High-Order Coupled Cluster Method (CCM) Formalism 1: Ground- and Excited-State Properties of Lattice Quantum Spin Systems with $s \geq \frac{1}{2}$

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(Dated: September 11, 2009)

Abstract

The coupled cluster method (CCM) is a powerful and widely applied technique of modern-day quantum many-body theory. It has been used with great success in order to understand the properties of quantum magnets at zero temperature. This is due largely to the application of computational techniques that allow the method to be applied to high orders of approximation using localised approximation schemes, e.g., such as the LSUBm scheme. In this article, the high-order CCM formalism for the ground and excited states of quantum magnetic systems are extended to those with spin quantum number $s \geq \frac{1}{2}$. Solution strategies for the ket- and bra-state equations are also considered. Aspects of extrapolation of CCM expectation values are discussed and future topics regarding extrapolations are presented.
I. INTRODUCTION

The coupled cluster method (CCM) is a well-known method of quantum many-body theory (QMBT). The CCM has been applied with much success over the last fifteen or so years in order to study quantum magnetic systems at zero temperature (see Refs. ). In particular, the use of computer-algebraic implementations of the CCM for quantum systems of large or infinite numbers of particles has largely been found to be very effective with respect to these spin-lattice problems. This approach uses localised approximation schemes, such as the LSUBm approximation. For the LSUBm scheme, the extent of the locale over which multi-spin correlations are explicitly included in the approximation is defined by the index m. The ground- and excited-state expectation values are often extrapolated in the limit \( m \to \infty \). In this article we focus on the development of new high-order CCM formalism for the ground and excited states of lattice quantum spin systems with spin quantum number \( s \geq \frac{1}{2} \). The solution the ket- and bra-state equations is also considered. Various aspects of the extrapolation of CCM expectation values are considered and future topics regarding extrapolations are described. The high-order CCCM code is freely available online.

II. CCM GROUND-STATE FORMALISM

The ket and bra ground-state energy eigenvectors, \( |\Psi\rangle \) and \( \langle \tilde{\Psi}| \), of a general many-body system described by a Hamiltonian \( H \), are given by

\[
|\Psi\rangle = e^{S} |\Phi\rangle \quad \text{and} \quad \langle \tilde{\Psi}| = \langle \Phi| e^{-S} \tilde{S}.
\]

Furthermore, the ket and bra states are parametrised within the single-reference CCM as follows:

\[
|\Psi\rangle = e^{S} |\Phi\rangle \quad \text{and} \quad \langle \tilde{\Psi}| \tilde{S} e^{-S} |\Phi\rangle = 0, \quad \forall I \neq 0.
\]

It may be proven from Eqs. and in a straightforward manner that the ket- and bra-state equations are thus given by

\[
\langle \Phi| C_I^- e^{-S} H e^S |\Phi\rangle = 0, \quad \forall I \neq 0.
\]


\[ \langle \Phi | S \tilde{e}^{-S}[H, C_I^+] e^S | \Phi \rangle = 0, \quad \forall I \neq 0. \quad (4) \]

The index \( I \) refers to a particular choice of cluster from the set of \( (N_F) \) fundamental clusters that are distinct under the symmetries of the crystallographic lattice and the Hamiltonian and for a given approximation scheme at a given level of approximation. We note that these equations are equivalent to the minimization of the expectation value of \( \tilde{H} = \langle \tilde{\Psi} | H | \Psi \rangle \) with respect to the the CCM bra- and ket-state correlation coefficients \( \{ \tilde{S}_I, S_I \} \). We note that Eq. (3) is equivalent to \( \delta \tilde{H} / \delta \tilde{S}_I = 0 \), whereas Eq. (4) is equivalent to \( \delta \tilde{H} / \delta S_I = 0 \). Furthermore, we note that Eq. (3) leads directly to simple form for the ground-state energy given by

\[ E_g = E_g(\{ S_I \}) = \langle \Phi | e^{-S} H e^S | \Phi \rangle. \quad (5) \]

The full set \( \{ S_I, \tilde{S}_I \} \) provides a complete description of the ground state. For instance, an arbitrary operator \( A \) will have a ground-state expectation value given as

\[ \tilde{A} \equiv \langle \tilde{\Psi} | A | \Psi \rangle = \langle \Phi | \tilde{S} e^{-S} A e^S | \Phi \rangle = \tilde{A} \left( \{ S_I, \tilde{S}_I \} \right). \quad (6) \]

