High-Order Coupled Cluster Calculations Via Parallel Processing: An Illustration For CaV$_4$O$_9$

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The coupled cluster method (CCM) is a method of quantum many-body theory that may provide accurate results for the ground-state properties of lattice quantum spin systems even in the presence of strong frustration and for lattices of arbitrary spatial dimensionality. Here we present a significant extension of the method by introducing a new approach that allows an efficient parallelization of computer codes that carry out “high-order” CCM calculations. We find that we are able to extend such CCM calculations by an order of magnitude higher than ever before utilized in a high-order CCM calculation for an antiferromagnet. Furthermore, we use only a relatively modest number of processors, namely, eight. Such very high-order CCM calculations are possible only by using such a parallelized approach. An illustration of the new approach is presented for the ground-state properties of a highly frustrated two-dimensional magnetic material, CaV$_4$O$_9$. Our best results for the ground-state energy and sublattice magnetization for the pure nearest-neighbor model are given by $E_0/N = -0.5534$ and $M = 0.19$, respectively, and we predict that there is no Néel ordering in the region $0.2 \leq J_2/J_1 \leq 0.7$. These results are shown to be in excellent agreement with the best results of other approximate methods.

A new procedure for carrying out high-order coupled cluster method (CCM) calculations via parallel processing is presented here. The CCM may be applied to systems demonstrating strong frustration and for arbitrary spatial dimension of the lattice. We illustrate our new approach by applying it to CaV$_4$O$_9$ at zero temperature. An increasing number of insulating quantum magnetic systems for lattices of low spatial dimensionality are being studied experimentally. Indeed, the calcium vanadium oxide (CAVO) materials are one particularly useful example. They exhibit strong frustration for a number of different crystallographic lattices with respect to varying chemical composition, and they may demonstrate “novel” ground-state ordering. Indeed, theoretical evidence suggests that CAVO systems may contain a number of differing quantum ground states (see, e.g., Refs. 9, 11) at zero temperature as a function of varying nearest-neighbor next-nearest-neighbor bond strengths. The relevant Hamiltonian is given by

$$H = J_1 \sum_{(i,j)} s_i s_j + J_2 \sum_{(i,k)} s_i s_k,$$

where $i$ runs over all lattice sites, and $j$ and $k$ run over all nearest-neighbor and next-nearest-neighbor sites to $i$, respectively, counting each bond once and once only. The bond strengths are given by $J_1$ and $J_2$ for nearest-neighbor and next-nearest-neighbor terms, respectively. The lattice and exchange “bonds” are illustrated graphically in Fig. 1.

For this model, collinear Néel ordering is observed with respect to nearest neighbors at $J_1 > 0$ and $J_2 = 0$ and with respect to next-nearest neighbors at $J_2 > 0$ and $J_1 = 0$. Exact diagonalizations predict an intermediate phase (or phases) for $0.2 < J_2/J_1 < 0.7$, which is also in good agreement with results of a “spin-wave theory-like” treatment that predict such an intermediate regime for $0.25 < J_2/J_1 < 0.8$. Strong evidence has been gathered using approximate approaches that the ground state in this "intermediate" regime demonstrate dimer or plaquette ordering. However, conclusive evidence for the nature of the ground state across the whole of this regime remains to be yet determined. The ground state of the pure nearest-neighbor model at $J = 1$ and $J_2 = 0$ demonstrates semi-classical Néel order with a sublattice magnetization of 0.178(8) via QMC.

