Magnetic order on a frustrated spin-$\frac{1}{2}$ Heisenberg antiferromagnet on the Union Jack lattice

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Abstract

We use the coupled cluster method (CCM) to study the zero-temperature phase diagram of a two-dimensional frustrated spin-half antiferromagnet, the so-called Union Jack model. It is defined on a square lattice such that all nearest-neighbor pairs are connected by bonds with a strength $J_1 > 0$, but only half the next-nearest-neighbor pairs are connected by bonds with a strength $J_2 \equiv \kappa J_1 > 0$. The bonds are arranged such that on the $2 \times 2$ unit cell they form the pattern of the Union Jack flag. Alternating sites on the square lattice are thus 4-connected and 8-connected.

We find strong evidence for a first phase transition between a Néel antiferromagnetic phase and a canted ferrimagnetic phase at a critical coupling $\kappa_{c_1} = 0.66 \pm 0.02$. The transition is an interesting one, at which the energy and its first derivative seem continuous, thus providing a typical scenario of a second-order transition (just as in the classical case for the model), although a weakly first-order transition cannot be excluded. By contrast, the average on-site magnetization approaches a nonzero value $M_{c_1} = 0.195 \pm 0.005$ on both sides of the transition, which is more typical of a first-order transition. The slope, $dM/d\kappa$, of the order parameter curve as a function of the coupling strength $\kappa$, also appears to be continuous, or very nearly so, at the critical point $\kappa_{c_1}$, thereby providing further evidence of the subtle nature of the transition between the Néel and canted phases. Our CCM calculations provide strong evidence that the canted ferrimagnetic phase becomes unstable at large values of $\kappa$, and hence we have also used the CCM with a model collinear semi-stripe-ordered ferrimagnetic state in which alternating rows (and columns) are ferromagnetically and antiferromagnetically ordered, and in which the spins connected by $J_2$-bonds are antiparallel to one another. We find tentative evidence, based on the relative energies of the two states, for a second zero-temperature phase transition between the canted and semi-stripe-ordered ferrimagnetic states at a large value of the coupling parameter around $\kappa_{c_2} \approx 125 \pm 5$. This prediction, however, is based on an extrapolation of the CCM results for the canted state into regimes where the solutions have already become unstable and the CCM equations based on the canted state at any level of approximation beyond the lowest have no solutions. Our prediction for $\kappa_{c_2}$ is hence less reliable than that for $\kappa_{c_1}$. Nevertheless, if this second transition at $\kappa_{c_2}$ does exist, our results clearly indicate it to be of first-order type.

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I. INTRODUCTION

Quantum magnetism at zero temperature for lattices in two spatial dimensions\(^3\) is an important and fascinating subject because such systems display a wide variety of behavior, including semi-classical Néel ordering, two-dimensional quantum “spirals”, valence-bond crystals/solids, and spin liquids. The behavior of these systems is driven by the nature of the underlying crystallographic lattice, the number and range of bonds on this lattice, and the spin quantum numbers of the atoms localised to the sites on the lattice. There are very few exact results for quantum spin systems on two-dimensional (2D) lattices, and so the application of approximate methods is crucial to their understanding. The theoretical investigation of these models has been strongly mirrored by the discovery and experimental investigation of new quasi-2D magnetic materials. It seems clear that we can only form a complete picture of such 2D quantum spin-lattice systems by considering a wide range of possible scenarios that are often inspired (or followed shortly afterwards) by experimental studies.

A prototypical case is presented by the spin-half square-lattice Heisenberg antiferromagnet (HAF) model. This model has been studied extensively via a range of approximate techniques\(^3\). Its basic properties have been well-established, where, for example, approximate results for the order parameter indicate that about 61% of the classical Néel ordering persists in the quantum limit at zero temperature. A review of the properties of the spin-half square-lattice HAF is given in Ref. \(^7\). The most accurate results for this model are provided by quantum Monte Carlo (QMC) simulations\(^4\). Indeed, QMC techniques generally provide the benchmark for quantum magnets in two spatial dimensions. However, its use is severely limited by the “sign problem,” which is often a consequence of quantum frustration in the context of lattice spin systems.

A common theme has also begun to emerge recently when frustrating next-nearest-neighbor (NNN) bonds with strength \(J_2 > 0\) are added to the basic spin-half square-lattice HAF with nearest-neighbor (NN) bonds with strength \(J_1 > 0\). The frustrating \(J_2\) bonds may be added on some or all of the square plaquettes of the lattice and/or across both or only one of the diagonals of each plaquette. Perhaps the prototypical such model is the so-called spin-half \(J_1-J_2\) model in which all possible NNN bonds are included. Recent interest in this model has been reinvigorated by the discovery of various layered magnetic materials, such
as Li$_2$VOSiO$_4$, Li$_2$VOGeO$_4$, VOMoO$_4$, and BaCdVO(PO$_4$)$_2$. Several approximate methods have been used to simulate the properties of this system including the coupled cluster method (CCM)\textsuperscript{8–12} series expansion (SE) techniques\textsuperscript{13–17} exact diagonalization (ED) methods\textsuperscript{18–20} and hierarchical mean-field (MF) calculations\textsuperscript{21} These approximate techniques have established conclusively that there are two phases exhibiting magnetic long-range order (LRO) at small and at large values of $\kappa \equiv J_2/J_1$ respectively. For $\kappa < \kappa_{c1} \approx 0.4$ the ground-state (gs) phase exhibits (NN) Néel magnetic LRO, whereas for $\kappa > \kappa_{c2} \approx 0.6$ it exhibits collinear striped LRO in which alternating rows (or columns) of the square lattice have opposite spins, with the spins on each row (or columns) aligned, so that the Néel order is between NNN pairs. The intermediate region consists of a quantum paramagnetic state without magnetic LRO.

Several other models in this general class of spin-half models with both NN and NNN interactions have prompted recent interest. They all involve the removal of some of the NNN $J_2$-bonds from the fundamental $J_1$–$J_2$ model. One such example is the Shastry-Sutherland model,\textsuperscript{22–24} realized experimentally by the magnetic material SrCu(BO$_3$)$_2$, which involves the removal of three-quarters of the $J_2$-bonds. Whereas the $J_1$–$J_2$ model on the 2D square lattice has each of the sites connected by 8 bonds (4 NN $J_1$-bonds and 4 NNN $J_2$-bonds) to other sites, the Shastry-Sutherland model has each of the sites connected by 5 bonds (4 NN $J_1$-bonds and 1 NNN $J_2$-bond). A second example is the HAF on the anisotropic triangular lattice model (also known as the interpolating square-triangle model),\textsuperscript{25} realized experimentally by the magnetic material Cs$_2$CuCl$_4$, which involves the removal of half the $J_2$-bonds from the original $J_1$–$J_2$ model. In this model each of the sites on the 2D square lattice is connected by 6 bonds (4 NN $J_1$-bonds and 2 NNN $J_2$-bonds) to other sites, such that the remaining $J_2$-bonds connect equivalent NNN sites in each square plaquette. Although all of the models mentioned above show antiferromagnetic Néel ordering for small $J_2$, their phase diagrams for larger $J_2$ display a wide variety of behavior, including, two-dimensional quantum “spirals”, valence-bond crystals/solids, and spin liquids. Thus, in the absence of any definitive theoretical argument, the best way to understand this class of NN/NNN models on the square lattice is to treat each one on a case-by-case basis.