The similarity transform of \( A \) is given by,

\[ \tilde{A} \equiv e^{-S} A e^S = A + [A, S] + \frac{1}{2!}[[A, S], S] + \cdots. \quad (7) \]

Finally, we remark that the CCM provides exact results in the limit of inclusion of all possible clusters in \( S \) and \( \tilde{S} \). However, this problem is often impossible to solve in a practical sense. Hence, we generally make approximations in both \( S \) and \( \tilde{S} \). The three most commonly employed approximation schemes previously utilised have been: (1) the SUBn scheme, in which all correlations involving only \( n \) or fewer spins are retained, but no further restriction is made concerning their spatial separation on the lattice; (2) the SUBn-m sub-approximation, in which all SUBn correlations spanning a range of no more than \( m \) adjacent lattice sites are retained; and (3) the localised LSUBm scheme, in which all multi-spin correlations over all distinct locales on the lattice defined by \( m \) or fewer contiguous sites are retained.

III. HIGH-ORDER GROUND-STATE OPERATORS AND COMMUTATIONS

We begin the treatment of high-order CCM by introducing the ket-state correlation operator given, as usual, by

\[ S = \sum_l \sum_{i_1, \ldots, i_l} S_{i_1, \ldots, i_l} s_{i_1}^+ \cdots s_{i_l}^+. \quad (8) \]
However, it is important point to note that each of the indices \{i_1, i_2, \ldots, i_l\} runs over all lattice sites. Furthermore, we assume that there are (l!) orderings of these indices (even for \( s > \frac{1}{2} \)), although we never need to work out these factors explicitly in practice. The index \( I \) corresponds to one of the choices of \{\( i_1, \ldots, i_l \)\} for the fundamental set of configurations. We may now write a set of high-order CCM ket-state operators, given by

\[
\begin{align*}
F_k &\equiv \sum_i \sum_{i_2, \ldots, i_l} lS_{k, i_2, \ldots, i_l} s_{i_2}^+ \cdots s_{i_l}^+ \\
G_{km} &\equiv \sum_{l>1} \sum_{i_3, \ldots, i_l} l(l-1)S_{k, m, i_3, \ldots, i_l} s_{i_3}^+ \cdots s_{i_l}^+ \\
M_{knn} &\equiv \sum_{l>2} \sum_{i_4, \ldots, i_l} l(l-1)(l-2)S_{k, m, n, i_4, \ldots, i_l} s_{i_4}^+ \cdots s_{i_l}^+ \\
N_{kmnp} &\equiv \sum_{l>3} \sum_{i_5, \ldots, i_l} l(l-1)(l-2)(l-3)S_{k, m, n, p, i_5, \ldots, i_l} s_{i_5}^+ \cdots s_{i_l}^+. 
\end{align*}
\]

The indices \( k, m, n, \) and \( p \) depend on those sums in the Hamiltonian or of another given operator. We note that \( s^\pm = s^x \pm is^y \), \( [s^x, s^\pm] = \pm s^\pm \), and \( [s^-, s^+] = -2s^z \). Hence, the following commutation relations may be proven:

\[
\begin{align*}
[s^z_k, S] &= F_k s^+_k, \\
[s^-_k, S] &= -2F_k s^z_k - G_{kk} s^+_k, \\
[s^z_k, F_m] &= G_{km} s^+_k, \\
[s^z_k, G_{mn}] &= M_{kmn} s^+_k, \\
[s^z_k, F^2_m] &= 2F_m G_{km} s^+_k, \\
[s^-_k, F_m] &= -2G_{km} s^z_k - M_{kkm} s^+_k, \\
[s^-_k, F^2_m] &= -2G_{km}^2 s^z_k - 2F_m M_{kkm} s^+_k - 4F_m G_{km} s^z_k, \\
[s^z_k, M_{mnp}] &= N_{kmnp} s^+_k, \\
[s^-_k, G_{mn}] &= -2M_{kmn} s^z_k - N_{kkmn} s^+_k.
\end{align*}
\]

We may now write the similarity-transformed expressions of the single-spin operators \( s^\alpha ; \alpha \equiv \{+, -, z\} \), as

\[
\begin{align*}
e^{-S} s^+_k e^S &\equiv \tilde{s}^+_k = s^+_k \\
e^{-S} s^z_k e^S &\equiv \tilde{s}^z_k = s^z_k + F_k s^+_k \\
e^{-S} s^-_k e^S &\equiv \tilde{s}^-_k = s^-_k - 2F_k s^z_k - G_{kk} s^+_k - F^2_k s^+_k.
\end{align*}
\]

