

High-Order Coupled Cluster Calculations for the Ground- and Excited-State Properties of Lattice Quantum Spin Systems: High-Order Formalism; Solution of Equations; and Parallelisation

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Abstract

Please feel free to use this L^AT_EXdocument in any publications for high-order CCM that you wish to write. I have checked the equations, but please note that there may be a few errors remaining – especially relating to definitions of bra- and ket-state coefficients and expectation values. Expansions of spin operators in terms of the high-order operators ought to be correct. Please let me know if you see any errors!

I. 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

$$H|\Psi\rangle = E_g|\Psi\rangle ; \quad \langle\tilde{\Psi}|H = E_g\langle\tilde{\Psi}| . \quad (1)$$

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

$$\begin{aligned} |\Psi\rangle &= e^S|\Phi\rangle \quad ; \quad S = \sum_{I \neq 0} \mathcal{S}_I C_I^+ , \\ \langle\tilde{\Psi}| &= \langle\Phi|\tilde{S}e^{-S} \quad ; \quad \tilde{S} = 1 + \sum_{I \neq 0} \tilde{\mathcal{S}}_I C_I^- . \end{aligned} \quad (2)$$

It may be proven from Eqs. (1) and (2) in a straightforward manner that the ket- and bra-state equations are thus given by

$$\langle\Phi|C_I^- e^{-S} H e^S |\Phi\rangle = 0, \quad \forall I \neq 0 ; \quad (3)$$

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

We note that these equations are equivalent to the minimization of the expectation value of $\bar{H} = \langle\tilde{\Psi}|H|\Psi\rangle$ with respect to the the CCM bra- and ket-state correlation coefficients $\{\tilde{\mathcal{S}}_I, \mathcal{S}_I\}$. We note that Eq. (3) is equivalent to $\delta\bar{H}/\delta\tilde{\mathcal{S}}_I$, whereas Eq. (4) is equivalent to $\delta\bar{H}/\delta\mathcal{S}_I$. Furthermore, we note that Eq. (3) leads directly to simple form for the ground-state energy given by

$$E_g = E_g(\{\mathcal{S}_I\}) = \langle\Phi|e^{-S} H e^S |\Phi\rangle . \quad (5)$$

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

$$\bar{A} \equiv \langle\tilde{\Psi}|A|\Psi\rangle = \langle\Phi|\tilde{S}e^{-S} A e^S |\Phi\rangle = \bar{A}(\{\mathcal{S}_I, \tilde{\mathcal{S}}_I\}) . \quad (6)$$

The similarity transform of A is given by,

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

II. 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

$$S = \sum_l \sum_{i_1, \dots, i_l} \mathcal{S}_{i_1, \dots, i_l} s_{i_1}^+ \cdots s_{i_l}^+ \quad (8)$$

However, it is important point to note that each of the indices $\{i_1, i_2, \dots, i_l\}$ runs over *all* lattice sites, and that there are $(l!)$ orderings of these indices. Thus, we now introduce explicitly a single ordering to the $\{i_1, i_2, \dots, i_l\}$ that we shall indicate by the symbol $[i_1, i_2, \dots, i_l]$. We do this so that we never need to determine explicitly the factors of type $(l!)$ in the later “pattern-matching” exercise used in order to find the CCM equations. Thus, we rewrite the CCM ket-state correlation coefficient \mathcal{S}_I as $x_I = (l!)\mathcal{S}_I$, such that

$$S = \sum_l \sum_{[i_1, \dots, i_l]} x_{i_1, \dots, i_l} s_{i_1}^+ \cdots s_{i_l}^+ \quad (9)$$

We may now write a set of high-order CCM ket-state operators, given by

$$\left. \begin{aligned} F_k &\equiv \sum_l \sum_{[i_2, \dots, i_l]} x_{k, i_2, \dots, i_l} s_{i_2}^+ \cdots s_{i_l}^+ \\ G_{km} &\equiv \sum_{l>1} \sum_{[i_3, \dots, i_l]} x_{k, m, i_3, \dots, i_l} s_{i_3}^+ \cdots s_{i_l}^+ \\ M_{kmn} &\equiv \sum_{l>2} \sum_{[i_4, \dots, i_l]} x_{k, m, n, i_4, \dots, i_l} s_{i_4}^+ \cdots s_{i_l}^+ \\ N_{kmnp} &\equiv \sum_{l>3} \sum_{[i_5, \dots, i_l]} x_{k, m, n, p, i_5, \dots, i_l} s_{i_5}^+ \cdots s_{i_l}^+ \end{aligned} \right\} \quad (10)$$