In this article we study another frustrated spin-half model that has both NN ($J_1$) and NNN ($J_2$) bonds on the square lattice, where these bonds form a pattern that resembles the “Union Jack” flag. Just as for the anisotropic triangular HAF described above, the Union
Jack model on the 2D square lattice also has only one frustrating NNN bond per square plaquette, but these $J_2$-bonds are now arranged such that half the sites are connected by 8 bonds (4 NN $J_1$-bonds and 4 NNN $J_2$-bonds) to other sites, while the other half are connected only by 4 $J_1$-bonds to their NN sites, as described more fully below in Sec. II. This model has previously been studied using SWT\textsuperscript{26,27} and SE techniques.\textsuperscript{28} As in the case of the spin-half interpolating square-triangle model, it was shown\textsuperscript{26–28} that NN Néel order for the Union Jack model persists until a critical value of the frustrating NNN ($J_2$) bonds. However, in contrast to the interpolating square-triangle model, there exists a ferrimagnetic ground state in which spins on the eight-connected sites cant at a nonzero angle with respect to their directions in the corresponding Néel state. This model thus exhibits an overall magnetic moment in this regime, which is quite unusual for spin-half 2D materials with only Heisenberg bonds and which therefore preserve (spin) rotational symmetries in the Hamiltonian. This model also presents us with a difficult computational task in order to simulate its properties. Here we wish to study this model using the CCM, which has consistently been shown to yield insight into a wide range of problems in quantum magnetism, and which we now hope will hence shed yet more light on the whole class of NN/NNN models as mentioned before. The dual associated features of a model with two sorts of sites with differing connectivities, and its consequent ferrimagnetic phase, are just those that are attracting the interest of the community now.

II. THE MODEL

In this paper we now apply the CCM to the spin-half Union Jack model that has been studied recently by other means.\textsuperscript{26–28} Its Hamiltonian is written as

$$H = J_1 \sum_{\langle i,j \rangle} \mathbf{s}_i \cdot \mathbf{s}_j + J_2 \sum_{[i,k]} \mathbf{s}_i \cdot \mathbf{s}_k,$$

where the operators $\mathbf{s}_i \equiv (s^x_i, s^y_i, s^z_i)$ are the quantum spin operators on lattice site $i$ with $s_i^2 = s(s + 1)$ and $s = 1/2$. On the square lattice the sum over $\langle i, j \rangle$ runs over all distinct NN bonds with strength $J_1$, while the sum over $[i, k]$ runs over only half of the distinct NNN diagonal bonds having strength $J_2$ and with only one diagonal bond on each square plaquette as arranged in the pattern shown explicitly in Fig. I. The unit cell is thus the $2 \times 2$ square shown in Fig. Ia. (We note that, by contrast, the $J_1$–$J_2$ model discussed above, includes all
of the diagonal NNN bonds on the square lattice.) We consider here the case where both sorts of bonds are antiferromagnetic, \( J_1 > 0 \) and \( J_2 \equiv \kappa J_1 > 0 \), and are hence acting to compete against (or to frustrate) each other. Henceforth we set \( J_1 \equiv 1 \). We consider the model equivalently defined by the Union Jack geometry in which there are two sorts of sites, namely the A sites with 8 NN sites and the B sites with 4 NN sites, as shown in Fig. 1a.

Considered classically rather than quantum-mechanically, (and thus corresponding to the quantum case in the limit where the spin quantum number \( s \to \infty \)), the Union Jack model has only two gs phases as the parameter \( \kappa \) is varied over the range \((0, \infty)\). A simple variational analysis for the classical model reveals that for \( 0 < \kappa < 1/2 \) the gs phase is Néel-ordered, exactly as for the full \( J_1-J_2 \) model. Thus the Néel ordering induced by the \( J_1 \)-bonds acting alone is preserved as the strength of the competing \( J_2 \)-bonds is increased, until the critical value \( \kappa_c = 0.5 \) is reached. For \( \kappa > \kappa_c \) a new phase of lower energy emerges, just as in the full \( J_1-J_2 \) model. However, whereas for the full \( J_1-J_2 \) model that new phase is a classical striped state in which alternate rows (or columns) of spins are arranged antiparallel to one another, the new classical gs phase for the Union Jack model is the canted ferrimagnetic state shown in Fig. 1a in which the spins on each of the alternating \( A_1 \) and \( A_2 \) sites of the A-sublattice are canted respectively at angles \( (\pi \mp \phi) \) with respect to those on the B-sublattice, all of the latter of which point in the same direction. On the A-sublattice each site \( A_1 \) has 4 NN sites \( A_2 \), and vice versa. The angle between the NN spins on the A-sublattice is thus \( 2\phi \).

FIG. 1: (Color online) Union Jack model; — \( J_1 \); - - - \( J_2 \). (a) Canted state; (b) Semi-striped state. The unit cell is a square of side length 2.
The classical energy of the above canted state is thus

\[ E = N s^2 (\kappa \cos 2\phi - 2 \cos \phi) , \tag{2} \]

where \( J_1 \equiv 1 \) and \( N \to \infty \) is the number of sites. Clearly the energy is extremized when

\[ \sin \phi (1 - 2\kappa \cos \phi) = 0 . \tag{3} \]

When \( \kappa < \kappa_{cl}^c \equiv 0.5 \), the lowest energy corresponds to \( \sin \phi = 0 \) and hence to the Néel state. By contrast, when \( \kappa > \kappa_{cl}^c \equiv 0.5 \) the lowest energy solution is the canted state with

\[ \phi_{cl} = \cos^{-1} \left( \frac{1}{2\kappa} \right) . \tag{4} \]

Thus the classical gs energy is given by

\[ E^{cl} = \begin{cases} N s^2 (\kappa - 2) ; & \kappa < \kappa_{cl}^c \equiv 0.5 . \\ N s^2 \left( -\frac{1}{2\kappa} - \kappa \right) ; & \kappa > \kappa_{cl}^c \equiv 0.5 . \end{cases} \tag{5} \]

The classical phase transition at \( \kappa = \kappa_{cl}^c \equiv 0.5 \) is of continuous (second-order) type with the gs energy and its first derivative both continuous functions of \( \kappa \), although there are finite discontinuities in the second- and higher-order derivatives at \( \kappa = \kappa_{cl}^c \).

In the classical canted phase the total magnetization per site is

\[ m_{cl} = \frac{1}{2}s \left[ 1 - (2\kappa)^{-1} \right] , \]

and the model thus exhibits ferrimagnetism in this phase. Whereas ferrimagnetism more commonly occurs when the individual ionic spins have different magnitudes on different sublattices, it arises here in a case where when the spins all have the same magnitude and all the interactions are antiferromagnetic in nature, but the frustration between them acts to produce an overall magnetization. The total magnetization \( m \) vanishes linearly as \( \kappa \to \kappa_{cl}^c \) from the canted phase and then remains zero in the Néel phase for \( \kappa < \kappa_{cl}^c \). The spontaneous breaking of the spin rotation symmetry is also reflected by the vanishing of the energy gap on both sides of the transition. Clearly on both sides of the transition the translation symmetry of the lattice is also broken.

One of the aims of the present paper is to give a fully microscopic analysis of the Union Jack model for the quantum case where the spins all have spin quantum number \( s = 1/2 \). We are interested to map out the zero-temperature \((T = 0)\) phase diagram of the model, including the positions and orders of any quantum phase transitions that emerge. In particular we investigate the quantum analogs of the classical Néel and canted phases and calculate the
effect of quantum fluctuations on the position and nature of the transition between them. We also aim to investigate, for particular regions of the control parameter $\kappa$, whether the quantum fluctuations may favor other phases, which have no classical counterparts. One such possible candidate is discussed below.

In the limit of $\kappa \to \infty$ the above classical limit corresponds to a canting angle $\phi \to \frac{1}{2}\pi$, such that the spins on the A-sublattice become Néel-ordered, as is expected. The spins on the antiferromagnetically-ordered A-sublattice are orientated at 90$^\circ$ to those on the ferromagnetically-ordered B-sublattice in this limit. In reality, of course, there is complete degeneracy at the classical level in this limit between all states for which the relative ordering directions for spins on the A- and B- sublattices are arbitrary. Clearly the exact spin-$1/2$ limit should also comprise decoupled antiferromagnetic and ferromagnetic sublattices. However, one might now expect that this degeneracy in the relative spin orientations between the two sublattices is lifted by quantum fluctuations by the well-known phenomenon of order by disorder. Just such a phase is known to exist in the full spin-$1/2$ $J_1-J_2$ model for values of $J_2/J_1 \gtrsim 0.6$, where it is the so-called collinear striped phase in which, on the square lattice, spins along (say) the rows in Fig. 1 order ferromagnetically while spins along the columns and diagonals order antiferromagnetically. We have also shown how such a striped state is stabilized by quantum fluctuations for values of $J'_2/J'_1 \gtrsim 1.8$ for the spin-$1/2$ $J_1-J'_2$ model defined on an anistropic 2D lattice as discussed in Sec. 1 above.