We see that there is a repeated index in \( G_{kk} \) in the similarity transformed version of \( s^- \). Clearly, this term contributes only for systems with spin quantum number \( s > \frac{1}{2} \).
IV. DERIVING AND SOLVING THE CCM GROUND-STATE EQUATIONS

We now wish to determine and solve the CCM ket-state equations, where the $I$-th such equation is given by

$$E_I \equiv \langle \Phi | C_I^\dagger e^{-S} He^S | \Phi \rangle = 0 \quad \forall I \neq 0 \quad .$$  \hfill (12)

(Note that we assume that $\langle \Phi | C_I^\dagger C_I^\dagger | \Phi \rangle = 1$ in the above equation). Specific terms in the Hamiltonian are now explicitly written in terms of the high-order CCM operators as:

TERM 1 : $\tilde{s}_i^+ \tilde{s}_j^+$ = $s_i^+ s_j^+ + F_j s_j^+ s_i^+ + F_i s_i^+ s_j^-$ + $G_{ij} s_i^+ s_j^+$ + $F_i F_j s_i^+ s_j^+$

TERM 2 : $\tilde{s}_i^+ s_j^+$ = $s_i^+ s_j^+$ + $F_i s_i^+ s_j^+$

TERM 3 : $\tilde{s}_i^+ s_j^+$ = $-2F_j s_i^+ s_j^+ - 2G_{ij} s_i^+ s_j^+ - G_{jj} s_j^+ s_i^- - M_{ij} s_i^+ s_j^+ - 2F_i F_j s_i^+ s_j^+$

TERM 4 : $\tilde{s}_i^+ s_j^+$ = $s_i^+ s_j^+ + F_j s_i^+ s_j^+$

TERM 5 : $\tilde{s}_i^- s_j^-$ = $-2F_j s_i^+ s_j^- - G_{jj} s_j^+ s_i^- - G_{ij} s_i^+ s_j^- - M_{ij} s_i^+ s_j^+ - 2F_i F_j s_i^+ s_j^-$

TERM 6 : $\tilde{s}_i^- s_j^-$ = $-2F_j s_i^+ s_j^- - G_{jj} s_j^+ s_i^- - F_j^2 s_i^+ s_j^-$

TERM 7 : $\tilde{s}_i^- s_j^-$ = $-2F_i s_j^- s_i^- - G_{ii} s_i^+ s_j^- - F_i^2 s_i^+ s_j^-$

TERM 8 : $\tilde{s}_i^+ s_j^+$ = $s_i^+ s_j^+$

TERM 9 : $\tilde{s}_i^- s_j^+$ = $4G_{ij} s_i^+ s_j^+ + 2M_{ii} s_i^+ s_j^+ + 2M_{jj} s_j^+ s_i^+ + N_{ii} s_i^+ s_j^+ + N_{jj} s_j^+ s_i^+$

TERM 10 : $\tilde{s}_i^+ = s_i^+ + F_i s_i^+$

TERM 11 : $\tilde{s}_i^- = -2F_i s_i^+ - G_{ii} s_i^+ - (F_i)^2 s_i^+$

TERM 12 : $(\tilde{s}_i^+)^2 = (s_i^+)^2 + 2F_i s_i^+ s_i^- + G_{ii}(s_i^+)^2 + F_i(s_i^+)^2 + F_i^2(s_i^+)^2$

TERM 13 : $\tilde{s}_i^+ = s_i^+$

(Note that $s^- | \Phi \rangle = 0$ is implicitly assumed in Eq. \hfill (13) above.) We now “pattern-match” the $C_i^-$ operators to those the relevant terms in the Hamiltonian from Eq. \hfill (13) above in order to form the CCM equations $E_I = 0$ of Eq. \hfill (12) at a given level of approximation.
We now define the following new set of CCM bra-state correlation coefficients given by $x_I \equiv S_I$ and $\tilde{x}_I \equiv N_B/N(!)\nu_I\tilde{S}_I$ and we assume again that $\langle \Phi|C_I^-C_I^+|\Phi \rangle = 1$. Note that $N_B$ is the number of Bravais lattice sites. Note also that for a given cluster $I$ then $\nu_I$ is a symmetry factor which is dependent purely on the point-group symmetries (and not the translational symmetries) of the crystallographic lattice and that $l$ is the number of spin operators. We note that the factors $\nu_I$, $N$, $N_B$, and $(!)$ never need to be explicitly determined. The CCM bra-state operator may thus be rewritten as