Again, we note that this approach means that we *never* need to determine explicitly the factors $(l!)$ in the high-order operators, which actually was the case in earlier versions of the high-order CCM code. In practice, this speeds up the CCM code greatly. The indices k , m , n , and p depend on sums in the Hamiltonian, and so (at least) one of these indices might possibly still run over all lattice sites. Furthermore, the following commutation relations

may be proven:

$$\left. \begin{aligned} [s_k^z, S] &= F_k s_k^+ , \\ [s_k^-, S] &= -2F_k s_k^z - G_{kk} s_k^+ , \\ [s_k^z, F_m] &= G_{km} s_k^+ , \\ [s_k^z, G_{mn}] &= M_{kmn} s_k^+ , \\ [s_k^z, F_m^2] &= 2F_m G_{km} s_k^+ , \\ [s_k^-, F_m] &= -2G_{km} s_k^z - M_{kkm} s_k^+ , \\ [s_k^-, F_m^2] &= -2G_{km}^2 s_k^+ - 2F_m M_{kkm} s_k^+ - 4F_m G_{km} s_k^z , \\ [s_k^z, M_{mnp}] &= N_{kmnp} s_k^+ , \\ [s_k^-, G_{mn}] &= -2M_{kmn} s_k^z - N_{kkmn} s_k^+ . \end{aligned} \right\} \quad (11)$$

We may now write the similarity-transformed expressions of the single-spin operators s^α ; $\alpha \equiv \{+, -, z\}$, where $s^- \equiv (s^+)^\dagger = s^x - i s^y$, as

$$\left. \begin{aligned} e^{-S} s_k^+ e^S &\equiv \tilde{s}_k^+ = s_k^+ \\ e^{-S} s_k^z e^S &\equiv \tilde{s}_k^z = s_k^z + F_k s_k^+ \\ e^{-S} s_k^- e^S &\equiv \tilde{s}_k^- = s_k^- - 2F_k s_k^z - G_{kk} s_k^+ - F_k^2 s_k^+ . \end{aligned} \right\} \quad (12)$$

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 $s > \frac{1}{2}$.

III. 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

$$E_I \equiv \langle \Phi | C_I^- e^{-S} H e^S | \Phi \rangle = 0 , \forall I \neq 0 . \quad (13)$$

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

$$\begin{aligned} \text{TERM 1 : } \tilde{s}_i^z \tilde{s}_j^z &= s_i^z s_j^z + F_j s_j^+ s_i^z + F_i s_i^+ s_j^z + G_{ij} s_i^+ s_j^+ + F_i F_j s_i^+ s_j^+ \\ \text{TERM 2 : } \tilde{s}_i^z \tilde{s}_j^+ &= s_j^+ s_i^z + F_i s_i^+ s_j^+ \\ \text{TERM 3 : } \tilde{s}_i^z \tilde{s}_j^- &= -2F_j s_i^z s_j^z - 2G_{ij} s_i^+ s_j^z - G_{jj} s_j^+ s_i^z - M_{ijj} s_i^+ s_j^+ - 2F_i F_j s_i^+ s_j^z \\ &\quad - 2F_j G_{ij} s_i^+ s_j^+ - F_i G_{jj} s_i^+ s_j^+ - F_i F_j^2 s_i^+ s_j^+ - F_j^2 s_j^+ s_i^z \\ \text{TERM 4 : } \tilde{s}_i^+ \tilde{s}_j^z &= s_i^+ s_j^z + F_j s_i^+ s_j^+ \end{aligned}$$

