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2009 J. Phys.: Condens. Matter 21 474204

(http://iopscience.iop.org/0953-8984/21/47/474204)

J. Phys.: Condens. Matter **21** (2009) 474204 (10pp) doi:10.1088/0953-8984/21/47/474204

Arrow diagram theory for non-orthogonal electronic groups: the continued fractions method

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Received 26 February 2009
Published 5 November 2009
Online at stacks.iop.org/JPhysCM/21/474204

Abstract

The group function theory by Tolpygo and McWeeny is a useful tool in treating quantum systems that can be represented as a set of localized electronic groups (e.g. atoms, molecules or bonds). It provides a general means of taking into account intra-correlation effects inside the groups without assuming that the interaction between the groups is weak. For non-orthogonal group functions the arrow diagram (AD) technique provides a convenient procedure for calculating matrix elements $\langle\Psi|\hat{O}|\Psi\rangle$ of arbitrary symmetrical operators $\hat{O}$ which are needed, for example, for calculating the total energy of the system or its electron density. The total wavefunction of the system $\Psi = \hat{A}\prod_I \Phi_I$ is represented as an antisymmetrized product of non-orthogonal electron group functions Φ_I of each group I in the system. However, application of the AD theory to extended (e.g. infinite) systems (such as biological molecules or crystals) is not straightforward, since the calculation of the mean value of an operator requires that each term of the diagram expansion be divided by the normalization integral $S = \langle\Psi|\Psi\rangle$ which is given by an AD expansion as well. In our previous work, we cast the mean value $\langle\Psi|\hat{O}|\Psi\rangle$ of a symmetrical operator $\hat{O}$ in the form of an AD expansion which is a linear combination of linked (connected) ADs multiplied by numerical pre-factors. To obtain the pre-factors, a method based on power series expansion with respect to overlap was developed and tested for a simple 1D Hartree–Fock (HF) ring model. In the present paper this method is first tested on a 2D HF model, and we find that the power series expansion for the pre-factors converges extremely slowly to the exact solution. Instead, we suggest another, more powerful, method based on a continued fraction expansion of the pre-factors that approaches the exact solution much faster. The method is illustrated on the calculation of the electron density for the 2D HF model. It provides a powerful technique for treating extended systems consisting of a large number of strongly localized electronic groups.

1. Introduction

The idea of localized electronic groups (EG) [1–5] has been widely applied in various areas of quantum chemistry, e.g. core and valence electrons in molecules or crystals, electrons localized on atoms or ions in atomic or ionic solids, core and bond electrons in strongly covalent materials, etc [2, 6–11]. Similar partition ideas can also be used to derive a particular embedding potential for the quantum cluster, when the electronic system of the entire system is divided into an infinite number of groups: one associated with the quantum cluster and others with the environment region surrounding the cluster [3, 12, 13]. Provided that the partition scheme applied to the given system is physically (or chemically) appropriate, one can assume that electrons belonging to the given group spend most of their time in the spatial region of this group. Therefore, to a good approximation, the wavefunction of the whole system consisting of M electron groups can be represented as an antisymmetrized product of wavefunctions $\Phi_I(X_I)$ of every individual group I:

$$\Psi(X_1, \ldots, X_M) = \hat{A}\prod_{I=1}^{M}\Phi_I(X_I). \tag{1}$$

Here $X_I = (x_1, \ldots, x_{N_I})$ is the coordinate set of N_I electrons in the Ith group. Note that the single-electron coordinates in

general include the spin index as well. The antisymmetrization operator above is defined as

$$\hat{A} = \frac{1}{N!} \sum_{P \in S_N} \epsilon_P \hat{P}, \tag{2}$$

where the sum runs over all $N!$ elements $\hat{P}$ (permutations) of the symmetry group S_N and $N = N_1 + N_2 + \cdots + N_M$ is the total number of electrons in the whole system. The factor $\epsilon_P = \pm 1$ corresponds to the parity of the permutation $\hat{P}$. It is assumed that the group functions $\Phi_I(X_I)$ are individually antisymmetrized.

In a general case when group functions $\Phi_I(X_I)$ are non-orthogonal to each other, the mean value of a symmetrical (with respect to electron permutations) operator $\hat{O} \equiv \hat{O}(x_1, \ldots, x_N)$ is defined as

$$\bar{O} = \frac{\langle \Psi | \hat{O} | \Psi \rangle}{\langle \Psi | \Psi \rangle}. \tag{3}$$

Using an explicit expression for the operator $\hat{A}$ above, the total wavefunction Ψ, normalization integral $S = \langle \Psi | \Psi \rangle$ and any matrix element $\langle \Psi | \hat{O} | \Psi \rangle$ are represented as a sum of $N!$ terms. In particular, the normalization integral becomes a sum of overlap integrals between group functions of different groups. Earlier attempts to simplify such expansions and associate each term with a diagram have been generalized in [14–16] where the arrow diagram (AD) theory was developed. Firstly, by exploiting a double coset decomposition of the symmetrical group S_N, all terms in the expansion are grouped into many distinct families; secondly, each distinct family is represented by a well-defined picture (arrow diagram) and simple rules were developed to associate an analytical expression with each AD.

When applying equation (3) to an extended system, the ratio of two AD expansions, corresponding to $\langle \Psi | \hat{O} | \Psi \rangle$ and $\langle \Psi | \Psi \rangle$, is obtained. Both expansions contain a very large (infinite for an infinite system) number of terms and thus the average $\bar{O}$ is difficult to calculate in practice. It was argued in [17] that $\bar{O}$ can be represented as a sum of all linked (connected) ADs only, $\bar{O} = \langle \Psi | \hat{O} | \Psi \rangle_c$, thereby solving the problem. However, it has been found recently [18] that this representation for the mean value is only approximate, and one has to multiply each linked diagram by a numerical pre-factor. In general, these pre-factors differ from unity, and only when the overlap between different groups is insignificant (i.e. the group functions are extremely well localized in space), then the pre-factors can be set to unity and the 'linked-AD' theorem of [17] is recovered.

To obtain the pre-factors in a general case of arbitrary overlap between the group functions, a method based on the power series expansion of the pre-factors with respect to overlap has been proposed recently [18]. The suggested approach is quite general and can be applied to any systems (including extended ones). The method was successfully applied to a simple 1D ring Hartree–Fock (HF) model. It seemed that a practical application of the AD theory to arbitrary extended systems became a not-very-distant possibility.

However, in the present paper we introduce a more general 2D HF problem and show that the pre-factors obtained by the power series expansion method converge extremely slowly to the exact solution that is obtained by a numerical calculation. We find that very many terms are required in the expansion of the pre-factors if the overlap is significant. This makes the method inconvenient and cumbersome in practice. The main purpose of this work is to suggest another, in our view, much more powerful method, based on the continued fraction expansion, to determine the pre-factors. We apply both methods to an electron density of a 2D HF lattice system, and compare their results with the well-converged numerical solutions. We show that the pre-factors obtained by the continued fraction method approach the exact values promptly as the level of the fraction is increased.

