The frustrated Heisenberg antiferromagnet on the honeycomb lattice: J_1 – J_2 model

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Abstract

We study the ground-state phase diagram of the frustrated spin- $\frac{1}{2}$ J_1 – J_2 antiferromagnet with $J_2 = xJ_1 > 0$ ($J_1 > 0$) on the honeycomb lattice, using the coupled-cluster method. We present results for the ground-state energy, magnetic order parameter and plaquette valence-bond crystal (PVBC) susceptibility. We find a paramagnetic PVBC phase for $x_{c1} < x < x_{c2}$, where $x_{c1} \approx 0.207 \pm 0.003$ and $x_{c2} \approx 0.385 \pm 0.010$. The transition at x_{c1} to the Néel phase seems to be a continuous deconfined transition (although we cannot exclude a very narrow intermediate phase in the range $0.21 \lesssim x \lesssim 0.24$), while that at x_{c2} is of first-order type to another quasiclassical antiferromagnetic phase that occurs in the classical version of the model only at the isolated and highly degenerate critical point $x = \frac{1}{2}$. The spiral phases that are present classically for all values $x > \frac{1}{6}$ are absent for all $x \lesssim 1$.

(Some figures may appear in colour only in the online journal)

1. Introduction

Two-dimensional (2D) frustrated quantum spin–lattice systems have become of huge interest both theoretically and experimentally [1–3]. Attention has particularly focused on the rich panoply of (zero temperature, $T = 0$) quantum phase transitions that they exhibit [3, 4]. Without thermal fluctuations the transitions are driven solely by the interplay of quantum fluctuations and any frustration due to inherent competition between the interactions. Such frustration can arise either dynamically or geometrically. A prototypical example of the former is the well-studied J_1 – J_2 Heisenberg antiferromagnet (HAFM) on the bipartite square lattice (see, e.g., [5–7] and references cited therein), where nearest-neighbour (NN) spins interact via a Heisenberg interaction with strength parameter $J_1 > 0$, which competes with a Heisenberg interaction with strength parameter $J_2 > 0$ between next-nearest-neighbour (NNN) pairs. Similar

prototypical models exhibiting geometrical frustration are the pure NN HAFMs on the triangular [8] and kagome lattices [9]. For either form of frustration special interest then centres on the possible appearance of novel quantum ground-state (GS) phases without the long-range order (LRO) that typifies the classical GS phases of the corresponding models taken in the limit $s \rightarrow \infty$ of the spin quantum number s of the lattice spins. Examples include various valence-bond crystalline solid phases and spin-liquid phases.

Quantum fluctuations tend to be largest for the smallest values of s , for lower dimensionality D of the lattice, and for the smallest coordination of the lattice. Thus, for spin- $\frac{1}{2}$ models in $D = 2$, the honeycomb lattice plays a special role. Frustration is easily incorporated via competing NNN and maybe also next-next-nearest-neighbour (NNNN) bonds. Such models and their experimental realizations have been much studied in recent years [10–18]. Additional interest has also sprung from the recent synthesis of graphene

monolayers and other magnetic materials with a honeycomb structure. Theoretical interest was spurred by the discovery of a spin-liquid phase in the exactly solvable Kitaev model [19], in which spin- $\frac{1}{2}$ particles reside on a honeycomb lattice. Hubbard models on the honeycomb lattice may also describe many of the relevant physical properties of graphene. For example, evidence has been found [20] that quantum fluctuations are sufficiently strong to establish an insulating spin-liquid phase between the nonmagnetic metallic phase and the antiferromagnetic (AFM) Mott insulator phase, when the Coulomb repulsion parameter U becomes moderately strong. For large values of U the latter phase corresponds to the pure NN HAFM on the bipartite honeycomb lattice, whose GS phase exhibits Néel LRO. However, higher-order terms in the t/U expansion of the Hubbard model will surely lead to frustrating exchange couplings in the corresponding spin-lattice limiting model (and see, e.g., [21]), in which the HAFM with NN exchange couplings is the leading term in the large- U expansion. There is a growing consensus [10, 11, 13–16] that the frustrated spin- $\frac{1}{2}$ HAFM on the honeycomb lattice undergoes a frustration-induced quantum phase transition to a paramagnetic phase showing no magnetic LRO.

Indirect experimental backup for such theoretical findings comes from recent observations of spin-liquid-like behaviour in the layered compound $\text{Bi}_3\text{Mn}_4\text{O}_{12}(\text{NO}_3)$ (BMNO) at temperatures below its Curie–Weiss temperature [12]. In BMNO the Mn^{4+} ions reside on the sites of (weakly coupled) honeycomb lattices, but they have spin quantum number $s = \frac{3}{2}$. The successful replacement of the Mn^{4+} ions in BMNO by V^{4+} ions could lead to the realization of a corresponding $s = \frac{1}{2}$ model on the honeycomb lattice. Other recent realizations of HAFMs on a honeycomb lattice include compounds such as $\text{Na}_3\text{Cu}_2\text{SbO}_6$ [22] and $\text{InCu}_{2/3}\text{V}_{1/3}\text{O}_3$ [23], in both of which the Cu^{2+} ions in the copper oxide layers form a spin- $\frac{1}{2}$ HAFM on a (distorted) honeycomb lattice. Others include the family of compounds $\text{BaM}_2(\text{XO}_4)_2$ ($\text{M} = \text{Co}, \text{Ni}; \text{X} = \text{P}, \text{As}$) [24], in which the magnetic ions M are disposed in weakly coupled layers where they reside on the sites of a honeycomb lattice. The Co ions have spins $s = \frac{1}{2}$ and the Ni ions have spins $s = 1$. Finally, recent calculations of the low-dimensional material $\beta\text{-Cu}_2\text{V}_2\text{O}_7$ [25] show that its magnetic properties can be described in terms of a spin- $\frac{1}{2}$ model on a distorted honeycomb lattice.