The existence of the striped state as a stable phase for large values of the frustration parameter for both the spin-$1/2$ $J_1-J_2$ and $J_1-J'_2$ models above is a reflection of the well-known fact that quantum fluctuations favor collinear ordering. In both cases the order-by-disorder mechanism favors the collinear state from the otherwise infinitely degenerate set of available states at the classical level. For the present Union Jack model the corresponding collinear state that might perhaps be favored by the order by disorder mechanism is the so-called semi-striped state shown in Fig. 1b where the A-sublattice is now Néel-ordered in the same direction as the B-sublattice is ferromagnetically ordered. Alternate rows (or columns) are thus ferromagnetically and antiferromagnetically ordered in the same direction. We investigate the possibility below that such a semi-stripe-ordered phase may be stabilized by quantum fluctuations at larger values of $\kappa$. 

8
III. THE COUPLED CLUSTER METHOD

The CCM (see, e.g., Refs. [30–32] and references cited therein) that we employ here is one of the most powerful and most versatile modern techniques available to us in quantum many-body theory. It has been applied very successfully to various quantum magnets (see Refs. [8–12,23–25,32–36] and references cited therein). The method is particularly appropriate for studying frustrated systems, for which some of the main alternative methods either cannot be applied or are sometimes only of limited usefulness, as explained below. For example, QMC techniques are particularly plagued by the sign problem for such systems, and the ED method is restricted in practice by available computational power, particularly for \( s > 1/2 \), to such small lattices that it is often insensitive to the details of any subtle phase order present.

The method of applying the CCM to quantum magnets has been described in detail elsewhere (see, e.g., Refs. [30–35] and references cited therein). It relies on building multispin correlations on top of a chosen gs model state \( |\Phi\rangle \) in a systematic hierarchy of LSUBn approximations (described below) for the correlation operators \( S \) and \( \tilde{S} \) that parametrize the exact gs ket and bra wave functions of the system respectively as \( |\Psi\rangle = e^S|\Phi\rangle \) and \( \langle \tilde{\Psi}| = \langle \Phi|\tilde{S}e^{-S}. \) In the present case we use three different choices for the model state \( |\Phi\rangle \), namely either of the classical Néel and canted states, as well as the semi-striped state. Note that for the canted phase we perform calculations for arbitrary canting angle \( \phi \) [as shown in Fig. 1a], and then minimize the corresponding LSUBn approximation for the energy with respect to \( \phi \), \( E_{\text{LSUBn}}(\phi) \to \min \iff \phi = \phi_{\text{LSUBn}}. \) Generally (for \( n > 2 \)) the minimization must be carried out computationally in an iterative procedure, and for the highest values of \( n \) that we use here the use of supercomputing resources was essential. Results for the canting angle \( \phi_{\text{LSUBn}} \) will be given later. We choose local spin coordinates on each site in each case so that all spins in \( |\Phi\rangle \), whatever the choice, point in the negative \( z \)-direction (i.e., downwards) by definition in these local coordinates.

Then, in the LSUBn approximation all possible multi-spin-flip correlations over different locales on the lattice defined by \( n \) or fewer contiguous lattice sites are retained. The numbers of such distinct (i.e., under the symmetries of the lattice and the model state) fundamental configurations of the current model in various LSUBn approximations are shown in Table I. We note that the distinct configurations given in Table I are defined with respect to the Union
TABLE I: Number of fundamental LSUBn configurations ($N_f$) for the semi-striped and canted states of the spin-1/2 Union Jack model, based on the Union Jack geometry defined in the text.

| Method | $N_f$ (semi-striped) | $N_f$ (canted) |
|--------|----------------------|----------------|
| LSUB2  | 3                    | 5              |
| LSUB3  | 5                    | 42             |
| LSUB4  | 41                   | 199            |
| LSUB5  | 194                  | 1259           |
| LSUB6  | 1159                 | 8047           |
| LSUB7  | 6862                 | 56442          |

Jack geometry described in Sec. II in which the B-sublattice sites of Fig. 1a are defined to have 4 NN sites and the A-sublattice sites are defined to have the 8 NN sites joined to them either by $J_1$- or $J_2$-bonds. If we chose instead to work in the square-lattice geometry every site would have 4 NN sites. The coupled sets of equations for these corresponding numbers of coefficients in the operators $S$ and $\tilde{S}$ are derived using computer algebra and then solved using parallel computing. We note that such CCM calculations using up to about $10^5$ fundamental configurations or so have been previously carried out many times using the CCCM code and heavy parallelization. A significant extra computational burden arises here for the canted state due to the need to optimize the quantum canting angle $\phi$ at each LSUB$n$ level of approximation as described above. Furthermore, for many model states the quantum number $s^z_T \equiv \sum_{i=1}^{N} s^z_i$ in the original global spin-coordinate frame, may be used to restrict the numbers of fundamental multi-spin-flip configurations to those clusters that preserve $s^z_T$ as a good quantum number. This is true for the Néel state where $s^z_T = 0$ and for the semi-striped state for which $s^z_T = N/4$, where $N$ is the number of lattice sites. However, for the canted model state that symmetry is absent, which largely explains the significantly greater number of fundamental configurations shown in Table I for the canted state at a given LSUB$n$ order. Hence, the maximum LSUB$n$ level that we can reach here for the canted state, even with massive parallelization and the use of supercomputing resources, is LSUB7. For example, to obtain a single data point for a given value of $\kappa$ (i.e., for a given
value of $J_2$, with $J_1 = 1$) for the canted phase at the LSUB7 level typically required about 0.3 hours computing time using 600 processors simultaneously. However, for values of $\kappa$ near to termination points at which CCM solutions using that model state disappear (as described more fully below), the computing time typically increased significantly.

At each level of approximation we may then calculate a corresponding estimate of the gs expectation value of any physical observable such as the energy $E$ and the magnetic order parameter, $M \equiv -\frac{1}{N} \sum_{i=1}^{N} \langle \tilde{\Psi} | s_i^z | \Psi \rangle$, defined in the local, rotated spin axes, and which thus represents the average on-site magnetization. Note that $M$ is just the usual sublattice (or staggered) magnetization per site for the case of the Néel state as the CCM model state, for example.

It is important to note that we never need to perform any finite-size scaling, since all CCM approximations are automatically performed from the outset in the infinite-lattice limit, $N \to \infty$, where $N$ is the number of lattice sites. However, we do need as a last step to extrapolate to the $n \to \infty$ limit in the LSUB$n$ truncation index $n$. We use here the well-tested empirical scaling laws

$$E/N = a_0 + a_1 n^{-2} + a_2 n^{-4},$$

$$M = b_0 + b_1 n^{-1} + b_2 n^{-2}.$$  

IV. RESULTS

We report here on CCM calculations for the present spin-1/2 Union Jack model Hamiltonian of Eq. (1) for given parameters ($J_1 = 1, J_2$), based respectively on the Néel, canted and semi-striped states as CCM model states. Our computational power is such that we can perform LSUB$n$ calculations for each model state with $n \leq 7$. We note that, as has been well documented in the past, the LSUB$n$ data for both the gs energy per spin $E/N$ and the average on-site magnetization $M$ converge differently for the even-$n$ and the odd-$n$ sequences, similar to what is frequently observed in perturbation theory. Since, as a general rule, it is desirable to have at least $(n + 1)$ data points to fit to any fitting formula that contains $n$ unknown parameters, we prefer to have at least 4 results for different values of the LSUB$n$ truncation index $n$ to fit to Eqs. (6) and (7). However, for all of our extrapolated results below we perform separate extrapolations using the even and odd LSUB$n$ sequences.
FIG. 2: (Color online) Ground-state energy per spin of the spin-1/2 Union Jack Hamiltonian of Eq. (1) with \( J_1 \equiv 1 \), using the LSUB6 approximation of the CCM with the canted model state, versus the canting angle \( \phi \), for some illustrative values of \( J_2 \) in the range \( 0 \leq J_2 \leq 1.5 \) for Fig. 2a and \( 0.65 \leq J_2 \leq 0.75 \) for Fig. 2b. For \( J_2 \lesssim 0.68 \) in this approximation the minimum is at \( \phi = 0 \) (Néel order), whereas for \( J_2 \gtrsim 0.68 \) the minimum occurs at \( \phi = \phi_{\text{LSUB6}} \neq 0 \), indicating a phase transition at \( J_2 \approx 0.68 \) in this LSUB6 approximation.

with \( n = \{2, 4, 6\} \) and \( n = \{3, 5, 7\} \).