$$\tilde{S} \equiv 1 + N \sum_{I=1}^{N_F} \tilde{x}_I C_I^-,$$

such that we have a particularly simple form for $\tilde{H}$, given by

$$\tilde{H} = N \sum_{I=0}^{N_F} \tilde{x}_I E_I,$$

where $\tilde{x}_0 = 1$. We note that the $E_0$ is defined by $E_0 = \frac{1}{N}\langle \Phi|e^{-S}He^S|\Phi \rangle$ (and, thus, $E_0 = \frac{1}{N}E_g$) and that $E_I$ is the $I$-th CCM ket-state equation defined by Eq. (12). The CCM ket-state equations are easily re-derived by taking the partial derivative of $\tilde{H}/N$ with respect to $\tilde{x}_I$, where

$$\frac{\delta(\tilde{H}/N)}{\delta \tilde{x}_I}(\equiv 0) = E_I.$$

We now take the partial derivative of $\tilde{H}/N$ with respect to $x_I$ such that the bra-state equations take on a particularly simple form, given by

$$\frac{\delta(\tilde{H}/N)}{\delta x_I} = \frac{\delta E_0}{\delta x_I} + \sum_{J=1}^{N_F} \tilde{x}_J \frac{\delta E_J}{\delta x_I}(\equiv 0) = \tilde{E}_I.$$

The coupled non-linear equations for the ket state $E_I = 0$ are solved readily, e.g, by using the Newton-Raphson method, in order to find the coefficients $\{x_I\}$. By contrast, the equations for the bra state $\tilde{E}_I = 0$ are easily solved via LU decomposition, although this may only be carried out once the CCM ket-state equations have been determined and solved. The numerical values of the coefficients $\{\tilde{x}_I\}$ may thus be obtained. We note that this approach greatly simplifies the task of determining the bra-state equations because we infer the bra-state equations directly from those of the ket-state equations via Eq. (17). Thus, we never need to evaluate Eq. (4) explicitly.

We may also solve the ket- and bra-state equations (i.e., $E_I = 0$ and $\tilde{E}_I = 0$, respectively) via direct iteration. For the case of the ket-state equations this is slightly more complicated.
because there are non-linear terms with respect the ket-state correlation coefficients \( \{ x_I \} \). We rearrange the ket-state equations such that the linear terms for \( x_I \) for the \( i \)th CCM ket-state equation are on the left of the new equation and all other terms are on the right. The right-hand side of this new equation is denoted by \( E'_I \) after dividing through by the factor on the left-hand-side for the ket-state correlation coefficients. We may carry out exactly the same procedure for the bra-state in order to find \( \tilde{E}'_I \), although the problem is linear with respect to \( \{ \tilde{x}_I \} \) in this case. These equations are thus rewritten conveniently for the ket state as

\[
x_I = E'_I(x_1, x_2, \ldots, x_{I-1}, x_{I+1}, \ldots, x_{N_F}, x^2_1, x^2_2, \ldots, x^4_1, \ldots, x^4_{N_F}) ,
\]

and for the bra state as,

\[
\tilde{x}_I = \tilde{E}'_I(\tilde{x}_1, \tilde{x}_2, \ldots, \tilde{x}_{I-1}, \tilde{x}_{I+1}, \ldots, \tilde{x}_{N_F} ; x_1, x_2, \ldots, x_{N_F}, x^2_1, x^2_2, \ldots, x^3_1, \ldots, x^3_{N_F}) .
\]

Clearly, these equations may be solved for \( x_I \) and \( \tilde{x}_I \) by iterating them “directly” until convergence. Indeed, the local memory usage is vastly reduced because we do not need to store any Jacobian or other large matrix that scales in size with \( N_F^2 \). This simple “brute force” approach of direct iteration has actually been found to be surprisingly successful. However, in practice, it needs to be implemented for large numbers of CPUs used in parallel for very large numbers of clusters (e.g., \( 10^6 \)) used in \( S \) and \( \tilde{S} \). Clearly, more sophisticated solvers for the bra- and ket-state equations that do not demand the memory requirements of Newton-Raphson for the ket state and LU decomposition for the bra state and are quicker than direct iteration may be implemented. However, this remains a task for the future.