We now turn our attention to the application of the CCM to this model. We choose two model states that are the classical ground states of this model in the limits $J_2/J_1 \to 0$ and $J_2/J_1 \to \infty$ (with both $J_2$ and $J_1$ positive). These are, namely, one in which nearest-neighbor spins are antiparallel, henceforth denoted the

![Unit Cell](image-url)
FIG. 3: CCM results for the ground-state energy per spin at 
$J_1 = 1$ and $J_2 = 0$ using the LSUB$m$ approximation with 
$m = \{6, 8, 10\}$. We see that our results converge 
rapidly with $m$ across a wide range of $J_2$ up to and including 
the point $J_2 = 0$. Extrapolations using Padé approximants 
are also shown.

nearest-neighbor Néel model state; and a second model 
state in which next-nearest-neighbor spins are anti-parallel, 
henceforth denoted the next-nearest-neighbor Néel 
model state. We use four spins in the crystallographic 
primitive cell in both cases. This unit cell is indicated 
in Fig. 1. We perform a notational rotation of the “up” 
spins to “down” spins, as in Refs. 2, 3, 4, 5, 6, 7, 8, 9, 10 for example. We use the full symmetries of the lattice 
to further reduce the computational problem of finding 
the fundamental CCM configurations and in determining 
and solving the CCM equations in both cases.

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

\[ H|\Psi\rangle = E_g |\Psi\rangle ; \quad \langle\tilde{\Psi}|H = E_g \langle\tilde{\Psi}| , \]

are parameterized within the single-reference CCM as fol-

\[ |\Psi\rangle = e^S |\Phi\rangle ; \quad S = \sum_{I \neq 0} S_I C_I^\dagger , \]

\[ \langle\tilde{\Psi}| = \langle\Phi|\tilde{S} e^{-S} ; \quad \tilde{S} = 1 + \sum_{I \neq 0} \tilde{S}_I C_I^- . \]

The single model or reference state $|\Phi\rangle$ is required to have 
the property of being a cyclic vector with respect to two 
well-defined Abelian subalgebras of multi-configurational 
creation operators $\{C_I^\dagger\}$ and their Hermitian-adjoint de-
struction counterparts $\{C_I^\dagger = (C_I^\dagger)^\dagger\}$. The determination 
of the correlation coefficients $\{S_I, \tilde{S}_I\}$ is achieved by 
by requiring the ground-state energy expectation func-
tional $\tilde{H}([S_I, \tilde{S}_I])$, to be stationary with respect to vari-
ations in each of the (independent) variables of the full 
set. We thereby easily derive the following coupled set of 
equations,

\[ \delta \tilde{H}/\delta \tilde{S}_I = 0 \Rightarrow \langle\Phi|\tilde{S} e^{-S} H e^S |\Phi\rangle = 0, \; I \neq 0 ; \quad (4) \]

\[ \delta \tilde{H}/\delta S_I = 0 \Rightarrow \langle\tilde{\Psi}|\tilde{S} e^{-S} [H, C_I^\dagger] e^S |\Phi\rangle = 0, \; I \neq 0 \] 

Equation 4 also shows that the ground-state energy at 
the stationary point has the form

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

It is important to realize that this (bi-)variational for-
mulation does not lead to an upper bound for $E_g$ when 
the summations for $S$ and $\tilde{S}$ in Eq. 6 are truncated, 
due to the lack of exact Hermiticity when such approx-
imations are made. However, one can prove that the 
important Hellmann-Feynman theorem is preserved in 
all such approximations. We utilize various approxima-
tion schemes within $S$ and $\tilde{S}$. The three most commonly 
employed schemes previously utilized have been: (1) the 
SUB$m$ 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 lat-
tice; (2) the SUB$m$-m sub-approximation, in which all 
SUB$m$ correlations spanning a range of no more than 
m adjacent lattice sites are retained; and (3) the localized 
LSUB$m$ scheme, in which all multi-spin correlations over 
distinct locales on the lattice defined by $m$ or fewer 
contiguous sites are retained.

We now solve the CCM equations in parallel by re-
arranging Eqs. 4 and 5 for the ket and bra states, re-
spectively, where

\[ S_I = f_I (S_1, \ldots, S_{I-1}, S_{I+1}, \ldots, S_{N_f}) ; \]

\[ \tilde{S}_I = \tilde{f}_I (S_1, \ldots, S_{N_f} ; \]

and where $N_f$ is the number of fundamental config-
urations. There are therefore $N_f$ equations for both $f_I$ 
and $\tilde{f}_I$, which refer to the ket and bra states, respec-
tively. Each equation contains a finite-number of terms

\[ \langle\tilde{S}_I| H = \sum_{I \neq 0} \tilde{S}_I C_I^- . \]
and that, indeed, this is always the case for the CCM for Hamiltonians that contain a finite number of creation and destruction operators. Our approach is to iterate directly the set of equations in (7) to convergence first in order to obtain the ket-state correlation coefficients. We then iterate directly the set of equations in (8) to convergence in order to obtain values for the bra-state correlation coefficients.

