a}_\alpha^\dagger \mathsf{a}_m \mathsf{a}_\beta^\dagger \mathsf{a}_n\} \quad m > n, \ \alpha > \beta \ , \tag{41}$$

and so on.

Different truncations of $\mathbf{h}$ will lead to approximate expressions of the PP. In particular, the choice $\mathbf{h} = \{\mathbf{h}_2, \mathbf{h}_4\}$ suffices to determine the principal propagator consistent to third order in the residual potential V, provided that the reference ground state is also appropriately approximated [18].

This choice of the manifold, $\mathbf{h}$, together with a ground state exact to first order in Rayleigh–Schrödinger perturbation theory and a few second-order terms origi-

nating from single excitations, ensures a PP consistent to second order in V (SOPPA [7, 20]).

The principal propagator $\mathbf{P}(E)$ in a second-order theory takes the form [20]

$$\mathbf{P}(E)^{-1} = \begin{bmatrix} \langle\langle \mathbf{q}; \tilde{\mathbf{q}}^\dagger \rangle\rangle_E & \langle\langle \mathbf{q}; \tilde{\mathbf{q}} \rangle\rangle_E \\ \langle\langle \mathbf{q}^\dagger; \tilde{\mathbf{q}}^\dagger \rangle\rangle_E & \langle\langle \mathbf{q}^\dagger; \tilde{\mathbf{q}} \rangle\rangle_E \end{bmatrix} = \begin{pmatrix} E\mathbf{S} - \mathbf{A} - \tilde{\mathbf{C}}[E\mathbf{1} - \mathbf{D}]^{-1}\mathbf{C} & -\mathbf{B} \\ -\mathbf{B} & -E\mathbf{S} - \mathbf{A} - \tilde{\mathbf{C}}[-E\mathbf{1} - \mathbf{D}]^{-1}\mathbf{C} \end{pmatrix}^{-1} , \tag{42}$$

where the matrices involved are asumed to be real and are defined as

$$\mathbf{A}^{(0,1,2)} = (\mathbf{q}^\dagger|\hat{\mathsf{H}}|\tilde{\mathbf{q}}^\dagger) = (\mathbf{q}|\hat{\mathsf{H}}|\tilde{\mathbf{q}}) \ , \tag{43}$$

$$\mathbf{B}^{(1,2)} = (\mathbf{q}|\hat{\mathsf{H}}|\tilde{\mathbf{q}}^\dagger) = (\mathbf{q}^\dagger|\hat{\mathsf{H}}|\tilde{\mathbf{q}}) \ , \tag{44}$$

$$\mathbf{C}^{(1)} = (\mathbf{q}^\dagger\mathbf{q}^\dagger|\hat{\mathsf{V}}|\tilde{\mathbf{q}}^\dagger) \ , \tag{45}$$

$$\mathbf{D}^{(0)} = (\mathbf{q}^\dagger\mathbf{q}^\dagger|\hat{\mathsf{F}}|\widetilde{\mathbf{q}^\dagger\mathbf{q}^\dagger}) \ , \tag{46}$$

where the superscripts denote the order in the electronic repulsion and explicit expressions can be found in Ref. [10]; the expression for $\mathbf{C}(1)$ in Ref. [10] has the wrong sign. The metric used at this level of approximation is given by

$$(\mathbf{h}|\tilde{\mathbf{h}}) = \begin{pmatrix} \mathbf{S}^{(0,2)} & \mathbf{0} & \mathbf{0} & \mathbf{0} \\ \mathbf{0} & -\mathbf{S}^{(0,2)} & \mathbf{0} & \mathbf{0} \\ \mathbf{0} & \mathbf{0} & \mathbf{1} & \mathbf{0} \\ \mathbf{0} & \mathbf{0} & \mathbf{0} & -\mathbf{1} \end{pmatrix} . \tag{47}$$

