First-order Néel-cVBS transition in a model square lattice $S = 1$ antiferromagnet

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We study the Néel to four-fold columnar valence bond solid (cVBS) quantum phase transition in a sign free $S = 1$ square lattice model. This is the same kind of transition that for $S = 1/2$ has been argued to realize the prototypical deconfined critical point. Extensive numerical simulations of the square lattice $S = 1/2$ Néel-VBS transition have found consistency with the DCP scenario with no direct evidence for first order behavior. In contrast to the $S = 1/2$ case, in our quantum Monte Carlo simulations for the $S = 1$ model, we present unambiguous evidence for a direct conventional first-order quantum phase transition. Classic signs for a first order transition demonstrating co-existence including double peaked histograms and switching behavior are observed. The sharp contrast from the $S = 1/2$ case is remarkable, and is a striking demonstration of the role of the size of the quantum spin in the phase diagram of two dimensional lattice models.

I. INTRODUCTION

The destruction of Néel order by quantum fluctuations is a hotly studied issue in quantum magnetism inspired originally by the parent compounds of cuprate high temperature superconductors. In the cuprates, the Néel order appears for $S = 1/2$ moments on the square lattice. In this case, many theoretical arguments and extensive unbiased numerical calculations have put forth evidence for a four-fold degenerate columnar valence bond solid (VBS) phase on the destruction of Néel order, possibly separated by the novel deconfined critical point.¹⁻⁵ More recently, inspired by the iron pnictide superconductors, a number of studies of the destruction of Néel order in $S = 1$ square lattice systems have appeared,⁶⁻⁹ building on previous studies of the phase diagram of square lattice $S = 1$ systems, (see¹⁰⁻¹⁴ and references therein). It is thus interesting to extend the success of unbiased quantum Monte Carlo (QMC) studies of the destruction of Néel order in square lattice $S = 1/2$ systems¹⁵ to the $S = 1$ case. In previous QMC studies the phase transitions in coupled $S = 1$ chains¹⁵ and the transition to the Haldane nematic have been considered.¹⁶ Here we will study the transition between the Néel state and a columnar Valence bond solid. A cartoon wavefunction for such a cVBS can be simply written down since two $S = 1$ spins can form a singlet from the elementary rules of the addition of angular momentum; these singlets can then be arranged in the standard columnar pattern.

The role of the microscopic value of spin on the phase diagrams of one-dimensional spin chains is now well established. Most famously Heisenberg models with integer spins realize a ground state with a gap to all excitations called the Haldane gap, whereas half integer spin chains realize an interesting gapless ground state described at long distances by the SU(2) Wess-Zumino-Witten field theory.¹⁷ It is interesting to ask what the role of the size of the spin is in two dimensions? While the square lattice Heisenberg model is Néel ordered for all spin-$S$, the nature of the accessible non-magnetic phases and the theory of critical phenomena at the destruction of Néel order has been argued to depend sensitively on the value of the spin.¹ Since the subtle quantum effects that arise from Berry phase terms depend crucially the microscopic value of the spin,¹⁸ one can expect striking differences between $S = 1/2$ and $S = 1$ even for phase transitions that appear identical with respect to the Landau-Ginzburg-Wilson criteria of dimensionality, symmetry and order parameters. We will study this interesting issue here by focusing on the square lattice Néel-cVBS phase transition in $S = 1$ magnets. The identical phase transition for $S = 1/2$ is described by deconfined criticality which has argued for a single continuous phase transition.

We note that a field theoretical study has taken up a related issue previously.¹⁹ Extending their results in a straightforward manner would suggest that a $S = 1$ Néel-cVBS transition could possibly be described by an anisotropic CP² field theory with quadrupled monopoles. That this implies a continuous deconfined transition in our microscopic model requires a litany of additional
assumptions, including that the field theory has an anisotropic fixed point, quadrupled monopoles are irrelevant at this fixed point and that our microscopic model crosses the critical surface so we can flow into the fixed point. As we shall see below in our microscopic model we find a first order transition, but it is unclear yet which of these assumptions fails. Further work on both microscopic models and field theory could shed light on this subtle detail in the future.