This paper is organized as follows. In the subsequent section we shall briefly review the AD theory for the reader's convenience and also introduce the necessary notations. More detailed explanations can be found elsewhere [14, 15, 17]. In section 3, we shall introduce the 2D HF lattice model and apply the method of the power series expansion to it. In section 4, we suggest an alternative method which allows presentation of the pre-factors via continued fractions. Finally, conclusions are drawn in section 5.

2. The arrow diagram theory

Let us consider a symmetric group S_{N_I}. Its elements permute electronic coordinates belonging only to the Ith group. Joining together all such groups we obtain a subgroup $S_0 = S_{N_1} \cup S_{N_2} \cup \cdots \cup S_{N_M}$ of S_N. Any element of S_0 interchanges electronic coordinates only within the groups, i.e. performs only intra-group permutations. The algebraic foundation of the AD theory [14] is based on the *double coset* (DC) decomposition of the complete symmetric group S_N with respect to S_0, which enables us to identify all inequivalent *inter-group* permutations $\hat{P}_{qT}$:

$$S_N = \frac{1}{N_0} \sum_{qT} \mu_{qT} S_0 \hat{P}_{qT} S_0. \tag{4}$$

Here the sum runs over all distinct types, q, of operations for inter-group permutations as well as all distinct ways, T, of labelling actual groups (EGs) involved in it, and $N_0 = N_1!N_2! \cdots \cdots N_M!$ is a numerical factor. $\hat{P}_{qT}$ is a DC generator, which contains only inter-group permutations and, in general, can be constructed as a product of some *primitive* cycles, each involving no more than one electron from each group.

Each cycle is represented as a directed closed loop connecting all groups involved in it. Thus, in general, each permutation $\hat{P}_{qT}$ can be drawn as an arrow diagram (AD) containing a collection of closed directed loops. Several loops may pass through the same group (depending on the number of electrons in the group involved in the permutation $\hat{P}_{qT}$). If the groups involved in the diagram cannot be separated without destroying the directed loops, the AD is called *linked* or *connected*. If, however, this separation is possible then the diagram is called *non-linked* or *disconnected* and it can be

represented as a collection of linked parts. The decomposition coefficients μ_{qT} can be calculated merely by counting arrows entering and leaving each group in the AD [14].

Using the DC decomposition of the symmetrical group S_N of permutations, the following expansion of the normalization integral is obtained [15]:

$$S = \langle \hat{A}\Phi | \hat{A}\Phi \rangle = \Lambda \sum_{qT} \epsilon_q \mu_{qT} \langle \Phi | \hat{P}_{qT} | \Phi \rangle \tag{5}$$

where $\Lambda = N_0/N!$ is a numerical pre-factor and $\Phi = \prod_I \Phi_I(X_I)$ is a product of all group functions. Equation (5) gives an expansion of S in terms of diagrams identical to that for S_N. The matrix elements $\langle \Phi | \hat{P}_{qT} | \Phi \rangle$ are represented as a product of reduced density matrices (RDMs) of electronic groups involved in the permutation $\hat{P}_{qT}$ integrated over corresponding electronic coordinates. Note that a total contribution of a non-linked AD is exactly equal to the product of contributions associated with each of its linked parts [15].

Getting rid of the common factor Λ, the expansion for S can also be written as a sum of contributions coming from different *clusters* of groups:

$$\tilde{S} = \frac{S}{\Lambda} = 1 + \sum_{K=2}^{M} \sum_{A_1 < \cdots < A_K} S_K(A_1, A_2, \ldots, A_K) \tag{6}$$

where $S_K(A_1, A_2, \ldots, A_K)$ is the contribution of *all* ADs that contain K different groups with the particular labelling $A_1, A_2, \ldots, A_K$. The unity in the equation above corresponds to the trivial permutation.

For convenience, the expansion in equation (6) will be referred to as the normalization integral expansion. In principle, in this expansion all groups of the entire system participate. It has also been proven useful to introduce a derivative object, $\tilde{S}(T)$, that is obtained from the above expansion by retaining all the ADs which are associated only with the groups from a manifold $T = \{A_1, \ldots, A_K\}$ of groups (with labels $A_1, \ldots, A_K$), where K may be either finite or infinite. If T comprises the whole system, we arrive at $\tilde{S}$ of equation (6). It is also convenient to introduce a reciprocal manifold $T_r = [T]$, i.e. an artificial system obtained by 'removing' from the entire system all groups comprising the set T [17, 18]. Then, $\tilde{S}(T_r) = \tilde{S}([T]) \equiv \tilde{S}[T]$ is obtained from equation (6) by retaining only ADs in which any of the groups belonging to T is *not* present. Obviously, $\tilde{S}$ can be considered as a particular case of $\tilde{S}[T]$ when $T \equiv \emptyset$ is empty. Occasionally, to indicate explicitly which particular groups are excluded, we shall use $\tilde{S}[A_1, \ldots, A_K]$ instead of $\tilde{S}[T]$, where $T = \{A_1, \ldots, A_K\}$.

The *unnormalized* RDM-1 of the whole system, i.e. the non-diagonal electron density:

$$\bar{\rho}(x; x') = N \int \Psi(x, x_2, \ldots, x_N) \times \Psi^*(x', x_2, \ldots, x_N) \mathrm{d}x_2 \cdots \mathrm{d}x_N \tag{7}$$

can also be written as a matrix element of a certain symmetrical one-particle operator [1, 2, 15] and thus represented via a special diagrammatic expansion [15]. The latter expansion can be constructed by considering the AD expansion for $\tilde{S}$ and then modifying each diagram by placing a small open circle, representing the variables (x, x'), in either of the three following ways: (i) on a group not involved in the diagram; (ii) on a group involved in the diagram (i.e. on the corresponding vertex), and, finally, (iii) on an arrow in the diagram. Thus, each AD in the $\tilde{S}$ expansion serves as a reference in building up the entire AD expansion for the RDM-1.

As in the case of the normalization integral, the contribution of any non-linked diagram for the RDM-1 is given by a product of the contributions of all its linked parts. Since only one open circle is used in each AD, any non-linked AD of the RDM-1 is equal to the product of the contributions from the single linked part with the circle and other non-linked parts that do not contain the circle. The latter are the same as in the reference expansion for the normalization integral. Therefore, in general, the following representation for the RDM-1 is valid [17]:

$$\tilde{\rho}(x; x') = \frac{\bar{\rho}(x; x')}{\Lambda} = \sum_{K=1}^{M} \sum_{A_1 < \ldots < A_K} \tilde{S}[A_1, \ldots, A_K] \times \sum_t \rho_K^t(A_1, \ldots, A_K \parallel x; x') \tag{8}$$

where $\rho_K^t(A_1, \ldots, A_K \parallel x; x')$ is the sum of contributions of all linked ADs with an open circle which are constructed using the particular group labelling $A_1, A_2, \ldots, A_K$. The sum over t takes account of the two positions of the open circle on the reference AD: either on its arrow (case (iii) above) or its group (case (ii)). Note that case (i) is also accounted for since it corresponds to the case when $K = 1$. Thus, each linked AD with the open circle, constructed from the set T of groups, is to be multiplied by the sum of all possible normalization integral ADs $\tilde{S}[T]$, constructed using the groups from the rest of the system $[T]$.