A. Néel state versus the canted state

We report first on results obtained using the Néel and canted model states. While classically we have a second-order phase transition from Néel order (for \( \kappa < \kappa_{c1}^{\text{cl}} \)) to canted order (for \( \kappa > \kappa_{c1}^{\text{cl}} \)), where \( \kappa \equiv J_2/J_1 \), at a value \( \kappa_{c1}^{\text{cl}} = 0.5 \), using the CCM we find strong indications of a shift of this critical point to a higher value \( \kappa_{c1} \approx 0.66 \) in the spin-1/2 quantum case as we explain in detail below. Thus, for example, curves such as those shown in Fig. 2 show that the Néel model state (\( \phi = 0 \)) gives the minimum gs energy for all values of \( \kappa < \kappa_{c1} \) where \( \kappa_{c1} = \kappa_{c1}^{\text{LSUBn}} \) is also dependent on the level of LSUBn approximation, as we see clearly in Fig. 3.

By contrast, for \( \kappa > \kappa_{c1} \) the minimum in the energy is found to occur at a value \( \phi \neq 0 \). If we consider the canting angle \( \phi \) itself as an order parameter (i.e., \( \phi = 0 \) for Néel order and \( \phi \neq 0 \) for canted order) a typical scenario for a first-order phase transition would be the
FIG. 3: (Color online) The angle $\phi_{\text{LSUB}n}$ that minimizes the energy $E_{\text{LSUB}n}(\phi)$ of the spin-1/2 Union Jack Hamiltonian of Eq. (1) with $J_1 \equiv 1$, in the LSUB$n$ approximations with (a) $n = \{2, 4, 6\}$ and (b) $n = \{3, 5, 7\}$, using the canted model state, versus $J_2$. The corresponding classical result $\phi_{\text{cl}}$ from Eq. (4) is shown for comparison. We find in the LSUB$n$ quantum case with $n > 2$ a weakly first-order phase transition or second-order phase transition (e.g., for LSUB6 at $J_2 \approx 0.680$ and LSUB7 at $J_2 \approx 0.646$). By contrast, in the classical case there is a second-order phase transition at $J_2 = 0.5$.

appearance of a two-minimum structure for the gs energy as a function of $\phi$. If we therefore admit such a scenario, in the typical case one would expect various special points in the transition region, namely the phase transition point $\kappa_{c_1}$ itself where the two minima have equal depth, plus one or two instability points $\kappa_{i_1}$ and $\kappa_{i_2}$ where one or other of the minima (at $\phi = 0$ and $\phi \neq 0$ respectively) disappears. By contrast, a second-order phase transition might manifest itself via a one-minimum structure for the gs energy as a function of $\phi$, in which the single minimum moves smoothly and continuously from the value $\phi = 0$ for all values of $\kappa < \kappa_{c_1}$ to nonzero value $\phi \neq 0$ for $\kappa > \kappa_{c_1}$.

We show in Fig. 2 our results for the LSUB6 approximation based on the canted (or Néel) state as the CCM model state. Very similar curves occur for other LSUB$n$ approximations. A close inspection of curves such as those shown in Fig. 2 for the LSUB6 case shows that what happens for this model at this level of approximation is that for $\kappa \lesssim 0.68$ the only minimum in the gs energy is at $\phi = 0$ (Néel order). As this value is approached from below the LSUB6 energy curves become extremely flat near $\phi = 0$, indicating the disappearance at $\phi = 0$ of the second derivative $d^2E/d\phi^2$ (and possibly also of one or more of the higher
derivatives $d^n E/d\phi^n$ with $n \geq 3$), as well as of the first derivative $dE/d\phi$. Then, for all values $\kappa \gtrsim 0.68$ the LSUB6 curves develop a minimum at a value $\phi \neq 0$ which is also the global minimum. The state for $\phi \neq 0$ is thus the quantum analog of the classical canted phase. The fact that the antiferromagnetic Néel order survives into the classically unstable regime is another example of the well-known phenomenon that quantum fluctuations tend to promote collinear order in magnetic spin-lattice systems, as has been observed in many other such cases (see e.g., Ref. [34,40]). Thus, this collinear Néel-ordered state survives into a region where classically it becomes unstable with respect to the non-collinear canted state.

A close inspection of the curves shown in Fig. 3 for various LSUB$n$ approximation shows that the crossover from one minimum ($\phi = 0$, Néel) solution to the other ($\phi \neq 0$, canted) appears to be continuous for the odd-$n$ sequence, thus indicating a second-order transition according to the above scenario. By contrast, for the even-$n$ sequence with $n > 2$ the curves in Fig. 3 become very steep in the crossover region just above $\kappa_{L\text{SUB}n}^{c1}$ and due to the extremely flat nature of the gs energy curves as a function of $\phi$ in this region, as shown in Fig. 2, it is impossible to rule out a small but finite discontinuity in the curves of Fig. 2a for the even-$n$ LSUB$n$ sequence at $\kappa = \kappa_{L\text{SUB}n}^{c1}$. However, if the phase transition is, in fact first-order, it is certainly only very weakly so according to this criterion.

Thus, based on the evidence presented so far of the gs energies of the Néel and canted phases, it would appear that the transition at $\kappa = \kappa_{c1}$ between these two phases is either second-order, as in the classical phase, or weakly first-order. Such a situation where the quantum fluctuations change the nature of a phase transition qualitatively from a classical second-order type to a quantum first-order type has also been seen previously in the comparable spin-1/2 HAF models that interpolate continuously between the square and triangular lattices, respectively. In the present spin-1/2 Union Jack model, however, the CCM gs energy results appear to favor a second-order transition, although the extreme insensitivity of the gs energy to the canting angle $\phi$ near the crossover region, especially for the even-$n$ LSUB$n$ sequence with $n > 2$, means that we cannot rule out a weakly first-order transition. The evidence to date indicates, however, that the quantum phase transition at $\kappa_{c1}$ is a subtle one. Furthermore, the present spin-1/2 Union Jack model appears, on the evidence to date, to behave somewhat differently (viz., in some senses “more classically”) than its corresponding spin-1/2 interpolating square-triangle Heisenberg antiferromagnet counterpart. Further evidence from Fig. 3 appears to back up
TABLE II: The critical value $\kappa_{c_1}^{\text{LSUB}n}$ at which the transition between the Néel phase ($\phi = 0$) and the canted phase ($\phi \neq 0$) occurs in the LSUB$n$ approximation using the CCM with (Néel or) canted state as model state.

| Method | $\kappa_{c_1}^{\text{LSUB}n}$ |
|--------|-------------------------------|
| LSUB2  | 0.740                         |
| LSUB4  | 0.696                         |
| LSUB6  | 0.680                         |
| LSUB$\infty$ $^a$ | 0.651 $\pm$ 0.001.          |
| LSUB$\infty$ $^b$ | 0.676 $\pm$ 0.004.          |
| LSUB3  | 0.597                         |
| LSUB5  | 0.630                         |
| LSUB7  | 0.645                         |
| LSUB$\infty$ $^a$ | 0.681 $\pm$ 0.001.          |
| LSUB$\infty$ $^b$ | 0.653 $\pm$ 0.004.          |

$^a$ Based on $\kappa_{c_1}^{\text{LSUB}n} = a_0 + a_1n^{-1}$, with $n = \{2, 4, 6\}$ or $n = \{3, 5, 7\}$.

