Valence-bond crystalline order in the $s = 1/2 J_1–J_2$ model on the honeycomb lattice

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Abstract

Using the coupled cluster method we study the phase diagram of the spin-$1/2$ Heisenberg antiferromagnet on a honeycomb lattice with nearest-neighbour exchange coupling $J_1 > 0$ and frustrating next-nearest-neighbour coupling $J_2 \equiv x J_1 > 0$. In the range $0 < x < 1$ we find four phases exhibiting respectively Néel, 6-spin plaquette, staggered dimer and Néel-II orderings, with quantum critical points at $x_c^1 \approx 0.207(3)$, $x_c^2 \approx 0.385(10)$ and $x_c^3 \approx 0.65(5)$. The transitions at $x_c^1$ and $x_c^3$ appear to be continuous (and hence deconfined) ones, while that at $x_c^2$ appears to be a direct first-order one.

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

1. Introduction

Frustrated quantum spin models on the two-dimensional (2D) honeycomb lattice have become the objects of intense study. Quantum fluctuations on spin lattices are generally larger for lower dimensionality $D$ and smaller values of the coordination number $z$ of the lattice, as well as for smaller values of the spin quantum number $s$ of the lattice spins. Spin-1/2 models on the honeycomb lattice (with $D = 2$ and $z = 3$) are thus expected to have large quantum fluctuations, which, in turn, open up the theoretical possibility of realizing exotic ground-state (GS) phases with novel magnetic properties and/or novel ordering.

Additional impetus for studying 2D honeycomb models came from the reported presence of a quantum spin-liquid (QSL) phase in both the exactly soluble (albeit somewhat artificial) Kitaev model of spin-1/2 particles on a honeycomb lattice [1], and the half-filled Fermi–Hubbard (FH) model on a honeycomb lattice [2]. Thus, Meng et al [2] reported in a quantum Monte Carlo (QMC) calculation, free of the usual fermion sign problems, the presence in the honeycomb FH model of a QSL phase, at moderate values of the onsite Coulomb repulsion strength ($U$), situated between the nonmagnetic metallic insulator (or semi-metal) phase at low $U$ and the antiferromagnetic (AFM) Mott insulator phase for large $U$. Since the $U \to \infty$ limit corresponds to the pure Heisenberg antiferromagnet (HAFM), i.e., with nearest-neighbour (NN) interactions (of strength $J_1 > 0$) only, the Mott insulator phase of the Hubbard model corresponds to the Néel-ordered phase of the HAFM spin–lattice model. Higher-order terms in the $t/U$ expansion of the FH model (where $t$ is the strength parameter of the NN hopping term) lead to frustrating exchange couplings in the corresponding spin–lattice model in which the HAFM with NN exchange couplings is the leading term in the large-$U$ expansion. The simplest such frustrated model is the $J_1–J_2$ model studied here, where the next-nearest-neighbour (NNN) spin pairs have an additional exchange coupling of strength $J_2 > 0$.

A later study of the FH model, using a Schwinger boson mean field theory (SB-MFT) approach [3], provided some corroborating evidence for a $\mathbb{Z}_2$ QSL state; and a Schwinger fermion representation of the same model [4] gave some evidence for both a $\mathbb{Z}_2$ QSL phase and a chiral antiferromagnetic phase. However, later numerically exact QMC calculations by Sorella et al [5], with much larger clusters than those used by Meng et al [2], have cast considerable doubt on their original finding of an intermediate
QSL phase. We note in this context that the presence of magnetically ordered phases is difficult to detect by standard QMC techniques when the ordering is small, since the usual quantity measured is the squared of the order parameter. As a consequence, in addition to the usual problem of finding an appropriate finite-size extrapolation formula, very large clusters are required with high precision. It is this effect that has apparently caused the controversy between [2] and [5] regarding the existence or not of an intermediate QSL phase in the FH model on the honeycomb lattice. In a very recent paper [6] this controversy has effectively been resolved by using a novel QMC technique that measures the local magnetic order parameter $M$ directly, rather than its square, $M^2$. Use of this technique leads [6] to the rather firm conclusion that in the FH model on the honeycomb lattice there is a single continuous quantum phase transition between the nonmagnetic semi-metal and AFM Mott insulator phases, with no intermediate QSL phase.