The *true* RDM-1 should be calculated according to equation (3) as [17]

$$\rho(x; x') = \frac{\bar{\rho}(x; x')}{\langle \Psi | \Psi \rangle} = \sum_{K=1}^{M} \sum_{A_1 < \ldots < A_K} f_{[T]} \times \sum_t \rho_K^t(A_1, \ldots, A_K \parallel x; x') \tag{9}$$

where

$$f_{[T]} = f_{[A_1, \ldots, A_K]} = \frac{\tilde{S}[A_1, \ldots, A_K]}{\tilde{S}} \tag{10}$$

are numerical pre-factors which depend only on the chosen set of groups T. Since the normalization integral $\tilde{S}$ is represented via an AD expansion, equation (6), one can see that the pre-factors are given as a ratio of two AD expansions, each containing a very large (infinite) number of terms for a large (infinite) system. Thus, the true RDM-1 is represented as a sum of all linked ADs with the open circle, $\rho_K^t(A_1, \ldots, A_K \parallel x; x')$, multiplied by numerical pre-factors, $f_{[A_1, \ldots, A_K]}$ [18]. For systems containing a small number of groups, the calculation of the pre-factors can be done explicitly since the AD expansions would contain a finite, relatively small, number of terms. However, the calculation of the pre-factors possesses the main problem in applying the AD theory to extended systems containing a large (or infinite) number of groups.

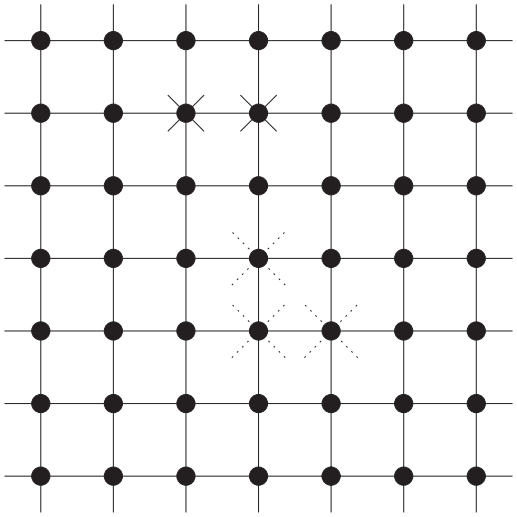

Figure 1. An extended 2D lattice of equally spaced one-electron groups. The two solid crosses indicate a *removed* pair of neighbouring groups. The dashed crosses indicate a *removed* cluster of three adjacent groups in the shape of an angle.

A power expansion method for calculating the pre-factors has been suggested recently [18]. It was successfully applied to a 1D ring HF system for which the exact analytical solution can be obtained. In the next section we shall check the applicability of this method for a more general 2D lattice HF model.

3. 2D Hartree–Fock lattice model

3.1. The model

Let us consider an infinite lattice of equally spaced one-electron groups as shown in figure 1. Each group is described by a single real s-type normalized wavefunction $\psi_i(x)$ ($i = 1, 2, \ldots$) localized around the group centre. Spin is ignored in this toy model that serves merely to help to analyse calculation of the pre-factors.

All groups are identical, i.e. every localized function $\psi_i(x)$ can be obtained by a spatial translation of other functions. Within the HF theory, the total wavefunction of the lattice is represented by a single Slater determinant $\Psi \propto \det|\psi_1(x_1)\psi_2(x_2)\cdots|$. We assume that only the nearest-neighbour functions overlap. Due to the symmetry of the model, only one parameter, i.e. the overlap integral $\sigma = \int \psi_i(x)\psi_j(x)\mathrm{d}x$ between any neighbouring group pairs, is required to characterize the system. Thus, the actual form of $\psi_i(x)$ is not needed.

3.2. Arrow diagram description of the electron density

Since the expressions for the pre-factors are the same for the diagonal ($x = x'$) and non-diagonal ($x \neq x'$) elements of the electron density (9), we shall consider only the former case for simplicity. Then, for the 2D lattice, its AD expansion has an infinite number of terms. The first few terms of the expansion are given as follows:

$$\rho(x) = \frac{\tilde{S}[\bullet]}{\tilde{S}} \cdot \circ + \frac{\tilde{S}[-]}{\tilde{S}} \cdot \diamond + \frac{\tilde{S}[\square]}{\tilde{S}} \left(\square + \square \right) + \cdots . \tag{11}$$

The first term is an open circle AD multiplied by a pre-factor $f_{[\bullet]} = \tilde{S}[\bullet]/\tilde{S}$. If the open circle AD is associated with group i, its contribution is given by $\rho_i^{(\circ)}(x) = \psi_i^2(x)$. In the pre-factor $\tilde{S}$ is the normalization integral of the total wavefunction of the lattice, while $\tilde{S}[\bullet]$ is the normalization integral for an *artificial* system in which one group (denoted by a dot $\bullet$) is *removed*. Finally, the index i should run over all groups in the lattice (corresponding to all positions of the open circle). Hence, the first term contributes $f_{[\bullet]} \sum_i \rho_i^{(\circ)}(x)$ to the density $\rho(x)$.

The second term consists of a bubble diagram with an open circle times a common pre-factor $f_{[-]} = \tilde{S}[-]/\tilde{S}$. The bubble diagram contributes $\rho_{\{i,j\}}^{(\mathrm{bubble})}(x) = -\sigma\psi_i(x)\psi_j(x)$ provided that the open circle is located on an arrow connecting two *nearest*-neighbour groups i and j; otherwise, $\rho_{\{i,j\}}^{(\mathrm{bubble})}(x) = 0$. Then, $\tilde{S}[-]$ is the normalization integral for an *artificial* system in which the group pair $\{i, j\}$ (denoted by a dash) is *removed*. Thus, the total contribution of the second term in equation (11) becomes $2f_{[-]} \sum_{\{i,j\}} \rho_{\{i,j\}}^{(\mathrm{bubble})}(x)$, where we sum over all pairs of nearest groups, corresponding to all possible positions of the open circle on the bubble ADs. The factor of two is due to that each bubble diagram has two possible positions of the open circle.

Similar explanations apply to other terms. Each term consists of a sum of polygon diagrams with all possible positions of the open circle times the corresponding common pre-factor $f_{[T]} = \tilde{S}[T]/\tilde{S}$. Each diagram with the circle appears twice due to two possible directions of the arrows. T is a *'cluster'* of electronic groups involved in the polygon diagram. The cluster is visualized as a polygon with every vertex corresponding to the lattice site. $\tilde{S}[T]$ is the normalization integral for the system in which all groups comprising the cluster are removed. Due to the fact that there is only overlap between nearest neighbours in our model, adjacent vertices in the polygon must correspond to the nearest neighbours. Thus, the removed groups forming the cluster are the nearest groups. Note that for simplicity only two diagrams are shown in the third term in equation (11) inside the brackets corresponding to opposite directions of the arrows; in reality, however, each diagram is equal to the sum of four diagrams with four possible positions of the open circle (for the given labelling of the vertices).