$$\begin{aligned}
\text{TERM 5 : } \tilde{s}_i^- \tilde{s}_j^z &= -2F_i s_i^z s_j^z - 2G_{ij} s_j^+ s_i^z - G_{ii} s_i^+ s_j^z - M_{ij} s_i^+ s_j^+ - 2F_i F_j s_j^+ s_i^z \\
&\quad - 2F_i G_{ij} s_i^+ s_j^+ - F_j G_{ii} s_i^+ s_j^+ - F_j F_i^2 s_i^+ s_j^+ - F_i^2 s_i^+ s_j^z \\
\text{TERM 6 : } \tilde{s}_i^+ \tilde{s}_j^- &= -2F_j s_i^+ s_j^z - G_{jj} s_i^+ s_j^+ - F_j^2 s_i^+ s_j^+ \\
\text{TERM 7 : } \tilde{s}_i^- \tilde{s}_j^+ &= -2F_i s_j^+ s_i^z - G_{ii} s_i^+ s_j^+ - F_i^2 s_i^+ s_j^+ \\
\text{TERM 8 : } \tilde{s}_i^+ \tilde{s}_j^+ &= s_i^+ s_j^+ \\
\text{TERM 9 : } \tilde{s}_i^- \tilde{s}_j^- &= 4G_{ij} s_i^z s_j^z + 2M_{ij} s_i^+ s_j^z + 2M_{ijj} s_j^+ s_i^z + N_{ijj} s_i^+ s_j^+ \\
&\quad + 2G_{ij}^2 s_i^+ s_j^+ + 2F_j M_{ij} s_i^+ s_j^+ + 4F_j G_{ij} s_j^+ s_i^z + 4F_i F_j s_i^z s_j^z \\
&\quad + 4F_i G_{ij} s_i^+ s_j^z + 2F_i G_{jj} s_i^+ s_j^+ + 2F_i M_{ijj} s_i^+ s_j^+ + 4F_i F_j G_{ij} s_i^+ s_j^+ \\
&\quad + 2F_i F_j^2 s_j^+ s_i^z + 2F_j G_{ii} s_i^+ s_j^z + G_{ii} G_{jj} s_i^+ s_j^+ + F_j^2 G_{ii} s_i^+ s_j^+ \\
&\quad + 2F_i^2 F_j s_i^+ s_j^z + F_i^2 G_{jj} s_i^+ s_j^+ + F_i^2 F_j^2 s_i^+ s_j^+ \\
\text{TERM 10 : } \tilde{s}_i^z &= s_i^z + F_i s_i^+ \\
\text{TERM 11 : } \tilde{s}_i^- &= -2F_i s_i^z - G_{ii} s_i^+ - (F_i)^2 s_i^+ \\
\text{TERM 12 : } (\tilde{s}_i^z)^2 &= (s_i^z)^2 + 2F_i s_i^+ s_i^z + G_{ii} (s_i^+)^2 + F_i (s_i^+)^2 + F_i^2 (s_i^+)^2 \\
\text{TERM 13 : } \tilde{s}_i^+ &= s_i^+
\end{aligned} \tag{14}$$

(Note that $s^-|\Phi\rangle = 0$ is implicitly assumed in Eq. (14) above.) We now “pattern-match” the C_i^- operators to those the relevant terms in the Hamiltonian from Eq. (14) above in order to form the CCM equations $E_I = 0$ of Eq. (13 at a given level of approximation. These coupled non-linear equations are then solved readily by using, e.g, the Newton-Raphson method. However, these are solved via direct iteration for larger values of the approximation level because the cost of storing the Jacobian in local memory for the Newton-Raphson (or other) method becomes prohibitive. This may be parallelised also to achieve very high orders of approximation and this is discussed below.

We now define the following *new* set of CCM bra-state correlation coefficients given by $\tilde{x}_I \equiv N_B/N(l!)\nu_I\tilde{S}_I$ 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 ν_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 coefficients ν_I , and $(l!)$ etc. never need to be explicitly determined. The CCM

bra-state operator may thus be rewritten as

$$\tilde{S} \equiv 1 + N \sum_{I=1}^{N_F} \tilde{x}_I C_I^- , \quad (15)$$

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

$$\bar{H} = N \sum_{I=0}^{N_F} \tilde{x}_I E_I , \quad (16)$$

where $\tilde{x}_0 = 1$. We note that the E_0 is defined by $E_0 = \frac{1}{N} \langle \Phi | e^{-S} H e^S | \Phi \rangle$ (and, thus, $E_0 = \frac{1}{N} E_g$) and that E_I is the I -th CCM ket-state equation defined by Eq. (13). The CCM ket-state equations are easily rederived by taking the partial derivative of \bar{H}/N with respect to \tilde{x}_I , where