V. THE EXCITED-STATE FORMALISM

We now consider how the excited state may be treated using the CCM via a high-order approach. We begin by remarking that the excited-state wave function is given by

\[
| \Psi_e \rangle = X^e e^S | \Phi \rangle .
\]

The Schrödinger equation, \( E_e | \Psi_e \rangle = H | \Psi_e \rangle \) and the equivalent equation for the ground state lead (after some simple algebra) to

\[
\epsilon_e X^e | \Phi \rangle = e^{-S[H, X^e]} e^S | \Phi \rangle (\equiv \hat{R} | \Phi \rangle) ,
\]

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\[
\epsilon_e X^e | \Phi \rangle = e^{-S[H, X^e]} e^S | \Phi \rangle (\equiv \hat{R} | \Phi \rangle) ,
\]

\[
(21)
\]
where $\epsilon_e \equiv E_e - E_g$ is the excitation energy. We note that the excited-state correlation operator is written as,

$$X^e = \sum_{I \neq 0} \lambda^e_I C^+_I ,$$

Equation (22) implies the overlap relation

$$\langle \Phi | \Psi_e \rangle = \langle \Phi | X^e e^S | \Phi \rangle \Rightarrow \langle \Phi | \Psi_e \rangle = 0 .$$

We may now form the basic equations for the excited state, given by

$$\epsilon_e \lambda^e_I = \langle \Phi | C_I^e e^{-S}[H, X^e]e^S | \Phi \rangle , \forall I \neq 0 ,$$

which is a generalized set of eigenvalue equations with eigenvalues $\epsilon_e$ and corresponding eigenvectors $\lambda^e_I$. We note that the choice of clusters for the excited-state may be different from those for the ground state. For example, the ground state for the Heisenberg model on bipartite lattices is in the subspace $s^z = 0$, whereas the excited state has $s^z = +1$. The number of excited-state “fundamental” clusters that are distinct under the translational and point-group symmetries of the lattice and Hamiltonian is given by $N_f$.

VI. HIGH-ORDER EXCITED-STATE OPERATORS AND COMMUTATIONS

In a similar manner as for the ground-state, we now define excited state operator via

$$X^e = \sum_{I} \sum_{i_1, \ldots, i_l} \lambda^e_{i_1, \ldots, i_l} s^+_{i_1} \cdots s^+_{i_l}$$

where the indices $\{i_1, \ldots, i_l\}$ run over all lattice sites. We assume explicitly again that there are $(l!)$ orderings of the indices (even for $s > \frac{1}{2}$). The index $I$ corresponds to one of the choices of $\{i_1, \ldots, i_l\}$ for the fundamental set of configurations for the excited state, such that $\lambda^e_I \equiv \lambda^e_{i_1, \ldots, i_l}$. We now also define the further high-order operators for the excited state, given by

$$P_k = \sum_{I} \sum_{i_1, \ldots, i_l} \lambda^e_{i_1, \ldots, i_l} s^+_{i_1} \cdots s^+_{i_l}$$

$$Q_{km} = \sum_{l>1} \sum_{i_1, \ldots, i_l} l(l-1) \lambda^e_{i_1, \ldots, i_l} s^+_{i_1} \cdots s^+_{i_l}$$

$$R_{kmn} = \sum_{l>2} \sum_{i_1, \ldots, i_l} l(l-1)(l-2) \lambda^e_{i_1, \ldots, i_l} s^+_{i_1} \cdots s^+_{i_l}$$

$$T_{kmnp} = \sum_{l>3} \sum_{i_1, \ldots, i_l} l(l-1)(l-2)(l-3) \lambda^e_{i_1, \ldots, i_l} s^+_{i_1} \cdots s^+_{i_l}$$

(26)
The following commutation relations may also be proven:

\[
\begin{align*}
[s^+_k, X^e] &= P_k s^+_k , \\
[s^-_k, X^e] &= -2P_k s^-_k - Q_{kk} s^+_k , \\
[s^+_k, P_m] &= Q_{km} s^+_k , \\
[s^+_k, Q_{mn}] &= R_{km} s^+_k , \\
[s^+_k, P^2_m] &= 2P_m Q_{km} s^+_k , \\
[s^-_k, P_m] &= -2Q_{km} s^-_k - R_{km} s^+_k , \\
[s^-_k, P^2_m] &= -2Q_{km} s^-_k - 2P_m R_{km} s^+_k - 4P_m Q_{km} s^+_k , \\
[s^+_k, R_{mn}] &= T_{kmn} s^+_k , \\
[s^-_k, Q_{mn}] &= -2R_{kmn} s^-_k - T_{kmn} s^+_k .
\end{align*}
\]