Our parallelization technique is now to split the problem of determining and solving the $N_f$ equations for both the bra and ket states of Eqs. (7) and (8) between each processor in the parallel cluster. Thus, each equation for $I = \{1, \cdots, N_f\}$ is ever dealt with by one node in the cluster only and the results of each node are collected together at each iteration. (Again, we remember that we iterate the ket-state equations first and then the bra-state equations.) Thus, the set of equations for the ket and bra states are solved independently and in parallel. Eight Compaq “Wildfire” processors were used here, each with 2 GIGs of RAM and CPU speeds of 731 MHz. The run for LSUB10 with the nearest-neighbor model state with 22395 fundamental configurations took 11 days for 18 distinct values of $J_2/J_1$.

We now wish only to illustrate this approach by considering the the CAVO system of Eq. (1). The ground-state energies of the pure nearest-neighbor antiferromagnet (at $J_1 = 1$ and $J_2 = 0$) are shown in Table I. Fig. 2 shows LSUBm ground-state energy plotted against $m^{-2}$ for $m = \{4, 6, 8, 10\}$. “Linear extrapolations” of data points have previously been used to great effect for isotropic nearest-neighbor antiferromagnets [2, 3, 4, 5, 6]. In this case of the CAVO model studies here, we obtain a value of $E_g/N = -0.55310(5)$. We also carry out extrapolations using Padé approximants [1] for $E_g/N$ plotted against $m^{-2}$ for $m = \{4, 6, 8, 10\}$. $l$ is the order of the polynomial in the denominator and that we perform an exact fit of the curve to the data. Hence, in contrast to the simple linear fit, there is no “curve-fitting” error. Extrapolations using Padé approximants for the ground-state energy at $J_1 = 1$ and $J_2 = 0$ are found to be in excellent agreement with a variational QMC result [13] of $E_g/N = -0.5501$ and an ED result [10] of $E_g/N = -0.5533$. We conclude that our results are converged to at least 3 decimal places, and we believe that these results constitute one of the most accurate estimations of the ground-state energy for this system ever found.

The number of fundamental configurations for the SUB10-10 approximations were 22340 and 39434 for the nearest-neighbor and next-nearest-neighbor model states, respectively, and this calculation for the next-nearest-neighbor model state is the largest CCM calculation as yet performed for quantum antiferromagnetic spin systems by an order of magnitude compared to the previous highest [5]. The results for the ground-state energies for both model states are shown in Fig. 3 with respect to $J_2/J_1$. We see that these results converge quickly with truncation index $m$ for the LSUB$m$ approximation over a wide range of $J_2/J_1$. The wide area without results is an indication ground-state ordering (e.g., dimer or plaquette) not accessible with our Néel model states.

TABLE I: CCM LSUB$m$ results for the CAVO antiferromagnet. The numbers of fundamental configurations nearest-neighbor and next-nearest-neighbor Néel model states are given by $N_f^{n,n}$ and $N_f^{n,n,n}$, respectively. Results for the points at which the sublattice magnetization goes to zero for the nearest-neighbor and next-nearest-neighbor Néel model states are indicated by $J_2/J_1|_1$ and $J_2/J_1|_2$, respectively. (Errors for the critical points are in the last decimal place shown.) Ground-state energies and sublattice magnetizations for nearest-neighbor bonds only, namely at $J_1 = 1$ and $J_2 = 0$, are extrapolated using Padé approximants for $m = \{4, 6, 8, 10\}$, where $l$ is the order of the polynomial in the denominator. By way of comparison, results of exact diagonalizations (ED) for $E_g/N$ and $M$ of Ref. [16] and values for $J_2/J_1|_1$ and $J_2/J_1|_2$ of Ref. [11] are also given.