It is also pertinent to ask whether the $J_1-J_2$ model actually does represent well the low-energy physics of the FH model on the honeycomb lattice. While this is undoubtedly true for small enough values of the Hubbard parameter $t/U$, it is interesting to explore more deeply and quantitatively about this question. In particular, two recent studies [7, 8] have thrown considerable light on the relationship between the physics of FH and $J_1-J_2$ models on the honeycomb lattice. Thus, in the first place, it has been shown [7] that the ratio $x = J_2/J_1$ actually stays quite small over a large range of values of $t/U$. More specifically, it is always smaller than the value $x_{c1}$, which is the point at which the Néel order, present at $x = 0$, first vanishes as $x$ is increased, as we discuss below. Secondly, in a very interesting paper [8] that studied in detail the full low-energy spin model arising from the FH model on the honeycomb lattice, it was shown that six-spin interactions on hexagonal plaquettes are the most important leading correction to the NN $J_1$ bonds, rather than the NNN $J_2$ bonds.

Despite all of the above caveats of the relevance of the $J_1-J_2$ model on the honeycomb lattice to describe the low-energy physics of the corresponding FH honeycomb model, it remains of great interest in its own right. This has possibly even been heightened by the considerable uncertainty that has existed until very recently, as discussed above, as to whether or not a QSL phase exists for the FH model. For this and other reasons, this spin–lattice model and its generalizations (specifically to include also next-next-nearest-neighbour (NNNN) bonds with strength $J_3$), have been much studied [9–20] recently.

2. The model

The Hamiltonian of the model studied here is given by

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

where index $i$ runs over all honeycomb lattice sites, and indices $j$ and $k$ run over all NN and NNN sites to $i$, respectively, counting each bond once only. Each lattice site $i$ carries a particle with spin $s = \frac{1}{2}$ and a spin operator $\mathbf{s}_i = (s_x^i, s_y^i, s_z^i)$.

The lattice and exchange bonds are illustrated in figure 1. We are interested in the case where both NN and NNN bonds are AFM in nature, and henceforth we put $J_1 = 1$ to set the energy scale, and define the frustration parameter $x \equiv J_2/J_1$.

The classical ($s \to \infty$) ground state of the model is Néel-ordered for $0 \leq x < \frac{1}{2}$, whereas for all values $x > \frac{1}{2}$ the spins are spirally ordered. In this latter regime the classical model has a one-parameter family of degenerate incommensurate ground states where the spiral wavevector can orient in any direction. At leading order, i.e., $O(1/s)$, spin-wave fluctuations lift this accidental degeneracy in favour of particular wavevectors [9]. For the extreme quantum case, $s = \frac{1}{2}$, considered here, we expect quantum fluctuations to be strong enough to destroy the spiral order over a wide range of values of $x$. In a recent paper [15] that used the coupled cluster method (CCM), we have verified that expectation for all values in the range $0 \leq x \leq 1$ considered here.

We showed too [15] that quantum fluctuations preserve the Néel order to higher values of $x$ than in the classical model. Thus, we found that the GS phase of the $s = \frac{1}{2}$ model is Néel-ordered for $0 \leq x < x_{c1} \approx 0.207(3)$. At $x = x_{c1}$ there appears to be a continuous deconfined phase transition to a GS paramagnetic phase exhibiting plaquette valence-bond crystalline (PVBC) order. Furthermore, we found the PVBC state to be the stable GS phase in the regime $x_{c1} < x < x_{c2}$, where $x_{c2} \approx 0.385(10)$. Our aim now is to investigate further the transition at $x = x_{c2}$ and the nature of the GS phase(s) for $x > x_{c2}$.

3. Coupled cluster method

The CCM [21–23, 26], that we will employ here, has been very successfully applied to many models in quantum magnetism, including models on the honeycomb lattice [10, 15, 16] of interest here. It provides a well-structured means of studying various candidate GS phases and their regimes of stability, for each of which the description is systematically improvable in terms of well-defined truncation hierarchies for the quantum multispin correlations. We now briefly describe the method and refer the reader to the literature (see, e.g., [21–30]) for further details.

