Journal of Statistical Physics, Vol. 108, Nos. 3/4, August 2002 (© 2002)

Coupled Cluster Method Calculations of Quantum Magnets with Spins of General Spin Quantum Number

D. J. J. Farnell,[1] R. F. Bishop,[2] and K. A. Gernoth[2]

Received January 3, 2002; accepted March 14, 2002

We present a new high-order coupled cluster method (CCM) formalism for the ground states of lattice quantum spin systems for general spin quantum number, s. This new "general-s" formalism is found to be highly suitable for a computational implementation, and the technical details of this implementation are given. To illustrate our new formalism we perform high-order CCM calculations for the one-dimensional spin-half and spin-one antiferromagnetic XXZ models and for the one-dimensional spin-half/spin-one ferrimagnetic XXZ model. The results for the ground-state properties of the isotropic points of these systems are seen to be in excellent quantitative agreement with exact results for the special case of the spin-half antiferromagnet and results of density matrix renormalization group (DMRG) calculations for the other systems. Extrapolated CCM results for the sublattice magnetization of the spin-half antiferromagnet closely follow the exact Bethe Ansatz solution, which contains an infinite-order phase transition at $\Delta = 1$. By contrast, extrapolated CCM results for the sublattice magnetization of the spin-one antiferromagnet using this same scheme are seen to go to zero at $\Delta \approx 1.2$, which is in excellent agreement with the value for the onset of the Haldane phase for this model. Results for sublattice magnetizations of the ferrimagnet for both the spin-half and spin-one spins are non-zero and finite across a wide range of Δ, up to and including the Heisenberg point at $\Delta = 1$.

KEY WORDS: CCM; quantum magnets; phase transitions.

1. INTRODUCTION

In this article we consider lattice quantum spin systems at zero temperature. Since the inception of this subject via the introduction of the Heisenberg

[1] School of Mechanical Engineering, University of Leeds, Woodhouse Lane, Leeds LS2 9JT, United Kingdom; e-mail: d.j.j.farnell@leeds.ac.uk

[2] Department of Physics, University of Manchester Institute of Science and Technology (UMIST), P.O. Box 88, Manchester M60 1QD, United Kingdom.

0022-4715/02/0800-0401/0 © 2002 Plenum Publishing Corporation

model it has undergone a period of steady development. Although the exact Bethe Ansatz solution of the Heisenberg model for the spin-half linear chain[1–4] followed quickly after its initial introduction, interest in lattice quantum spin systems has not faltered ever since. In particular, exact solutions, such as those typified by the Bethe Ansatz, are generally restricted to unfrustrated systems of low-dimensional lattices and to low spin quantum number. Thus, much effort has been expended over the last fifty or so years in trying to understand the properties of more general lattice quantum spin systems at zero temperature via approximate methods.

A recent and exciting topic in this field has been the development of the density matrix renormalization group (DMRG)[5–9] method. This technique allows one to perform highly accurate calculations for both frustrated *and* unfrustrated systems of general spin quantum number. As long as one is able to partition a one-dimensional quantum system into "system" and "environment" subsystems then one ought in principle to be able to apply this powerful method of modern-day quantum theory. In particular, DMRG calculations have been used to demonstrate conclusively that the spin-one Heisenberg linear chain antiferromagnet contains an excitation gap.[10] This is in stark contrast to both its classical behaviour and the behaviour of its quantum spin-half counterpart. However, a major restriction on the DMRG method is that it has, so far, only been conclusively applied to one-dimensional or quasi-one-dimensional systems, although isolated cases of highly successful DMRG calculations for various two-dimensional lattices have been performed.

By contrast, quantum Monte Carlo (QMC) calculations[12–15] have been already applied with much success to spin systems of spatial dimensionality both equal to and greater than one. In particular, QMC calculations for the zero-temperature properties of the spin-half, square-lattice[16–19] Heisenberg model present a very accurate and valuable benchmark against which other approximate methods may test themselves. However, although QMC is not as limited by spatial dimensionality as the DMRG method, it is limited in its range of applicability by the existence of the well-known "minus-sign problem." For lattice quantum spin systems the minus sign problem is, in turn, often a consequence of frustration. We note however that for non-frustrated systems one can often, but not always, determine a "sign rule"[28, 29] which effectively circumvents the minus-sign problem of QMC.

Other approximate methods that have been applied to lattice quantum spin problems (at zero temperature) include spin-wave theory (SWT),[20–22], exact diagonalizations of finite-sized lattices,[23, 24] cumulant series expansions,[25–27] and the coupled cluster method (CCM).[30–59] For SWT one maps the spin system onto an exact (or effective) bosonic system (for example, via the Holstein–Primakov transformation) which may then be

solved. The advantage of *linear* SWT is that it is simple to apply and an exact solution generally exists via the Bogoliubov transformation. Higher-order SWT becomes rapidly more complicated and may only be solved perturbatively for these higher orders. Also, it is difficult to increase the level of approximation of this method in a structured and well-defined manner. By contrast, for the QMC method one may systematically increase the lattice size and still obtain an "exact" result to within a well-defined statistical error. Exact diagonalizations of finite-sized systems[23, 24] have also been applied to quantum spin models with much success. However, such calculations rapidly become limited as the size of the lattice is increased by the amount of the computational resources available. By contrast, this problem is not so evident for the exact cumulant series expansion[25–27] calculations. Indeed, such calculations have provided very accurate results for the ground- and excited-state properties of various spin systems. However, in order to determine such expectation values a resummation of the otherwise rapidly divergent perturbation series must be performed using Padé approximants or related techniques. Finally, a technique of quantum many-body theory (QMBT) called the coupled cluster method (CCM)[30–38] has also been applied[39–59] to both unfrustrated and frustrated lattice quantum spin systems with considerable success, and it is this method upon which we shall concentrate henceforth.

The first application[39] of the CCM to the lattice quantum spin systems was to the spin-half Heisenberg model and related models on various bipartite lattices using several localised approximation schemes. This work was quickly generalised[40, 41] to include anisotropy within the Hamiltonian via the spin-half XXZ model. In particular, critical points as a function of the anisotropy were discovered in an approximation scheme denoted as SUB2 (in which *all* possible two-spin correlations are retained). It was furthermore found at these critical points that the CCM excitation spectra became "soft" and that the behaviour with respect to spatial separation of the spin-spin correlation functions changes from exponential to algebraic decay. Both of these features were clearly strong indicators that these critical points were reflections of quantum phase transitions in the real system, which in turn were driven by the anisotropy parameter within the Hamiltonian. (An interesting application of the *extended* coupled cluster method (ECCM) to this model was also performed, and the interested reader is referred to ref. 54.) Frustrated quantum spin systems, such as the J_1–J_2 model[43–45, 52] and the triangular lattice antiferromagnet,[50, 51] have also been studied using the CCM with equal success. Indeed, the CCM has been utilised not only to provide accurate values of the ground-state properties of these systems and very accurate predictions of their phase diagrams, but also to simulate the nodal surfaces[52, 53] of these

models. We note that such approximately determined nodal surfaces are potentially of very great use to QMC calculations in which the "sign problem" is present. Other such applications of note of the CCM have been to the biquadratic model[42] and to the one-dimensional spin-half J_1–J_2 model using a "dimerised" model state.[44]

Furthermore, recent high-order CCM calculations[46, 47–49, 51, 52, 55–59] for spin-half systems using localised approximation schemes have been shown to be very effective in simulating the ground- and excited-state properties of quantum spin systems. The strength of this approach is that one can systematically and rigorously increase the approximation level as a function of some parameter which increases the size of the "locale" over which one retains *all* multi-spin correlations. The approximation schemes become exact in the limit of this parameter going to infinity, although this is generally impossible to achieve in a practical application. One can however extrapolate[55] a series of such results to this limit, and thus obtain very accurate approximate results for the exact properties of even highly frustrated systems. Indeed, the results produced for both unfrustrated systems such as the spin-half XXZ,[46–48, 51, 55] XY,[49] and transverse Ising models,[56] and frustrated systems such as the spin-half J_1–J_2 model,[52] the spin-half triangular[51] and kagomé[58] lattice antiferromagnets, and the spin-half square-lattice Heisenberg antiferromagnet with two types of bonds,[57] have all been seen to be in excellent agreement with the results of the best of other approximate methods, where such results exist. A recent calculation has been performed for the spin-one square-lattice XXZ model[59] which utilised the new "general-s" formalism presented in this article, and the results for the ground-state properties of this model were found to be in superb agreement with previous SWT and cumulant series expansion results.

We note that although spin problems are conceptually simple, they often demonstrate rich and unusual phase diagrams which are the direct result of the strong influence of quantum fluctuations in these strongly correlated systems. Indeed, these phase diagrams can become richer for systems with spin quantum, s, greater than $1/2$, although we note that the limit $s \to \infty$ coincides with the pure classical model. For example, the spin-one biquadratic model[42] contains antiferromagnetic, dimerised, and trimerised phases, and a "Haldane" phase as a function of the ratio of the bond strengths of the linear and biquadratic terms. Thus, lattice quantum spin problems open a wide window on to the rapidly developing field of quantum phase transitions. This subject becomes even more interesting when one also considers the non-zero temperature behaviour of such systems, and the respective roles of quantum and thermal fluctuations in driving phase transitions. For the present, however, we restrict ourselves to the zero-temperature case.