Owing to the translational symmetry of the 2D lattice model, the pre-factors do not depend on the particular choice of the removed cluster (e.g. its orientation), only on its *shape*. Two simplest clusters, a nearest pair (denoted by a dash in equation (11)) and an 'angle' consisting of three nearest groups (to be denoted as $\square$), are shown in figure 1 as examples. The rest of the system will therefore be given in these two cases as $[-]$ and $[\square]$, respectively.

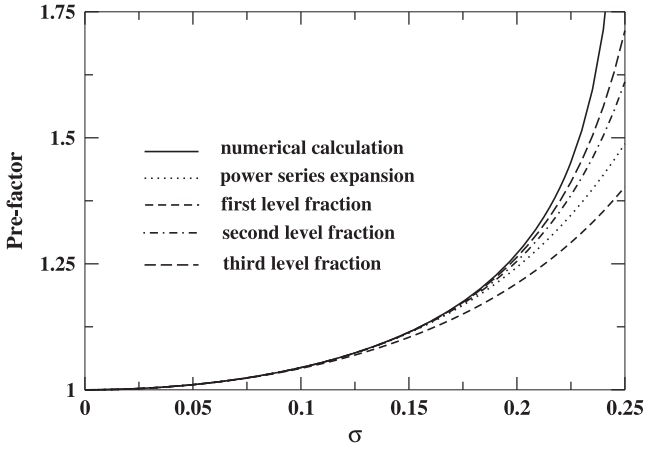

Figure 2. The first pre-factor $f_{[\bullet]} = \tilde{S}[\bullet]/\tilde{S}$ in equation (11) calculated numerically (solid line), using a power series expansion up to the sixth order (dotted) and the continued fractions of up to the first (dashed), second (dotted–dashed) and the third (long dashed) level approximations.

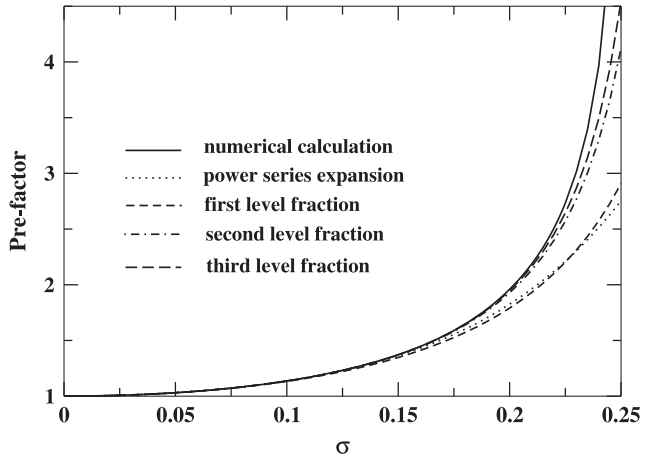

Figure 4. The third pre-factor in equation (11) calculated numerically (solid line), using a power series expansion up to the sixth order (dotted) and the continued fractions of up to the first (dashed), second (dotted–dashed) and the third (long dashed) level approximations.

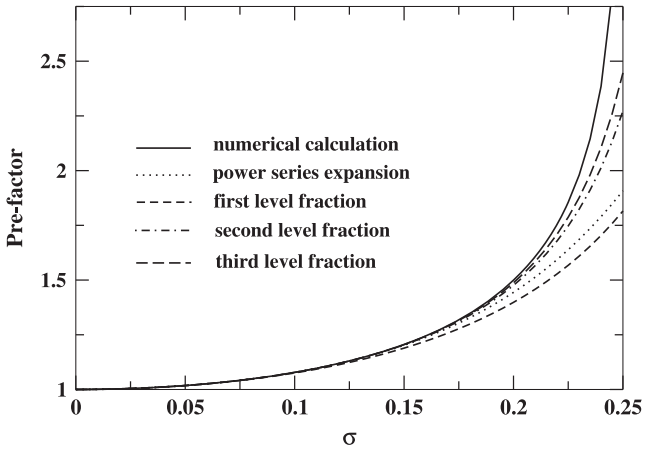

Figure 3. The second pre-factor $f_{[-]} = \tilde{S}[-]/\tilde{S}$ in equation (11) calculated numerically (solid line), using a power series expansion up to the sixth order (dotted) and the continued fractions of up to the first (dashed), second (dotted–dashed) and the third (long dashed) level approximations.

3.3. Numerical solution

We have already mentioned that the pre-factors $f_{[T]}$ only depend on the topology (picture) of the cluster T of groups, not their particular position in the infinite lattice. Any pre-factor is given as a ratio of two normalization integrals $\tilde{S}[T]$ and $\tilde{S}$, one for the system $[T]$ and another for the whole system. Within the HF theory, both normalization integrals can actually be calculated analytically for the given overlap σ. Indeed, $\tilde{S}$ in the HF theory can be recast in the form of a determinant of the overlap matrix $\mathbf{M}_0 = \|\langle\psi_i|\psi_j\rangle\|$ [1]. This can be calculated, employing the periodic symmetry, in the **k** space as a product of all the eigenvalues of the overlap matrix $\mathbf{M}_0$. For an *artificial* system in which a set of neighbouring groups $T = \{A_1, A_2, \ldots, A_K\}$ is *removed*, the normalization integral $\tilde{S}[T]$ can also be presented as a determinant of a matrix **M** that can be obtained from $\mathbf{M}_0$ by replacing some of its nearest-neighbour overlaps with zero. Since **M** differs from $\mathbf{M}_0$ by a defect matrix $\mathbf{D} = \mathbf{M} - \mathbf{M}_0$ of a finite rank, its determinant can also be calculated exactly via $\mathbf{M}^{-1} = (\mathbf{1} + \mathbf{M}_0^{-1}\mathbf{D})^{-1}\mathbf{M}_0^{-1}$. This calculation is similar to the one used in the resolvent theory of defects in crystals (see, e.g., [19]).

Although the route outlined above is in principle feasible, in this paper we have used a simpler method based on a numerical calculation of the necessary determinants for a finite square system containing $K \times K$ lattice sites. Then $\tilde{S}$ is calculated as a determinant of the matrix $\mathbf{M}_0$ for the system with all sites present, while $\tilde{S}[T]$ can be calculated in a similar way by taking away the necessary sites in the centre of the square system. To calculate the pre-factors for the first few terms in the expansion of the density (11), small clusters T containing not more than six nearest removed sites are to be considered only. Therefore, systems of relatively modest sizes are only needed. We find in our calculations that the 21×21 lattice system is sufficient to simulate the extended 2D lattice.

The numerical solutions for the first three pre-factors are shown in figures 2–4 by solid lines. These can be considered as exact since a further increase of the size of the square lattice system does not give any noticeable change for the pre-factors. One can see that the pre-factors are some smooth functions of σ. For small overlap (well-localized group functions) the pre-factors are close to unity. With the increase of the overlap between the neighbouring group functions, the pre-factors quickly grow, shooting to infinity at $\sigma = \frac{1}{4}$. This particular behaviour of the pre-factors is the consequence of our model in which only the nearest-neighbour overlap is accounted for. A similar effect was also observed in the case of the 1D ring model in our previous work [18].