The starting point for any CCM calculation is the selection of a suitable normalized model (or reference) state $|\Phi\rangle$. For spin systems it is often convenient to take a classical

![Figure 1. The $J_1-J_2$ model on the honeycomb lattice (with $J_1 = 1$), showing (a) the Néel and (b) Néel-II states. The arrows represent spins located on lattice sites.](image-url)
employs the exponential parametrizations, \(z\) also easily seen to break the lattice rotational symmetry. The \(\text{N} \rightarrow \text{II}\) state is thus called striped states (see, e.g., [16]). The \(\text{N} \rightarrow \text{II}\) state is also AFM states on the honeycomb lattice that have also been avoided this name here since it is open to confusion with other AFM states on the honeycomb lattice that have also been called striped states (see, e.g., [16]). The \(\text{N} \rightarrow \text{II}\) state is thus also easily seen to break the lattice rotational symmetry.

It is convenient to perform a mathematical rotation of the local axes of the spins such that all spins in the reference state align along the negative \(z\)–axis. The Schrödinger ground-state ket and bra CCM equations are \(H(\Psi) = E(\Psi)\) and \(\langle \tilde{\Psi}|H = E(\tilde{\Psi})\) respectively. The CCM employs the exponential parametrizations, \(\Psi = e^s|\Phi\rangle\) and \(\langle \tilde{\Psi}| = \langle \Phi|e^{-s}|\Psi\rangle\). The correlation operator \(S\) is expressed as \(S = \sum_{I \neq j}S_I C_I^+\) and its counterpart is \(\tilde{S} = 1 + \sum_{I \neq j}S_I C_I^-\) where, by definition, \(C_I^+|\Phi\rangle = 0 = \langle \Phi|C_I^-\rangle, \forall I \neq 0\). Thus we have the normalization condition \(\langle \tilde{\Psi}|\Psi\rangle = \langle \Phi|\Phi\rangle = 1\). The multispin creation operators \(C_I^+\equiv (C_I^0)^\dagger\), with \(C_I^0 = 1\), are written as \(C_I^+ = s_i^+s_j^+\cdots s_n^+\) in terms of the single-site spin-raisers operators \(s_i^+ = s_i^x + i s_i^y\). The GS energy is \(E = \langle\Phi|e^{-H}\tilde{H}|\Phi\rangle\); and the local average onsite magnetization \(M\) in the rotated spin coordinates is \(M = -\frac{1}{N}\langle\tilde{\Psi}|\sum_{j=1}^N s_j^z|\Psi\rangle\).

The ket- and bra-state correlation coefficients \((S_I, \tilde{S}_I)\) are calculated by requiring the expectation value \(\tilde{H} = \langle \tilde{\Psi}|H|\Psi\rangle\) to be a minimum with respect to all parameters \((S_I, \tilde{S}_I)\), and hence \(\langle \Phi|C_I^+ e^{-H}H e^S|\Phi\rangle = 0\) and \(\langle \Phi|S(e^{-S}He^S - E_0)C_I^+|\Phi\rangle = 0; \forall I \neq 0\).

The CCM formalism is exact if all spin configurations are included in the \(S\) and \(\tilde{S}\) operators. In practice, however, truncations are needed. We employ here the well-studied localized (lattice-animal-based subsystem) LSUBm scheme [24–30], in which all possible multispin-flip correlations over different locales on the lattice defined by \(m\) or fewer contiguous lattice sites are retained. 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 interested reader is referred to the literature (see, e.g., [24]) for figures illustrating the LSUBm scheme in detail. The numbers \(Nf_i\) of such fundamental configurations that are distinct under the (space- and point-group) symmetries of the lattice and the model state increase rapidly with the LSUBm truncation index \(m\). Thus the highest LSUBm level that we can reach here, even with massive parallelization and the use of supercomputing resources\(^3\), is LSUB12, for which \(Nf = 293309\) for the \(\text{N} \rightarrow \text{II}\) state.

Since, in any truncation, CCM parametrizations automatically satisfy the Goldstone linked cluster theorem, we may work from the outset in the thermodynamic limit, \(N \rightarrow \infty\). Nevertheless, the raw LSUBm data still need to be extrapolated to the exact \(m \rightarrow \infty\) limit. Thus, for the GS energy per spin, \(E/N\), we use (see, e.g., [26–29])

\[
E(m)/N = a_0 + a_1m^{-2} + a_2m^{-4},
\]

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

\[
M(m) = b_0 + b_1m^{-1} + b_2m^{-3/2},
\]

for systems showing no or only slight frustration (see, e.g., [25, 27]), or the scheme

\[
M(m) = c_0 + c_1m^{-1/2} + b_2m^{-3/2},
\]

for more strongly frustrated systems or ones showing a GS order–disorder transition (see, e.g., [28, 29]).