2. The model

In recent papers [26, 27] we have studied the frustrated spin- $\frac{1}{2}$ J_1 – J_2 – J_3 model on the honeycomb lattice [10, 11, 13–18]. Its Hamiltonian is given by

$$H = J_1 \sum_{\langle i,j \rangle} \mathbf{s}_i \cdot \mathbf{s}_j + J_2 \sum_{\langle\langle i,k \rangle\rangle} \mathbf{s}_i \cdot \mathbf{s}_k + J_3 \sum_{\langle\langle\langle i,l \rangle\rangle\rangle} \mathbf{s}_i \cdot \mathbf{s}_l, \quad (1)$$

where index i runs over all honeycomb lattice sites, and indices j, k , and l run over all NN, NNN and NNNN sites to i , respectively, counting each bond once and once only. Each lattice site i carries a particle with spin $s = \frac{1}{2}$ and a spin operator $\mathbf{s}_i = (s_i^x, s_i^y, s_i^z)$. The lattice and exchange bonds are illustrated in figure 1.

Figure 1. The J_1 – J_2 model on the honeycomb lattice (with $J_1 = 1$), showing (a) the Néel, (b) spiral, and (c) anti-Néel states. The arrows represent spins located on lattice sites $\bullet$.

In earlier work we restricted ourselves to the case where $J_3 = J_2 > 0$, but we dealt both with the AFM case with $J_1 > 0$ [26] and the ferromagnetic (FM) case with $J_1 < 0$ [27]. Here we put $J_3 = 0$, and hence consider the J_1 – J_2 model. We restrict ourselves to the case where both bonds are of AFM type, $J_1 > 0$ and $J_2 \equiv xJ_1 > 0$.

In order to set the scene and to aid later understanding of the spin- $\frac{1}{2}$ GS phase diagram for the J_1 – J_2 model, it is appropriate to consider first the corresponding classical phase diagram. However, since, as we shall see below, phases that are not present in the classical J_1 – J_2 model, but which are present in the classical J_1 – J_2 – J_3 model for values $J_3 < 0$ only, it is very convenient to consider first the classical case with $J_3 \neq 0$ (i.e., without making the restriction $J_3 = 0$.)

The classical ($s \rightarrow \infty$) GS phase diagram of the J_1 – J_2 – J_3 model on the honeycomb lattice [11, 28] comprises six different phases when $J_1 > 0$ and the other two bonds, J_2 and J_3 , can take either sign. Three are collinear AFM phases, one is the FM phase, and the other two are different helical phases (and see, e.g., figure 2 of [11]). The AFM phases are the Néel phase (N) shown in figure 1(a), the striped (S) phase discussed in our earlier paper [26], and the anti-Néel (aN) phase shown in figure 1(c). The S, aN, and N states have, respectively, 1, 2, and all 3 NN spins to a given spin antiparallel to it. Equivalently, if we consider the sites of the honeycomb lattice as comprising a set of parallel sawtooth (or zigzag) chains (in any one of the three equivalent directions), the S state comprises alternating FM chains, while the aN state comprises AFM chains in which NN spins on adjacent chains are parallel. Although there are infinite manifolds of non-coplanar states degenerate in energy with each of the S and aN states at $T = 0$, both thermal and quantum fluctuations [11] select the collinear configurations.

When $J_3 > 0$ there is a region in which the spiral state shown in figure 1(b) is the stable GS phase. It is characterized by a pitch vector perpendicular to one of the J_1 bond directions, and a single spiral angle defined so that as we move along the parallel sawtooth chains (drawn in the horizontal direction in figure 1(b)) the spin angle increases by $\pi + \phi$ from one site to the next, and with NN spins on adjacent chains antiparallel. The classical GS energy is minimized for this spiral state when the pitch angle $\phi = \cos^{-1}[\frac{1}{4}(J_1 - 2J_2)/(J_2 - J_3)]$, when the energy per spin takes the value,

$$\frac{E_{\text{spiral}}^{\text{cl}}}{N} = \frac{s^2}{2} \left(-J_1 - 2J_2 + J_3 - \frac{1}{4} \frac{(J_1 - 2J_2)^2}{(J_2 - J_3)} \right). \quad (2)$$