2. THE COUPLED CLUSTER METHOD (CCM)

2.1. The Ground-State Formalism

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

$$H\,|\Psi\rangle = E_g\,|\Psi\rangle; \qquad \langle\tilde{\Psi}|\,H = E_g\,\langle\tilde{\Psi}| \tag{1}$$

are parametrised within the single-reference CCM as follows:

$$\begin{aligned} |\Psi\rangle &= \mathrm{e}^{S}\,|\Phi\rangle; & S &= \sum_{I\neq 0} \mathscr{S}_I C_I^+, \\ \langle\tilde{\Psi}| &= \langle\Phi|\,\tilde{S}\mathrm{e}^{-S}; & \tilde{S} &= 1 + \sum_{I\neq 0} \tilde{\mathscr{S}}_I C_I^- \end{aligned} \tag{2}$$

The single model or reference state $|\Phi\rangle$ is normalised ($\langle\Phi\,|\,\Phi\rangle = 1$), and is required to have the property of being a cyclic vector with respect to two well-defined Abelian subalgebras of *multi-configurational* creation operators $\{C_I^+\}$ and their Hermitian-adjoint destruction counterparts $\{C_I^- \equiv (C_I^+)^\dagger\}$. Thus, $|\Phi\rangle$ plays the role of a vacuum state with respect to a suitable set of (mutually commuting) many-body creation operators $\{C_I^+\}$,

$$C_I^-\,|\Phi\rangle = 0, \qquad I \neq 0 \tag{3}$$

with $C_0^- \equiv 1$, the identity operator. These operators are complete in the many-body Hilbert (or Fock) space,

$$1 = |\Phi\rangle\langle\Phi| + \sum_{I\neq 0} \frac{C_I^+\,|\Phi\rangle\langle\Phi|\,C_I^-}{\langle\Phi|\,C_I^- C_I^+\,|\Phi\rangle} \tag{4}$$

We note that although the manifest hermiticity, ($\langle\tilde{\Psi}|^\dagger = |\Psi\rangle/\langle\Psi\,|\,\Psi\rangle$), is lost in these parametrizations, the intermediate normalization condition $\langle\tilde{\Psi}\,|\,\Psi\rangle = \langle\Phi\,|\,\Psi\rangle = \langle\Phi\,|\,\Phi\rangle \equiv 1$ is explicitly imposed. The *correlation coefficients* $\{\mathscr{S}_I, \tilde{\mathscr{S}}_I\}$ are regarded as being independent variables, even though formally we have the relation

$$\langle\Phi|\,\tilde{S} = \frac{\langle\Phi|\,\mathrm{e}^{S^\dagger}\mathrm{e}^{S}}{\langle\Phi|\,\mathrm{e}^{S^\dagger}\mathrm{e}^{S}\,|\Phi\rangle} \tag{5}$$

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

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

We note that the exponentiated form of the ground-state CCM parametrization of Eq. (2) ensures the correct counting of the *independent* and excited correlated many-body clusters with respect to $|\Phi\rangle$ which are present in the exact ground state $|\Psi\rangle$. It also ensures the exact incorporation of the Goldstone linked-cluster theorem, which itself guarantees the size-extensivity of all relevant extensive physical quantities.[37]

The determination of the correlation coefficients $\{\mathscr{S}_I, \tilde{\mathscr{S}}_I\}$ is achieved by taking appropriate projections onto the ground-state Schrödinger equations of Eq. (1). Equivalently, they may be determined variationally,

$$\delta \bar{H}/\delta \tilde{\mathscr{S}}_I = 0 \Rightarrow \langle \Phi| C_I^- e^{-S} H e^{S} |\Phi\rangle = 0, \qquad \forall I \neq 0; \tag{7}$$

$$\delta \bar{H}/\delta \mathscr{S}_I = 0 \Rightarrow \langle \Phi| \tilde{S} e^{-S} [H, C_I^+] e^{S} |\Phi\rangle = 0, \qquad \forall I \neq 0 \tag{8}$$

Equation (7) also shows that the ground-state energy at the stationary point has the simple form

$$E_g = E_g(\{\mathscr{S}_I\}) = \langle \Phi| e^{-S} H e^{S} |\Phi\rangle \tag{9}$$

We note that Eq. (7) represents a coupled set of nonlinear multinomial equations for the c-number correlation coefficients $\{\mathscr{S}_I\}$. The nested commutator expansion of the similarity-transformed Hamiltonian,

$$\tilde{H} \equiv e^{-S} H e^{S} = H + [H, S] + \frac{1}{2!}[[H, S], S] + \cdots \tag{10}$$

together with the fact that all of the individual components of S in the sum in Eq. (2) commute with one another, imply that each element of S in Eq. (2) is linked directly to the Hamiltonian in each of the terms in Eq. (10). Thus, each of the coupled equations (7) is of linked-cluster type. Furthermore, each of these equations is of finite length when expanded, since the otherwise infinite series of Eq. (10) will always terminate at a finite order, provided (as is usually the case) only that each term in the second-quantised form of the Hamiltonian H contains a finite number of single-body destruction operators, defined with respect to the reference

(vacuum) state $|\Phi\rangle$. Therefore, the CCM parametrization naturally leads to a workable scheme which can be efficiently implemented computationally.

The CCM formalism is exact in the limit of inclusion of all possible multi-spin cluster correlations within S and $\tilde{S}$, although in any real application this is usually impossible to achieve. It is therefore necessary to utilise various approximation schemes within S and $\tilde{S}$. The three most commonly employed schemes 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 contiguous lattice sites are retained; and (3) the localised LSUBm scheme, in which all multi-spin correlations over distinct locales on the lattice defined by m or fewer contiguous sites are retained. We also make the specific restriction that the creation operators $\{C_I^+\}$ in S preserve any additional symmetries of the Hamiltonian. Thus, the approximate CCM ground-state wave function is constrained to lie in the appropriate subspace defined by the additional quantum numbers corresponding to these additional symmetries. For the XXZ model illustrated later the additional symmetry is provided by the total z-component of spin $s_T^z = \sum_i s_i^z$, which commutes with the Hamiltonian. The ground state lies in the sector $s_T^z = 0$. We denote as distinct configurations those in such appropriately defined subspace which are inequivalent under the point- and space-group symmetries of both the lattice and the Hamiltonian. The number of such distinct (or fundamental) configurations for the ground state at a given level of approximation is labelled by N_F.

2.2. The High-Order Formalism for General Quantum Spin Number

In order to determine the CCM ground-state ket configurations we fundamentally need to *pattern-match* the configurations in the set $\{C_I^-\}$ to the spin-raising operators contained in $\tilde{H}|\Phi\rangle$. We note that for small values of the truncation indices $\{m, n\}$ mentioned in the previous section (and thus for low orders of approximation) this may readily be performed analytically. However, for higher orders of approximation we must use computational methods (see, for example, also refs. 46, 47, 51, and 55) in order to do this. For the cases of interest here, we begin by defining a set of local spin axes in which all of the spins in the chosen model state $|\Phi\rangle$ point along the respective negative z-axes, namely

$$|\Phi\rangle = \bigotimes_{i=1}^{N} |\downarrow\rangle_i; \qquad \text{in the local quantization axes} \tag{11}$$

where $|\downarrow\rangle_i \equiv |s, -s\rangle_i$. This is achieved by an appropriate set of local rotations. Since such rotations are canonical transformations, the underlying spin algebra is preserved, and the energy spectrum of the transformed Hamiltonian (i.e., written in the rotated local spin coordinate scheme) is unchanged.

The next step in the computational implementation of the CCM for lattice quantum spin systems of general spin quantum number, s, is to define a suitable set of multi-spin creation and destruction operators with respect to this model state. We thus define the CCM ket-state correlation operator S in terms of sums of products of single spin-raising operators, $s_k^+ \equiv s_k^x + \mathrm{i}s_k^y$, (again with respect to their local spin axes), such that

$$S = \sum_{i_1}^{N} \mathscr{S}_{i_1} s_{i_1}^+ + \sum_{i_1, i_2}^{N} \mathscr{S}_{i_1, i_2} s_{i_1}^+ s_{i_2}^+ + \cdots \tag{12}$$

The coefficients $\mathscr{S}_{i_1}$, $\mathscr{S}_{i_1, i_2}$, and so on, now represent the spin-correlation coefficients specified by the sets of site indices, $\{i_1\}$, $\{i_1, i_2\}$ and so on, on the regular lattices under consideration. We note that these indices run over *all* lattice sites, and that different indices may thus indicate the same lattice site. For the case of general spin quantum number s we note that we have a maximum number of spin-raising operators at any specific site l which is $2s_l$, where s_l is the spin quantum number of the spin situated at site l. For the spin-half case, we are thus limited to only *one* spin-raising operator per lattice site as required, and we note that in this manner we build in a previous spin-half high-order CCM formalism (see refs. 46, 47, 51, and 55) into the new high-order formalism for general-s directly from the outset. In order to simplify the high-order CCM formalism, it is also found to be useful to define the following operators:

$$\left.\begin{aligned}
F_k &\equiv \sum_{l} \sum_{i_2, \ldots, i_l} l\, \mathscr{S}_{k, i_2, \ldots, i_l}\, s_{i_2}^+ \cdots s_{i_l}^+ \\
G_{k,m} &\equiv \sum_{l>1} \sum_{i_3, \ldots, i_l} l(l-1)\, \mathscr{S}_{k, m, i_3, \ldots, i_l}\, s_{i_3}^+ \cdots s_{i_l}^+ \\
M_{k,m,n} &\equiv \sum_{l>2} \sum_{i_4, \ldots, i_l} l(l-1)(l-2)\, \mathscr{S}_{k, m, n, i_4, \ldots, i_l}\, s_{i_4}^+ \cdots s_{i_l}^+ \\
N_{k,m,n,p} &\equiv \sum_{l>3} \sum_{i_5, \ldots, i_l} l(l-1)(l-2)(l-3)\, \mathscr{S}_{k, m, n, p, i_5, \ldots, i_l}\, s_{i_5}^+ \cdots s_{i_l}^+
\end{aligned}\right\} \tag{13}$$

Hence, we determine the similarity-transformed expressions of the single-spin operators s^α; $\alpha \equiv \{+, -, z\}$, where $s^- \equiv (s^+)^\dagger = s^x - \mathrm{i}s^y$, by using Eq. (6) and the usual spin commutation relations, such that

$$\left.\begin{aligned} e^{-S}s_k^+e^S &\equiv \tilde{s}_k^+ = s_k^+ \\ e^{-S}s_k^ze^S &\equiv \tilde{s}_k^z = s_k^z + F_k s_k^+ \\ e^{-S}s_k^-e^S &\equiv \tilde{s}_k^- = s_k^- - 2F_k s_k^z - G_{kk}s_k^+ - (F_k)^2\, s_k^+ \end{aligned}\right\} \tag{14}$$

For the specific case of $s = 1/2$ at site k we note that $G_{kk} = 0$ because "double occupancy" of the lattice site k is prohibited in this case. In order to determine the similarity transformed version of a given Hamiltonian, we also need to know the commutation relations of the operators defined in Eq. (13) with the single-spin operators s^α; $\alpha \equiv \{+, -, z\}$, and these are stated in Appendix A.