$^b$ Based on $\kappa_{c_1}^{\text{LSUB}n} = b_0 + b_1n^{-2}$, with $n = \{2, 4, 6\}$ or $n = \{3, 5, 7\}$.

this observation. Thus, we see from Fig. 3 that the quantum canting angle $\phi$ approaches its asymptotic value $\pi/2$ as $\kappa \to \infty$ slightly faster than does the corresponding classical value. By contrast, in the case of the spin-1/2 interpolating square-triangle Heisenberg antiferromagnet$^{25}$ the corresponding pitch angle $\phi$ of the spiral phase (that is the analog of the canted phase for the present model) approaches its similar asymptotic value $\pi/2$ as $\kappa \to \infty$ very much faster than does the classical value. We also discuss this difference more fully below, where we find further evidence that quantum fluctuations modify the classical behavior of the Union Jack model rather less than they do for its corresponding spin-1/2 interpolating square-triangle Heisenberg antiferromagnet counterpart.

We show in Table III the critical values $\kappa_{c_1}^{\text{LSUB}n}$ at which the transition between the Néel and canted phases occurs in the various LSUB$n$ approximations shown in Fig. 3. In the past we have found that a simple linear extrapolation, $\kappa_{c_1}^{\text{LSUB}n} = a_0 + a_1n^{-1}$, yields a good fit to such critical points, as seems to be the case here too. The corresponding “LSUB$\infty$”
estimates from the LSUBn data in Table II are $\kappa_{c1} = 0.651 \pm 0.001$ based on $n = \{2, 4, 6\}$ and $\kappa_{c1} = 0.681 \pm 0.001$ based on $n = \{3, 5, 7\}$ where the quoted errors are simply the standard deviations from the two fits. Similar estimates based on an extrapolation $\kappa_{c1}^{\text{LSUBn}} = b_0 + b_1 n^{-2}$ are also shown in Table III for which the standard deviations are clearly greater. The fact that the two estimates based on the even-$n$ and odd-$n$ LSUBn sequences differ slightly from one another is a reflection of the extreme insensitivity of the gs energy to the canting angle $\phi$ near $\kappa_{c1}^{\text{LSUBn}}$, and the difference between the two estimates is a rough indication of our real error bars on $\kappa_{c1}$. We also present other independent estimates of $\kappa_{c1}$ below.

We note from Fig. 2 that for certain values of $J_2$ with $J_1 \equiv 1$ (or, equivalently, $\kappa$) CCM solutions at a given LSUBn level of approximation (viz., LSUB6 in Fig. 2) exist only for certain ranges of the canting angle $\phi$. For example, for the pure square-lattice HAF ($\kappa = 0$) the CCM LSUB6 solution based on a canted model state only exists for $0 \leq \phi \lesssim 0.161\pi$. In this case, where the Néel solution is the stable ground state, if we attempt to move too far away from Néel collinearity the CCM equations themselves become “unstable” and simply do not have a real solution. Similarly, we see from Fig. 2 that for $\kappa = 1.5$ the CCM LSUB6 solution exists only for $0.308\pi \lesssim \phi \leq 0.445\pi$. In this case the stable ground state is a canted phase, and now if we attempt either to move too close to Néel collinearity or to increase the canting angle too close to its asymptotic value of $\pi/2$, the real solution terminates.

Such terminations of CCM solutions are very common and are very well documented. In all such cases a termination point always arises due to the solution of the CCM equations becoming complex at this point, beyond which there exist two branches of entirely unphysical complex conjugate solutions. In the region where the solution reflecting the true physical solution is real there actually also exists another (unstable) real solution. However, only the shown branch of these two solutions reflects the true (stable) physical ground state, whereas the other branch does not. The physical branch is usually easily identified in practice as the one which becomes exact in some known (e.g., perturbative) limit. This physical branch then meets the corresponding unphysical branch at some termination point (with infinite slope on Fig. 2) beyond which no real solutions exist. The LSUBn termination points are themselves also reflections of the quantum phase transitions in the real system, and may be used to estimate the position of the phase boundary although we do not do so for this first critical point since we have more accurate criteria discussed above as well as below.

Before doing so, however, we wish to give some further indication of the accuracy of our
TABLE III: Ground-state energy per spin $E/N$ and magnetic order parameter $M$ (i.e., the average on-site magnetization) for the spin-1/2 square-lattice HAF. We show CCM results obtained for the Union Jack model with $J_1 = 1$ and $J_2 = 0$ using the Néel model state in various CCM $L_{SUBn}$ approximations defined on the Union Jack geometry described in Sec. III. We compare our extrapolated ($n \to \infty$) results using Eqs. (6) and (7) with the odd-$n$ and even-$n$ $L_{SUBn}$ data sets with other calculations.

| Method  | $E/N$     | $M$  |
|---------|-----------|------|
| $L_{SUB2}$ | -0.64833  | 0.4207 |
| $L_{SUB3}$ | -0.65044  | 0.4151 |
| $L_{SUB4}$ | -0.66366  | 0.3821 |
| $L_{SUB5}$ | -0.66398  | 0.3795 |
| $L_{SUB6}$ | -0.66703  | 0.3630 |
| $L_{SUB7}$ | -0.66724  | 0.3606 |
| Extrapolations |          |      |
| $L_{SUB\infty}^a$ | -0.6698  | 0.316 |
| $L_{SUB\infty}^b$ | -0.6704  | 0.304 |
| QMC $^c$ | -0.669437(5) | 0.3070(3) |
| SE $^d$ | -0.6693(1)  | 0.307(1) |

$^a$ Based on $n = \{2, 4, 6\}$  
$^b$ Based on $n = \{3, 5, 7\}$  
$^c$ QMC (Quantum Monte Carlo) for square lattice  
$^d$ SE (Series Expansion) for square lattice

results. Thus in Table III we show data for the case of the spin-1/2 HAF on the square lattice (corresponding to the case $\kappa = 0$ of the present Union Jack model). We present our CCM results in various $L_{SUBn}$ approximations (with $2 \leq n \leq 7$) based on the Union Jack geometry using the Néel model state. Results are given for the gs energy per spin $E/N$, and the magnetic order parameter $M$. We also display our extrapolated ($n \to \infty$) results using the schemes of Eqs. (6) and (7) with the data sets $n = \{2, 4, 6\}$ and $n = \{3, 5, 7\}$. The results are clearly seen to be robust and consistent, and for comparison purposes we
FIG. 4: (Color online) Ground-state energy per spin versus $J_2$ for the Néel and canted phases of the spin-1/2 Union Jack Hamiltonian of Eq. (11) with $J_1 \equiv 1$. The CCM results using the canted model state are shown for various $\text{LSUB}n$ approximations with (a) $n = \{2, 4, 6\}$ and (b) $n = \{3, 5, 7\}$) with the canting angle $\phi = \phi_{\text{LSUB}n}$ that minimizes $E_{\text{LSUB}n}(\phi)$. We also show the $n \to \infty$ extrapolated result from using Eq. (6).

also show the corresponding results using a QMC technique and from a linked-cluster series expansion (SE) method. We note that for the square-lattice HAF no dynamic (or geometric) frustration exists and the Marshall-Peierls sign rule applies and may be used to circumvent the QMC “minus-sign problem”. The QMC results are thus extremely accurate for this limiting ($\kappa = 0$) case only, and represent the best available results in this case. Our own extrapolated results are in good agreement with these QMC benchmark results, as found previously (see, e.g., Ref. [38] and references cited therein) for CCM calculations performed specifically using the square lattice geometry, as well as for other CCM calculations for which the square-lattice HAF is a limit, such as for the spin-1/2 interpolating square-triangle $J_1-J'_2$ model for which the triangular lattice geometry was employed. It is gratifying to note in particular that although the individual $\text{LSUB}n$ results for the spin-1/2 square-lattice HAF depend upon which geometry is used to define the configurations, the corresponding $\text{LSUB}\infty$ extrapolations are in excellent agreement with one another.