$$\frac{\delta(\bar{H}/N)}{\delta \tilde{x}_I} (\equiv 0) = E_I . \quad (17)$$

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

$$\frac{\delta(\bar{H}/N)}{\delta x_I} = \frac{\delta E_0}{\delta x_I} + \sum_{J=1}^{N_F} \tilde{x}_J \frac{\delta E_J}{\delta x_I} (\equiv 0) = \tilde{E}_I . \quad (18)$$

The equation $\tilde{E}_I = 0$ is easily solved computationally via LU decomposition for low to medium orders of approximation or via direct iteration (which may be parallised – discussed below) for even higher orders of approximation, 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. (18). Thus, we never need to evaluate Eq. (4) explicitly.

IV. GENERALISED GROUND-STATE EXPECTATION VALUES

Expectation values of spin operators may be treated in an analogous manner to that of the expectation value of the Hamiltonian, given by \bar{H} . We write:

$$A_I = \langle \Phi | C_I^- e^{-S} A e^S | \Phi \rangle \quad (19)$$

and with $C_0^- = 1$. The similarity transform of A is defined by Eq. (6) and once again this process results in terms such as those shown in Eq. (14) may again be employed. However,

we do not constrain k and m in two-body terms to be nearest-neighbours although in this case. The expectation value of the generalised may again be written in a particularly simple form as

$$\begin{aligned}\bar{A} &= \langle \tilde{\Psi} | A | \Psi \rangle \\ &= N \sum_{I=0}^{N_F} \tilde{x}_I \langle \Phi | C_I^- e^{-S} A e^S | \Phi \rangle \\ \Rightarrow \bar{A} &= N \sum_{I=0}^{N_F} \tilde{x}_I A_I\end{aligned}\tag{20}$$

where $\tilde{x}_0 = 1$ also and $A_0 = \frac{1}{N} \langle \Phi | e^{-S} A e^S | \Phi \rangle$. The same code used to find ground-state equations may be used to find the generalised expectation values. Again we note that the index I in Eq. (20) runs from zero to N_f . Again, we note that factors such as N_B or ν_I or $l!$ in F_I do not need to be determined explicitly because they cancel because of the definition of $\{\tilde{x}_I\}$ given above.

V. DIRECT ITERATION OF THE GROUND-STATE EQUATIONS AND PARALLELIZATION

The parallelization of the ground-state CCM problem for very high-order CCM is achieved by solving 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 collect the linear terms for x_I for the i^{th} CCM ket-state equation on the left of the equation and all other terms on the right. We carry out exactly the same procedure for the bra-state, although in this case the problem is linear with respect to $\{\tilde{x}_I\}$. This is written conveniently for the ket state as

$$x_I = E_I(x_1, x_2, \dots, x_{I-1}, x_{I+1}, \dots, x_{N_f}, x_1^2, x_2^2, \dots, x_1^4, \dots, x_{N_f}^4) \quad , \tag{21}$$

and for the bra state as,

$$\tilde{x}_I = \tilde{E}_I(\tilde{x}_1, \tilde{x}_2, \dots, \tilde{x}_{I-1}, \tilde{x}_{I+1}, \dots, \tilde{x}_{N_f} ; x_1, x_2, \dots, x_{N_f}, x_1^2, x_2^2, \dots, x_1^3, \dots, x_{N_f}^3) \quad . \tag{22}$$

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 .

Furthermore, the computational problem posed by solving Eqs. (21) and (22) via direct iteration may be solved using parallel processing. The different equations of Eqs. (21) and (22) for different values of the index I are determined separately on different processors. The equations for the different values of I are then stored locally to each processor. As each iteration of the “direct iteration” method we find the right-hand sides of those relevant values of I allocated to each processor. We then collect the right-hand side into a single array and this forms our values for x_I or \tilde{x}_I for the next iteration. Again, we note that we must solve the ket-state equations of Eq. (21) first and then these values for the ket-state coefficients are used in the bra-state equations of Eq. (22). This approach is a simple “brute-force” method, although it has been found to be surprisingly successful at going to very high orders of approximation. Indeed, we may now treat of order 10^6 fundamental clusters using this approach and for approximately 10^3 processors used in parallel. Clearly, a similar approach may also be used to find the “generalised” expectation values via parallel processing.