We now define the set of CCM destruction operators $\{C_I^-\}$ (of l number of spin-lowering operators), as follows

$$C_I^- \equiv s_{j_1}^- s_{j_2}^- \cdots s_{j_l}^- \tag{15}$$

where the indices $j_1, j_2, \ldots, j_l$ represent any given lattice site. We choose only *one* of the $N_B(l!)\, \nu_I$ symmetry-equivalent configurations to *pattern-match* with the terms within $\tilde{H}$ in order to determine the CCM ket-state equations of Eq. (7). Note that N_B is the number of Bravais lattice sites and that, for a given cluster I, ν_I is a symmetry factor dependent purely on the point-group symmetries (and *not* the translational symmetries) of the crystallographic lattice.

The process of the enumeration of all possible fundamental clusters and the process of "pattern-matching" are both ideally suited to an efficient computational implementation, and a full description of these processes is also given in Appendix A. Furthermore, it is a simple matter to determine and solve the CCM bra-state equations, once the ket-state equations have been obtained and solved, as described in Appendix A, where we also explain the technicalities involved in obtaining ground-state expectation values.

In the remainder of this paper we now demonstrate the new formalism for various models of interest.

3. THE ONE-DIMENSIONAL HEISENBERG MODEL

The 1D XXZ model is defined by the following Hamiltonian,

$$H = \sum_{i=1}^{N} \{s_i^x s_{i+1}^x + s_i^y s_{i+1}^y + \Delta s_i^z s_{i+1}^z\} \tag{16}$$

where the index i in Eq. (16) runs over all N lattice sites on the linear chain. We are interested specifically in the infinite chain, $N \to \infty$. The system is by now extremely well understood for the spin-half case via the existence of the Bethe Ansatz[1–4] exact solution for this case. The exact ground-state energy for the infinite chain at $\Delta = 1$ was shown to be given by the expression, $E_g/N = 1/4 + \ln(1/2)$ (≈ -0.44314718). Indeed, the long-range Néel ordering inherent in the solution in the limit $\Delta \to \infty$ is completely destroyed by quantum fluctuations at the phase transition point at $\Delta = 1$. By contrast, no exact solution for the spin-one Heisenberg model on the infinite linear chain has as yet been found, although extremely accurate DMRG calculations have been performed[8] for this model giving a value for the ground-state energy of $E_g/N = -1.401484038971(4)$ at $\Delta = 1$. Furthermore, these calculations conclusively showed that there is an excitation energy gap of magnitude 0.41050(2) in this system, which was first postulated by Haldane.[10] This is in stark contrast to the spin-half model which is gapless, and we note that conventional spin-wave theory predicts that the excitation energy of the Heisenberg model on the infinite linear chain for both the spin-half *and* spin-one cases is gapless. For the spin-one anisotropic Heisenberg model, the isotropic Heisenberg point is in the Haldane phase, in which the amount of long-range Néel-ordering is zero and the excitation spectrum is gapped. The phase transition from the Néel-like phase occurs for a value of anisotropy $\Delta = 1.167 \pm 0.007$[11] for the spin-one XXZ model.

Interest has also been expressed recently in one-dimensional ferrimagnetic Heisenberg systems, for which case the Lieb–Mattis theorem states that the system may demonstrate a macroscopic lattice (and not sublattice) magnetization. For example, we note that for the spin-half/spin-one ferrimagnets in which alternating spins on the chain have, respectively, spin quantum numbers of $1/2$ and 1, recent DMRG calculations[9] predict that the ground-state energy of the Heisenberg model ($\Delta = 1$) is given by $E_g/N \approx -0.72704$ and the *sublattice* magnetization on the spin-one sublattice is given by 0.79248 and the *sublattice* magnetization (multiplied by a factor of 2 here – see below) on the spin-half sublattice is given by 0.58496. The phase transition is also believed to be at (or near to) the Heisenberg point ($\Delta = 1$).

The model state that we shall use in our CCM calculations here is the one-dimensional Néel state. We perform a rotation of the local spin axes of 180° about the y-axis of the spins on one sublattice only, such that the model state now appears mathematically to consist entirely of spins which points along the negative z-axis, as in Eq. (11). We note however that this unitary transformation does not affect the eigenvalue spectrum of this

problem. The Hamiltonian may now be written in terms of these local spin axes, as

$$H = -\sum_{i=1}^{N} \{\Delta s_i^z s_{i+1}^z + \tfrac{1}{2}(s_i^+ s_{i+1}^+ + s_i^- s_{i+1}^-)\} \tag{17}$$

where $s^{\pm} = s^x \pm \mathrm{i}s^y$. For the spin-half and spin-one antiferromagnets, we define the sublattice magnetization in terms of these local spin-axes as,

$$M = -\frac{1}{sN} \sum_{i=1}^{N} \langle \tilde{\Psi}| \, s_i^z \, |\Psi\rangle \tag{18}$$

where the index i runs over all N lattice sites. This reflects the fact that for the spin-half and spin-one Heisenberg models we have a unit cell which contains one single lattice site only. By contrast, for the spin-half/spin-one ferrimagnet we have a unit cell which contains two nearest-neighbour spins of quantum spin numbers $1/2$ and 1. We thus define the sublattice magnetizations M_1, on the A (spin-half) sublattice sites, and M_2, on the B (spin-one) sublattice sites, separately, in the local spin axes by

$$M_1 = -\frac{2}{N_1} \sum_{i_1}^{N_1} \langle \tilde{\Psi}| \, s_{i_1}^z \, |\Psi\rangle \qquad \text{and} \tag{19}$$

$$M_2 = -\frac{1}{N_2} \sum_{i_2}^{N_2} \langle \tilde{\Psi}| \, s_{i_2}^z \, |\Psi\rangle \tag{20}$$

where i_1 runs over all $N_1 = N/2$ spin-half sites and i_2 runs over all $N_2 = N/2$ spin-one sites. Furthermore, we note that the Lieb–Mattis theorem states that $|M_1/2 - M_2| = 1/2$ (after rotation of the local spin axes) thus allowing an overall macroscopic magnetic moment. The technicalities of determining M, M_1, and M_2 are explained in Appendix A.

We may now perform the similarity transform of the Hamiltonian in order to write that Hamiltonian, in terms of the operators F_k, $G_{k,m}$, $M_{k,m,n}$, and $N_{k,m,n,p}$ given above. By making use of Eqs. (14) and (A.8) we find that $\tilde{H}\,|\Phi\rangle \equiv e^{-S} H e^{S}\,|\Phi\rangle = (\tilde{H}_1 + \tilde{H}_2 + \tilde{H}_3)\,|\Phi\rangle$, where

$$\begin{aligned} \tilde{H}_1 \equiv -\sum_i \{ & \Delta(G_{i,i+1} + F_i F_{i+1}) + \tfrac{1}{2}(1 + N_{i,i,i+1,i+1} + 2G_{i,i+1}^2 \\ & + 2M_{i,i,i+1} F_{i+1} + 2F_i M_{i,i+1,i+1} + 4F_i F_{i+1} G_{i,i+1} + G_{i,i} G_{i+1,i+1} \\ & + G_{i,i} F_{i+1}^2 + F_i^2 G_{i+1,i+1} + F_i^2 F_{i+1}^2)\} \, s_i^+ s_{i+1}^+ \end{aligned} \tag{21}$$

$$\tilde{H}_2 \equiv -\sum_i \{(\Delta F_i + M_{i,i,i+1} + 2F_i G_{i,i+1} + G_{i,i} F_{i+1} + F_i^2 F_{i+1})\, s_i^+ s_{i+1}^z + (\Delta F_{i+1} + M_{i,i+1,i+1} + 2G_{i,i+1} F_{i+1} + F_i G_{i+1,i+1} + F_i F_{i+1}^2)\, s_i^z s_{i+1}^+ \} \tag{22}$$

$$\tilde{H}_3 \equiv -\sum_i \{\Delta + 2G_{i,i+1} + 2F_i F_{i+1}\}\, s_i^z s_{i+1}^z \tag{23}$$

Again, we note that repeated indices in the operators $G_{k,m}$, $M_{k,m,n}$, and $N_{k,m,n,p}$ operators are prohibited for the spin-half case. Hence, the general spin quantum number formalism reduces to the previous formalism determined for the spin-half case in this limit.[46, 47, 51, 55] A general feature of CCM calculations for spin systems is that we often solve the CCM at different values of some tunable parameter within the Hamiltonian. For the case of the Hamiltonian of Eq. (17) this is the anisotropy parameter, Δ, and we note that we "track" the solution from the trivial limit $\Delta \to \infty$ to lower values of Δ. Another feature of CCM calculations is that often the real solution to the CCM equations is seen to terminate at some critical value of this parameter (e.g., denoted Δ_c here), and this behaviour is taken to be a signal of a phase transition in the real system. This illustrates a strong advantage of this method: namely, that the CCM is able to make predictions for the positions of quantum phase transition points from within a fully *ab initio* framework.