In Fig. 4 we show the CCM results for the gs energy per spin in various $\text{LSUB}n$ approximations based on the canted (and Néel) model states, with the canting angle $\phi_{\text{LSUB}n}$ chosen to minimize the energy $E_{\text{LSUB}n}(\phi)$, as shown in Fig. 3. We also show separately the extrapolated ($\text{LSUB}\infty$) results obtained from Eq. (6) using the separate data sets $n = \{2, 4, 6\}$

18
FIG. 5: (Color online) Ground-state magnetic order parameter (i.e., the average on-site magnetization) versus $J_2$ for the Néel and canted phases of the spin-1/2 Union Jack Hamiltonian of Eq. (11) with $J_1 \equiv 1$. The CCM results using the canted model state are shown for various LSUBn approximations with (a) $n = \{2, 4, 6\}$ and (b) $n = \{3, 5, 7\}$ with the canting angle $\phi = \phi_{LSUBn}$ that minimizes $E_{LSUBn}(\phi)$. We also show the $n \to \infty$ extrapolated result from using Eq. (7).

and $n = \{3, 5, 7\}$ as shown. As is expected from our previous discussion the energy curves themselves show very little evidence of the phase transition at $\kappa = \kappa_{c1}$, with the energy and its first derivative seemingly continuous.

Much clearer evidence for the transition between the Néel and canted phases is observed in our corresponding results for the gs magnetic order parameter $M$ (the average on-site magnetization) shown in Fig. 5. For the raw LSUBn data we display the results for the Néel phase only for values of $\kappa < \kappa_{c1}^{LSUBn}$ for clarity. However, the extrapolated (LSUB\infty) results for the Néel phase are shown for all values of $\kappa$ in the range shown and where $M > 0$, using the extrapolation scheme of Eq. (7) and the LSUBn results based on the Néel model state. Once again we extrapolate the odd-$n$ and even-$n$ LSUBn results separately. It is interesting to note from Fig. 5 that the raw LSUBn data show the transition at $\kappa_{c1}^{LSUBn}$ more clearly for even values of $n$ than for odd values of $n$. For the canted phase (for which $\phi_{LSUBn} \neq 0$) we can clearly only show the extrapolated (LSUB\infty) results using Eq. (7), for regions of $\kappa$ for which we have data for all of the set $n = \{2, 4, 6\}$ or $n = \{3, 5, 7\}$. We see from Table II that for the even-$n$ values we are limited (by the LSUB2 results) to values $\kappa > \kappa_{c1}^{LSUB2} \approx 0.740$, whereas for the odd-$n$ values we are limited (by the LSUB7 results) to values $\kappa > \kappa_{c1}^{LSUB7} \approx 0.645$. 19
The separate odd-$n$ LSUB$\infty$ extrapolation curves for $M$ for the Néel and canted phases are seen from Fig. 5b to be extremely close at the value $\kappa = 0.645$ and the curves appear to be about to meet at an angle which is either zero or very close to zero. A straightforward LSUB$\infty$ extrapolation using Eq. (7) for the three whole LSUB$n$ curves (Néel plus canted) with $n = \{3, 5, 7\}$ shows a value $\kappa_{c_1} \approx 0.637$ at which the extrapolated curve diverges (at zero or very small angle) from the corresponding LSUB$\infty$ estimate for the Néel state shown in Fig. 5b. For the corresponding even-$n$ data shown in Fig. 5a, simple extrapolations of the LSUB$\infty$ curve to lower values of $\kappa < \kappa_{c_1} \approx 0.637$ at which the extrapolated curve diverges (at zero or very small angle) from the corresponding LSUB$\infty$ estimate for the Néel state shown in Fig. 5b. For the corresponding even-$n$ data shown in Fig. 5a, simple extrapolations of the LSUB$\infty$ curve to lower values of $\kappa < \kappa_{c_1} \approx 0.637$ at which the extrapolated curve diverges (at zero or very small angle) from the corresponding LSUB$\infty$ estimate for the Néel state shown in Fig. 5b. Thus the evidence from the behavior of the order parameter is that the transition at $\kappa_{c_1}$ is a first-order one, in the sense that the order parameter does not go to zero at $\kappa_{c_1}$, although it is certainly continuous at this point, and with every indication that its derivative as a function of $\kappa$ is also continuous (or very nearly so) at $\kappa = \kappa_{c_1}$.

We also show in Fig. 6 the corresponding extrapolated (LSUB$\infty$) results for the average on-site magnetization as a function of $J_2$ (with $J_1 \equiv 1$), or hence equivalently as a function of $\kappa$, for both the A sites ($M_A$) and the B sites ($M_B$) of the Union Jack lattice. We recall that, as shown in Fig. 1a, each of the A and B sites is connected to 4 NN sites on the square lattice by $J_1$-bonds, whereas each of the A sites is additionally connected to 4 NNN sites on the square lattice by $J_2$-bonds. The extrapolations are shown in exactly the same regions, and for the same reasons, as those shown in Fig. 5.

In Fig. 7 we also show the total gs magnetization per site, $m = \frac{1}{2}(M_B - M_A \cos \phi)$, from using our CCM LSUB$n$ results with $n = \{2, 4, 6\}$ and with the canting angle $\phi = \phi_{LSUBn}$ that minimizes $E_{LSUBn}(\phi)$. Clearly $m = 0$ in the Néel phase where $M_A = M_B$ and $\phi = 0$. We also show in Fig. 7 the corresponding classical result $m^{cl} = \frac{1}{2}s(1 - \cos \phi_{cl}) = \frac{1}{4}[1 - (2\kappa)^{-1}]$ in the canted phase (with $s = \frac{1}{2}$). A comparison of the extrapolated (LSUB$\infty$) CCM curve with its classical counterpart shows very clearly that the quantum fluctuations for this spin-half Union Jack model modify the classical behavior only relatively modestly, providing further evidence to what we also noted earlier in relation to Fig. 3.

We also comment briefly on the large-$J_2$ behaviour of our results for the canted phase. (We note that for computational purposes it is easier to re-scale the original Hamiltonian of
FIG. 6: (Color online) Extrapolated curves (LSUB∞) for the ground-state magnetic order parameters (i.e., the on-site magnetizations) $M_A$ at sites A (joined by 8 bonds to other sites) and $M_B$ at sites B (joined by 4 bonds to other sites) of the Union Jack lattice [and see Fig. 1a] versus $J_2$ for the Néel and canted phases of the spin-1/2 Union Jack Hamiltonian of Eq. (1) with $J_1 \equiv 1$. The CCM results using the canted model state are shown for various LSUBn approximations ($n = \{2, 4, 6\}$ and $n = \{3, 5, 7\}$) with the canting angle $\phi = \phi_{LSUBn}$ that minimizes $E_{LSUBn}(\phi)$.

Eq. (1) by putting $J_2 \equiv 1$ and considering small values of $J_1$. The most interesting feature of the CCM results using the canted state as model state is that in all LSUBn approximations with $n > 2$ a termination point $\kappa_{LSUBn}^t$ is reached, beyond which no real solution can be found, very similar to the termination points shown in Fig. 2. For the even-$n$ sequence the values are $\kappa_{LSUB4}^t \approx 80$ and $\kappa_{LSUB6}^t \approx 80$, whereas for the odd-$n$ sequence the values are $\kappa_{LSUB3}^t \approx 250$, $\kappa_{LSUB5}^t \approx 85$, and $\kappa_{LSUB7}^t \approx 55$. This is a first indication that the canted state becomes unstable at very large values of $\kappa$ against the formation of another (as yet unknown) state, as we discuss further in Sec. IV B below.

Extrapolations of the gs energy using the data before the terminations points $\kappa_{LSUBn}^t$ show that at large $J_2$ values we have $E/N \to -0.3349J_2$ using the even-$n$ LSUBn series and $E/N \to -0.3352J_2$ using the odd-$n$ series. These numerical coefficients are precisely half of the values quoted in Table III for the case $J_2 = 0$. This is exactly as expected since both the $\kappa \to 0$ and the $\kappa \to \infty$ limits of the Union Jack model are the square-lattice HAF, where in the latter case the square lattice contains only half the original sites, namely the A sites. Similarly, the extrapolated LSUB∞ values at larger values of $\kappa$ before the termination point...
FIG. 7: (Color online) The total ground-state magnetization per site, \( m = M_B - M_A \cos \phi \), of the Union Jack lattice versus \( J_2 \) of the spin-1/2 Union Jack Hamiltonian of Eq. (1) with \( J_1 \equiv 1 \). The CCM results using the canted model state are shown for various \( \text{LSUB}_n \) approximations \( (n = \{2, 4, 6\}) \) with the canting angle \( \phi = \phi_{\text{LSUB}_n} \) that minimizes \( E_{\text{LSUB}_n}(\phi) \). We also show the \( n \rightarrow \infty \) extrapolated result from using Eq. (7) and compare it with the classical value \( m^{\text{cl}} = \frac{1}{4}(1 - \cos \phi_{\text{cl}}) \).

for the on-site magnetization on the A sites are \( M_A \rightarrow 0.317 \) for the even-\( n \) \( \text{LSUB}_n \) series and \( M_A \rightarrow 0.306 \) for the odd-\( n \) series. Both values are again remarkably consistent with those shown in Table IIII for the \( J_2 = 0 \) limit. The corresponding asymptotic values for the B-site magnetization are consistent with \( M_B \rightarrow 0.5 \), as expected for large values of \( J_2 \).