VI. 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

$$|\Psi_e\rangle = X^e e^S |\Phi\rangle . \quad (23)$$

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

$$\epsilon_e X^e |\Phi\rangle = e^{-S} [H, X^e] e^S |\Phi\rangle (\equiv \hat{R} |\Phi\rangle) , \quad (24)$$

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

$$X^e = \sum_{I \neq 0} \mathcal{X}_I^e C_I^+ , \quad (25)$$

Eq. (25) implies the overlap relation

$$\begin{aligned} \langle \Phi | \Psi_e \rangle &= \langle \Phi | \tilde{X} e^S | \Phi \rangle \\ \Rightarrow \langle \Phi | \Psi_e \rangle &= 0 . \end{aligned} \quad (26)$$

We now apply $\langle \Phi | C_I^-$ to Eq. (24) in order to form the basic equation for the excited state, given by

$$\epsilon_e \mathcal{X}_I^e = \langle \Phi | C_I^- e^{-S} [H, X^e] e^S | \Phi \rangle, \forall I \neq 0, \quad (27)$$

which is a generalised set of eigenvalue equations with eigenvalues ϵ_e and corresponding eigenvectors \mathcal{X}_I^e . 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 model considered here is in the subspace $s_T^z \equiv \sum_i s_i^z = 0$, whereas the excited state has $s_T^z \equiv \sum_i s_i^z = +1$. 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_e} .

VII. HIGH-ORDER EXCITED-STATE OPERATORS AND COMMUTATIONS

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

$$X = \sum_l \sum_{[i_2, \dots, i_l]} X_{i_i, \dots, i_l}^e s_{i_2}^+ \cdots s_{i_l}^+ \quad (28)$$

where the expression $[i_1, \dots, i_l]$ indicates that we choose one of the $(l!)$ possible orderings of the spin-raising operators and $X_I^e \equiv X_{i_i, \dots, i_l}^e = (l!) \mathcal{X}_I^e$. We now also define the further high-order operators for the excited state, given by

$$\left. \begin{aligned} P_k &\equiv \sum_l \sum_{[i_2, \dots, i_l]} X_{k, i_2, \dots, i_l}^e s_{i_2}^+ \cdots s_{i_l}^+ \\ Q_{km} &\equiv \sum_{l>1} \sum_{[i_3, \dots, i_l]} X_{k, m, i_3, \dots, i_l}^e s_{i_3}^+ \cdots s_{i_l}^+ \\ R_{kmn} &\equiv \sum_{l>2} \sum_{[i_4, \dots, i_l]} X_{k, m, n, i_4, \dots, i_l}^e s_{i_4}^+ \cdots s_{i_l}^+ \\ T_{kmnp} &\equiv \sum_{l>3} \sum_{[i_5, \dots, i_l]} X_{k, m, n, p, i_5, \dots, i_l}^e s_{i_5}^+ \cdots s_{i_l}^+ \end{aligned} \right\} \quad (29)$$

Again, we note that this approach means that we *never* need to determine explicitly the factors $(l!)$ in the high-order operators. The following commutation relations may also be

proven:

$$\left. \begin{aligned}
[s_k^z, X] &= P_k s_k^+ , \\
[s_k^-, X] &= -2P_k s_k^z - Q_{kk} s_k^+ , \\
[s_k^z, P_m] &= Q_{km} s_k^+ , \\
[s_k^z, Q_{mn}] &= R_{kmn} s_k^+ , \\
[s_k^z, P_m^2] &= 2P_m Q_{km} s_k^+ , \\
[s_k^-, P_m] &= -2Q_{km} s_k^z - R_{kkm} s_k^+ , \\
[s_k^-, P_m^2] &= -2Q_{km}^2 s_k^+ - 2P_m R_{kkm} s_k^+ - 4P_m Q_{km} s_k^z , \\
[s_k^z, R_{mnp}] &= T_{kmnp} s_k^+ , \\
[s_k^-, Q_{m,n}] &= -2R_{kmn} s_k^z - T_{kkmn} s_k^+ .
\end{aligned} \right\} \quad (30)$$