Another interesting observation which one can make is that the pre-factors become generally larger with the increase of the number of removed sites, i.e. one can notice that for any σ we have

$$f_{[\bullet]} < f_{[-]} < f_{[\square]} < \cdots. \tag{12}$$

3.4. Method of power series expansion

In general, each pre-factor is a rational function of overlap σ, since it is a ratio of two polynomials, and thus is analytical about $\sigma = 0$. Hence we can expand pre-factors in a power series with respect to σ as suggested in [18]. Then we can determine the expansion coefficients using special *diagram equations* that can be formulated according to the spatial structure of the system and as described below.

The idea of the method is based on a representation of the normalization integral, $\tilde{S}[T]$, of the system $[T]$ (where $T = \{A_1, A_2, \ldots, A_K\}$ comprises a set of groups removed from the initial system) via a series of terms containing ascending numbers of connections of a chosen single group $A \in [T]$ with its neighbours:

$$\tilde{S}[T] = \tilde{S}[T + A] + \sum_{T_1 \subset [T+A]} \tilde{D}(AT_1)\tilde{S}[T + A + T_1]. \quad (13)$$

The first term on the right-hand side contains all the ADs that can be constructed out of the groups of the set $[T + A]$, i.e. the group A is not involved in either of them. The sum in the second term contains all the ADs which are formed by the group A and any groups from the rest of the system $[T + A]$. This sum picks out various manifolds $T_1 \subset [T + A]$ containing one, two, three, etc, groups. Then, a sum of all possible ADs constructed using A and *every* group from the manifold T_1 is denoted $\tilde{D}(AT_1)$. Note that ADs in $\tilde{D}(AT_1)$ may contain non-linked diagrams as well. Then, every AD in $\tilde{D}(AT_1)$ is multiplied by all possible ADs formed out of the rest of the groups, $\tilde{S}[T + A + T_1]$, i.e. the latter is obtained by all ADs left after 'removing' the set T_1 from the set $[T + A]$.

Let us now apply the above equation to the 2D lattice model for which the exact solution is known from the numerical calculation as explained in the previous subsection. We start by choosing $T = \emptyset$ (empty) and A being an arbitrary group (say, group 1). Then, we obtain from equation (13)

$$\tilde{S} = \tilde{S}[\bullet] - 4\sigma^2\tilde{S}[-] - 8\sigma^4\tilde{S}[\square] + \cdots. \quad (14)$$

Here the first term consists of all ADs, $\tilde{S}[\bullet]$, based on groups from the rest of the system, $[T + A] \equiv [\bullet]$, which is an *artificial* lattice with the chosen group 1 (denoted by the dot) *removed*. The second term in the series is constructed of all bubble ADs (each contributing $-\sigma^2$) between group 1 and all its four neighbours times the sum of all normalization integral ADs, $\tilde{S}[-]$, corresponding to an *artificial* lattice in which a pair of neighbouring groups (denoted by a *dash*) is *removed*. The third term in the series is based on all square ADs (each contributing $-2\sigma^4$, the factor of two corresponding to two possible directions of the loops) connecting group 1 to its neighbouring three groups times all possible normalization integral ADs, $\tilde{S}[\square]$, in which groups from the rest of the system are involved. There are four squares possible having group 1 at one of its corners connected to three nearest neighbours, which accounts for an additional factor of four. Similarly other terms in the series can be constructed. They consist of N-vertex polygons in which each side connects nearest neighbours on the lattice. The sites of the polygons will be shown graphically by a picture without particular labelling of the sites since, as was mentioned previously, the result depends only on the *shape* of the corresponding clusters of sites, not their actual position in the lattice.

Dividing both sides of equation (14) by $\tilde{S}$, we obtain the corresponding equation for the pre-factors:

$$1 = f_{[\bullet]} - 4\sigma^2 f_{[-]} - 8\sigma^4 f_{[\square]} + \cdots. \quad (15)$$

Repeating the above procedure for the system $T = \bullet$ and a group A being the nearest neighbour of the group 1 (that is, of the dot), we similarly obtain

$$f_{[\bullet]} = f_{[-]} - 2\sigma^2 f_{[\_\_]} - \sigma^2 f_{[-\bullet-]} - 4\sigma^4 f_{[\square\_]} + \cdots, \quad (16)$$

where pictures in the square brackets denote the removed clusters, i.e. the sets $T + A + T_1$ in equation (13).

Continuing this procedure, one can derive an infinite series of equations in which every time a new set of pre-factors appear. For instance

$$f_{[-]} = f_{[\_\_]} - \sigma^2 f_{[\_\_]} - \sigma^2 f_{[\_\_]} - \sigma^2 f_{[\square]} + \cdots \quad (17)$$

$$f_{[-]} = f_{[-\bullet-]} - 2\sigma^2 f_{[\_\_]} - \sigma^2 f_{[-\bullet-\bullet-]} + \cdots \quad (18)$$

$$f_{[\_\_]} = f_{[\square]} - 2\sigma^2 f_{[\square\_]} + \cdots. \quad (19)$$

Next, we expand every pre-factor in a power series with respect to σ:

$$f_{[T]} = 1 + \sum_{n=1}^{\infty} C_n[T]\sigma^n \quad (20)$$

where T denotes the removed cluster. Note that the expansion starts from unity for any $[T]$ since $f_{[T]} = 1$ for zero overlap ($\sigma = 0$). Substituting these expansions into the above diagram equations, one can recursively determine the expansion coefficients up to a certain order by comparing terms with the same power of σ. For instance, it follows from equation (15) that $C_1[\bullet] = 0$, $C_2[\bullet] = 4$, $C_3[\bullet] = 4C_1[-]$, $C_4[\bullet] = 4C_2[-] + 8$, etc. Similarly from equation (16) we obtain $C_1[-] = C_1[\bullet] = 0$, $C_2[-] = C_2[\bullet] + 2 + 1 = 7$, etc. Combining these relations, new coefficients can be obtained, e.g. $C_3[\bullet] = 0$, $C_4[\bullet] = 36$, and so on.

Equations (17)–(19) enable us to calculate more coefficients. Following this line of reasoning, we obtain the power series expansions up to sixth order for the first three pre-factors in equation (11):

$$f_{[\bullet]} = 1 + 4\sigma^2 + 36\sigma^4 + 400\sigma^6 + \cdots \quad (21)$$

$$f_{[-]} = 1 + 7\sigma^2 + 70\sigma^4 + 807\sigma^6 + \cdots \quad (22)$$

$$f_{[\square]} = 1 + 12\sigma^2 + 144\sigma^4 + 1804\sigma^6 + \cdots. \quad (23)$$

To obtain higher-order terms in the series expansions by this method, one has to consider more diagram equations which bring more pre-factors in.

The series expansions obtained above are compared with the exact numerical solutions in figures 2–4, respectively. One can see that, in the cases of small and intermediate overlap, reasonable approximations to the exact solutions for the pre-

factors are obtained by this method. It is also obvious that one has to go to higher orders in the series expansions when the overlap is much larger. Indeed, for large values of σ, pre-factors obtained by the power series expansion method converge extremely slowly to the exact solution. Therefore, the method becomes impractical in this case as more terms in the expansions are to be derived. In general, this calculation is extremely cumbersome.