4. RESULTS

In this article we utilise two approximation schemes in order to determine CCM expectation values for the ground-state energy and the sublattice magnetization. The latter gives a measure of the amount of sublattice ordering. The two schemes are, namely, the LSUB*m* and SUB*m*-*m* approximation schemes, both of which include correlations in a systematic and structured manner and provide exact results in the asymptotic limit $m \to \infty$. We note that the LSUB*m* and SUB*m*-*m* schemes are equivalent for the ferrimagnet, if the truncation index m is an even number. This is due to the restriction that $s_T^z = \sum_i s_i^z = 0$ in the ground state thus ruling out any clusters in the LSUB*m* approximation which contain more than m spin flips. This is *not* the case for the spin-one antiferromagnet, in which case the restriction $s_T^z = \sum_i s_i^z = 0$ may be satisfied for various clusters in LSUB*m* which do contain more than m spin flips.

However, we note that it is usually impossible in a practical application of this method to exactly solve for the LSUB*m* and SUB*m*-*m* schemes in the limit $m \to \infty$, and so we extrapolate the "raw" results as a function

of m in this limit. Although no rigorous extrapolation scaling laws exist, we may attempt to extrapolate these results "heuristically." Thus, for example, extrapolation of these results in the limit $m \to \infty$ at each value of Δ independently may be achieved by assuming a "power-law" dependence, hereforth denoted as the "extrapolated CCM" results, given by

$$y_i = a + bx_i^y \tag{24}$$

where y_i is a CCM expectation value at a given level of LSUBm or SUBm-m approximation and $x_i = 1/m$, and there are p data points (i.e., for the p different values of m for which calculations have been performed). The *best fit* of this data to the parametric form given in the equation above is found and the extrapolated result is thus given by a. The interested reader will find a comprehensive explanation of the extrapolation process of CCM LSUBm expectation values for the spin-half XXZ model for a variety of lattices in refs. 55 and 56.

Results for the XXZ model are presented in Figs. 1–5, and results for the Heisenberg model ($\Delta = 1$) on the linear chain are given in Tables I–III.

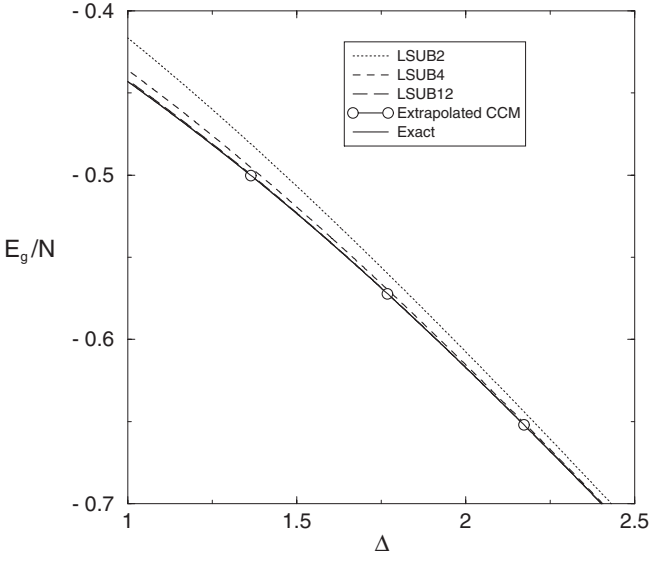

Fig. 1. CCM results for the ground-state energy per spin of the spin-half XXZ model on the linear chain using the LSUBm approximation scheme with $m = \{2, 4, 12\}$. The LSUBm results for $m = \{6, 8, 10, 12\}$ are extrapolated in the limit $m \to \infty$ for this case and are compared to exact results of the Bethe Ansatz.[1-4] (Note that the LSUBm and SUBm-m approximation schemes are equivalent for this model.)

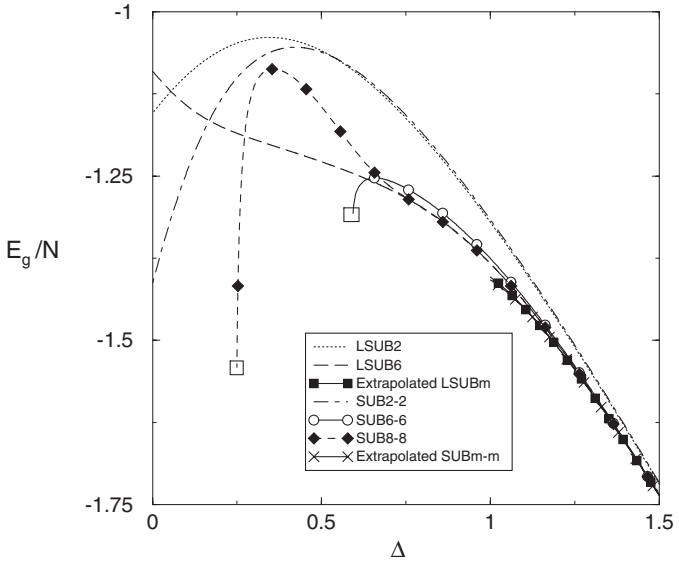

Fig. 2. CCM results for the ground-state energy per spin of the spin-one XXZ model on the linear chain using the SUBm-m approximation scheme with $m = \{2, 6, 8\}$ and the LSUBm approximation scheme with $m = \{2, 6\}$. LSUBm and SUBm-m results are extrapolated in the limit $m \to \infty$ for $\Delta \geqslant 1$. Note that boxes indicate the CCM critical points.

Table I. Results Obtained for the Spin-Half Heisenberg Model on the Linear Chain Using the CCM LSUBm Approximation Scheme with $m = \{2, 4, 6, 8, 10, 12\}$. N_F Denotes the Number of Fundamental Configurations for the Ground State. The Ground-State Energy per Spin, E_g/N, and the Sublattice Magnetization, M Are Shown. The LSUBm Results for $m = \{6, 8, 10, 12\}$ Are Extrapolated in the Limit $m \to \infty$ for this Case and Are Compared to Exact Results of the Bethe Ansatz.[1–4] (Numbers in Brackets for the Extrapolated CCM Results Indicate the Estimated Error in the Last Significant Figure Shown. Note that the LSUBm and SUBm-m Approximation Schemes Are Equivalent for this Model.)

	N_F	E_g/N	M
LSUB2	1	−0.416667	0.666667
LSUB4	3	−0.436270	0.496776
LSUB6	9	−0.440024	0.415771
LSUB8	26	−0.441366	0.365943
LSUB10	81	−0.441995	0.331249
LSUB12	267	−0.442340	0.305254
Extrapolated CCM	–	−0.44315084(6)	−0.01876(3)
Bethe Ansatz	–	−0.443147	0.0

Table II. Results Obtained for the Spin-One Heisenberg Model on the Linear Chain Using the LSUBm Approximation Scheme with $m=\{2, 4, 6\}$ and SUBm-m Approximation Scheme with $m=\{2, 4, 6, 8, 10\}$. N_F Denotes the Number of Fundamental Configurations for the Ground State. The Ground-State Energy per Spin, E_g/N, the Sublattice Magnetization, M, and the Critical Values of Δ_c Are Shown. The SUBm-m Results for $m=\{4, 6, 8\}$ and LSUBm Results for $m=\{2, 4, 6\}$ Are Extrapolated in the Limit $m\to\infty$ in this Case, and Are Compared to Results†† of the DMRG Method.[8] The ** Symbol Indicates that Extrapolated CCM Results for the Sublattice Magnetization, M, Go to Zero at Δ_c=1.19 and Δ_c=1.18 for the SUBm-m and LSUBm Approximations, Respectively, († Also Given in this Table) and Are Thus Ill-Defined Below These Points. The Result* for the Position of the Phase Transition Point Using the Large-Cluster-Decomposition Monte Carlo Method[11] Is Also Quoted

	N_F	E_g/N	M	Δ_c
SUB2-2	1	-1.316625	0.809068	–
SUB4-4	7	-1.360084	0.694610	–
SUB6-6	37	-1.375607	0.607339	0.593
SUB8-8	247	-1.383466	0.533252	0.249
Extrapolated SUBm-m	–	-1.408039	**	$1.19^{\dagger}$
LSUB2	2	-1.320608	0.800702	–
LSUB4	11	-1.369428	0.646536	–
LSUB6	63	-1.383292	0.532198	-0.670
Extrapolated LSUBm	–	-1.403737	**	$1.18^{\dagger}$
cf.	–	$-1.401484038971(4)^{\dagger\dagger}$	0.0	1.167(7)*

We may see from Figs. 1 and 2 for the spin-half and spin-one XXZ models that our results for the ground-state energy are highly converged, and analogous behaviour is seen for the ground-state energies of the spin-half/spin-one ferrimagnet. Indeed, we may see from Tables I–III that our CCM results for the Heisenberg model ($\Delta=1$) converge rapidly in each case, and that they agree well with their respective exact or DMRG results. We note that the extrapolated CCM results for the spin-one Heisenberg model seem to agree with DMRG results to only two decimal places, whereas the extrapolated CCM results for the spin-half Heisenberg model appear to agree with exact Bethe Ansatz results to about five decimal places. The apparent discrepancy is understood by noting that we are well into the Haldane phase at $\Delta=1$ for the spin-one model and the fact that we obtain results at this point at all is a testament to the power of the CCM. We note however that other model states might be employed at the Heisenberg point for the spin-one model in order to obtain even more accurate results for this model. Inspection of Table III (for $\Delta=1$) indicate that our "raw" (unextrapolated) CCM results for the ground-state energy of the spin-half/spin-one ferrimagnet appear already to be converged to *at*

Table III. Results Obtained for the Spin-Half/Spin-One Ferrimagnetic HAF on the Linear Chain Using the CCM SUB*m*-*m* Approximation Scheme with $m=\{2, 4, 6, 8, 10\}$ Compared to Results of DMRG[9] Calculations. CCM Results for $m=\{6, 8, 10\}$ Are Extrapolated in the Limit $m \to \infty$. N_F Denotes the Number of Fundamental Configurations for the Ground State. The Ground-State Energy per Spin, E_g/N, the Sublattice Magnetization on the Spin-Half Sites, M_1, the Sublattice Magnetization on the Spin-One Sites, M_2 Are Shown. (Note that the LSUB*m* and SUB*m*-*m* Approximation Schemes Are Equivalent at the Levels of Approximation Shown for this Model.)