VIII. DERIVING AND SOLVING THE EXCITED STATE EQUATIONS

We now wish to determine and solve the CCM excited-state equations given by Eq. (27). Specific terms in the Hamiltonian are now explicitly written in terms of the new excited-state high-order CCM operators as:

$$\begin{aligned}
\text{TERM 1 : } e^{-S}[s_i^z s_j^z, X^e]e^S &= P_i s_i^+ s_j^z + P_i F_j s_i^+ s_j^+ + P_j s_j^+ s_i^z + P_j F_i s_i^+ s_j^+ + Q_{ij} s_i^+ s_j^+ \\
\text{TERM 2 : } e^{-S}[s_i^z s_j^+, X^e]e^S &= P_i s_i^+ s_j^+ \\
\text{TERM 3 : } e^{-S}[s_i^z s_j^-, X^e]e^S &= -2P_i F_j s_i^+ s_j^z - P_i G_{jj} s_i^+ s_j^+ - P_i F_j^2 s_i^+ s_j^+ - 2P_j s_i^z s_j^z - 2P_j F_j s_j^+ s_i^z \\
&\quad - 2P_j F_i s_i^+ s_j^z - 2P_j G_{ij} s_i^+ s_j^+ - 2P_j F_i F_j s_i^+ s_j^+ - 2Q_{ij} s_i^+ s_j^z \\
&\quad - 2F_j Q_{ij} s_i^+ s_j^+ - Q_{jj} s_j^+ s_i^z - F_i Q_{jj} s_i^+ s_j^+ - R_{ijj} s_i^+ s_j^+ \\
\text{TERM 4 : } e^{-S}[s_i^+ s_j^z, X^e]e^S &= P_j s_i^+ s_j^+ \\
\text{TERM 5 : } e^{-S}[s_i^- s_j^z, X^e]e^S &= -2P_j F_i s_j^+ s_i^z - P_j G_{ii} s_i^+ s_j^+ - P_j F_i^2 s_j^+ s_i^+ - 2P_i s_i^z s_j^z - 2P_i F_j s_j^+ s_i^z \\
&\quad - 2P_i F_i s_i^+ s_j^z - 2P_i G_{ij} s_i^+ s_j^+ - 2P_i F_i F_j s_i^+ s_j^+ - 2Q_{ij} s_j^+ s_i^z \\
&\quad - 2F_i Q_{ij} s_i^+ s_j^+ - Q_{ii} s_i^+ s_j^z - F_j Q_{ii} s_i^+ s_j^+ - R_{iij} s_i^+ s_j^+ \\
\text{TERM 6 : } e^{-S}[s_i^+ s_j^-, X^e]e^S &= -2P_j s_i^+ s_j^z - Q_{jj} s_i^+ s_j^+ - 2P_j F_j s_i^+ s_j^+ \\
\text{TERM 7 : } e^{-S}[s_i^- s_j^+, X^e]e^S &= -2P_i s_j^+ s_i^z - Q_{ii} s_i^+ s_j^+ - 2P_i F_i s_i^+ s_j^+ \\
\text{TERM 8 : } e^{-S}[s_i^+ s_j^+, X^e]e^S &= 0 \\
\text{TERM 9 : } e^{-S}[s_i^- s_j^-, X^e]e^S &= 4P_i F_j s_i^z s_j^z + 4P_i G_{ij} s_i^+ s_j^z + 2P_i G_{jj} s_j^+ s_i^z + 2P_i M_{ijj} s_i^+ s_j^+ \\
&\quad + 4P_i F_i F_j s_i^+ s_j^z + 4P_i F_j G_{ij} s_i^+ s_j^+ + 2P_i F_i G_{jj} s_i^+ s_j^+ + 2P_i F_i F_j^2 s_i^+ s_j^+
\end{aligned}$$