We note that as $\phi \rightarrow 0$ this spiral state becomes the collinear N state with energy per spin,

$$\frac{E_N^{\text{cl}}}{N} = \frac{s^2}{2} (-3J_1 + 6J_2 - 3J_3), \quad (3)$$

and there is a continuous (second-order) phase transition between these two states on the boundary $y = \frac{3}{2}x - \frac{1}{4}$, for $\frac{1}{6} < x < \frac{1}{2}$, where $y \equiv J_3/J_1$ and $x \equiv J_2/J_1$. Similarly, as $\phi \rightarrow \pi$ the spiral state becomes the collinear S state, and there is another continuous (second-order) phase transition between the two states on the boundary line $y = \frac{1}{2}x + \frac{1}{4}$, for $x > \frac{1}{2}$. There is a first-order phase transition between the collinear N and S states along the boundary line $x = \frac{1}{2}$, for $y > \frac{1}{2}$. These three phases (N, S, and spiral) meet at the tricritical point $(x, y) = (\frac{1}{2}, \frac{1}{2})$. As $x \rightarrow \infty$ (for fixed finite y), the spiral pitch angle $\phi \rightarrow \frac{2}{3}\pi$. In this limit the model becomes two HAFMs on weakly connected interpenetrating triangular lattices, with the usual classical ordering of NN spins oriented at an angle $\frac{2}{3}\pi$ to each other on each sublattice.

The above three states are the only classical GS phases when $y > 0$. For $y < 0$ the N state persists in a region bounded by the same boundary line as above, $y = \frac{3}{2}x - \frac{1}{4}$, for $-\frac{1}{2} < x < \frac{1}{6}$, on which it continuously meets a second spiral state, and by the boundary line $y = -1$, for $x < -\frac{1}{2}$, at which it undergoes a first-order transition to the FM state, which itself is the stable GS phase in the region $x < -\frac{1}{2}$ and $y < -1$. Another collinear AFM state, the aN state shown in figure 1(c), with energy per spin,

$$\frac{E_{\text{aN}}^{\text{cl}}}{N} = \frac{s^2}{2} (-J_1 - 2J_2 + 3J_3), \quad (4)$$

becomes the stable GS phase in the region $x > \frac{1}{2}$, for $y < \frac{1}{2}\{x - [x^2 + 2(x - \frac{1}{2})^2]^{1/2}\}$. On the boundary it undergoes a first-order transition to the spiral state shown in figure 1(b). For $\frac{1}{6} < x < \frac{1}{2}$ the spiral state shown in figure 1(b) meets another spiral GS phase on the boundary line $y = 0$, at which point there is a first-order transition. This second spiral phase is characterized by a pitch vector parallel to one of the J_1 bond directions and by two spiral angles θ and ϕ . Within a unit cell the spin directions deviate from those in the N state by an angle ϕ , while as one advances from one unit cell to the next there is a twist by angle θ . The two pitch angles of this second spiral phase smoothly approach the value zero along the above boundary with the N state, and the value π along a second boundary curve that joins the points $(x, y) = (-\frac{1}{2}, -1)$ and $(\frac{1}{2}, 0)$, on which it meets the aN state. Both transitions are continuous (i.e., second-order) ones. This second spiral meets the three collinear states N, aN, and FM at the tetracritical point $(x, y) = (-\frac{1}{2}, -1)$.

Henceforth we restrict consideration to the J_1 - J_2 model where $J_3 = 0$ and where we define $J_1 \equiv 1$ to set the overall energy scale. The classical ($s \rightarrow \infty$) model thus has the N state as its GS for $J_2 < \frac{1}{6}$, whereas for $J_2 > \frac{1}{6}$ the GS comprises an infinite family of degenerate coplanar states with spiral order (including that shown in figure 1(b)), in which the spiral wavevector can point in any direction [11, 28, 29]. It is found [29] that, to leading order in $1/s$, spin-wave fluctuations

lift this degeneracy by the well-known order-by-disorder mechanism, in favour of specific wavevectors. However, these spiral states for the J_1 - J_2 model are expected to be very fragile against quantum fluctuations, and indeed to leading order in $1/s$ the spin-wave correction to the spiral order parameter has been shown to diverge as $\log N$, where N is the number of lattice sites [29], although it is still possible that higher-order terms in $1/s$ involving spin-wave interactions could stabilize the spiral order for large enough values of s . In view of the close proximity of the classical collinear (AFM) aN state for small values of the NNNN coupling J_3 in the J_1 - J_2 - J_3 model, it seems very likely that spiral order in the spin- $\frac{1}{2}$ J_1 - J_2 model might well be totally absent.

To investigate this question further, and more generally to consider the entire $T = 0$ phase diagram of the J_1 - J_2 model, we utilize the coupled-cluster method (CCM) [30–32] as in our work for the analogous J_1 - J_2 - J_3 model with $J_3 = J_2$ [26]. When used at high orders in the systematic approximation schemes developed for it, the CCM is an accurate approach to tackling a wide variety of quantum spin systems [5–7, 33–39]. In particular, it can accurately locate the quantum critical points (QCPs) in such frustrated systems [6, 7, 34, 35, 37, 39], as well as helping to determine the nature of the phases involved, including any quantum paramagnetic phases without magnetic order [7].