B. Canted state versus the semi-striped state

We turn finally to our CCM results based on the use of the semi-striped state shown in Fig. 1b as the model state. Unlike in the case of the corresponding use of the canted state as model state, the results based on the semi-striped state do not terminate at a high value of \( J_2 \) (with \( J_1 \equiv 1 \)). We found no indication of such a termination value at any \( \text{LSUB}_n \) level of approximation for \( 2 \leq n \leq 7 \) for values of \( J_2 < 1000 \). All indications are thus that the semi-striped state is stable out to the \( J_2 \rightarrow \infty \) limit. Indeed for these \( \text{LSUB}_n \) levels the CCM solutions based on the semi-striped state as model state exist for all values \( J_2 > 1 \). For example, the \( \text{LSUB}_6 \) solution based on the semi-striped state terminates at a lower end-point \( J_2 \approx 0.41 \).
FIG. 8: (Color online) Comparison of the extrapolated (LSUB∞) curves for the
ground-state energy per spin of the spin-1/2 Union Jack model, using the LSUBn data
with \(n = \{2, 4, 6\}\) fitted to Eq. (6), for the CCM based on the canted and semi-stripped
states as model states. The LSUB4 and LSUB6 approximations (and hence also the
extrapolated curve) for the canted model state terminate at \(J_2 \approx 80\).

In Fig. 8 we compare the extrapolated (LSUB∞) values of the gs energy per spin, based
in each case on the LSUBn results with \(n = \{2, 4, 6\}\), for our CCM results using the canted
and semi-stripped states. Although results for the canted model state become unavailable for
\(J_2 \gtrsim 80\) for the LSUB4 and LSUB6 approximations, the results based on the canted state lie
lower in energy than those based on the semi-stripped state for all values of \(J_2 \lesssim 80\) for which
both sets of solutions exist. Although this is disappointing at first sight, the two sets of curves
become extremely close for larger values of \(J_2\) as can be seen from Fig. 8. Furthermore, we
have also attempted a simple power-law extrapolation of the quantity \(E/(NJ_2)\) for the gs
energy of the canted state in powers of \(1/J_2\), beyond the large-\(J_2\) LSUBn termination points
(viz. at \(J_2 \approx 80\) for the LSUB6 approximation). Fits to sixth-, seventh-, and eighth-order
polynomials give virtually identical results for values of \(J_2\) in the range \(80 \lesssim J_2 \lesssim 500\) and
these extrapolated curves do indicate that there is a second phase transition at \(\kappa_{c2} \approx 125\pm 5\)
between the canted and semi-stripped phases, such that for values \(\kappa > \kappa_{c2}\) the semi-stripped
phase becomes lower in energy.

The gs energy in both the canted and semi-stripped phases approaches the asymptotic
value \(E/N \approx -0.3349J_2\) for large values of \(J_2\) as \(J_2 \to \infty\) (with \(J_1 \equiv 1\)). The corresponding
asymptotic (\(J_2 \to \infty\)) values for the average on-site magnetization of the semi-stripped state
are $M_A \to 0.317$ for the A sites and $M_B \to 0.5$ for the B sites.

V. DISCUSSION AND CONCLUSIONS

In this paper we have used the CCM to study the influence of quantum fluctuations on the zero-temperature gs phase diagram of a frustrated spin-half Heisenberg antiferromagnet (HAF) defined on the 2D Union Jack lattice. We have studied the case where the NN $J_1$ bonds are antiferromagnetic ($J_1 > 0$) and the competing NNN $J_2 \equiv \kappa J_1$ bonds in the Union Jack array have a strength in the range $0 \leq \kappa < \infty$. On the underlying bipartite square lattice there are thus two types of sites, viz. the A sites that are connected to the 4 NN sites on the B-sublattice with $J_1$-bonds and to the 4 NNN on the A-sublattice with $J_2$-bonds, and the B sites that are connected only to the 4 NN sites on the A-sublattice with $J_1$-bonds. The $\kappa = 0$ limit of the model thus corresponds to the spin-half HAF on the original square lattice (of A and B sites), while the $\kappa \to \infty$ limit corresponds to the spin-half HAF on the square lattice comprised of only A sites. We have seen that at the classical level (corresponding to the case where the spin quantum number $s \to \infty$) this Union Jack model has only two stable gs phases, one with Néel order for $\kappa < \kappa_{c1}^{cl} = 0.5$ and another with canted ferrimagnetic order for $\kappa > \kappa_{c1}^{cl}$. We have therefore first used these two classical states as CCM model states to investigate the effects of quantum fluctuations on them.

For the spin-half model we find that the phase transition between the Néel antiferromagnetic phase and the canted ferrimagnetic phase occurs at the higher value $\kappa_{c1} = 0.66 \pm 0.02$. The evidence from our calculations is that the transition at $\kappa_{c1}$ is a subtle one. From the energies of the two phases it appears that the transition is either second-order, as in the classical case, or possibly, weakly first-order. However, on neither side of the transition at $\kappa_{c1}$ does the order parameter $M$ (i.e., the average on-site magnetization) go to zero. Instead as $\kappa \to \kappa_{c1}$ from either side, $M \to 0.195 \pm 0.005$, which is more indicative of a first-order transition. Furthermore, the slope $dM/d\kappa$ of the average on-site magnetization as a function of $\kappa$ also seems to be either continuous or to have only a very weak discontinuity at $\kappa = \kappa_{c1}$.

Before continuing with the possibility of a further phase we compare our results with those from previous calculations of the same model using spin-wave theory (SWT)\textsuperscript{26,27} and the linked-cluster series expansion (SE) method.\textsuperscript{28} Collins et al.\textsuperscript{26,27} used linear (or leading-order) spin-wave theory (LSWT) to show that on the basis of a comparison of the gs energies
of the two phases, the phase transition between the Néel and canted phases is of first-order type and occurs at $\kappa_{c1} \approx 0.84$. In LSWT the Néel staggered magnetization per site $M$ remains substantial at this estimate of $\kappa_{c1} \approx 0.84$. However, it is well known that LSWT results become unreliable near the transition region, and they surmised that the Néel order parameter $M$ might vanish at or before this point, yielding a possible scenario where a second-order Néel transition might occur at a value $\kappa_{c1} \lesssim 0.84$, followed by a possible intermediate spin-liquid phase (as in the pure $J_1-J_2$ model, as discussed in Sec. I), and then a first-order transition to the canted phase at a somewhat larger value of $\kappa$. Our own results provide no evidence at all for such an intermediate spin-liquid phase between the Néel antiferromagnetic and canted ferrimagnetic phases.

Although LSWT is known to give a reasonable description of the spin-1/2 Heisenberg antiferromagnet on the square lattice ($\kappa = 0$), it is surely unable to model the frustrated or intermediate regime accurately. Similar shortcomings of spin-wave theory (SWT) have been noted by Igarashi in the context of the related spin-1/2 $J_1-J_2$ model on the square lattice, discussed briefly in Sec. I. He showed that whereas its lowest-order version (LSWT) works well when $J_2 = 0$, it consistently overestimates the quantum fluctuations as the frustration $J_2/J_1$ increases. In particular he showed, by going to higher orders in SWT in powers of $1/s$ where $s$ is the spin quantum number and LSWT is the leading order, that the expansion converges reasonably well for $J_2/J_1 \lesssim 0.35$, but for larger values of $J_2/J_1$, including the point $J_2/J_1 = 0.5$ of maximum classical frustration, the series loses stability. He also showed that the higher-order corrections to LSWT for $J_2/J_1 \lesssim 0.4$ make the Néel-ordered phase more stable than predicted by LSWT. He concluded that any predictions from SWT for the spin-1/2 $J_1-J_2$ model on the square lattice are likely to be unreliable for values $J_2/J_1 \gtrsim 0.4$. It is likely that a similar analysis of the SWT results for the spin-1/2 Union Jack model studied here would reveal similar shortcomings of LSWT as the frustration parameter $\kappa \equiv J_2/J_1$ is increased.