(27)

VII. DERIVING AND SOLVING THE EXCITED STATE EQUATIONS

We now wish to determine and solve the CCM excited-state equations given by Eq. (24)
Specific terms in the Hamiltonian are now explicitly written in terms of the new excited-state
high-order CCM operators as:

TERM 1 : \( e^{-S}[s^+_i s^+_j, X^e] e^S = P_i s^+_i s^+_j + P_i F_i s^+_i s^+_j + P_j s^+_j s^+_i + P_j F_i s^+_i s^+_j + Q_{ij} s^+_i s^+_j \)

TERM 2 : \( e^{-S}[s^+_i s^+_j, X^e] e^S = P_i s^+_i s^+_j \)

TERM 3 : \( e^{-S}[s^+_i s^-_j, X^e] e^S = -2P_i F_i s^+_i s^-_j - P_i G_{ij} s^+_i s^-_j - P_i F^2_i s^+_i s^-_j - 2P_i s^+_i s^-_j - 2P_i F_i s^-_i s^+_j \\
-2P_j F_j s^+_j s^-_i - 2P_j G_{ij} s^+_j s^-_i - 2P_j F_j s^-_j s^+_i - 2Q_{ij} s^+_i s^-_j \\
-2Q_{ij} F_j s^+_i s^-_j - Q_{jj} s^-_j s^+_i - Q_{ij} F_i s^+_i s^-_j - R_{ij} s^+_i s^-_j \)

TERM 4 : \( e^{-S}[s^+_i s^-_j, X^e] e^S = P_j s^+_j s^-_i \)

TERM 5 : \( e^{-S}[s^+_i s^-_j, X^e] e^S = -2P_i F_i s^+_i s^-_j - P_i G_{ij} s^+_i s^-_j - P_i F^2_i s^+_i s^-_j - 2P_i s^+_i s^-_j - 2P_i F_j s^-_j s^+_i \\
-2P_j F_j s^+_j s^-_i - 2P_j G_{ij} s^+_j s^-_i - 2P_j F_j s^-_j s^+_i - 2Q_{ij} s^+_j s^-_i \\
-2Q_{ij} F_i s^+_j s^-_j - Q_{ii} s^-_i s^+_j - Q_{ij} F_i s^+_i s^-_j - R_{ij} s^+_i s^-_j \)

TERM 6 : \( e^{-S}[s^+_i s^-_j, X^e] e^S = -2P_j s^+_j s^-_i - Q_{jj} s^+_i s^-_j - 2P_j F_i s^+_i s^-_j \)

TERM 7 : \( e^{-S}[s^+_i s^-_j, X^e] e^S = -2P_i s^+_j s^-_i - Q_{ii} s^+_i s^-_j - 2P_i F_i s^+_i s^-_j \)

TERM 8 : \( e^{-S}[s^+_i s^-_j, X^e] e^S = 0 \)