3. The coupled-cluster method

Since the CCM is a size-extensive method it provides results in the limit $N \rightarrow \infty$ from the outset. However, it requires us to input a model (or reference) state, with respect to which the quantum correlations may, in principle, be exactly included (and see, e.g., [5, 40, 41] and references cited therein). We use here the Néel (N), spiral, and anti-Néel (aN) states shown in figure 1 as our CCM model states. The CCM then incorporates multispin correlations on top of the chosen GS model state $|\Phi\rangle$ for the correlation operators S and $\tilde{S}$ that parametrize the exact GS ket and bra wavefunctions of the system in the respective exponentiated forms $|\Psi\rangle = e^S|\Phi\rangle$ and $\langle\tilde{\Psi}| = \langle\Phi|\tilde{S}e^{-\tilde{S}}$, where $\langle\tilde{\Psi}|\Psi\rangle \equiv 1$. The Schrödinger ket and bra equations are $H|\Psi\rangle = E|\Psi\rangle$ and $\langle\tilde{\Psi}|H = E\langle\tilde{\Psi}|$ respectively. The correlation operators are defined as $S = \sum_{I \neq 0} \tilde{S}_I C_I^+$ and $\tilde{S} = 1 + \sum_{I \neq 0} \tilde{S}_I C_I^-$ respectively. The operators $C_I^+ \equiv (C_I^-)^\dagger$ and C_I^- are the creation and destruction operators respectively, where $C_0^+ \equiv 1$ and $\langle\Phi|C_I^+ = 0; \forall I \neq 0$. The set $\{C_I^+\}$ forms a complete set of multispin creation operators with respect to the model state $|\Phi\rangle$ as a generalized vacuum.

In order to treat each lattice site in the spin system on an equal basis it is extremely convenient to rotate the local spin axes on each site in such a way that all the spins of each CCM reference state used point along the negative z -direction. Such rotations in spin space are obviously canonical transformations that have no effect on the fundamental $SU(2)$ commutation relations. The spins of our system are then represented entirely by these locally defined spin coordinate frames. The multispin creation operators may be written as linear sums of products of the individual spin raising operators $s_k^+ \equiv s_k^x + is_k^y$, i.e., $C_I^+ \equiv s_{k_1}^+ s_{k_2}^+ \cdots s_{k_n}^+$.

We then calculate the correlation coefficients $\{\mathcal{S}_i, \tilde{\mathcal{S}}_i\}$ by minimizing the GS energy expectation value $\bar{H} \equiv \langle \Psi | H | \Psi \rangle$ with respect to each of them. This yields the coupled sets of equations $\langle \Phi | C_I^- e^{-S} H e^S | \Phi \rangle = 0$ and $\langle \Phi | \tilde{S} (e^{-S} H e^S - E) C_I^+ | \Phi \rangle = 0; \forall I \neq 0$, which are used to calculate the ket- and bra-state correlation coefficients within a specified systematic approximation hierarchy that is a truncation scheme for the retained multispin configuration set $\{I\}$, as described below. In practice it is necessary to use parallel computing routines for the high-order approximations used here [40, 42]. After calculating the correlation coefficients ($\mathcal{S}_i, \tilde{\mathcal{S}}_i$) at a given level of approximation, we can then compute the GS energy using $E = \langle \Phi | e^{-S} H e^S | \Phi \rangle$, and the magnetic order parameter, which is defined to be the average on-site magnetization, $M = -\frac{1}{N} \langle \Psi | \sum_{i=1}^N \hat{S}_i^z | \Psi \rangle$, with respect to the local rotated spin coordinates defined above.

For the $s = \frac{1}{2}$ case considered here we use the well-tested localized (lattice-animal-based subsystem) LSUB m truncation scheme which includes all multispin correlations in the CCM correlation operators over all regions (or lattice animals) on the (here, honeycomb) lattice defined by m or fewer contiguous lattice sites. Such clusters are defined to be contiguous in this sense if every site in the cluster is adjacent (as a nearest neighbour) to at least one other site in the cluster. The number N_f of such fundamental configurations that are distinct under the (space and point-group) symmetries of the lattice and the model state in various LSUB m approximations increases rapidly with the truncation index m . For example, the highest LSUB m level that we can reach, even with massive parallelization and the use of supercomputing resources, is LSUB12, for which $N_f = 293\,309$ for the aN state. The raw LSUB m data still need to be extrapolated to the exact $m \rightarrow \infty$ limit. Although there are no exact extrapolation rules we have a great deal of experience in doing so. Thus, for the GS energy per spin, E/N , we use (see, e.g., [6, 7, 34, 35, 37, 41])

$$E(m)/N = a_0 + a_1 m^{-2} + a_2 m^{-4}; \quad (5)$$

while for the magnetic order parameter, M , defined above, we use either the scheme

$$M(m) = b_0 + b_1 m^{-1} + b_2 m^{-2}, \quad (6)$$

for systems showing no or only slight frustration (see, e.g., [33, 34]), or the scheme

$$M(m) = c_0 + c_1 m^{-1/2} + c_2 m^{-3/2}, \quad (7)$$

for more strongly frustrated systems or ones showing a GS order-disorder transition (see, e.g., [6, 7, 37, 39]). Since the hexagon is an important structural element of the honeycomb lattice we perform the extrapolations only for LSUB m data with $m \geq 6$. Thus, we present below CCM results with $m = \{6, 8, 10, 12\}$ and the corresponding extrapolations using this data set. However, we have also performed separate extrapolations using data sets with $m = \{4, 6, 8, 10\}$, $\{6, 8, 10\}$, $\{2, 4, 6, 8\}$, and $\{4, 6, 8\}$. They yield very similar results in all cases, which gives credence to our results and demonstrates their robustness with respect to the extrapolation schemes used. Such sensitivity checks also provide us with a means to estimate the error bars associated with our results. The error bars quoted later in section 4 are obtained in this fashion.