In principle one may always test for the correct leading exponent in the LSUBm extrapolation scheme for any physical quantity \(Z\) by first fitting to the formula \(Z(m) = d_0 + d_1m^{-\nu}\). For the GS energy, \(E/N\), we generally find \(\nu \approx 2\) for a wide variety of spin systems, both non-frustrated and frustrated. For the magnetic order parameter, \(M\), on the other hand we generally find \(\nu \approx 1\) for unfrustrated systems (or for ones with very small frustration), and \(\nu \approx 0.5\) for more strongly frustrated systems. We discuss this more fully in section 4 in the context of the present model. These general results for the leading exponents then provide the basis for equations (2)–(4).

Finally, we note that since the hexagon is an important structural element of the honeycomb lattice we never use LSUBm data with \(m < 6\) to perform the extrapolations. Furthermore, in any CCM calculation using the LSUBm scheme, we always need to check whether the lowest-order potentially useable approximation, namely LSUB6 here, is actually useable in the sense of fitting the extrapolation scheme to be used. Although it generally does do so, there are also (relatively rare) occasions when it does not, presumably due either to the result being too far removed from the asymptotic \(m \rightarrow \infty\) limit or to the fact that for the particular CCM model state used these lowest-order approximants omit one or more of the most important multispin correlations.

### 4. Results and discussion

In figures 2 and 3 we show our results for the GS energy per spin, \(E/N\), and magnetic order parameter, \(M\), using both the \(\text{N} \rightarrow \text{II}\) states as CCM model states.

Figure 2 shows clearly that the CCM LSUBm results for the GS energy extrapolate extremely rapidly with increasing

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\(^3\) We use the program package CCM of Farnell DJ J and Schulenburg J see www-e.uni-magdeburg.de/jschulen/ccm/index.html.
order $m$ of approximation to the exact LSUB∞ limit. It also shows clearly how the LSUB$m$ results based on both the Néel and Néel-II model states naturally terminate at some critical values of the frustration parameter $x$, which themselves depend on the order parameter $m$ of the particular LSUB$m$ approximation, beyond which no real CCM solution can be found. Such termination points of CCM solutions are well studied (and see, e.g., [22, 23]) and well understood. They are simply reflections of the quantum phase transitions in the system and, as such, may themselves be used to estimate the positions of the corresponding quantum critical points [22]. We do not, however, examine the extrapolation properties of the termination points further here, since we have more accurate criteria available to us to determine the quantum critical points, as we discuss more fully below.

Nevertheless, figure 2 shows clearly that the CCM LSUB$m$ results based on both the Néel and Néel-II model states for finite values of $m$ extend beyond the corresponding LSUB∞ transition points into unphysical regions where such states in the real (LSUB∞) case have ceased to exist. Such unphysical regimes diminish in size to zero as $m \to \infty$. Figure 2 shows that there are no energy crossings between the Néel and Néel-II phases at any LSUB$m$ level of approximation, and that there is a clear range of values of the frustration parameter, $x_{c1} < x < x_{c2}$, in which neither the Néel nor the Néel-II states provide a physical GS phase. The simple unextrapolated LSUB12 estimates for the two termination points, namely $x_{c1} \approx 0.23$ and $x_{c2} \approx 0.35$ already provide remarkably good estimates for the corresponding quantum critical points, as we shall see below.

We note from figure 2 that the LSUB$m$ estimates for the GS energy approach the asymptotic LSUB∞ limit very rapidly, and hence the extrapolations are rather insensitive to both the fitting scheme and data set used. Nevertheless, a fit of the form $E(m)/N = e_0 + e_1 m^{-\nu}$ for the Néel-II LSUB$m$ results gives the usual expected result $\nu \approx 2$ for the data set $m = \{8, 10, 12\}$, whereas the inclusion of the LSUB6 result leads to a spurious value $\nu \approx 1$. By contrast, both data sets $m = \{6, 8, 10, 12\}$ and $m = \{8, 10, 12\}$ yield a value $\nu \approx 2$ for the corresponding LSUB$m$ Néel results. The anomalous nature of the LSUB6 Néel-II approximation is discussed further below with regard to the magnetic order parameter $M$, for which its behaviour is more critical and more pronounced.