	N_F	E_g/N	M_1	M_2
SUB2-2	1	−0.70710678	0.70710678	0.85355339
SUB4-4	5	−0.72582592	0.59865621	0.79932811
SUB6-6	21	−0.72697237	0.58611255	0.79305628
SUB8-8	93	−0.72704344	0.58503667	0.79251834
SUB10-10	427	−0.72704696	0.58497261	0.79248630
Extrapolated CCM	–	−0.7270474	0.5849641	0.7924820
DMRG	–	−0.72704	0.58496	0.79248

least five decimal places. We expect that the CCM results are even more accurate for $\Delta > 1$ because quantum fluctuations are (comparatively) weaker in this regime than at the isotropic Heisenberg point at $\Delta = 1$. We furthermore expect that our extrapolated CCM value at $\Delta = 1$ given in Table III represents an even better result than the raw SUB*m*-*m* results. Indeed, we note that all of our CCM results are in excellent agreement with those result of the DMRG method[9] at this point.

Figures 3 and 4 present results for the sublattice magnetization, M, for the spin-half and spin-one XXZ models in the Néel-ordered regime. The extrapolated CCM results for the spin-half XXZ model are clearly in excellent agreement with exact results and we note in particular that the sublattice magnetization goes to zero at $\Delta = 1.07$. This is in good agreement with the exact behaviour of the model and we note that there is an infinite-order phase transition at $\Delta = 1$ to disordered phase for $\Delta \leqslant 1$ in the "real" system. Similarly, the extrapolated CCM results for both the SUB*m*-*m* and LSUB*m* approximation schemes for the spin-one XXZ model go to zero at $\Delta \approx 1.19$ and $\Delta \approx 1.18$, respectively. This is in excellent agreement with the position for the onset of the Haldane phase of $\Delta = 1.167 \pm 0.007$.[11] We furthermore note that even greater accuracy for the CCM extrapolated results for both the spin-half and spin-one antiferromagnets is expected with higher levels of approximation. We also note that these results are a clear indication that the CCM is capturing the qualitative difference in the behaviour of the sublattice magnetizations between the spin-half and spin-one antiferromagnetic XXZ models.

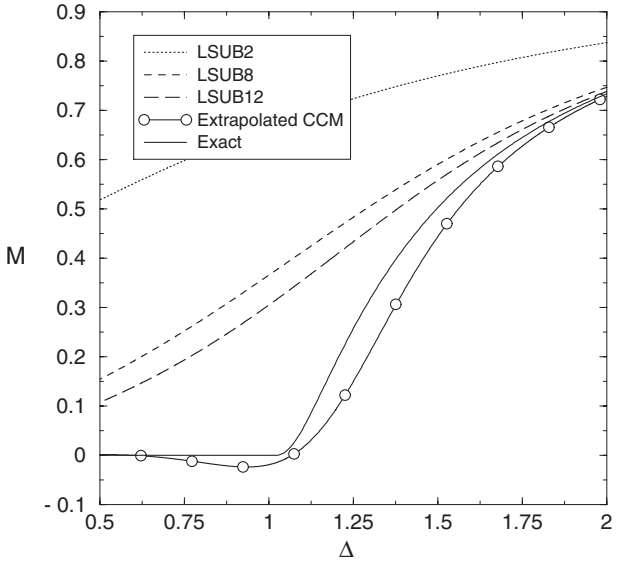

Fig. 3. CCM results for the sublattice magnetization of the spin-half XXZ model on the linear chain using the LSUBm approximation scheme with $m = \{2, 8, 12\}$. The LSUBm results for $m = \{6, 8, 10, 12\}$ are extrapolated in the limit $m \to \infty$ and are compared to exact results of Bethe Ansatz.[1–4] Note that the extrapolated CCM results follow the qualitative behaviour of the exact results quite closely and that the extrapolated results go to zero at $\Delta = 1.07$.

Furthermore, an overall magnetic moment is possible for the case of the spin-half/spin-one ferrimagnet and we may see from Fig. 5 and Table III that the CCM results for the amount of sublattice ordering on both sublattices, namely, M_1 and M_2, are extremely well converged over the whole of the Néel-like regime. Indeed, we note that even though the values for M_1 and M_2 are determined separately using the CCM, they still obey relationship $|M_1/2 - M_2| = 1/2$ specified by the Lieb–Mattis theorem. It is furthermore not *a priori* evident that this ought to be the case as we have not artificially constrained this to be true in our CCM calculation at any point. It is thus a reflection that the CCM is detecting the underlying nature of the ground state from within an *ab initio* framework. We also see from Table III that the CCM results agree with DMRG results to (at least) about five decimal places for the isotropic model. Furthermore, we believe that the extrapolated CCM results for the sublattice magnetizations at the Heisenberg point at $\Delta = 1$ present even more accurate estimates of these quantities. We note that the results for the sublattice magnetizations of the spin-half/spin-one ferrimagnet remain non-zero and finite over the whole of the regime $\Delta \geqslant 1$, although it is possible that a phase transition to a

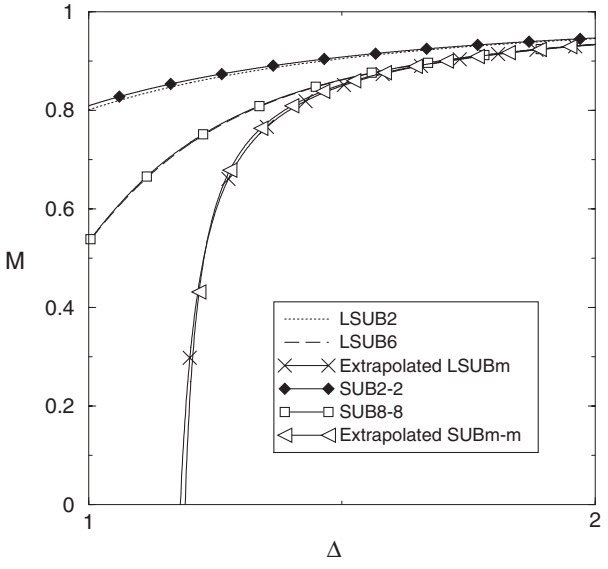

Fig. 4. CCM results for the sublattice magnetization of the spin-one XXZ model on the linear chain using the LSUBm approximation scheme with $m = \{2, 6\}$ and SUBm-m approximation scheme with $m = \{2, 8\}$. Results for the LSUBm approximation scheme with $m = \{2, 4, 6\}$ and SUBm-m approximation scheme with $m = \{2, 8\}$ are extrapolated in the limit $m \to \infty$. Note that the extrapolated CCM results go to zero at $\Delta \approx 1.19$ and $\Delta \approx 1.18$ for the SUBm-m and LSUBm approximation schemes, respectively.

regime in which spins lie in the xy-plane (and for which $M_1 = M_2 = 0$) occurs for $\Delta \approx 1$ (for example, see refs. 48 and 51). (We note that we would probably employ a model state in which spin lie in the xy-plane for the CCM in the region $\Delta \leqslant 1$.) Our highly converged CCM results thus indicate that if the phase transition does indeed occur near to $\Delta \approx 1$ then this might involve a stepwise change in the sublattice magnetizations from a non-zero (and finite) value to zero at this point. If this conjecture were true then this behaviour would furthermore be consistent with a first-order phase transition at this point.