TERM 9 : \( e^{-S}[s^+_i s^-_j, X^e] e^S = 4P_i F_j s^+_i s^-_j + 4P_i G_{ij} s^+_i s^-_j + 2P_i G_{jj} s^+_j s^-_i + 2P_i M_{jj} s^+_i s^-_j \\
+4P_i F_j s^+_i s^-_j + 4P_i F_j G_{ij} s^+_i s^-_j + 2P_i F_i G_{jj} s^+_i s^-_j + 2P_i F_j F^2_i s^+_i s^-_j \)
+2P_iF_i^2s_i^+s_i^z + 2Q_{ii}F_is_i^+s_j^z + Q_{ii}G_{jj}s_j^+s_j^z + Q_{ii}F_j^2s_i^+s_j^z \\
+4Q_{ij}F_is_j^+s_j^z + 4Q_{ij}F_is_j^+s_j^z + 4Q_{ij}G_{ij}s_j^+s_j^z + 4Q_{ij}G_{ij}s_j^+s_j^z \\
+4Q_{ij}F_jF_is_i^+s_j^z + 2R_{iij}F_is_j^+s_j^z + 2R_{iij}F_jF_is_i^+s_j^z + 2R_{iij}F_jF_is_i^+s_j^z \\
+2R_{iij}F_iF_is_j^+s_j^z + 2R_{iij}F_iF_jF_is_j^+s_j^z + 4P_jG_{ij}s_j^+s_i^z + 2P_jM_{ijj}s_j^+s_j^z \\
+4P_jF_is_j^+s_i^z + 4P_jF_iF_jF_is_j^+s_i^z + 2P_jG_{ij}s_j^+s_j^z + 2P_jG_{ij}s_j^+s_j^z \\
+2P_jF_i^2F_is_i^+s_i^z + 2P_jF_i^2F_jF_is_i^+s_i^z + 4P_jF_iG_{ij}s_i^+s_j^z + 2Q_{jjj}F_is_j^+s_j^z \\
+Q_{jjj}G_{ij}s_j^+s_j^z + Q_{jjj}F_i^2s_j^+s_j^z + e^{−S}[s_i^z, X^e]e^S = P_is_i^z \\
-2P_is_i^z - Q_{ii}s_i^+ - 2P_iF_is_i^+ \\
2P_iF_is_i^+s_i^z + Q_{ii}(s_i^+)^2 + P_i(s_i^+)^2 + 2P_iF_i(s_i^+)^2 \\
= e^{−S}[s_i^z, X^e]e^S = 0 \tag{28}

(Nota e s^−|\Phi\rangle = 0 is again implicitly assumed in Eq. \tag{28} above.) Again, now “pattern-match” the \( C^−_f \) operators (this time with respect to the fundamental set of the clusters in the excited state) to those the relevant terms in the Hamiltonian from Eq. \tag{24} above in order to form the CCM excited-state equations at a given level of approximation. By contrast to the case for the ground state, we see that the high-order operators of Eq. \tag{26} are in linear in those terms in Eq. \tag{28}. We choose the eigenvalue of lowest value to be our result, and this method was found to provide good results in regions of the parameter space for which the model state was a good choice. Again we note that we have formed an eigenvalue problem, which is readily solved using a standard eigenvalue solver. However, the computational problem thus formed uses local memory that scales with the number of fundamental clusters used in the excited state, i.e., as \( N^2_f \).

The eigenvalue equations of Eq. \tag{24} may be iterated directly in order to solve them. We denote the matrix for the eigenvalue problem of Eq. \tag{24} by \( B \) and we denote the eigenvectors by \( y = (\mathcal{X}_i^e, \cdots, \mathcal{X}_{N_f}^e)^T \). Hence, we iterate directly the eigenvalue equation given by

\[ By = \lambda y \ . \tag{29} \]

This is just the well-known “power iteration” method and the ratios of \( \mathcal{X}_i^e \) in successive iterations yields the relevant eigenvalue. However, the eigenvalue determined in this manner is the eigenvalue of largest magnitude, \( \lambda_{\text{MAX}} \), rather than the lowest (generally the one of
smallest magnitude $\lambda_{\text{MIN}}$ for our purposes) that we wish to obtain here. Thus, we use find the eigenvalue of smallest magnitude by using the “shifted” power iteration method. Once $\lambda_{\text{MAX}}$ has been found, we then solve the following eigenvalue equation by direct iteration:

$$ (B - \lambda_{\text{MAX}}I)y' = \lambda' y' . $$

This process ought to converge to an eigenvalue $\lambda' = \lambda_{\text{MIN}} - \lambda_{\text{MAX}}$. Indeed, this was found to be the case for those spin models for which the model state was a “good choice”. Indeed, the lowest-valued eigenvalue obtained in this manner agreed perfectly with those results for the eigenvalue of lowest values obtained via a complete diagonalization of the matrix eigenvalue problem at every level of approximation.