II. MODEL

Our first goal is to design a \( S = 1 \) sign free model in which the Néel-cVBS transition can be studied using Monte-Carlo simulations. We start with the square lattice \( S = 1 \) Heisenberg model,

\[
H_f = J \sum_{\langle ij \rangle} \vec{S}_i \cdot \vec{S}_j
\]

This model is well known to be Néel ordered. Because we are working with \( S = 1 \), it is possible to square the biquadratic operator and obtain an independent “biquadratic operator,” \( (\vec{S}_i \cdot \vec{S}_j)^2 \), also amenable to QMC.\(^{20,21}\) Using this term we can construct a Sandvik-like four spin interaction,

\[
H_{QK} = -Q_K \sum_{ijkl \in \Box} \left( (\vec{S}_i \cdot \vec{S}_j)^2 - 1 \right) \left( (\vec{S}_k \cdot \vec{S}_l)^2 - 1 \right)
\]

We note that \( H_{QK} \) has a higher staggered SU(3) symmetry because it is constructed from the biquadratic interaction, of which the physical SU(2) is a subgroup. However the model we study here \( H_{QK} = H_f + H_{QK} \) has only the generic SU(2) symmetry obtained by rotating the \( \vec{S} \) vector in the usual way. Previous numerical studies have established that \( H_{QK} \) on the square lattice has four-fold columnar VBS order.\(^{22-24}\) Thus the single tuning parameter in \( H_{QK} \) gives us unbiased numerical access to the Néel-VBS transition in a \( S = 1 \) system, as desired.

Since our model is constructed to be Marshall sign positive, it can be simulated without a sign problem using the stochastic series expansion method (SSE).\(^{25}\) We have used two different algorithms as described in Sec. A1 which produce the same results within errors. Our simulations are carried out on \( L \times L \) square lattices at an inverse temperature \( \beta \) – all the data presented here has been checked to be in the \( T = 0 \) limit as shown in A3. We work in units in which \( J = 1 \), and define the tuning parameter \( g \equiv Q_K/J \) to access the phase transition. We study the Fourier transform of the Néel and VBS correlation functions, \( S^N_k = \frac{1}{L^2} \sum_{\mathbf{r}} e^{i \mathbf{k} \cdot \mathbf{r}} \langle \vec{S}(\mathbf{r}) \vec{S}(0) \rangle \) and \( S^V_k = \frac{1}{L^2} \sum_{\mathbf{r}} e^{i \mathbf{k} \cdot \mathbf{r}} \langle \vec{S}(\mathbf{r}) \cdot \vec{S}(\mathbf{r} + \hat{x}) \vec{S}(0) \cdot \vec{S}(0 + \hat{x}) \rangle \). We

This close to the phase transition on system sizes up to \( L = 32 \). The extrapolations to a finite value for both Néel and VBS order parameters at a common coupling \( g = 32 \). The extrapolations to a finite value for both Néel and VBS order parameters at a common coupling \( g = 32 \). The extrapolations to a finite value for both Néel and VBS order parameters at a common coupling \( g = 32 \).
define the order parameters as $O^2_N = S^N_{\pi,\pi}$ and $O^2_V = S^V_{\pi,0}$. For each of the order parameters we define ratios $R = 1 - \frac{S_{Kx}^2 + S_{Ky}^2}{S_k^2}$ (with $K$ the ordering momentum); $R$ goes to 1 in a phase with long range order and 0 in a disordered phase. In the SSE method we map the quantum partition function of our model to a classical loop model in one higher dimension. The winding number of these loops is also a useful quantity to detect the magnetic phase. The spin stiffness defined as Eq. A9 is related to the square of the winding number of these loops, $\langle W^2 \rangle$, as shown in Eq. A10. The magnetic phase is characterized by long loops with $\langle W^2 \rangle$ diverging linearly with $L$ while the VBS phase has short loops with $\langle W^2 \rangle$ going to zero.

III. NUMERICAL RESULTS

Fig. 1 shows the ratios $R$ for the Néel and VBS order parameters as a function of $g$ for different $L$. The data (see inset for finite size scaling) provides strong evidence that the Néel-VBS transition is direct with a $g_c = 0.588(2)$ – we can safely rule out co-existence or an intermediate phase. We note that this study does not by itself indicate whether the transition is first order or continuous.

The ratio data leaves open the possibility of a direct continuous transition. The first indication that this does not occur is shown in Fig. 2. In this finite size scaling plot of both order parameters, we have reasonable evidence that at the transition both order parameters are finite. We have carried out extrapolations on system sizes up to $L = 32$. While it is not fully reliable quantitatively to extrapolate the order parameter data with such a limited system size range, there is little doubt that both Néel and cVBS order parameters are finite at $g = 0.587$. This would indicate a first order transition or a co-existence between Néel and cVBS phases.