By evaluating the left upper block of the inverse in Eq. (42), the self-energy associated with the SOPPA p-h propagator $\langle\langle \mathbf{q}; \tilde{\mathbf{q}}^\dagger \rangle\rangle_E$ can be expressed as

$$\mathbf{\Sigma}(E) = -E\mathbf{S}^{(2)} + \mathbf{A}^{(1,2)} + \tilde{\mathbf{C}}(E\mathbf{1} - \mathbf{D})^{-1}\mathbf{C} + \tilde{\mathbf{B}}\left[-E\mathbf{S} - \mathbf{A} - \tilde{\mathbf{C}}(-E\mathbf{1} - \mathbf{D})^{-1}\mathbf{C}\right]^{-1}\mathbf{B} \ . \tag{48}$$

4.2 The FEIM analysis of the p-h propagator

However, another approach using the FEIM method is also possible. Using the metric of Eq. (47) and partitioning $\mathbf{h} = \{\mathbf{h}_2, \mathbf{h}_4\}$ into a primary p-h manifold, $\mathbf{q}^\dagger$, and a secondary space, $\mathbf{f}$, orthogonal to it, which groups the h − p, 2p − 2h and 2h − 2p manifolds, the p-h propagator may be expressed in matrix form from Eq. (38) as

$$\langle\langle \mathbf{q}; \tilde{\mathbf{q}}^\dagger \rangle\rangle_E = (\mathbf{S}\ \mathbf{0}) \begin{pmatrix} E\mathbf{S} - (\mathbf{q}^\dagger|\hat{\mathsf{H}}|\tilde{\mathbf{q}}^\dagger) & -(\mathbf{q}^\dagger|\hat{\mathsf{V}}|\tilde{\mathbf{f}}) \\ -(\mathbf{f}|\hat{\mathsf{V}}|\tilde{\mathbf{q}}^\dagger) & (\mathbf{f}|E\hat{\mathsf{I}} - \hat{\mathsf{H}}|\tilde{\mathbf{f}}) \end{pmatrix}^{-1} \times \begin{pmatrix} \mathbf{S} \\ \mathbf{0} \end{pmatrix} , \tag{49}$$

where $\mathbf{f}$ is used as an eigenstate of $\hat{\mathsf{F}}$.

It is readily seen that only the upper left block of the inverse is relevant and

$$\langle\langle \mathbf{q}; \mathbf{q}^\dagger \rangle\rangle_E^{-1} = E\mathbf{S} - (\mathbf{q}^\dagger|\hat{\mathsf{H}}|\tilde{\mathbf{q}}^\dagger) - (\mathbf{q}^\dagger|\hat{\mathsf{V}}|\tilde{\mathbf{f}})(\mathbf{f}|E\hat{\mathsf{I}} - \hat{\mathsf{H}}|\tilde{\mathbf{f}})^{-1} \times (\mathbf{f}|\hat{\mathsf{V}}|\tilde{\mathbf{q}}^\dagger) \tag{50}$$

is obtained, where $\mathbf{S}$ was taken as the identity matrix on both sides of the brackets in Eq. (50) to perform an analysis consistent with Eq. (48). A Dyson-like equation can thus be extracted in the form

$$\langle\langle \mathbf{q}; \tilde{\mathbf{q}}^\dagger \rangle\rangle_E^{-1} = \mathbf{G}_0(E)^{-1} - \mathbf{\Sigma}(E) \ , \tag{51}$$

where

$$\mathbf{G}_0(E)^{-1} = E\mathbf{1} - \mathbf{A}^{(0)} \ , \tag{52}$$

$$\mathbf{\Sigma}(E) = -E\mathbf{S}^{(2)} + \mathbf{A}^{(1,2)} + (\mathbf{q}^\dagger|\hat{\mathsf{V}}|\tilde{\mathbf{f}})(\mathbf{f}|E\hat{\mathsf{I}} - \hat{\mathsf{H}}|\tilde{\mathbf{f}})^{-1} \times (\mathbf{f}|\hat{\mathsf{V}}|\tilde{\mathbf{q}}^\dagger) \ . \tag{53}$$