Finally, we note that the CCM critical points, Δ_c, are observed for the spin-one antiferromagnetic XXZ model (illustrated in Table II) at which the real (i.e., physical) solution to the CCM equations breaks down at given level of LSUBm or SUBm-m approximation level. This behaviour is an indication that a phase transition occurs in the "real" system. We note that an alternative and independent indication of the position of the phase transition is afforded by the value of Δ_c at which the sublattice magnetization goes to zero (discussed above). No equivalent results for the critical

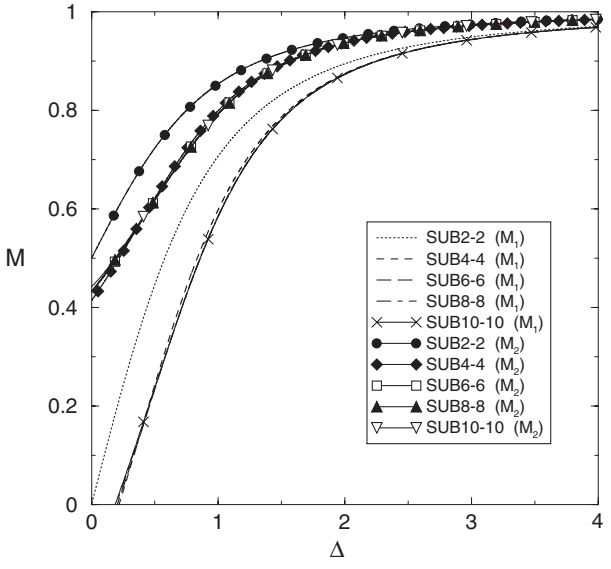

Fig. 5. CCM results for the sublattice magnetization of the spin-half spins, M_1, and of the spin-one spins, M_2, for the spin-half/spin-one XXZ ferrimagnet on the linear chain using SUBm-m approximation scheme with $m = \{2, 4, 6, 8, 10\}$. Note that the LSUBm and SUBm-m approximation schemes are equivalent for this model at the levels of truncation index m shown.

points of the ferrimagnetic XXZ model were obtained, although CCM calculations for this model show characteristic peaks in the derivatives of the ground-state energy for $\Delta \leqslant 1$. This behaviour might possibly again be interpreted as a signature of a phase transition. It is also quite possible however that this typical breakdown of the real solution of the CCM equations might however occur at higher orders of approximation.[48]

5. CONCLUSIONS

In this article a high-order CCM ground-state formalism for lattices comprised of spins with general spin quantum number s was presented. We have shown that this new formalism is highly suitable for computational implementation for localised approximation schemes (namely, the LSUBm and the SUBm-m approximation schemes). In order to demonstrate the effectiveness of the formalism, successful applications of the general-s formalism were made to the one-dimensional XXZ model for the spin-half and spin-one antiferromagnetic cases and the spin-half/spin-one ferrimagnetic case. Our extrapolated results were seen to agree extremely well with

exact results for the spin-half antiferromagnetic XXZ model, up to and including the phase transition point at $\Delta = 1$. Our extrapolated results for the spin-one antiferromagnetic XXZ model predicted that the sublattice magnetization goes to zero at $\Delta \approx 1.2$. This result is in excellent agreement with the value for the onset of the Haldane phase of $\Delta = 1.167 \pm 0.007$[11] for this model. Finally, CCM results for the sublattice magnetization (on both sublattices) of the spin-half/spin-one ferrimagnet were seen to be extremely well converged as a function of the truncation index m, and these results were finite and non-zero over a wide range of Δ, up to and including the Heisenberg point at $\Delta = 1$.

Further applications of the new high-order general-s CCM formalism to the zero-temperature properties of lattice quantum spin systems are also envisaged for the future. In particular, it is expected that such techniques will be applied to highly frustrated cases with spatial dimensionality greater than one which are difficult (if not impossible) to treat using other approximate theories. It is also straightforward to extend the ground-state formalism presented here to deal with excited states, as has been done previously for the spin-half case.[55, 57]

APPENDIX A

A.1. Commutation Relations and the High-Order General-s CCM Formalism

In this article we present a new formalism and results for high-order ground-state CCM calculations for general spin quantum number, s, based on a model state in which all spins on the crystallographic lattice point downwards along the local z-axes. A large part of the new formalism relies on the new "high-order" CCM operators defined by Eq. (13) and also their commutation relations with the single-spin operators in order to determine the similarity transforms of various operators, such as the Hamiltonian for example. In order to determine these commutation relations we firstly remind ourselves that the ket-state correlation operator S is given by Eq. (2) with $C_I^+ \equiv s_{i_1}^+ s_{i_2}^+ \cdots s_{i_l}^+$ and $\mathscr{S}_I \equiv \mathscr{S}_{i_1, i_2, \ldots, i_l}$, and hence

$$S = \sum_l \sum_{i_1, i_2, \ldots, i_l} \mathscr{S}_{i_1, i_2, \ldots, i_l} \, s_{i_1}^+ s_{i_2}^+ \cdots s_{i_l}^+ \tag{A.1}$$

where each of the indices $\{i_1, i_2, \ldots, i_l\}$ runs over all lattice sites with the condition that there can be no more than $2s$ of them at any particular

lattice site. The usual spin commutation relations of the spin operators also apply,

$$[s_l^+, s_{l'}^-] = 2s_l^z \delta_{l,l'}; \qquad [s_l^z, s_{l'}^\pm] = \pm s_l^\pm \delta_{l,l'} \tag{A.2}$$

We also note that the commutation of a given operator with S must be distributive, such that

$$[s_k^\alpha, S] = \sum_l \sum_{i_1, i_2, \ldots, i_l} \mathscr{S}_{i_1, i_2, \ldots, i_l} \{[s_k^\alpha, s_{i_1}^+]\, s_{i_2}^+ \cdots s_{i_l}^+ + s_{i_1}^+ [s_k^\alpha, s_{i_2}^+]\, s_{i_3}^+ \cdots s_{i_l}^+ + \cdots + s_{i_1}^+ s_{i_2}^+ \cdots [s_k^\alpha, s_{i_l}^+]\} \tag{A.3}$$

where $\alpha = \{z, +, -\}$. As pairs of spin-raising operators always commute, we may therefore state that $[s_k^+, S] = 0$. Furthermore, for the case of $[s_k^z, S]$ we note again that each index runs over all lattice sites, which implies that each term on the right-hand side of Eq. (A.3) is equivalent and that, as there are l such terms, we may write this expression as

$$[s_k^z, S] = \sum_l \sum_{i_2, \ldots, i_l} l\, \mathscr{S}_{k, i_2, \ldots, i_l} s_{i_2}^+ \cdots s_{i_l}^+ s_k^+ = F_k s_k^+ \tag{A.4}$$

Note again that the "high-order" operators such as F_k are defined by Eq. (13). We lastly calculate the commutator $[s_k^-, S]$ in Eq. (A.3), and using the basic commutation relations of Eq. (A.2) we thus have

$$[s_k^-, S] = -2 \sum_l \sum_{i_1, i_2, \ldots, i_l} \mathscr{S}_{i_1, i_2, \ldots, i_l} \{\delta_{k, i_1} s_k^z s_{i_2}^+ \cdots s_{i_l}^+ + \delta_{k, i_2} s_{i_1}^+ s_k^z s_{i_3}^+ \cdots s_{i_l}^+ + \cdots + \delta_{k, i_l} s_{i_1}^+ s_{i_2}^+ \cdots s_k^z\} \tag{A.5}$$

We now commute the operator s_k^z past the strings of spin-raising operators in Eq. (A.5) using the basic commutation relations of Eq. (A.2). Thus, for example,

$$s_k^z s_{i_2}^+ s_{i_3}^+ \cdots s_{i_l}^+ = (\delta_{k, i_2} + \delta_{k, i_3} + \cdots \delta_{k, i_l})\, s_{i_2}^+ s_{i_3}^+ \cdots s_{i_l}^+ + s_{i_2}^+ s_{i_3}^+ \cdots s_{i_l}^+ s_k^z \tag{A.6}$$

By inserting Eq. (A.6) into Eq. (A.5) we find that

$$\begin{aligned} [s_k^-, S] &= -2 \sum_l \sum_{i_3, i_4, \ldots, i_l} \left(\sum_{n=1}^{l-1} \right) \mathscr{S}_{k, k, i_3, \ldots, i_l}\, s_{i_3}^+ s_{i_4}^+ \cdots s_{i_l}^+ s_k^+ \\ &\quad -2 \sum_l \sum_{i_2, i_3, \ldots, i_l} l \mathscr{S}_{k, i_2, \ldots, i_l}\, s_{i_2}^+ s_{i_3}^+ \cdots s_{i_l}^+ s_k^z \\ &= -G_{k,k} s_k^+ - 2F_k s_k^z \end{aligned} \tag{A.7}$$

using the definitions in Eq. (13). We note again that for the case $s = 1/2$ the operator $G_{k,k} \equiv 0$.

By making use of the nested commutator expansion for the similarity-transformed operators [cf., Eq. (10)], it is now a simple matter to verify the relations in Eq. (14), by using Eqs. (A.4) and (A.7).

In order to determine the similarity transform of the Hamiltonian however it is also necessary to also know the commutation relations of the single-spin operators with respect to F_k, F_k^2, $G_{k,m}$, and $M_{k,m,n}$. The proofs of these commutation relations follow a similar pattern to the proofs given above, and so we merely state them here:

$$\left.\begin{aligned}
[s_k^z, F_m] &= G_{k,m} s_k^+, \\
[s_k^z, G_{m,n}] &= M_{k,m,n} s_k^+, \\
[s_k^z, F_m^2] &= 2F_m G_{k,m} s_k^+, \\
[s_k^-, F_m] &= -2G_{k,m} s_k^z - M_{k,k,m} s_k^+, \\
[s_k^-, F_m^2] &= -2G_{k,m}^2 s_k^+ - 2F_m M_{k,k,m} s_k^+ - 4F_m G_{k,m} s_k^z, \\
[s_k^z, M_{m,n,p}] &= N_{k,m,n,p} s_k^+, \\
[s_k^-, G_{m,n}] &= -2M_{k,m,n} s_k^z - N_{k,k,m,n} s_k^+
\end{aligned}\right\} \qquad \text{(A.8)}$$

We note once more that the operators F_k, $G_{k,m}$, $M_{k,m,n}$, and $N_{k,m,n,p}$ are defined by Eq. (13).