Beyond system sizes of $L \approx 32$, it is very difficult to get QMC data with small error bars close to the critical point. As we now elaborate the reason for this is that we are encountering a first order Néel-cVBS transition. Fig. 3 shows histograms for the Néel and cVBS order parameter estimators which show clear double peaked behavior that gets pronounced as the system size is increased. The stiffness, which is finite in the Néel phase and goes to zero in the cVBS phase also shows clear double peaked behavior close to the transition. The double peaked behavior results from the system switching between Néel and cVBS phases during the simulation. This is shown in Fig. 5 in which we observe clearly that when one order is present when the other is absent. We thus conclude that the system switches between the two orders at the critical point which is characteristic of a first order transition.

IV. CONCLUSIONS

We have introduced a model for the transition from the Néel to the four fold degenerate columnar valence bond solid state which is amenable to sign free quantum Monte Carlo simulations. Previous field theoretic work extending the $S = 1/2$ deconfined criticality scenario to $S = 1$ has argued that this transition could be direct and continuous, and described by an anisotropic CP$^2$ field theory. Instead, a detailed numerical study of our model shows that this phase transition is direct but of first order in our model. With no known model that shows a continuous transition it is possible that one of the assumptions of the field theoretic scenario is itself incorrect, e.g. the existence of an anisotropic SU(3) fixed point. Clearly more field theoretic work is needed to further our understand-
Also shown are the so-called ratios $R$ parameter values for the extensive ground state energy, the Néel order state in the $S_l$ lattice, we project out the ground state from a random very large Hilbert space for this spin-1 model on a 4x4 lattice and $Q$ for the spin–1/2 model instead of a $S = 1$ model and project out states that only belong to the $S = 1$ subspace. 

Appendix A: Numerical Details

1. Algorithm

The numerical results presented in this work have been obtained using two different methods, both of which are some adaptation of the standard Stochastic Series Expansion (SSE)\textsuperscript{25} algorithm:

1. In the first method we work in the $S_z = -1, 0, 1$ basis for our $S = 1$ problem. To update the SSE configurations we use both local diagonal updates and the non-local directed loop algorithm\textsuperscript{26} that allows us to switch between the allowed vertices while respecting the $S_z$ conservation.

2. In the second method we use the split spin representation\textsuperscript{17,28} where each $S = 1$ is replaced by two $S = 1/2$s. We then simulate a $S = 1/2$ model instead of a $S = 1$ model and project out states that only belong to the $S = 1$ subspace.\textsuperscript{16}

2. Measurements and QMC-ED comparison:

We have tested our code by performing comparisons against exact diagonalization. For future reference, Tables I and II provide test comparisons between measurements obtained from a SSE study and exact diagonalization (ED) on a lattice of size $(L_x, L_y) = (4, 4)$, for various combinations of the bond and plaquette interactions $J$ and $Q_K$ for the $J - Q_K$ model under investigation in this work and for various combinations of the bond and plaquette interactions $J$ and $Q_f$ for the spin–1 version of Sandvik’s $J - Q_f$ model (described in A.4). Due to the very large Hilbert space for this spin-1 model on a 4x4 lattice, we project out the ground state from a random state in the $S^2 = 0$ subspace, thus avoiding the need to diagonalize the sparse Hamiltonian matrix. We list values for the extensive ground state energy, the Néel order parameter $O_N$, as well as the VBS order parameter $O_V$. Also shown are the so-called ratios $R_N$ and $R_V$. These quantities measured using both the algorithms described in A.1 have been checked to match. All observables are defined below.

Measurements: In order to simplify the QMC loop algorithm, we have shifted our $J$ bond operators by the identity, $J(S_i · S_j - 1)$. The extensive energy quoted in the tables includes this shift. In order to character-

![FIG. 6. All observables saturate as a function of inverse temperature ($\beta$) before $\beta = 6$ at $g = 0.59$](image)