In section 4, another method will be suggested for the calculation of the pre-factors based on continued fractions.

4. Method of continued fractions

4.1. General idea of the method

Our starting point is the following general expression for the normalization integral ADs written for the system $[T]$ from which some of the groups, comprising the manifold T, have been *removed* [18]:

$$\tilde{S}[T] = \tilde{S}(\Delta T)\tilde{S}[T+\Delta T] + \sum_{T_1\subset\Delta T}\sum_{T_2\subset[T+\Delta T]}\tilde{D}(T_1T_2) \times \tilde{S}(\Delta T - T_1)\tilde{S}[T+\Delta T+T_2]. \quad (24)$$

Note that equation (13) is a particular case of this equation. Here $\Delta T \subset [T]$ is some arbitrary set of groups from the system $[T]$ and $\tilde{S}(\Delta T)$ denotes normalization integral ADs for the group set ΔT, including the trivial term (unity). Thus the first term in the above equation corresponds to all ADs that can be made separately from the sets ΔT and $[T+\Delta T]$. In the next term on the right-hand side we sum over all possible sets T_1 and T_2 of groups, one taken from ΔT and another from $[T+\Delta T]$. Thus, all ADs in the second term are constructed using groups from *both* sets ΔT and $[T+\Delta T]$. The latter ADs are presented explicitly as a product (i.e. via non-linked ADs) of simpler ADs. Namely, the *'interaction'* term $\tilde{D}(T_1T_2)$ represents the sum of all possible ADs which can be constructed using *every* group from T_1 and T_2. Note that $\tilde{D}(T_1T_2)$ may contain non-linked ADs as well, in which case each part of it must contain at least one group from T_1 and one from T_2. Other terms have similar meaning: $\tilde{S}(\Delta T - T_1)$ contains all ADs constructed using groups from the set $\Delta T - T_1$ obtained by taking away (removing) groups of the set T_1 from the set ΔT, while $\tilde{S}[T+\Delta T+T_2]$ contains all ADs due to groups from the system $[T+\Delta T+T_2]$. The expression above is valid for any choice of ΔT.

According to the definition of $\tilde{D}(T_1T_2)$, its lowest order with respect to overlap is equal to the number of groups $n_{T_1+T_2}$ in the combined set T_1+T_2. Since two other factors $\tilde{S}(\Delta T-T_1)$ and $\tilde{S}[T+\Delta T+T_2]$ contain all ADs including unity, each term in the sum in equation (24) starts from terms that are of the order $n_{T_1+T_2}$ with respect to overlap. Effectively, equation (24) results in an expansion of $\tilde{S}[T]$ with respect to overlap as in section 3.4.

However, instead of following the series expansion route, we can divide both sides of equation (24) by $\tilde{S}[T]$ and then solve it for the fraction $\tilde{S}[T+\Delta T]/\tilde{S}[T]$ as shown below:

$$f_{[T]}^{[T+\Delta T]} \equiv \frac{\tilde{S}[T+\Delta T]}{\tilde{S}[T]} = \Big[\tilde{S}(\Delta T) + \sum_{T_1\subset\Delta T}\tilde{S}(\Delta T - T_1)\sum_{T_2\subset[T+\Delta T]}\tilde{D}(T_1T_2)f_{[T+\Delta T]}^{[T+\Delta T+T_2]}\Big]^{-1}. \quad (25)$$

In the left-hand side we have a ratio of two normalization integral AD expansions corresponding to the manifolds $[T+\Delta T]$ and $[T]$ that differ by a set of groups ΔT. On the right-hand side we have fractions corresponding to different manifolds $[T+\Delta T+T_2]$ and $[T+\Delta T]$, that differ by a set of groups T_2. In the second sum on the right-hand side of the above equation we take account of all possible choices of the set $T_2 \subset [T+\Delta T]$, and the summation starts from a single group.

Recall that the expression for $\tilde{D}(T_1T_2)$ scales with respect to overlap at least as the number of groups in the set T_1+T_2. The fraction $f_{[T+\Delta T]}^{[T+\Delta T+T_2]}$ is some well-defined function of the overlap that is equal to unity in the case of zero overlap. Therefore, the sum over T_2 above can be considered as an expansion in terms of the overlap, and only several first terms can normally be retained since the group functions are required to be well localized, e.g.

$$\sum_{T_2\subset[T+\Delta T]}\tilde{D}(T_1T_2)f_{[T+\Delta T]}^{[T+\Delta T+T_2]} = \sum_{B_1\subset[T+\Delta T]}\tilde{D}(T_1B_1)f_{[T+\Delta T]}^{[T+\Delta T+B_1]} + \sum_{\{B_1,B_2\}\subset[T+\Delta T]}\tilde{D}(T_1B_1B_2)f_{[T+\Delta T]}^{[T+\Delta T+B_1+B_2]} + \sum_{\{B_1,B_2,B_3\}\subset[T+\Delta T]}\tilde{D}(T_1B_1B_2B_3)f_{[T+\Delta T]}^{[T+\Delta T+B_1+B_2+B_3]} + \cdots. \quad (26)$$

Here B_1, B_2, etc, are groups from the system $[T+\Delta T]$, and the notations like $\{B_1, B_2\}$, $\{B_1, B_2, B_3\}$, etc, correspond to summing over all non-equivalent pairs, triplets, etc.

Expressions similar to equation (25) can now be written for each of the fractions that appear on the right-hand side of equation (25) by setting $T+\Delta T \to T$ and $T_2 \to \Delta T$ on the left-hand side of it. Note that in this case a smaller system, $[T+\Delta T]$, than initially (which was $[T]$) is considered. Substituting the expressions for the fractions $f_{[T+\Delta T]}^{[T+\Delta T+T_2]}$ back into the right-hand side of the original equation (25), we obtain an expression for the original fraction, $f_{[T]}^{[T+\Delta T]}$, at the second level. If the process is repeated n times, it leads to a *continued fraction* of a quite complicated structure with n levels. To terminate this process, one can set up all the fractions at the last level to unity. We shall return to this point later on. Note only that a better approximation can be achieved if the last level fractions are expanded in a power series with respect to the overlap up to a certain order.

When it is clear how the continued fraction can be constructed, we can turn our attention to calculating the pre-factors. Indeed, the required expressions for the pre-factors $f_{[\Delta T]}$ are obtained by simply choosing $T = \emptyset$ (empty) in the

initial expression (i.e. at the *first* level) for the fraction and then replacing $\Delta T \to T$:

$$f_{[T]} \equiv f_{[\emptyset]}^{[T]} = \Big[\tilde{S}(T) + \sum_{T_1 \subset T} \tilde{S}(T - T_1) \times \sum_{T_2 \subset [T]} \tilde{D}(T_1 T_2) f_{[T]}^{[T+T_2]}\Big]^{-1}. \quad (27)$$

The first-level approximation for the pre-factor $f_{[T]}$ is obtained if only a finite number of terms are considered in the T_2 sum above corresponding to a certain order with respect to overlap, and all fractions on the right-hand side, $f_{[T]}^{[T+T_2]}$, are set to unity. At the second level, one uses equation (25) to represent the fractions $f_{[T]}^{[T+T_2]}$. Again, only some finite numbers of terms are retained in the corresponding T_2 sum. This process is continued until the required number of levels is achieved. At the last level all fractions are set to unity.