The third term of the rhs of Eq. (53) is called $\mathbf{\Sigma}(E)'$, being expressed in matrix form as

$$\Sigma(E)' = (\mathbf{B}\ \tilde{\mathbf{C}} - \tilde{\mathbf{G}}) \begin{pmatrix} -E\mathbf{S} - \mathbf{A} & \tilde{\mathbf{G}} & \tilde{\mathbf{C}} \\ \mathbf{G} & E\mathbf{1} - \mathbf{D} & \mathbf{L} \\ \mathbf{C} & \tilde{\mathbf{L}} & -E\mathbf{1} - \mathbf{D} \end{pmatrix}^{-1} \times \begin{pmatrix} \mathbf{B} \\ \mathbf{C} \\ -\mathbf{G} \end{pmatrix} , \tag{54}$$

where Eqs. (43)–(47) and the property $(\mathsf{A}|\mathsf{H}|\mathsf{B}) = (\mathsf{A}^\dagger|\mathsf{H}|\mathsf{B}^\dagger)^*$ were used. Matrices $\mathbf{G}$ and $\mathbf{L}$ are defined through

$$\mathbf{G}^{(2)} = -(\mathbf{qq}|\hat{\mathsf{V}}|\tilde{\mathbf{q}}^\dagger) \ , \tag{55}$$

$$\mathbf{L}^{(2)} = -(\mathbf{q}^\dagger\mathbf{q}^\dagger|\hat{\mathsf{V}}|\widetilde{\mathbf{qq}}) \ . \tag{56}$$

Since we are using an approximate reference ground state correct to first order in perturbation theory, the hermiticity of $\hat{\mathsf{H}}$ is guaranteed through that order [18]. However, the particular form of matrices $\mathbf{G}$ and $\mathbf{L}$ ensures that they are also Hermitian through second order. From this, it is easy to see that the matrices in Eq. (54) are real and symmetric, and so the square one may be expanded using the FEIM method.

From Eqs. (9), (16), (17) and (20), the first terms of the FEIM expansion are easily obtained:

$$\mathbf{F}^{\mathrm{q,q}} = (-E\mathbf{S} - \mathbf{A})^{-1} \ , \tag{57}$$

$$\mathbf{F}^{\mathrm{q}^\dagger\mathrm{q}^\dagger,\mathrm{q}^\dagger\mathrm{q}^\dagger} = (E\mathbf{1} - \mathbf{D})^{-1} \ , \tag{58}$$

$$\mathbf{F}^{\mathrm{q,q}}(\mathbf{qq}) = \left[(-E\mathbf{S} - \mathbf{A} - \tilde{\mathbf{C}}(-E\mathbf{1} - \mathbf{D})^{-1}\mathbf{C}\right]^{-1} - (-E\mathbf{S} - \mathbf{A})^{-1} \ . \tag{59}$$

As stated in Sect. 2, $\mathbf{F}^{\mathrm{q,q}}$ could be regarded as the term which describes the propagation of the interaction of pairs h – p through elements of the same manifold; $\mathbf{F}^{\mathrm{q,q}}(\mathbf{qq})$ represents the same interaction, but is transmitted through the manifold 2h – 2p. The FEIM method thus provides a practical way to isolate the total interaction of two elements of the operator space transmitted through a given subset of electronic excitations.