Figure 2. Difference between the GS energies per spin ($e \equiv E/N$) of the spiral and anti-Néel phases ($\Delta e \equiv e^{\text{spiral}} - e^{\text{aN}}$) versus J_2 for the spin- $\frac{1}{2}$ J_1 - J_2 honeycomb model ($J_1 = 1$) in LSUB m approximations with $m = \{6, 8, 10\}$.

4. Results and discussion

We show in figure 2 the difference in the GS energies of the CCM LSUB m results based on the spiral and aN model states. For the spiral state the pitch angle at a given LSUB m level is chosen to minimize the corresponding estimate for the GS energy. Although the energy differences are small the results for all values of m , as well as the extrapolated results, show clearly that, as expected from our previous discussion, the spiral state that is the classical GS phase for all values $x \equiv J_2/J_1 > 0.5$ gives way to the collinear aN state to become the stable GS phase for the spin- $\frac{1}{2}$ model for a considerable range of values of x . If a quantum phase transition between the spiral and aN states does exist, figure 2 shows that it can occur only at a value $x > 1$. Our result that the spiral phase is disfavoured with respect to the aN state is in complete agreement with that of a recent large-scale exact diagonalization (ED) study [16], but in disagreement with that from a series expansion (SE) study [17]. Henceforth we concentrate on GS phases other than the spiral phase.

In figure 3 we show results for the GS energy per spin, E/N , using the N and aN states as model states. We observe that each of the energy curves based on a particular model state terminates at some critical value of x (that itself depends on the LSUB m approximation used), beyond which no real CCM solution can be found. We note that in figure 2 results are shown for each LSUB m case down to values of x at which real solutions based on the spiral model state cease to exist. In all cases the corresponding termination point at a given LSUB m level shown in figure 3 for aN model state is lower than that for the equivalent spiral model state case. Such terminations of the CCM solutions are well understood [32, 38]. They are simply manifestations of the quantum phase transitions in the real system, and may thus be used to estimate the positions of the corresponding QCPs [32], although we do not do so here since we have more accurate criteria available as discussed below. We note, however, that as is usually the case, the CCM LSUB m results for finite m values for both

Figure 3. CCM LSUB m results for the GS energy, E/N , of the Néel and anti-Néel phases of the spin- $\frac{1}{2}$ J_1 - J_2 honeycomb model ($J_1 = 1$), with $m = \{6, 8, 10, 12\}$ and the extrapolated LSUB ∞ result using this data set.

the N and aN phases shown in figure 3 extend beyond the corresponding LSUB ∞ transition points. For large values of m each LSUB m transition point is quite close to the actual QCP where that phase ends. For example, the LSUB12 termination points shown in figure 3 are at $x_t^N \approx 0.23$ for the N state and $x_t^{aN} \approx 0.35$ for the aN state. The CCM results show a clear intermediate regime in which neither of the quasiclassical AFM states (N or aN) is stable.

We now discuss the magnetic order parameter, M , in order to investigate the stability of quasiclassical magnetic LRO. Our CCM results for M are shown in figure 4. The extrapolated Néel order parameter goes to zero at a value x_{c1} that is very insensitive both to which extrapolation scheme is used of equations (6) or (7), and to whether or not we include the LSUB6 data point in the extrapolations. Our best estimate is $x_{c1} \approx 0.207 \pm 0.003$. This value can be considered as our first CCM estimate of the corresponding QCP of the model. It is in reasonable agreement with, but more precise than, similar estimates for x_{c1} of about 0.17–0.22 from an ED approach [16], about 0.2 from an alternative ED approach [13], about 0.2 from a Schwinger boson approach [10], about 0.13–0.17 from a pseudo-fermion functional renormalization group (PFFRG) approach [15], and about 0.20 from a SE approach [17], but in substantial disagreement with a recent variational Monte Carlo (VMC) estimate of about 0.08 [18]. As the authors admit, the VMC study seems to substantially underestimate the QCP x_{c1} at which Néel order disappears. Indeed, the VMC result is itself in substantial disagreement with a more recent variational calculation based on a very broad family of plaquette-entangled states [43] that we discuss more fully in section 5, and which gives an estimate $x_{c1} \approx 0.2$, in full agreement with us. As expected, our own estimate shows that, as usual, quantum fluctuations preserve the collinear Néel order to stronger frustrations than the corresponding classical transition to noncollinear spiral order at $x_{cl} = \frac{1}{6}$.