| Method | $N_f^{n,n}$ | $E_g/N$ | $M$ | $N_f^{n,n,n}$ | $J_2/J_1|_1$ | $J_2/J_1|_2$ |
|--------|-------------|---------|-----|-------------|-------------|-------------|
| LSUB2  | 2           | -0.52859| 0.393| 2           | 0.73        | 0.29        |
| LSUB4  | 14          | -0.54595| 0.339| 18          | 0.48        | 0.60        |
| LSUB6  | 130         | -0.54984| 0.311| 208         | 0.42        | 0.63        |
| LSUB8  | 1589        | -0.55129| 0.293| 2715        | 0.39        | 0.64        |
| LSUB10 | 22395       | -0.55200| 0.280| 39434       | 0.36        | 0.65        |
| Padé $l = 0$ | -0.55332 | 0.204 | - | 0.22 | 0.66 |
| Padé $l = 1$ | -0.55338 | 0.190 | - | 0.16 | 0.72 |
| Padé $l = 2$ | -0.55340 | 0.191 | - | 0.18 | 0.71 |
| ED [11, 16] | -0.5533 | 0.2303 | 0.2 | 0.2 | 0.7 |

FIG. 4: CCM results for the sublattice magnetization of the $J_1$-$J_2$ model using the LSUB$m$ approximation with $m = \{6, 8, 10\}$. Extrapolations using Padé approximants are also shown.
approximants in order to carry out the extrapolations. This gives values for \( M \) in the range 0.19 to 0.20, which is again in good agreement with the best of other approximante methods, such as results from QMC \(^{[10]}\) and ED \(^{[10]}\) of \( M = 0.178(8) \) and \( M = 0.2303 \), respectively. We therefore use Padé approximants for increasing values of \( J_2/J_1 > 0 \) as a reliable indicator of the position of the phase transition points is given by the position at which the sublattice magnetization goes to zero as a function of \( J_2/J_1 \). Fig. \(^{[4]}\) plots our high-order CCM results for the amount of sublattice magnetization. We may see from Table \(^{[1]}\) that our results for the positions of the phase transition points is in good agreement with results of exact diagonalizations \(^{[11]}\) that predict that plaquette and dimer ordering exists in the region \( 0.2 < J_2/J_1 < 0.7 \).

We have shown in this article that it is possible to simulate the properties of a CAVO material at zero temperature using parallelized high-order CCM techniques. The construction of CCM codes to such high orders of approximation is a non-trivial task. Furthermore, the parallelization of the CCM code adds an additional level of complexity to it. The 39434 fundamental configurations for the next-nearest-neighbor Néel model state is a massive leap forward in the practical application of the CCM, which is impossible without an efficient parallelization. However, the numbers of fundamental configurations of order \( 10^8 \) for quantum antiferromagnets are clearly within our reach via parallel processing for even larger clusters of processors than the relatively modest number of (eight) processors used here. We have therefore “proven the principle” that the CCM can be effectively parallelized, and we believe that we have also shown here that this, in turn, leads to great increases in accuracy of observed expectation values.

The extension of these calculations to treat quantum systems that demonstrate “novel ordering,” such as with plaquette or dimer solid ground states, is, in principle, straightforward. The basic building blocks for the CCM using dimer models states are already in place, and low-order calculations have already been attempted successfully for a one-dimensional frustrated spin system \(^{[1]}\). However, procedurally, the spatial dimension of the lattice makes no difference to the coding of the problem, although the computational expense grows with it and so efficient parallelized approaches, such as that shown here, would be very useful in this case. Extensions of the method to consider excitation energies have been very successful \(^{[4]}\), and it is again a straightforward matter to obtain excitation spectra using “localized” high-order CCM approximations. The CCM may be applied to consider systems even in the presence of strong frustration and for lattices of arbitrary spatial dimensionality, and the CCM now constitutes a very powerful and a useful tool in order to understand the basic properties of quantum spin systems.

\[ \text{References} \]

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