It is evident that the self-energy associated with the second-order PP (Eq. 48) is equivalent to considering in Eq. (54) the inverse matrix being approximated as

$$(\mathbf{f}|E\hat{\mathrm{I}} - \hat{\mathrm{H}}|\tilde{\mathbf{f}})^{-1} \simeq \begin{pmatrix} \mathbf{F}^{\mathrm{q,q}} + \mathbf{F}^{\mathrm{q,q}}(\mathbf{qq}) & \mathbf{0} & \mathbf{0} \\ \mathbf{0} & \mathbf{F}^{\mathrm{q}^\dagger\mathrm{q}^\dagger,\mathrm{q}^\dagger\mathrm{q}^\dagger} & \mathbf{0} \\ \mathbf{0} & \mathbf{0} & \mathbf{0} \end{pmatrix} . \quad (60)$$

Known approximations levels to the PP appear as special cases of the application of the FEIM method to the inverse matrix in Eq. (54): retaining only $\mathbf{F}^{\mathrm{q,q}}$ in Eq. (60) is equivalent to the RPA [10], and by including only $\mathbf{F}^{\mathrm{q,q}}$ and $\mathbf{F}^{\mathrm{q}^\dagger\mathrm{q}^\dagger,\mathrm{q}^\dagger\mathrm{q}^\dagger}$, one recovers the self-energy of the second-order p-h propagator written in partitioned form [18]. We notice that the FEIM expansion takes into account in a natural way collective effects [19], represented by diagrams summed to infinite order.

This expansion thus represents an alternative way to obtain approximate expressions for the PP where, for a given operator manifold, the criterion to include or not different terms in the expansion is not directly the order in the residual potential, since each $\mathbf{F}^{ij}(\cdots)$ includes terms summed to infinite order, but the type of interaction terms and transmission paths one wishes to include in the calculation.

This criterion, based on a selective choice of contributions, offers an interesting way to improve the results obtained with a PP consistent to a given order (excitation energies, response properties, etc.). This is the case, for example, in systems where the perturbative series of the PP converges slowly, since selected new $\mathbf{F}^{ij}(k_1,\ldots,k_l)$ terms may be simply added to the existing ones, without the need to calculate the propagator or its self-energy completely to the next order, and terms which are suspected of having negligible contributions may be omitted in a first attempt.

In the example of Eq. (60), inclusion of new terms, such as $\mathbf{F}^{\mathrm{qq,qq}}$, $\mathbf{F}^{\mathrm{q,q}}(\mathbf{q}^\dagger\mathbf{q}^\dagger)$, $\mathbf{F}^{\mathrm{q,q}^\dagger\mathrm{q}^\dagger}$, etc., is equivalent to adding in the perturbative scheme of the p-h propagator certain infinite sums of terms which begin beyond the third order [18]. Thus, a complete calculation of the p-h propagator to third (or higher) order is avoided, and instead selected infinite sums of diagrams are included.

The criterion for including terms in the FEIM expansion as outlined here should be based on the physical characteristics of the system under consideration and may be applied to other propagators or perturbative-like calculations.

5 Conclusions

Within the superoperator formalism, an alternative approach for the p-h propagator is presented, based on a FEIM which appears in the propagator formulation.

In the FEIM method, the terms that in the perturbative series of an N-dimensional inverse matrix $\mathbf{X}^{-1}$ involve elements $\mathbf{X}_{mn}$ (with $m,n \in K = \{i_1 < i_2 < \cdots < i_n\}_{1\le n\le N}$) are collected in only one term. Since the number of subsets of indices, K, is finite, this expansion is finite.

When applied to the p-h propagator, each contribution in the finite expansion can be interpreted as describing the propagation of the interaction through a given subset of electronic excitations. Since each contribution includes corrections to infinite order, collective effects are taken into account in a natural way.

For approximate schemes for the propagator, the criteria for the selection of terms are based on the type of interaction terms and the transmission paths one wishes to consider and not on the perturbative order in the electronic interaction. The results obtained with a given initial choice of contributions in the FEIM might be easily improved by adding more terms to the initial ones. Known approximation levels to the PP, such as the RPA, could be regarded as a special case of this approximation scheme.

Although the FEIM method can also be applied for numerical computation of inverse matrices, its main utility is as a theoretical tool. However, since this method is absolutely general, it can be applied to other types of propagators and perturbation-like problems where an inverse matrix is involved. Also, by comparing results obtained by including different terms in the FEIM, a powerful tool for the analysis of the transmission paths of the interaction is achieved.

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