By contrast with the situation at the lower QCP at x_{c1} , figure 4 shows that the corresponding QCP at x_{c2} at which

Figure 4. CCM LSUB m results for the GS order parameter, M , of the Néel and anti-Néel phases of the spin- $\frac{1}{2}$ J_1 - J_2 honeycomb model ($J_1 = 1$), with $m = \{8, 10, 12\}$, and the extrapolated LSUB ∞ (1) and LSUB ∞ (2) results using this data set and equations (6) and (7) respectively.

the anti-Néel order vanishes is considerably more difficult to estimate from the extrapolated CCM LSUB m values, with estimates that range from 0.47 to 0.64. We find a much more accurate estimate for x_{c2} below. Nevertheless it is clear already that a new quantum phase exists in the range $x_{c1} < x < x_{c2}$. It is also clear that, as suggested above, the two QCPs are very close to the corresponding CCM termination points seen in figure 3. In our previous work on the spin- $\frac{1}{2}$ J_1 - J_2 - J_3 model on the honeycomb lattice [26] with $J_1 = 1$, we found strong evidence for a nonmagnetic plaquette valence-bond crystal (PVBC) phase along the line $J_3 = J_2$ between the two quasiclassical AFM phases, namely the Néel (N) and striped (S) phases. It seems likely that the corresponding phase in the present $J_3 = 0$ case, which now intervenes between the N and aN phases, might also be the same PVBC phase.

In order to investigate the possibility of a PVBC phase we consider a generalized susceptibility χ_F that describes the response of the system to a ‘field’ operator F (see, e.g., [7]). A field term $F = \delta \hat{O}$ is added to the Hamiltonian (1), where $\hat{O}$ is an operator which corresponds here to the possible PVBC order illustrated in figure 5, and which thus breaks the translational symmetry of H . The energy per site $E(\delta)/N \equiv e(\delta)$ is then calculated in the CCM for the perturbed Hamiltonian $H + F$, using both the N and aN model states. The susceptibility is defined as $\chi_F \equiv -(\partial^2 e(\delta))/(\partial \delta^2)|_{\delta=0}$. Clearly, the GS phase becomes unstable against the perturbation F when χ_F^{-1} becomes zero. As in [26] we use the extrapolation scheme $\chi_F^{-1}(m) = d_0 + d_1 m^{-2} + d_2 m^{-4}$.

Thus, in the present case we now set the field term $F \rightarrow \delta \hat{O}_p$, where $\hat{O}_p$ is defined and shown diagrammatically in figure 5. Our CCM results for the inverse plaquette susceptibility, χ_p^{-1} , thus defined, are also shown in figure 5. The number of LSUB12 fundamental configurations for the plaquette susceptibility for the aN state is $N_f = 877\,315$. The extrapolated inverse susceptibility vanishes on the Néel side at $x \approx 0.24 \pm 0.01$ and on the anti-Néel side at $x \approx 0.385 \pm 0.010$.

Figure 5. Left: CCM LSUB m results for the inverse plaquette susceptibility, $1/\chi_p$, of the Néel and anti-Néel phases of the spin- $\frac{1}{2}$ J_1 - J_2 honeycomb model ($J_1 = 1$) with $m = \{6, 8, 10, 12\}$, and the extrapolated results LSUB ∞ (1) using $m = \{6, 8, 10, 12\}$ and LSUB ∞ (2) using $m = \{8, 10, 12\}$ (see text). Right: the fields $F \rightarrow \delta\hat{O}_p$ for the plaquette susceptibility, χ_p . Thick (red) and thin (black) lines correspond respectively to strengthened and weakened NN exchange couplings, where $\hat{O}_p = \sum_{\langle ij \rangle} a_{ij} \mathbf{s}_i \cdot \mathbf{s}_j$, and the sum runs over all NN bonds, with $a_{ij} = +1$ and -1 for thick (red) and thin (black) lines respectively.

The shape of the CCM curves where $\chi_p^{-1} \rightarrow 0$ on the Néel side is strongly suggestive of a continuous transition there, just as we found for the corresponding J_1 - J_2 - J_3 model [26]. The shallow slope of the χ_p^{-1} curves there makes it difficult to estimate accurately the point where it vanishes. Nevertheless it is certainly consistent with the much more accurate value we obtained for x_{c1} above from the point where the Néel order parameter M vanishes. On the other hand we cannot exclude the possibility of the transition between the Néel and PVBC states occurring via an intermediate phase that exists in the region $x_{c1} < x \lesssim 0.24$. Just such an intermediate (resonating valence-bond spin-liquid) state has been discussed in [11]. By contrast, the shape of the CCM curves for χ_p^{-1} on the anti-Néel side are much more indicative of a first-order transition, and the point where $\chi_p^{-1} \rightarrow 0$ on that side gives us our best estimate for $x_{c2} \approx 0.385 \pm 0.010$. Again, this value is in good agreement with, but much more accurate than, estimates of about 0.35–0.4 from two different ED studies [13, 16]. On both the N and aN sides it seems very likely that the PVBC phase occurs at, or very close to, the QCPs where the quasiclassical magnetic LRO in the N and aN phases vanishes. Since the N and PVBC phases break different symmetries, and our CCM results show that they appear to meet at $x_{c1} \approx 0.21$ at a continuous transition, they support the deconfinement scenario there. The possibility of deconfined quantum criticality for the frustrated honeycomb HAFM was pointed out in [1], and supporting evidence for the J_1 - J_2 - J_3 model has also been found both by us [26] and by others [16].