$$\begin{aligned}
& +2P_i F_j^2 s_j^+ s_i^z + 2Q_{ii} F_j s_i^+ s_j^z + Q_{ii} G_{jj} s_i^+ s_j^+ + Q_{ii} F_j^2 s_i^+ s_j^+ \\
& +4Q_{ij} s_i^z s_j^z + 4Q_{ij} F_j s_j^+ s_i^z + 4Q_{ij} F_i s_i^+ s_j^z + 4Q_{ij} G_{ij} s_i^+ s_j^+ \\
& +4Q_{ij} F_i F_j s_i^+ s_j^+ + 2R_{ii} s_i^+ s_j^z + 2R_{ii} F_j s_i^+ s_j^+ + 2R_{ij} s_j^+ s_i^z \\
& +2R_{ij} F_i s_i^+ s_j^+ + T_{ij} s_i^+ s_j^+ + 4P_j G_{ij} s_j^+ s_i^z + 2P_j M_{ij} s_i^+ s_j^+ \\
& +4P_j F_i s_i^z s_j^z + 4P_j F_i F_j s_j^+ s_i^z + 2P_j G_{ii} s_i^+ s_j^z + 2P_j G_{ii} F_j s_i^+ s_j^+ \\
& +2P_i F_i^2 s_i^+ s_j^z + 2P_j F_i^2 F_j s_i^+ s_j^+ + 4P_j F_i G_{ij} s_i^+ s_j^+ + 2Q_{jj} F_i s_j^+ s_i^z \\
& +Q_{jj} G_{ii} s_i^+ s_j^+ + Q_{jj} F_i^2 s_i^+ s_j^+
\end{aligned}$$

$$\text{TERM 10 : } e^{-S}[s_i^z, X^e]e^S = P_i s_i^+$$

$$\text{TERM 11 : } e^{-S}[s_i^-, X^e]e^S = -2P_i s_i^z - Q_{ii} s_i^+ - 2P_i F_i s_i^+$$

$$\text{TERM 12 : } e^{-S}[(s_i^z)^2, X^e]e^S = 2P_i s_i^+ s_i^z + Q_{ii} (s_i^+)^2 + P_i (s_i^+)^2 + 2P_i F_i (s_i^+)^2$$

$$\text{TERM 13 : } e^{-S}[s_i^+, X^e]e^S = 0 \quad (31)$$

(Note that $s^-|\Phi\rangle = 0$ is implicitly assumed in Eq. (31) above.) Again, we now “pattern-match” the C_I^- 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. (27) 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. (29) are in linear in those terms in Eq. (31). 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. Hence, once 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 $N_{f_e}^2$. Again, this becomes prohibitive computationally for extremely large values of N_{f_e} and so we again use direct iteration methods.

IX. DIRECT ITERATION OF THE EXCITED-STATE EQUATIONS AND PARALLELIZATION

The eigenvalue equations of Eq. (27) may be iterated directly in order to solve them. We denote the matrix for the eigenvalue problem of Eq. (27) by A and we denote the

eigenvectors by $y = (X_1^e, \dots, X_{N_{Fe}})^T$. Hence, we iterate directly the eigenvalue equation given by

$$Ay = \lambda y \quad . \quad (32)$$

This is just the well-known “power iteration” method and the ratios of X_I^e in successive iterations yields the relevant eigenvalue. However, the eigenvalue determined in this manner is the eigenvalue of largest magnitude, λ_{MAX} , rather than the lowest (generally the one of smallest magnitude λ_{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 λ_{MAX} has been found, we then solve the following eigenvalue equation by direct iteration:

$$(A - \lambda_{\text{MAX}}I)y' = \lambda'y' \quad . \quad (33)$$

This process ought to converge to an eigenvalue $\lambda' = \lambda_{\text{MIN}} - \lambda_{\text{MAX}}$. Indeed, this was found to be the case for the model studied here at all levels of approximation in those regions where the model state was a “good choice”. Furthermore, we saw that 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.

We note that the local memory usage is again far lower for this “power iteration” approach rather than the corresponding “complete diagonalization” of the matrix problem because we need store only the values for the set $\{X_I^e\}$ at each point. Finally, the direct iterative solution of the CCM excited-state problem may be parallelized readily. Again, we share the problem of finding the right-hand side of Eq. (27) over all processors for different values of I . We then collect the results together in order to form the left-hand side of Eq. (27). We iterate to find λ_{MAX} . A similar parallelization process is then used to find λ_{MIN} .