A.2. Enumeration of the Fundamental Clusters

At a given level of approximation, we choose only *one* of the $N_B(l!)\, v_I$ possible symmetry-equivalent configurations for a given *fundamental* configuration of l spin-raising operators, where N_B is the total number of Bravais lattice sites, and where v_I is a symmetry factor dependent purely on the point-group symmetries (and *not* the translational symmetries) for the crystallographic lattice in question and for fundamental configuration I. We note that there are N_F such *fundamental* configurations. The first part of the computational algorithm is to enumerate all of the "lattice animals" which define the "locale" in which the clusters must lie. For the levels of approximation shown in this article it is possible to do this by using a simple recursive algorithm which enumerates all possible lattice animals of m contiguous sites. This "locale" is explicitly assumed here to be the same for both the LSUBm and SUBn-m approximation schemes. Secondly, one then needs to enumerate all possible ways in which one can place $(2s)$ or less spin-raising operators on each of the positions of the m sites within each of these lattice animals. There are thus $(2s)^m$ possibilities for each

lattice animal. However, one must also restrict the total number of spin-raising operators to be less than or equal to n for the SUBn-m approximation scheme. We note however that there is no such restriction on the total number of spin-raising operators for the LSUBm approximation. This process thus enumerates all possible connected and disconnected clusters, and we make a restriction that we include only those clusters which are inequivalent under the point and space group symmetries of both the lattice and the Hamiltonian. A further restriction for the systems under consideration in this article is that we must restrict the set of fundamental clusters to include only those which preserve the relationship, $s_T^z = \sum_i s_i^z = 0$, with respect to the original ("unrotated") Néel model state since $[s_T^z, H] = 0$ and the ground state lies in the $s_T^z = 0$ sector.

A.3. The Ket-State Equations

We now wish to determine the CCM ket-state equations, where the Ith such equation is given by

$$E_I \equiv \frac{1}{A_I} \langle \Phi | C_I^- e^{-S} H e^S | \Phi \rangle = 0, \qquad \forall I \neq 0 \tag{A.9}$$

where A_I is a normalization factor given by $A_I \equiv \langle \Phi | C_I^- C_I^+ | \Phi \rangle = \langle \Phi | (s_{i_1}^- s_{i_2}^- \cdots s_{i_l}^-)(s_{i_1}^+ s_{i_2}^+ \cdots s_{i_l}^+) | \Phi \rangle$. We note once more that we choose only *one* of the $N_B(l!)\, v_I$ possible symmetry-equivalent configurations for a given *fundamental* configuration in C_I^- in order to pattern-match with the terms within $\tilde{H} | \Phi \rangle$ and thus determine the Ith CCM ket-state equation. We then computationally match the individual spin-lowering operators in C_I^-, defined by Eq. (15), to the spin-raising operators in $\tilde{H} | \Phi \rangle$. We therefore put constraints on the indices in the CCM ket-state correlation coefficients, $\{\mathscr{S}_{i_1, i_2, \ldots, i_l}\}$, and these constraints on the indices allow us to enumerate all possible terms which contribute to the CCM ket-state equations. For example, we may consider a specific term in the evaluation of the CCM ket-state equations, given by

$$\begin{aligned} &\langle \Phi | C_I^- \tilde{s}_k^z \tilde{s}_m^z | \Phi \rangle \\ &\quad = \langle \Phi | C_I^- (s_k^z s_m^z + F_m s_m^+ s_k^z + F_k s_k^+ s_m^z + G_{k,m} s_k^+ s_m^+ + F_k F_m s_k^+ s_m^+) | \Phi \rangle \end{aligned} \tag{A.10}$$

For the case of the linear chain Heisenberg model we let k run over all lattice sites on the 1D chain and we set $m = i + 1$. Now consider a specific term within Eq. (A.10), given by $\langle \Phi | C_I^- F_k F_m s_k^+ s_m^+ | \Phi \rangle$. We match the indices of

the spin-lowering operators in C_I^- to the spin raising-operators in F_k, F_m, s_k^+, and s_m^+. Hence, both k and m are fully constrained to take on site values dependent on those indices of the spin-lowering operators in the fundamental configuration chosen for C_I^-. We note however that one may not always have such complete constraints on the indices k and m in $\tilde{H}$. For example, we may attempt to evaluate such a term as, $\langle \Phi | C_I^- F_k F_m s_k^z s_m^z | \Phi \rangle$. In this case, both k and m are free to cover all lattice sites *independently* from the fundamental cluster utilised in C_I^-. However, we may retain only those configurations in the set $\{\mathscr{S}_{i_1, i_2, \ldots, i_l}\}$ in F_k and F_m which are equivalent to the fundamental set of configurations under the symmetries of the lattice. This condition is sufficient to render the computational problem both tractable and efficient. Finally, the resulting coupled, non-linear CCM ket-state equations are easily solved computationally (for example, via the Newton–Raphson method) at a given value of the anisotropy parameter Δ.

A.4. The Bra-State Equations

The bra-state coefficients $\mathscr{S}_I$ of Eq. (2) are formally determined by Eq. (8). However, this form of the bra-state equations is slightly cumbersome to use, and a simpler and more elegant approach is possible by defining the following *new* set of CCM correlation coefficients given by

$$\left.\begin{aligned} x_I &= \mathscr{S}_I \\ \tilde{x}_I &= \frac{N_B}{N} \tilde{\mathscr{S}}_I A_I v_I (l!) \end{aligned}\right\} \tag{A.11}$$

where again $A_I \equiv \langle \Phi | C_I^- C_I^+ | \Phi \rangle = \langle \Phi | (s_{i_1}^- s_{i_2}^- \cdots s_{i_l}^-)(s_{i_1}^+ s_{i_2}^+ \cdots s_{i_l}^+) | \Phi \rangle$. Note that N_B is the number of Bravais lattice sites. Note also that for a given cluster I then v_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 A_I, v_I, and N_B however do not need to be explicitly determined because they *always* cancel out when obtaining ground-state expectation values (see below). The CCM bra-state operator may thus be rewritten as

$$\tilde{S} \equiv 1 + N \sum_{I=1}^{N_F} \frac{\tilde{x}_I}{A_I} C_I^- \tag{A.12}$$

such that

$$\bar{H} = E_0 + N \sum_{I}^{N_F} \tilde{x}_I E_I \tag{A.13}$$

We note again that the ground-state energy expectation value is defined by $E_0 = \langle \Phi | \, e^{-S} H e^{S} \, | \Phi \rangle$ and that E_I is the Ith CCM ket-state equation defined by Eq. (A.9). The CCM ket-state equations are easily rederived by taking the partial derivative of $\bar{H}/N$ with respect to $\tilde{x}_I$, where

$$0 = \frac{\delta(\bar{H}/N)}{\delta \tilde{x}_I} \equiv E_I \tag{A.14}$$

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

$$0 = \frac{\delta(\bar{H}/N)}{\delta x_I} = \frac{\delta(E_0/N)}{\delta x_I} + \sum_{J=1}^{N_F} \tilde{x}_J \frac{\delta E_J}{\delta x_I} \tag{A.15}$$

This equation is easily solved computationally, once the CCM ket-state equations have been determined and solved, and 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 never need to explicitly determine the factors N_B, A_I, or v_I.

A.5. 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 $\bar{H}$. For example, the sublattice magnetization for the spin-half and spin-one antiferromagnets of Eq. (18) may be written as

$$\begin{aligned} M &= -\frac{1}{sN} \sum_{i=1}^{N} \langle \tilde{\Psi} | \, s_i^z \, | \Psi \rangle \\ &= 1 - \frac{1}{s} \sum_{I=1}^{N_F} \frac{\tilde{x}_I}{A_I} \langle \Phi | \, C_I^- \sum_{i=1}^{N} (F_i s_i^+) \, | \Phi \rangle \\ &= 1 - \frac{1}{s} \sum_{I=1}^{N_F} l(l!) \, \tilde{x}_I x_I \end{aligned} \tag{A.16}$$

where i runs over all lattice sites. We again note that the factors A_I and v_I in Eq. (A.16) have cancelled out. Equation (A.16) is easily evaluated once the ket- and bra-state equations have been solved at a given value of the anisotropy parameter Δ. (Results for both the ground-state energy and the sublattice magnetization are given in Section 3 of this article.)

The situation for the ferrimagnet is slightly different, because the unit cell now contains two spins. Thus the magnetization on N_1 the spin-half sites ($s = 1/2$) is given by,

$$\begin{aligned} M_1 &= -\frac{1}{sN_1} \sum_{i_1}^{N_1} \langle \tilde{\Psi}| \, s_{i_1}^z \, |\Psi\rangle \\ &= 1 - \frac{N}{sN_1} \sum_{I=1}^{N_F} \frac{\tilde{x}_I}{A_I} \langle \Phi| \, C_I^- \sum_{i_1=1}^{N_1} (F_{i_1} s_{i_1}^+) \, |\Phi\rangle \\ &= 1 - 4 \sum_{I=1}^{N_F} \frac{\tilde{x}_I}{A_I} \langle \Phi| \, C_I^- \sum_{i_1=1}^{N_1} (F_{i_1} s_{i_1}^+) \, |\Phi\rangle \end{aligned} \tag{A.17}$$

Note that i_1 runs over all $N_1 (= N/2)$ spin-half lattice sites, such that we have the factor $\frac{N}{sN_1} = 4$. It is a simple matter to explicitly enumerate all combinations of orderings of the l spin-raising operators in $\sum_{i_1=1}^{N_1} F_{i_1} s_{i_1}^+$ which match with spin-lowering operators in C_I^-, although we must also explicitly restrict i_1 to be a spin-half site. (We note that the factors A_I again cancel out with coefficients in $\langle \Phi| \, C_I^- \sum_{i_1=1}^{N_1} (F_{i_1} s_{i_1}^+) \, |\Phi\rangle$ at this point.) A similar expression may be obtained for M_2.