5. Summary

We have studied the influence of quantum spin fluctuations on the GS properties of the spin- $\frac{1}{2}$ J_1 - J_2 HAFM ($J_1 > 0, x \equiv J_2/J_1$) on the honeycomb lattice. We find a paramagnetic

PVBC phase in the regime $x_{c1} < x < x_{c2}$, where $x_{c1} \approx 0.207 \pm 0.003$ and $x_{c2} \approx 0.385 \pm 0.010$. The transition at x_{c1} to the Néel phase appears to be a continuous transition of the deconfinement variety, while that at x_{c2} is of first-order type to an anti-Néel-ordered AFM phase that does not occur in the classical version of the model except at the critical point $x = \frac{1}{2}$. Due to the difficulty in determining the lower critical value of x at which PVBC order is established as accurately as we determine the value $x = x_{c1}$ at which Néel order is destabilized, we cannot however exclude the possibility that the transition from the N phase to the PVBC phase might proceed via an intervening phase, possibly of an exotic spin-liquid type, in the narrow window $0.21 \lesssim x \lesssim 0.24$.

During the process of submitting this paper we became aware of a very recent study [43] of the same model as is studied here, using an entangled-plaquette variational (EPV) ansatz. Unlike the VMC calculations cited above [18], which are mainly based on a comparison of energy estimates from variational wavefunctions corresponding to GS phases with different order (or disorder), the EPV study uses a (single) very broad variational class of entangled-plaquette states. It is then found that Néel order persists for $x \lesssim 0.2$, in much closer agreement with our own estimate. The EPV study also finds a collinear phase with anti-Néel order for $x \gtrsim 0.4$, again in very good agreement with our own results. For values $0.2 \lesssim x \lesssim 0.4$ the EPV study finds that none of the examined order parameters remains finite in the infinite lattice limit, and concludes that the GS phase is disordered, whereas we find evidence for a PVBC phase in that region.

It is clear that the spin- $\frac{1}{2}$ J_1 - J_2 HAFM on the honeycomb lattice is a challenging model, with different theoretical methods giving differing results, as we have indicated above. Nevertheless, there appears to be a growing consensus that the model is Néel-ordered for $x < x_{c1}$, and anti-Néel-ordered for $x > x_{c2}$, with both a recent ED study [16] and an EPV study [43] concurring with us on these two phases. Both our CCM and two quite separate ED calculations [13, 16] indicate that the intervening phase has PVBC order, while the EPV calculations seem to favour a disordered (spin-liquid) phase. One of the ED studies [13] uses a NN singlet valence-bond basis and finds a staggered dimerized phase (which is also known in the literature as a lattice nematic phase [29]) in the region $x > x_{c2}$ beyond the melting of PVBC order. We note, however, that their results would be equally consistent with the identification of this phase as our aN phase.

As a concluding remark we note that the CCM has been used in the present paper with reference states that are classical states built from independent-spin product states. In view of the lack of agreement over what form of order (if any) is present in the intermediate quantum paramagnetic phase, it would be of interest, if that were possible, to repeat the CCM calculations with model states that are ideal valence-bond crystalline (VBC) states of the staggered dimer or plaquette kind. We note that indeed such a generalized CCM approach has recently been formulated [36]. It combines exact solutions for dimer or plaquette VBC ground states with the computational implementation described and used here for model states of the independent-spin product variety. Such

a calculation for the hexagonal plaquette of the honeycomb lattice has never yet been undertaken, however, and its use would be far outside the scope of the present paper.

Finally, we note that our results indicate that the spiral GS phases that exist classically for all values $x > \frac{1}{6}$ are absent for all values $x \lesssim 1$, but may exist for larger values. This finding is in accord with both two recent ED studies [13, 16] and a very recent EPV study [43]. Nevertheless, it is in disagreement with the contrary findings from a recent SE study [17] and from a recent PFFRG approach [15], both of which conclude that spiral order is present in the model for values of x beyond where the intervening (PVBC or disordered) state ceases to be the stable GS phase. To investigate this and other aspects of the model further we shall present results in a future paper on the phase diagram of the extended J_1 – J_2 – J_3 model, using the same CCM techniques.