We now turn our attention to the corresponding CCM LSUB$m$ results for the magnetic order parameter, as shown in figure 3, using both the Néel and Néel-II states as the CCM model states. For the present model we find that an extrapolation formula for the magnetic order parameter of the form $M(m) = d_0 + d_1 m^{-\nu}$ fits the data well on the Néel side with a leading exponent $\nu \approx 1$ for values of the frustration parameter $x$ equal to or very close to zero, whereas the value $\nu \approx 0.5$ accurately fits the data over most of the range $x \gtrsim 0.1$. Accordingly, in figure 3 on the Néel side we show extrapolations using both equations (3) and (4). Equation (3), which is appropriate when $J_2 = 0$, yields the value $M \approx 0.271(2)$ for the unfrustrated HAFM on the hexagonal lattice (i.e., with NN interactions only), in excellent agreement with the best available QMC estimate [31], $M = 0.2677(6)$. Our own error estimates are based on sensitivity checks using different LSUB$m$ data sets. Similarly we see from figure 3 that all extrapolations give essentially the same estimate $x_{c1} \approx 0.207(3)$ for the point where Néel order vanishes ($M \to 0$).

We showed previously [15] that the phase transition at $x = x_{c1}$ is a continuous deconfined one between states with Néel and PVBC order. Figure 3 also shows corresponding results for $M$ for a possible phase with Néel-II ordering. In this case we find (even by simple inspection by eye) that the LSUB6 results do not fit with a leading-order extrapolation scheme of the form $M(m) = d_0 + d_1 m^{-\nu}$ with any value of $\nu$. By contrast, the LSUB$m$ results with $m > 6$ are accurately fitted by this
the actual GS phase in this intermediate regime might share
terminate (as is clearly seen from figure 3), we expect that
the Néel-II state as model state, with \( m = \{ 6, 8, 10, 12 \} \). The
extrapolated curves LSB\( \times \infty \) (1) and LSB\( \times \infty \) (2) are derived from
fitting the perturbed energies (see text) as \( e(\delta) = e_0(\delta) + e_1(\delta)m^{-\nu} \),
and the data sets \( m = \{ 6, 8, 10, 12 \} \) and \( m = \{ 8, 10, 12 \} \)
respectively. Right: the field \( F \rightarrow \delta \hat{O}_d \) for the staggered dimer
susceptibility, \( \chi_d \). Thick (red) and thin (black) lines correspond
respectively to strengthened and unaltered NN exchange couplings,
where \( \hat{O}_d = \sum_{\langle i,j \rangle} n_i \cdot n_j \), and the sum runs over all NN bonds,
with \( n_i = +1 \) and 0 for thick (red) lines and thin (black) lines respectively.

form with a leading-order exponent \( \nu \approx 0.5 \) over the whole
range of values of the frustration parameter \( x \) shown. Precisely
why the LSB\( \times \) result should be anomalous in this case is
unclear, but as discussed in section 3 we must now discard it
for extrapolation purposes. For these reasons we show in
figure 3 only extrapolated results using equation (4) for
the Néel-II model state, based on \( m = \{ 8, 10, 12 \} \). The results
clearly show that Néel-II ordering is present, albeit with a
rather small value of the order parameter, \( M \lesssim 0.1 \), for \( x > x_{c3} \)
where \( x_{c3} \approx 0.65 \) (5), but where the error estimate is now more
uncertain.

In our previous work [15] we showed that the Néel-II
state becomes susceptible to PVBC ordering for \( x < x_{c2} \approx 0.385 \) (10),
but we now observe that the Néel-II state is itself
only stable as a magnetically ordered state for \( x > x_{c3} \). We
are thus led to enquire about the possible GS phase(s) of
the system in the range \( x_{c2} < x < x_{c3} \). In view of the persistence of
our CCM LSB\( \times m \) solutions based on the Néel-II model state,
with finite values of \( m \), well into the region \( x < x_{c1} \), before they
terminate (as is clearly seen from figure 3), we expect that
the actual GS phase in this intermediate regime might share
similarities with the Néel-II state. For example, just as the
Néel-II state breaks the lattice rotational symmetry, so does
another valence-bond solid state, namely the staggered dimer
valence-bond crystalline (SDVBC) (or lattice nematic) state.
This is formed from the Néel-II state by replacing all of the
parallel NN spin pairs by spin-zero dimers (and see figure 4).