We may also determine numerical values for other expectation values, such as the spin-spin correlation function, given by

$$M_r^{zz} \equiv \frac{1}{N} \sum_{k=1}^{N} \langle \tilde{\Psi}| \, \tilde{s}_k^z \tilde{s}_{k+r}^z \, |\Psi\rangle$$

We see from Eq. (A.10) above that this expression may be written in terms of our high-order CCM operators as

$$\begin{aligned} M_r^{zz} = \langle \Phi| \left\{ \frac{1}{N} + \sum_{I=1}^{N_F} \frac{\tilde{x}_I}{A_I} C_I^- \right\} \sum_{k=1}^{N} & (s_k^z s_{k+r}^z + F_m s_{k+r}^+ s_k^z + F_k s_k^+ s_{k+r}^z \\ & + G_{k,\,k+r} s_k^+ s_{k+r}^+ + F_k F_{k+r} s_k^+ s_{k+r}^+) \, |\Phi\rangle \end{aligned} \tag{A.18}$$

The right-hand-side of Eq. (A.18) may be evaluated computationally in exactly the same manner as for the ket-state equations, although no results for spin-spin correlation functions are quoted in this article.

ACKNOWLEDGMENTS

We thank Dr. Chen Zeng for his useful and interesting discussions. One of us (K.A.G.) gratefully acknowledges a research grant (GR/M45429)

from the Engineering and Physical Sciences Research Council (EPSRC) of Great Britain. This work has also been supported by the Deutsche Forschungsgemeinschaft (GRK 549, Graduiertenkolleg on "Accentric Crystals" and also Project No. RI 615/9-1).

REFERENCES

1. H. A. Bethe, *Z. Phys.* **71**:205 (1931).
2. L. Hulthén, *Ark. Mat. Astron. Fys. A* **26**:No. 11 (1938).
3. R. Orbach, *Phys. Rev.* **112**:309 (1958); C. N. Yang and C. P. Yang, *ibid.* **150**:321 (1966); *ibid.* **150**:327 (1966).
4. J. Des Cloiseaux and J. J. Pearson, *Phys. Rev.* **128**:2131 (1962); L. D. Faddeev and L. A. Takhtajan, *Phys. Lett. A* **85**:375 (1981).
5. S. R. White and R. M. Noack, *Phys. Rev. Lett.* **68**:3487 (1992).
6. S. R. White, *Phys. Rev. Lett.* **69**:2863 (1992).
7. S. R. White, *Phys. Rev. B* **48**:10345 (1993).
8. S. R. White and D. A. Huse, *Phys. Rev. B* **48** 3844 (1993).
9. S. K. Pati, S. Ramasesha, and D. Sen, *J. Phys.: Condens. Matter* **9**:8707 (1997).
10. F. D. M. Haldane, *Phys. Lett. A* **93**:464 (1983).
11. K. Nomura, *Phys. Rev. B* **40**:9142 (1989).
12. M. H. Kalos, *Phys. Rev.* **128**:1791 (1962).
13. D. M. Ceperley and M. H. Kalos, in *Monte Carlo Methods in Statistical Physics*, K. Binder, ed. (Springer Verlag, Berlin, 1979), p. 145.
14. K. Schmidt and M. H. Kalos, in *Applications of the Monte Carlo Method in Statistical Physics*, K. Binder, ed. (Springer Verlag, Berlin, 1984), p. 125.
15. R. Guardiola, in *Microscopic Quantum-Many-Body Theories and Their Applications*, J. Navarro and A. Polls, eds., by Lecture Notes in Physics, Vol. 510 (Springer-Verlag, Berlin, 1998), p. 269.
16. T. Barnes, D. Kotchan, and E. S. Swanson, *Phys. Rev. B* **39**:4357 (1989).
17. J. Carlson, *Phys. Rev. B* **40**:846 (1989); N. Trivedi and D. M. Ceperley, *ibid.* **41**:4552 (1990).
18. K. J. Runge, *Phys. Rev. B* **45**:12292 (1992); *ibid.* **45**:7229 (1992).
19. A. W. Sandvik, *Phys. Rev. B* **56**:11678 (1997).
20. P. W. Anderson, *Phys. Rev.* **86**:694 (1952); T. Oguchi, *Phys. Rev.* **117**:117 (1960).
21. C. J. Hamer, Z. Weihong, and P. Arndt, *Phys. Rev. B* **46**:6276 (1992).
22. N. B. Ivanov *Phys. Rev. B* **57**:14024R (1998).
23. S. Tang and J. E. Hirsch, *Phys. Rev. B* **39**:4548 (1989).
24. D. D. Betts and G. E. Stewart, *Can. J. Phys.* **75**:47 (1997).
25. R. R. P. Singh, *Phys. Rev. B* **39**:9760 (1989).
26. R. R. P. Singh and D. A. Huse, *Phys. Rev. B* **40**:7247 (1989).
27. Z. Weihong, J. Oitmaa, and C. J. Hamer, *Phys. Rev. B* **43**:8321 (1991).
28. R. F. Bishop, D. J. J. Farnell, and J. B. Parkinson, *Phys. Rev. B* **61**:6775 (2000).
29. R. F. Bishop and D. J. J. Farnell, in *Advances in Quantum Many-Body Theory*, Vol. 3, Raymond F. Bishop, Klaus A. Gernoth, Niels R. Walet, and Yang Xian, eds. (World Scientific, Singapore), in press.
30. F. Coester, *Nucl. Phys.* **7**:421 (1958); F. Coester and H. Kümmel, *ibid.* **17**:477 (1960).
31. H. Kümmel, K. H. Lührmann, and J. G. Zabolitzky, *Phys Rep. C* **36**:1 (1978).
32. R. F. Bishop and K. H. Lührmann, *Phys. Rev. B* **17**:3757 (1978).

33. J. S. Arponen, *Ann. Phys. (N.Y.)* **151**:311 (1983).
34. J. S. Arponen, R. F. Bishop, and E. Pajanne, *Phys. Rev. A* **36**:2519 (1987); *ibid.* **36**:2539 (1987); *ibid.* **37**:1065 (1988).
35. R. F. Bishop, *Theor. Chim. Acta* **80**:95 (1991).
36. J. S. Arponen, and R. F. Bishop, *Ann. Phys. (N.Y.)* **207**:171 (1991); *ibid.* **227**:275 (1993); *ibid.* **227**:2334 (1993).
37. R. F. Bishop, in *Microscopic Quantum-Many-Body Theories and Their Applications*, J. Navarro and A. Polls, eds., Lecture Notes in Physics, Vol. 510 (Springer-Verlag, Berlin, 1998), p. 1.
38. K. Emrich, *Nucl. Phys. A* **351**:379, 397 (1981).
39. M. Roger and J. H. Hetherington, *Phys. Rev. B* **41**:200 (1990); M. Roger and J. H. Hetherington, *Europhys. Lett.* **11**:255 (1990).
40. R. F. Bishop, J. B. Parkinson, and Y. Xian, *Phys. Rev. B* **44**:9425 (1991).
41. R. F. Bishop, J. B. Parkinson, and Y. Xian, *Phys. Rev. B* **46**:880 (1992).
42. R. F. Bishop, J. B. Parkinson, and Y. Xian, *J. Phys.: Condens. Matter* **5**:9169 (1993).
43. D. J. J. Farnell and J. B. Parkinson, *J. Phys.: Condens. Matter* **6**:5521 (1994).
44. Y. Xian, *J. Phys.: Condens. Matter* **6**:5965 (1994).
45. R. Bursill, G. A. Gehring, D. J. J. Farnell, J. B. Parkinson, T. Xiang, and C. Zeng, *J. Phys.: Condens. Matter* **7**:8605 (1995).
46. R. G. Hale, Ph.D. thesis, UMIST, Manchester, United Kingdom, (1995).
47. R. F. Bishop, R. G. Hale, and Y. Xian, *Phys. Rev. Lett.* **73**:3157 (1994).
48. R. F. Bishop, D. J. J. Farnell, and J. B. Parkinson, *J. Phys.: Condens. Matter* **8**:11153 (1996).
49. D. J. J. Farnell, S.A. Krüger, and J. B. Parkinson, *J. Phys.: Condens. Matter* **9**:7601 (1997).
50. R. F. Bishop, Y. Xian, and C. Zeng, in *Condensed Matter Theories*, Vol. 11, E.V. Ludeña, P. Vashishta, and R. F. Bishop, eds. (Nova Science, Commack, New York, 1996), p. 91.
51. C. Zeng, D. J. J. Farnell, and R. F. Bishop, *J. Stat. Phys.*, **90**:327 (1998).
52. R. F. Bishop, D. J. J. Farnell, and J.B. Parkinson, *Phys. Rev. B* **58**:6394 (1998).
53. R. F. Bishop, D. J. J. Farnell, and Chen Zeng, *Phys. Rev. B* **59**:1000 (1999).
54. J. Rosenfeld, N. E. Ligterink, and R. F. Bishop, *Phys. Rev. B* **60**:4030 (1999).
55. R. F. Bishop, D. J. J. Farnell, S. E. Krueger, J. B. Parkinson, J. Richter, and C. Zeng, *J. Phys.: Condens. Matter* **12**:6887 (2000).
56. R. F. Bishop, D. J. J. Farnell, and M. L. Ristig, *Int. J. Mod. Phys. B* **14**:1517 (2000).
57. S. E. Krüger, J. Richter, J. Schulenberg, D. J. J. Farnell, and R. F. Bishop, *Phys. Rev. B* **61**:14607 (2000).
58. D. J. J. Farnell, R. F. Bishop, and K. A. Gernoth, *Phys. Rev. B* **63**:220402R (2001).
59. D. J. J. Farnell, K. A. Gernoth, and R. F. Bishop, *Phys. Rev. B* **64**:172409 (2001).