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References

- [1] Senthil T, Vishwanath A, Balents L, Sachdev S and Fisher M P A 2004 *Science* **303** 1490
- [2] Moessner R and Ramirez A P 2006 *Phys. Today* **59** (Feb) 24
- [3] Schollwöck U, Richter J, Farnell D J J and Bishop R F (ed) 2004 *Quantum Magnetism (Springer Lecture Notes in Physics vol 645)* (Berlin: Springer)
- [4] Sachdev S 1999 *Quantum Phase Transitions* (Cambridge: Cambridge University Press)
- [5] Bishop R F, Farnell D J J and Parkinson J B 1998 *Phys. Rev. B* **58** 6394
- [6] Bishop R F, Li P H Y, Darradi R, Schulenburg J and Richter J 2008 *Phys. Rev. B* **78** 054412
- [7] Darradi R, Derzhko O, Zinke R, Schulenburg J, Krüger S E and Richter J 2008 *Phys. Rev. B* **78** 214415
- [8] Bernu B, Lhuillier C and Pierre L 1992 *Phys. Rev. Lett.* **69** 2590
- [9] Schnyder A P, Starykh O A and Balents L 2008 *Phys. Rev. B* **78** 174420
- [10] Mattsson A, Fröjd P and Einarsson T 1994 *Phys. Rev. B* **49** 3997
- [11] Fouet J B, Sindzingre P and Lhuillier C 2001 *Eur. Phys. J. B* **20** 241
- [12] Okubo S, Elmasry F, Zhang W, Fujisawa M, Sakurai T, Ohta H, Azuma M, Sumirnova O A and Kumada N 2010 *J. Phys.: Conf. Ser.* **200** 022042
- [13] Mosadeq H, Shahbazi F and Jafari S A 2011 *J. Phys.: Condens. Matter* **23** 226006
- [14] Cabra D C, Lamas C A and Rosales H D 2011 *Phys. Rev. B* **83** 094506
- [15] Reuther J, Abanin D A and Thomale R 2011 *Phys. Rev. B* **84** 014417
- [16] Albuquerque A F, Schwandt D, Hetényi B, Capponi S, Mambrini M and Läuchli A M 2011 *Phys. Rev. B* **84** 024406
- [17] Oitmaa J and Singh R R P 2011 *Phys. Rev. B* **84** 094424
- [18] Clark B K, Abanin D A and Sondhi S L 2011 *Phys. Rev. Lett.* **107** 087204
- [19] Kitaev A 2006 *Ann. Phys.* **321** 2
- [20] Meng Z Y, Lang T C, Wessel S, Assaad F F and Muramatsu A 2010 *Nature* **464** 847
- [21] Yang H Y and Schmidt K P 2011 *Europhys. Lett.* **94** 17004
- [22] Miura Y, Hiari R, Kobayashi Y and Sato M 2006 *J. Phys. Soc. Japan* **75** 084707
- [23] Kataev V, Möller A, Löw U, Jung W, Schittner N, Kriener M and Freimuth A 2005 *J. Magn. Magn. Mater.* **290/291** 310
- [24] Regnault L P and Rossat-Mignod J 1990 *Phase Transitions in Quasi-Two-Dimensional Planar Magnets* ed L J De Jongh (Dordrecht: Kluwer) p 271
- [25] Tsirlin A A, Janson O and Rosner H 2010 *Phys. Rev. B* **82** 144416
- [26] Farnell D J J, Bishop R F, Li P H Y, Richter J and Campbell C E 2011 *Phys. Rev. B* **84** 012403
- [27] Li P H Y, Bishop R F, Farnell D J J, Richter J and Campbell C E 2011 *Phys. Rev. B* **85** 085115
- [28] Rastelli E, Tassi A and Reatto L 1979 *Physica B+C* **97** 1
- [29] Mulder A, Ganesh R, Capriotti L and Paramakanti A 2010 *Phys. Rev. B* **81** 214419
- [30] Bishop R F 1991 *Theor. Chim. Acta* **80** 95
- [31] Bishop R F 1998 *Microscopic Quantum Many-Body Theories and Their Applications (Springer Lecture Notes in Physics vol 510)* ed J Navarro and A Polls (Berlin: Springer) p 1
- [32] Farnell D J J and Bishop R F 2004 *Quantum Magnetism (Springer Lecture Notes in Physics vol 645)* ed U Schollwöck, J Richter, D J J Farnell and R F Bishop (Berlin: Springer) p 307
- [33] Krüger S E, Richter J, Schulenburg J, Farnell D J J and Bishop R F 2000 *Phys. Rev. B* **61** 14607
- [34] Darradi R, Richter J and Farnell D J J 2005 *Phys. Rev. B* **72** 104425
- [35] Schmalfuß D, Darradi R, Richter J, Schulenburg J and Ihle D 2006 *Phys. Rev. Lett.* **97** 157201
- [36] Farnell D J J, Richter J, Zinke R and Bishop R F 2009 *J. Stat. Phys.* **135** 175
- [37] Richter J, Darradi R, Schulenburg J, Farnell D J J and Rosner H 2010 *Phys. Rev. B* **81** 174429
- [38] Bishop R F, Li P H Y, Farnell D J J and Campbell C E 2010 *Phys. Rev. B* **82** 024416
- [39] Reuther J, Wölfe P, Darradi R, Brenig W, Arlego M and Richter J 2011 *Phys. Rev. B* **83** 064416
- [40] Zeng C, Farnell D J J and Bishop R F 1998 *J. Stat. Phys.* **90** 327
- [41] Bishop R F, Farnell D J J, Krüger S E, Parkinson J B, Richter J and Zeng C 2000 *J. Phys.: Condens. Matter* **12** 6887
- [42] We use the program package of Farnell D J J and Schulenburg J see www-e.uni-magdeburg.de/jschulen/ccm/index.html.
- [43] Mezzacapo F and Boninsegni M 2012 *Phys. Rev. B* **85** 060402(R)