In a later paper by Zheng et al., SE techniques were applied to our spin-half Union Jack model and were compared with those from both LSWT for both the Néel and canted phases and modified second-order SWT for the Néel phase. Using the SE method for the Néel phase gave what these authors termed very clear evidence of a second-order phase transition at a critical coupling $\kappa_{c1} = 0.65 \pm 0.01$ at which the Néel staggered magnetization per site vanished. For higher couplings the system was seen to lie in the canted phase, with no sign
of any intermediate spin-liquid phase between these two magnetically ordered states. Use of the SE method in the canted phase produced a gs energy which continues smoothly from the Néel into the canted phase. Zheng et al.\textsuperscript{28} found, furthermore, that in the canted phase the staggered magnetizations per site in both the vertical and horizontal directions shown in Fig. 1a also appear to drop smoothly toward zero around the same value $\kappa_{c1} = 0.65 \pm 0.01$, albeit with very large error bars.

The above SE estimate for $\kappa_{c1}$ is clearly in excellent agreement with our own. However, whereas the evidence from the order parameter $M$ from the SE technique clearly favors a second-order transition at $\kappa_{c1}$ at which $M \to 0$ from both sides, our own CCM calculations clearly favor a first-order transition at which $M \to 0.195 \pm 0.005$. We note, however, that the errors on the SE estimates for $M$ become increasingly large as the phase transition at $\kappa_{c1}$ is approached from either side. We believe that this could easily account for the seeming discrepancy between our respective predictions for the order of the phase transition at $\kappa_{c1}$. We note too that Zheng et al.\textsuperscript{28} were themselves puzzled by the discrepancy between the prediction of SWT that the Néel magnetization per site $M$ does not vanish at $\kappa_{c1}$ and that of the SE technique that $M$ vanish does vanish there. While they recognized (as do we, as we discussed above) that SWT cannot be taken as an infallible quide, they found the huge difference with the prediction from the SE technique perturbing. Those authors ended by stating that, in their opinion, the nature of the transition from the Néel to the canted phase in the spin-half Union Jack model deserved further exploration. We believe that our own work reported here has considerably illuminated the transition at $\kappa_{c1}$.

Neither SWT nor SE techniques have been applied to the possible semi-striped state of Fig. 1b for the spin-half Union Jack model and so we have no results against which to compare our own. We were led to consider such a state as a possible gs phase of the model at large values of $\kappa$ as discussed in Sec. II. Thus, to recapitulate, the $\kappa \to \infty$ limit of the canted phase of the Union Jack model (for either the quantum $s = 1/2$ model considered here or the classical $s \to \infty$ case) gives a state in which the spins on the antiferromagnetically-ordered A-sublattice are orientated at $90^\circ$ to those on the ferromagnetically-ordered B-sublattice. The actual $\kappa \to \infty$ limit should, in either case, be decoupled antiferromagnetic (A) and ferromagnetic (B) sublattices, with complete degeneracy at the classical level for all angles of relative ordering directions between the two sublattices. We argued that quantum fluctuations could, in principle, lift this degeneracy by the well-known order by disorder phenomenon.\textsuperscript{29}
Since quantum fluctuations are also well known from many spin-lattice problems to favor
collinearity, there is a strong \textit{a priori} possibility that the semi-striped state of Fig. III might
be energetically favored at large values of \( \kappa \) over the non-collinear state which is the \( \kappa \to \infty \)
limit of the canted state in which \( \phi \to 90^\circ \).

Accordingly we repeated our CCM calculations using the semi-striped state as model
state. We found some evidence that at very large values of \( \kappa \) there might indeed be a second
phase transition at \( \kappa_{c_2} \approx 125\pm5 \), based on the relative energies of the canted and semi-striped
states. Such a prediction is based, however, on an extrapolation of the data on the canted
state into regimes where the CCM equations have no solution for LSUBn approximations
with \( n > 3 \), and hence cannot be regarded as being as reliable as our prediction for \( \kappa_{c_1} \).
If the phase transition at \( \kappa_{c_2} \) does exist it would be of first-order type according to our
results. It would be of considerable interest to explore the possible transition at \( \kappa_{c_2} \) between
the canted and semi-striped phases by other techniques, possibly including SWT and SE
methods.

As has been noted elsewhere, high-order CCM results of the sort presented here have
been seen to provide accurate and reliable results for a wide range of such highly frustrated
spin-lattice models. Many previous applications of the CCM to unfrustrated spin models
have given excellent quantitative agreement with other numerical methods (including exact
diagonalization (ED) of small lattices, quantum Monte Carlo (QMC), and series expansion
(SE) techniques). A typical example is the spin-half HAF on the square lattice, which is the
\( \kappa = 0 \) limit of the present model (and see Table III). It is interesting to compare for this \( \kappa = 0 \)
case, where comparison can be made with QMC results, the present CCM extrapolations
of the LSUBn data for the infinite lattice to the \( n \to \infty \) limit and the corresponding QMC
or ED extrapolations for the results obtained for finite lattices containing \( N \) spins that
have to be carried out to give the \( N \to \infty \) limit. Thus, for the spin-1/2 HAF on the square
lattice the “distance” between the CCM results for the ground-state energy per spin at the
LSUB6 (LSUB7) level and the extrapolated LSUB\( \infty \) value is approximately the same as the
distance of the corresponding QMC result for a lattice of size \( N = 12 \times 12 \) (\( N = 16 \times 16 \))
from its \( N \to \infty \) limit. The corresponding comparison for the magnetic order parameter
\( M \) is even more striking. Thus even the CCM LSUB6 result for \( M \) is closer to the LSUB\( \infty \)
limit than any of the QMC results for \( M \) for lattices of \( N \) spins are to their \( N \to \infty \) limit
for all lattices up to size \( N = 16 \times 16 \), the largest for which calculations were undertaken.
Such comparisons show, for example, that even though the “distance” between our \( \text{LSUB}n \) data points for \( M \) and the extrapolated \( (n \to \infty) \) \( \text{LSUB}\infty \) result shown in Fig. 5 may, at first sight, appear to be large, they are completely comparable to or smaller than those in alternative methods (where those other methods can be applied). Furthermore, where such alternative methods can be applied, as for the spin-1/2 HAF on the square lattice, the CCM results are in complete agreement with them.

By contrast, for frustrated spin-lattice models in two dimensions both the QMC and ED techniques face formidable difficulties. These arise in the former case due to the “minus-sign problem” present for frustrated systems when the nodal structure of the gs wave function is unknown, and in the latter case due to the practical restriction to relatively small lattices imposed by computational limits. The latter problem is exacerbated for incommensurate phases, and is compounded due to the large (and essentially uncontrolled) variation of the results with respect to the different possible shapes of clusters of a given size.

For highly frustrated spin-lattice models like the present Union Jack model, a powerful numerical method, complementary to the CCM, is the linked-cluster series expansion (SE) technique.\(^\text{13–17,44–48}\) The SE technique has also been applied to the present model.\(^\text{28}\) Our own results have shed considerable light on the nature of the phase transition at \( \kappa_{c1} \) observed by SE techniques and the discrepancies between the results from SE and SWT methods.

We end by remarking that it would also be of interest to repeat the present study for the case of the \( s > 1/2 \) Union Jack model. The calculations for this case are more demanding due to an increase at a given \( \text{LSUB}n \) level of approximation in the number of fundamental configurations retained in the CCM correlation operators. Nevertheless, we hope to be able to report results for this system in the future.

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