![FIG. 7. Shown is the ratio $R_N$ of the Néel order parameter of various values of the plaquette interaction coupling $Q_f$ with $J^2 + Q_f^2 = 1$ for systems of size $(L, L)$ with $L$ up to 32 lattice sites. $R_N$ appears to be independent from $Q_f$ and approaches 1 for increasingly large system sizes indicating a phase diagram consisting entirely of Néel order. The inset shows the Néel order parameter $O_N$.](image)
Confirming the findings from Fig. 7, we see that J\text{VBS} order in the Q model for spin-1 J – QK model. Additionally shown are the ratios RN and RV. For the SSE, errors are also shown. The MC data is computed with β = 40.

| Lx | Ly | J  | QK | E (ED) | E (MC) | \( \mathcal{O}_N^2 \) (ED) | \( \mathcal{O}_N^2 \) (MC) | \( \mathcal{O}_V^2 \) (ED) | \( \mathcal{O}_V^2 \) (MC) | RN (ED) | RN (MC) | RV (ED) | RV (MC) |
|----|----|----|----|--------|--------|----------------|----------------|----------------|----------------|----------|----------|----------|----------|
| 4  | 4  | 0.2 | 0.9 | -96.15381 | -96.147(8) | 0.13590 | 0.13592(2) | 0.50414 | 0.5044(5) | 0.49940 | 0.4993(1) | 0.75173 | 0.7570(7) |
| 4  | 4  | 0.5 | 0.2 | -99.02960 | -99.024(4) | 0.25596 | 0.25594(9) | 0.28370 | 0.2838(2) | 0.78769 | 0.7868(1) | 0.59012 | 0.5907(7) |
| 4  | 4  | 0.7 | 0.3 | -70.29052 | -70.288(5) | 0.24879 | 0.24867(8) | 0.29726 | 0.2971(2) | 0.77611 | 0.7760(1) | 0.60493 | 0.6054(6) |
| 4  | 4  | 0.8 | 0.4 | -85.17819 | -85.180(6) | 0.23283 | 0.23291(6) | 0.32728 | 0.3269(2) | 0.75040 | 0.7503(1) | 0.63436 | 0.6346(5) |
| 4  | 4  | 0.9 | 0.6 | -109.00470 | -109.001(7) | 0.20556 | 0.20562(3) | 0.37805 | 0.3777(2) | 0.69897 | 0.6989(1) | 0.67619 | 0.6761(4) |

This gives rise to a Bragg peak in the Fourier transform of the bond-bond correlator defined as

\[
\hat{C}^\alpha(q) = \frac{1}{N^2_{\text{site}}} \sum_{\bar{r},\bar{r}'} \sum_{\alpha} e^{i(\bar{r}-\bar{r}') \cdot q} \langle \bar{S}_{\bar{r}} \cdot \bar{S}_{\bar{r}+\hat{\alpha}} \rangle \cdot (A1)
\]

For a columnar VBS patterns, peaks appear at the momenta (π, 0) and (0, π) for x and y-orientated bonds, respectively. Thus, the VBS order parameter is given by

\[
\mathcal{O}_{VBS} = \frac{\hat{C}^x(\pi, 0) + \hat{C}^y(0, \pi)}{2} .
\]

Another useful quantity to locate a possible phase transitions is the above mentioned VBS ratio RV. We first distinguish between x- and y-orientated bonds:

\[
\mathcal{R}^x_V = 1 - \frac{\hat{C}^x(\pi, 2\pi/L) + \hat{C}^x(\pi, 0)}{2}
\]

\[
\mathcal{R}^y_V = 1 - \frac{\hat{C}^y(2\pi/L, \pi) + \hat{C}^y(0, \pi)}{2} .
\]

Subsequently, we average over x and y-orientations:

\[
\mathcal{R}_V = \frac{\mathcal{R}^x_V + \mathcal{R}^y_V}{2} .
\]

This quantity goes to 1 in a phase with long-range VBS order and it approaches 0 in a phase without VBS order present.

The Néel structure factor is,

\[
m_2^z(q) = \frac{1}{N^2_{\text{site}}} \sum_{\bar{r},\bar{r}'} e^{i(\bar{r}-\bar{r}') \cdot q} \langle \bar{S}_{\bar{r}} \bar{S}_{\bar{r}+\hat{\alpha}} \rangle .
\]

The Bragg peak appears at momentum (π, π) and thus the Néel order parameter is given by

\[
\mathcal{O}_N = m_2^z(\pi, \pi) .
\]
To additionally provide a quantity that goes to 1 in a Néel ordered phase and vanishes in a phase without, we study the Néel ratio:

\[
\mathcal{R}_N^x = 1 - m_z^2(\pi + 2\pi/L, \pi)/m_z^2(\pi, \pi)
\]
\[
\mathcal{R}_N^y = 1 - m_z^2(\pi, \pi + 2\pi/L)/m_z^2(\pi, \pi). \tag{A7}
\]