Let us analyse the obtained continued fraction expression for the pre-factor. In $f_{[T]} = \tilde{S}[T]/\tilde{S}$ two sets of groups are used: the whole system and the system $[T]$ obtained by removing all groups of the set T from it. We know from equation (12) that the ratios like $f_{[T]}$ differ more from unity if the difference in the two sets is greater. The continued fraction expansion allows one to re-express the pre-factor in question via fractions $f_{[R']}^{[R]} = \tilde{S}[R]/\tilde{S}[R']$ that contain *less difference* between the two sets R and R'. Since the sum over T_2 in equations (25) and (27) is terminated at sets T_2 that have a rather small number of groups, as equation (26) illustrates, the ratios $f_{[R']}^{[R]}$ will contain close sets R and R' even in the case when the initial set T in $f_{[T]}$ is large. By going to deeper levels while constructing the continued fraction, the sets R and R' become smaller and smaller, i.e. more and more groups are removed from the initial system. The small difference between the two sets ensures that setting the corresponding ratios $f_{[R']}^{[R]}$ to unity would be a good approximation. This general reasoning justifies the method we suggest to terminate the fraction by setting all such ratios at the last level simply to unity. A better approximation can be obtained if the last level fractions are expanded in a power series up to a certain finite order as was mentioned earlier.

As an example, let us work out explicitly an expression for $f_{[T]}$ up to the third level keeping terms at least of the third order with respect to overlap. At the first level we use the equation (27)

$$f_{[T]} = \Big\{\tilde{S}(T) + \sum_{A_1 \in T} \tilde{S}(T - A_1)\Big[\sum_{B_1 \in [T]} \tilde{D}(A_1 B_1) f_{[T]}^{[TB_1]} + \sum_{\{B_1, B_2\} \in [T]} \tilde{D}(A_1 B_1 B_2) f_{[T]}^{[TB_1B_2]} + \cdots\Big] + \sum_{\{A_1, A_2\} \in T} \tilde{S}(T - A_1 - A_2) \times \Big[\sum_{B_1 \in [T]} \tilde{D}(A_1 A_2 B_1) f_{[T]}^{[TB_1]} + \cdots\Big] + \ldots\Big\}^{-1} \quad (28)$$

where $\tilde{D}(A_1 B_1)$, $\tilde{D}(A_1 A_2 B_1)$, etc, are sums of ADs starting from the second and third orders with respect to overlap, respectively. We have also used a shorter notation for the *combined* sets such as $T + A + B \equiv TAB$.

It is seen from the above expression, written only at the first level, that we need now the corresponding expressions for the following two ratios, namely $f_{[T]}^{[TA]}$ and $f_{[T]}^{[TAB]}$, where $A, B \in [T]$. These can be worked out explicitly from equation (25), and we obtain

$$f_{[T]}^{[TA]} = \Big\{1 + \sum_{B_1 \in [TA]} \tilde{D}(AB_1) f_{[TA]}^{[TAB_1]} + \sum_{\{B_1, B_2\} \in [TA]} \tilde{D}(AB_1B_2) f_{[TA]}^{[TAB_1B_2]} + \cdots\Big\}^{-1} \quad (29)$$

$$f_{[T]}^{[TAB]} = \Big\{\tilde{S}(AB) + \sum_{C_1 \in [TAB]} [\tilde{D}(AC_1) + \tilde{D}(BC_1) + \tilde{D}(ABC_1)] f_{[TAB]}^{[TABC_1]} + \sum_{\{C_1, C_2\} \in [TAB]} [\tilde{D}(AC_1C_2) + \tilde{D}(BC_1C_2) + \tilde{D}(ABC_1C_2)] f_{[TAB]}^{[TABC_1C_2]} + \cdots\Big\}^{-1} \quad (30)$$

where $\tilde{S}(AB)$ contains all ADs (including the trivial one, i.e. the unity) between two groups A and B. If we wish to terminate the continued fraction at the third level, we still need expressions for the ratios in equations (29) and (30). These can all be obtained similarly to the ones given above. In fact, in all the ratios we have encountered above, namely $f_{[TA]}^{[TAB_1]}$, $f_{[TA]}^{[TAB_1B_2]}$, $f_{[TAB]}^{[TABC_1]}$ and $f_{[TAB]}^{[TABC_1C_2]}$, the two sets in the super- and subscripts differ by either one or two groups only, so that the same expressions (29) and (30) can formally be used again. The only difference is that at this level of the continued fraction we set all the ratios on the right-hand side to unity:

$$f_{[TA]}^{[TAB_1]} = \Big\{1 + \sum_{C_1 \in [TAB_1]} \tilde{D}(B_1C_1) + \sum_{\{C_1, C_2\} \in [TAB_1]} \tilde{D}(B_1C_1C_2) + \cdots\Big\}^{-1} \quad (31)$$

$$f_{[TAB]}^{[TABC_1]} = \Big\{1 + \sum_{D_1 \in [TABC_1]} \tilde{D}(C_1D_1) + \sum_{\{D_1, D_2\} \in [TABC_1]} \tilde{D}(C_1D_1D_2) + \cdots\Big\}^{-1}. \quad (32)$$

The corresponding expressions for the ratios $f_{[TA]}^{[TAB_1B_2]}$ and $f_{[TAB]}^{[TABC_1C_2]}$ are obtained similarly from equation (30). It is clearly seen that for any manifold T the ratios in the continued fraction expression do not differ by more than two groups in this approximation. In order to improve the approximation for the pre-factor $f_{[T]}$, one has to retain the corresponding ratios at the third level and work out the corresponding expressions for them. In addition, one has to keep more terms in the AD expression with respect to the overlap at each level which will result in more fractions appearing at each level. Generally, the AD expansion at each level may be terminated earlier as deeper levels are considered.

In section 4.2 we shall illustrate the continued fraction method on the 2D lattice model.

4.2. Continued fractions for the 2D lattice model

Let us consider the 2D lattice model. To illustrate how the continued fraction method works, we shall obtain a two-level fraction for $f_{[\bullet]}$ and a one-level fraction for $f_{[-]}$.

For the 2D lattice model with only nearest-neighbour overlap, the first three *possible* ADs in $\tilde{D}(T_1T_2)$ (restricted by the numbers of groups in sets T_1 and T_2), are: a bubble AD, a twin-bubble AD (non-linked) and a square AD, which contribute $-\sigma^2$, σ^4 and $-2\sigma^4$, respectively.