In order to investigate the possibility of an SDVBC
phase we first consider the response of the system to a field
operator \( F \) (and see, e.g., [29]). Thus, a field term \( F = \delta \hat{O}_d \) is added to the Hamiltonian of equation (1), where
\( \hat{O}_d \) is an operator corresponding to the possible SDVBC
order, illustrated in figure 4 and defined in its caption. The
energy per site, \( E(\delta)/N \approx e(\delta) \), is then calculated in
the CCM for the perturbed Hamiltonian \( H + F \), using the Néel-II
model state. We define the corresponding susceptibility as
\( \chi_d = -\left( \langle \delta^2 e(\delta) \rangle \right)/\left( \langle \delta e(\delta) \rangle \right) \rangle \). Clearly the GS phase becomes
unstable against SDVBC order when \( \chi_d^{-1} \) becomes zero. We
now use the LSB\( \times m \) extrapolation scheme \( e(\delta) = e_0(\delta) + e_1(\delta)m^{-\nu} \), with the exponent \( \nu \) also a fitting parameter,
rather than our standard energy extrapolation scheme of
equation (2), in order to calculate the extrapolated values of
\( \chi_d^{-1} \) shown in figure 4. For the same data set \( m = \{ 8, 10, 12 \} \)
used to calculate \( M \) for the Néel-II state above, the fitted value of
\( \nu \) is close to 2 over most of the range of the \( J_2 \) values shown,
except near the termination point of this phase, where it falls
sharply. By contrast, for the set \( m = \{ 8, 10, 12 \} \) also shown in
figure 4, \( \nu \) is closer to 1 over most of the range. This again
reinforces the anomalous nature of the LSB\( \times 6 \) results.

What we see from figure 4 is that the extrapolated value of
\( \chi_d^{-1} \) is close to zero over a range of values of \( x \) that extends
from \( x_{c2} \) below to an upper value of about 0.6, which is
completely compatible with the value \( x_{c3} \) obtained from the
order parameter \( M \) of the Néel-II state. Thus, by combining
our results, we conclude that in the region \( x_{c2} < x < x_{c3} \),
the GS phase has SDVBC order, while for \( x > x_{c3} \) the GS
phase has Néel-II order, although this latter ordering is weak
and quite fragile against the still strongly competing SDVBC
order. The shape of the CCM curves for \( \chi_d^{-1} \) in figure 4 are
indicative of a continuous (and hence deconfined) quantum
critical point at \( x_{c3} \), whereas the corresponding curves for
\( \chi_P^{-1} \), the inverse plaquette susceptibility, found in our earlier
work [15] were much more indicative of a direct first-order
transition at \( x_{c2} \). We see no signals at all of the spiral ordering
that is present classically for \( x > 1/6 \) for any value of \( x \) in the
range 0 < \( x < 1 \) examined.

5. Summary

In conclusion, over the range 0 < \( x < 1 \) we find that the
spin-1/2 \( J_1-J_2 \) HAFM on the honeycomb lattice has four
phases with, respectively, Néel, PVBC, SDVBC, and Néel-II
ordering. Our CCM estimate for the phase diagram is shown in
figure 5. We note that all of our most accurate estimates for
the three quantum critical points are based on evaluations of
the positions at which the relevant magnetic order parameters
and/or the inverse susceptibilities to the relevant forms of
valence-bond solid order vanish. Since there are no energy
crossings between the Néel and Néel-II states directly used
as CCM model states in our CCM calculations, the GS energy
data only give direct corroborating evidence for the transitions
at \( x_{c2} \) and \( x_{c3} \) from the corresponding termination points of the
CCM LSB\( \times m \) solutions based on the Néel and Néel-II model
states respectively, as discussed in section 4 and illustrated in
figure 2.