We can now average over both quantities:

\[
\mathcal{R}_N = \frac{\mathcal{R}_N^x + \mathcal{R}_N^y}{2}. \tag{A8}
\]

The spin stiffness, \(\rho_s\), is another quantity we use to detect the magnetic phase. It is defined as:

\[
\rho_s = \frac{\partial^2 E(\phi)}{\partial \phi^2} \bigg|_{\phi=0}. \tag{A9}
\]

Here \(E(\phi)\) is the energy of the system when you add a twist of \(\phi\) in the boundary condition in either the \(x\) or the \(y\) direction. In the QMC, this quantity is related to the winding number of loops in the direction that the twist has been added:

\[
\rho_s = \frac{\langle \mathcal{W}^2 \rangle}{\beta}. \tag{A10}
\]

where \(\beta\) is the inverse temperature. In the magnetic phase the stiffness extrapolates to a finite value in the thermodynamic limit, but goes to zero in the non-magnetic phase.

3. Ground state convergence:

We investigate the behavior of the observables described above in Eq. 2 \((\mathcal{O}_V^2, \mathcal{O}_N^2, \rho_s)\) when the SSE is carried out at different inverse temperatures \(\beta\). Fig. 6 shows that these quantities saturate as a function of inverse temperature \(\beta\) before \(\beta = 6\). However close to the transition, one needs to go lower in temperature for saturation. Therefore we do finite size scaling of histograms near the critical point for \(\beta = L/4\) in order to probe the first order behavior. One can see from Fig. 9 that decreasing the temperature to \(\beta > L/4\) does not significantly weaken the first order transition, so we can conclude that first order behavior persists at zero temperature.

4. \(J - Q_1\) Model for \(S = 1\)

We now briefly discuss another designer model Hamiltonian and compare the phase diagram for the two cases of a spin\(1/2\) system and a spin\(1\) system.

The so-called “\(J - Q\)” model was introduced by Sandvik in 2007\(^4\). The model consists of a Heisenberg interaction between nearest neighbor sites (see equation (1) in the main manuscript) on the square lattice and an additional plaquette term:

\[
H_Q = -Q \sum_{ijkl} (\vec{S}_i \cdot \vec{S}_j - \frac{1}{4}) (\vec{S}_k \cdot \vec{S}_l - \frac{1}{4}). \tag{A11}
\]

The spin\(1/2\) case of this model \(H = H_J + H_Q\) was shown to have a phase transition from Néel order to VBS order at a critical point \(J/Q \approx 0.04\).

We now subject the same term structure to a SSE-MC simulation in order to determine the phase diagram. We note that for the spin\(1\) case the constant \(\frac{1}{4}\) is replaced by 1 in order by make the plaquette term amenable to the SSE-MC study:

\[
H_{QJ} = -Q_J \sum_{ijkl} (\vec{S}_i \cdot \vec{S}_j - 1) (\vec{S}_k \cdot \vec{S}_l - 1). \tag{A12}
\]

The \(J - Q_J\) model spin\(1\) Hamiltonian is then \(H_{QJ} = H_J + H_{QJ}\). We analyzed the phase diagram for various couplings \(J\) and \(Q_J\) with the condition \(J^2 + Q_J^2 = 1\) and found that the phase diagram consists entirely of Néel order independent from the ratio of the two coupling strengths \(J\) and \(Q_J\). Fig. 7 shows the ratio of the Néel order parameter. The ratio appears to be independent from \(Q_J\) (with \(J\) fixed by \(J^2 + Q_J^2 = 1\)) further the ratio \(\mathcal{R}_N\) approaches 1 for increasingly large system sizes. This is a clear indicator that the entire phase diagram consists of Néel order. For completeness we also give the ratio \(\mathcal{R}_V\) of the VBS order parameter \(\mathcal{O}_V^2\). In compliance with our findings from Fig. 7, we see the ratio \(\mathcal{R}_V\) approaches zero for sufficiently large lattice sizes independent from the coupling \(Q_J\) (again with \(J\) fixed by \(J^2 + Q_J^2 = 1\)). This provides evidence for the absence of VBS order that was present in the spin\(1/2\) flavor of the model.