Noting that $\tilde{S}(\emptyset) = \tilde{S}(\bullet) = 1$, we start from $T = \bullet$ in equation (27) to obtain

$$f_{[\bullet]} = \{1 - 4\sigma^2\Gamma[-] - 8\sigma^4\Gamma[\square] + \cdots\}^{-1}. \tag{33}$$

where $\Gamma[\text{cluster}] = f^{[\text{cluster}]}_{[\bullet]}$. In fact, this equation is the same as equation (15), but written differently. If we assume that every ratio in the denominator of equation (33) equals unity and only terms up to fourth order with respect to overlap are retained, then the following one-level fraction approximation for $f_{[\bullet]}$ is obtained:

$$f_{[\bullet]} \simeq \frac{1}{1 - 4\sigma^2 - 8\sigma^4}.$$

Next, using equation (25), we can write the two fractions in the denominator of equation (33) in more detail as

$$\Gamma[-] = \{1 - 2\sigma^2\gamma_1[\,\square\,] - \sigma^2\gamma_1[\bullet\!-\!\bullet] - 4\sigma^4\gamma_1[\square\square] + \cdots\}^{-1}. \tag{34}$$

with $\gamma_1[\text{cluster}] = f^{[\text{cluster}]}_{[-]}$ and

$$\begin{aligned}\Gamma[\square] = \{&(1 - 2\sigma^2) - 4\sigma^2(1 - \sigma^2)\gamma_2[\square\,] - 2\sigma^2\gamma_2[\square\,] \\ &+ \sigma^4(2\gamma_2[\square\,] + 2\gamma_2[\square\square] + 2\gamma_2[\square\square] \\ &+ 4\gamma_2[\square] + 2\gamma_2[\square]) - 2\sigma^4(\gamma_2[\square] + 2\gamma_2[\square\square]) \\ &- 4\sigma^4(1 - \sigma^2)\gamma_2[\square] + \cdots\}^{-1}.\end{aligned} \tag{35}$$

with $\gamma_2[\text{cluster}] = f^{[\text{cluster}]}_{[\square]}$. Note that equation (34) can also be obtained directly from equation (16). After substituting equations (34) and (35) into equation (33), keeping only terms up to the fourth order with respect to overlap and assuming that $\gamma_1[\text{cluster}] = \gamma_2[\text{cluster}] = 1$, we obtain the following two-level fraction approximation for $f_{[\bullet]}$:

$$f_{[\bullet]} \simeq \frac{1}{1 - \frac{4\sigma^2}{1-3\sigma^2-4\sigma^4} - \frac{8\sigma^4}{1-8\sigma^2+6\sigma^4}}.$$

This process can be continued.

Similarly one can consider other pre-factors. For instance, the first level expression for the second pre-factor, $f_{[-]}$, is obtained from equation (27) by considering T to be two nearest lattice sites (i.e. a 'dash' in our cluster notation):

$$\begin{aligned}f_{[-]} = \{&(1 - \sigma^2) - 4\sigma^2\gamma_1[\,\square\,] - 2\sigma^2\gamma_1[\bullet\!-\!\bullet] \\ &+ \sigma^4(2\gamma_1[\square] + 2\gamma_1[\square] + 4\gamma_1[\square] + \gamma_1[\bullet\!-\!\bullet\!-\!\bullet]) \\ &- 2\sigma^4(4\gamma_1[\square\,] + 2\gamma_1[\square]) + \cdots.\}^{-1}\end{aligned} \tag{36}$$

where $\gamma_1[\text{cluster}]$ was introduced earlier in this subsection. Assuming $\gamma_1[\text{cluster}] = 1$ and keeping only terms up to fourth order with respect to overlap, we obtain the following one-level fraction approximation for $f_{[-]}$:

$$f_{[-]} \simeq \frac{1}{1 - 7\sigma^2 - 3\sigma^4}.$$

Following the scheme outlined in section 4.1, we have worked out explicit expressions for the first three pre-factors appearing in equation (11) up to the third-level fraction approximation. We shall not give here the derived expressions in detail as they are rather cumbersome. Using these expressions, the three pre-factors have been calculated as functions of the overlap integral σ.

The results of these calculations are shown in figures 2–4. Firstly, we see that for all three pre-factors the first-level fraction approximation performs only slightly worse than the power series expansion calculated up to sixth order. However, already the second-level fractions give a much better approximation than the power series expansion. Secondly, higher-level fractions give better results than the lower-level fractions. Finally, already the third-level fraction formulae give a reasonable approximation to the numerical solutions for all overlaps except near the singularity. Since the singularity in the pre-factors is an artefact of our nearest-neighbour approximation, we believe that the discrepancy would be much smaller if the overlap between more distant neighbours is also accounted for.

5. Conclusions

In this paper we have analysed the power series expansion method for the pre-factors in the AD expansion for the reduced density matrices (RDM) suggested in our previous paper [18]. By considering a 2D Hartree–Fock lattice model with the nearest-neighbour overlap, we find that the power series expansion converges to the correct result (obtained using a numerical calculation) very slowly, especially in the region of significant overlap. Although a better approximation can always be constructed employing more terms in the expansion, the calculation becomes very tedious and thus impractical.

Instead, we suggest a different method based on the continued fraction expansion for the pre-factors. Explicit expressions have been worked out for this method up to the third level of the fractions.

Applying this method to the same 2D lattice model, we demonstrate that the new method converges to the correct result much faster for any, even significant, overlap. The main reason why this method works better is this. A numerical calculation shows that the fraction $f^{[T_1]}_{[T_2]}$, equation (25), differs less from unity if the *smaller* manifold, $[T_1] = [T_2 + \Delta T]$, is closer to the other one, $[T_2]$. The more significant is overlap, the more terms are needed in the AD expansion of the ratio $f^{[T_1]}_{[T_2]}$. The whole point of representing the pre-factors $f_{[T]} \equiv f^{[T]}_{[\emptyset]}$ via a continued fraction is that it is expressed via ratios $f^{[T_1]}_{[T_2]}$ in which the manifolds $[T_1]$ and $[T_2]$ are closer to each other. As a result, if one goes to a sufficiently high level fraction, the AD expansion for the ratios at the final level may converge very quickly with the overlap, so that fewer terms will be required. In fact, in the above consideration, we just used the first term in the expansion of the ratios at the final level at which the fraction

is terminated, i.e. $f_{[T_2]}^{[T_1]} \simeq 1$, and found for our 2D lattice model that already at the second level the continued fractions converge better to the correct result for the first three pre-factors than their sixth-order power series expansions. Note that a similar consideration can also be applied to the much simpler 1D ring model considered in [18]. It can then be shown analytically, without doing any numerical calculations, that the continued fraction converges precisely to the exact result at any value of the overlap.

We believe that the proposed method of continued fractions is a practical tool in developing approximate expressions for the required pre-factors in the AD expansions of arbitrary matrix elements of symmetric operators (like density matrices) in many-body quantum chemistry. The formalism developed here can be used for developing corrections to the corresponding formulae developed previously in [17] where all the pre-factors were assumed to be equal to unity. One of the promising applications of the AD theory that is being looked at in our laboratory is the theory of cluster embedding.

Acknowledgments

YW acknowledges financial support from the China Scholarship Council and the K C Wong Education Foundation, Hong Kong.

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