VIII. EXTRAPOLATION OF EXPECTATION VALUES

In practice, we often need to extrapolate individual $L_{SB}m$ or $SU_{m^m}$ (etc.) expectation values $y(m)$ in the limit $m \to \infty$. Indeed, the extrapolation of CCM expectation values is pivotal to its practical use for quantum magnetic systems. However, there are many aspects of such extrapolations that we still do not understand and that have not been tested. Furthermore, the extrapolation of CCM expectation values poses two distinct problems: a) no provable exact rules of extrapolation are known (unlike, e.g., finite-size exact diagonalisations); and, b) we have only small numbers of data points (i.e., 7 or 8 at most for 1D systems). Hence, until now, only heuristic or “ad hoc” schemes have been used. These heuristic schemes are “parametric” in the sense that the data is fitted to specific functions (e.g., polynomials) that are defined prior to fitting the data and where the coefficients of these functions are the basic “parameters.” However, one should note that the results of such parametric/ad hoc extrapolation procedures have been shown to yield consistently valid results for an extremely wide range of quantum spin systems, see, e.g., Refs. [22, 26, 32]. Indeed, the use of such schemes has been shown to provide improved results in all cases where the model state might even be suspected to be a reasonable starting point. This is a very strong vindication of the use of these simple extrapolation procedures.

We now turn to specific instances of such extrapolation schemes, and we start by noting that a common scheme for extrapolating the ground-state energy is given by

$$ y(m)_{\text{SCHEME 1}} = a_0 + a_1 m^{-2} + b_2 m^{-4} . $$

(31)
Thus far, least-squares fits of the data to this scaling rule have been performed, where the extrapolated value \( y(m \to \infty)_{\text{SCHEME 1}} \) is given by \( a_1 \). For other expectation values, such as the sublattice magnetisation and excitation energy gap, a common scheme is given by

\[
y(m)_{\text{SCHEME 2}} = b_0 + b_1 m^{-1} + b_2 m^{-2}.
\] (32)

In this case, the extrapolated value \( y(m \to \infty)_{\text{SCHEME 2}} \) is given by \( b_0 \). Finally, another common scheme is given by

\[
y(m)_{\text{SCHEME 3}} = c_0 + c_1 m^{c_2}.
\] (33)

The extrapolated value \( y(m \to \infty)_{\text{SCHEME 3}} \) is given by \( c_0 \). Other schemes have included Padé approximants and similar schemes to Eqs. (31) and (32) for fixed, though non-integer, exponents have been used. For example, the sublattice magnetisation in Refs. 45–48 was found to scale as: \( y(m) = d_0 + d_1 m^{-0.5} + d_2 m^{-1.5} \).

In future, we would ideally like to establish exact rules of scaling of expectation values with approximation level and/or go to much higher orders of approximation so that we have more data points to extrapolate. Evidence for exact rules of scaling is supported by the fact that the ground-state energy of the Heisenberg model on bipartite lattices such as the linear chain and square lattice appear to follow the scheme of Eq. (31) quite well. By contrast, their magnetisations and excitation energy gaps appear to follow the scheme of Eq. (32). Few other general rules seem to exist and certainly no such rules have, as yet, been proven mathematically. Furthermore, we are must use computationally intensive methods in parallel to solve for levels of approximation currently available. Further increases in approximation level might be possible in future due to computational improvements, although we might still be restricted to fairly small numbers of data points for the current approximation schemes. Hence, both of these goals may be not be achieveable in practice. In the absence of either of these goals, one might wish to extrapolate using a variety of extrapolation schemes in order to determine (in broad terms only) the amount of their mutual agreement. Furthermore, odd and even series of (e.g., \( \text{LSUB} m \text{ expectation values} \)) ought to extrapolate to the same value (see Ref. 50). Again, this might yield a rough idea of errors of extrapolation.

Future research might also concentrate on the establishment of new approximation schemes that do not scale as quickly as \( \text{LSUB} m \text{ and SUB} m-m \text{ schemes} \) and yet still appear scalable with approximation level. However, it is likely that the number of fundamental
clusters in any such scheme will still increase exponentially with approximation level. It would be beneficial to carry out a fully “statistical” exploration of the topic of parametric extrapolations such as those of Eqs. (31) to (33) based on only small numbers of data points. This might yield better extrapolations, but perhaps more importantly it would hope to establish with more mathematical rigour the estimate of the error in the extrapolated values. Until now, the extrapolated figures have generally been presented with no estimated degree of error. Furthermore, this “statistical” approach might also include an exploration of, e.g., least-squares, weighted least-square and maximum likelihood methods fitting methods of the parametric scaling laws to the data and the effect that this would have on the extrapolated results. Indeed, hitherto, only ad hoc/parametric models or rules of the types shown in Eqs. (31) to (33) have been used to carry out extrapolation of CCM data using least-squared methods. Finally, a outline of “best practice” for extrapolating CCM data (i.e., which extrapolation schemes and methods to use and exactly what to report) for given types of expectation values might prove very useful. These topics remain for future study however.

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