Our first calculated critical point, \( x_{c1} \approx 0.207(3) \), at
which Néel order melts, agrees well with other recent
results, including \( x_{c1} \approx 0.195(25) \) from a large-scale exact
diagonalization (ED) study [11], \( x_{c3} \approx 0.26 \) [17] and
$x_c \approx 0.22$ [18] from two separate density-matrix renormalization group (DMRG) studies, and $x_c \approx 0.2075$ [19] and 0.21 [20] from two recent SB-MFT studies. Both DMRG studies [17, 18] and the ED study [11] concur with us that the transition at $x_c$ is probably a continuous deconfined one to a PVBC state, whereas both SB-MFT studies [19, 20] indicate a transition to a QSL state.

Our second calculated critical point, $x_{c_2} \approx 0.385(10)$, at which the PVBC order melts, is similarly in good agreement with the result $x_{c_2} \approx 0.375(25)$ from the ED study [11], and the results $x_c \approx 0.36$ [17] and $x_c \approx 0.35$ [18] from the two DMRG studies. We find that the transition at $x_c$ is probably a direct first-order one to a state with SDVBC order. Both DMRG studies [17, 18] and the ED study [11] concur with us that the transition at $x_c$ is to a state with SDVBC order, although Ganesh et al [18] find evidence for the surprising scenario that the transition at $x_c$ is also of the continuous deconfined type, as at $x_{c_1}$. The two SB-MFT studies [19, 20] find QSL states out to values $x \approx 0.3732$ [19] and $x \approx 0.43$ [20], respectively, beyond the point $x_c$, at which Néel order melts. They disagree, however, between themselves as to what is the nature of the GS phase for larger values of $x$, beyond the QSL phase. Thus, Zhang and Lamas [19] find the GS phase to be spirally ordered (just as in the classical, $s \to \infty$, version of the model) for $0.398 \lesssim x \lesssim 0.5$, and to have SDVBC order in the very narrow region $0.372 \lesssim x \lesssim 0.398$; whereas Yu et al [20] find that for $x \gtrsim 0.43$ the GS phase has Néel-II order. The ED study [11], by contrast, finds a first-order transition at $x_{c_2}$ to a state that cannot be distinguished between having either SDVBC or Néel-II order.

Finally, we find evidence for a third critical point at $x_{c_3} \approx 0.65(5)$ at which a continuous (and hence again deconfined) transition occurs to a state with weak Néel-II magnetic order. We note that such a transition is also compatible with the DMRG result of Ganesh et al [18], which could not rule out a melting of the SDVBC order for values $x \gtrsim 0.7$. It is interesting to speculate whether the weak Néel-II magnetic order observed by us for $x > x_{c_3}$ might be interpreted as, or arise from, a sort of ‘dressed’ SDVBC state in which spin-triplets now contribute on the spin-singlet dimer bonds. It is too far beyond the scope of the present analysis, however, to address such delicate questions authoritatively.

As a last remark, it is interesting to note that in a very recent study using a projector QMC technique [32] a very similar direct continuous quantum phase transition to what we observe here for the $J_1-J_2$ model at $x_{c_1}$, between states with Néel and PVBC order, has also been observed in a related spin-1/2 $J_1-Q$ model on the honeycomb lattice, of precisely the type suggested by Yang et al [8] to be more relevant to the low-energy physics of the FH model on the honeycomb lattice, as discussed previously in section 1. This $J_1-Q$ model also contains NN AFM exchange bonds of strength $J_1$, but with our competing NNN exchange bonds of strength $J_2$ replaced by a six-spin interaction term of strength $Q$ on hexagonal plaquettes, which by itself favours the formation of a state with PVBC order. It would clearly also be of interest to apply a comparable CCM study to the $J_1-Q$ model to that used here for the $J_1-J_2$ model, in order to investigate its GS phase diagram similarly.

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\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{phase_diagram.png}
\caption{Phase diagram of the spin-1/2 $J_1-J_2$ model on the honeycomb lattice (with $J_1 > 0$ and $x \equiv J_2/J_1 > 0$), as obtained by a CCM analysis. The quantum critical points are at $x_{c_1} \approx 0.207(3)$, $x_{c_2} \approx 0.385(10)$, and $x_{c_3} \approx 0.65(5)$, as shown in the diagram.}
\end{figure}
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