Generalized plaquette state in the anisotropic Shastry-Sutherland model

Zhang, Zhifeng; Sengupta, Pinaki

2015

Zhang, Z., & Sengupta, P. (2015). Generalized plaquette state in the anisotropic Shastry-Sutherland model. *Physical Review B*, 92, 094440-.

<https://hdl.handle.net/10356/81122>

<https://doi.org/10.1103/PhysRevB.92.094440>

© 2015 American Physical Society. This paper was published in *Physical Review B* and is made available as an electronic reprint (preprint) with permission of American Physical Society. The published version is available at:
[<http://dx.doi.org/10.1103/PhysRevB.92.094440>]. One print or electronic copy may be made for personal use only. Systematic or multiple reproduction, distribution to multiple locations via electronic or other means, duplication of any material in this paper for a fee or for commercial purposes, or modification of the content of the paper is prohibited and is subject to penalties under law.

Downloaded on 26 Apr 2023 15:21:39 SGT

Generalized plaquette state in the anisotropic Shastry-Sutherland model

Zhifeng Zhang and Pinaki Sengupta

School of Physical and Mathematical Sciences, Nanyang Technological University, Singapore

(Received 28 October 2014; revised manuscript received 18 May 2015; published 25 September 2015)

We use a generalized Schwinger boson approach to investigate the nature of the intermediate phase (separating the antiferromagnetic and dimer phases) in the Shastry-Sutherland model with exchange anisotropy. Our results confirm the plaquette nature of the intermediate phase and demonstrate that exchange anisotropy (both Ising-like and XY -like) weakens the plaquette order and ultimately suppresses it altogether for sufficiently strong anisotropy. Interestingly, we find that the low-energy excitation spectrum of the plaquette state changes dramatically near the boundary with the dimer phase—the location of the minimum in the (gapped) lowest excitation mode changes from $\mathbf{k} = (0,0)$ to four degenerate points.

DOI: [10.1103/PhysRevB.92.094440](https://doi.org/10.1103/PhysRevB.92.094440)

PACS number(s): 75.10.Jm, 75.40.Cx, 75.40.Mg

I. INTRODUCTION

The Shastry-Sutherland model (SSM) serves as a paradigm to study the emergence of novel quantum phases from the interplay among strong interactions, external magnetic field, and geometric frustration in two-dimensional quantum spin systems. The model has garnered extensive interest [1–8], especially after the discovery of the spin-gap compound $\text{SrCu}_2(\text{BO}_3)_2$ [9] and the observation of multiple field-induced magnetization plateaus therein. The compound consists of weakly coupled layers. The Cu^{2+} ions carrying $S = 1/2$ spins in each layer are arranged in an array topologically equivalent to the Shastry-Sutherland lattice (Fig. 1). The spins interact via isotropic Heisenberg interactions: a nearest-neighbor interaction J_1 along the x and y axes and next-nearest-neighbor interaction J_2 along alternating diagonal bonds. Figure 1 shows the phase diagram of the SSM. In the limit $J_1 \rightarrow 0$, the model is reduced to a set of isolated dimers on the diagonal bonds, and the ground state consists of singlets on the dimers. Remarkably, this remains the *exact* ground state up to a finite nonzero critical value of J_1 [10,11]. On the other hand, in the limit of $J_2 \rightarrow 0$, the ground state is a Néel state with long-range antiferromagnetic (AFM) order that persists for small but nonzero values of the diagonal interaction J_2 . Although the nature of the ground state in the two limits has been well known, the nature of its evolution with varying J_2/J_1 is not so clear. Whether there is a direct transition from the magnetic Néel state to the nonmagnetic singlet phase or there exists an intermediate phase had remained unresolved for a long time. Several studies based on a diverse array of approaches have variously predicted a direct transition or a number of different intermediate phases [8,12–17]. The general consensus has leaned towards an intermediate plaquette phase over a narrow range of J_2/J_1 [8,16,17]. The most convincing evidence for the plaquette phase was recently presented in Ref. [17] based on a tensor network study.

We explore the existence of an intermediate plaquette (PL) phase in the SSM with an exchange anisotropy. Effects of both Ising-like and XY -like anisotropies on the stability of the plaquette phase are considered. The choice of model is partly motivated by its relevance to the rare-earth tetraborides RB_4 ($R = \text{Tm}, \text{Er}, \text{Dy}, \text{Tb}, \text{and Ho}$)—a family of layered quantum magnets whose low-energy magnetic properties can be explained in terms of an effective $S = 1/2$ XXZ model with Ising-like exchange anisotropy (albeit with additional

longer-range interactions). But the real motivation is to gain deeper insight into the plaquette phase by studying its properties in an expanded parameter space. The model is described by the Hamiltonian,

$$H = \sum_{i,j} J_{ij} \left[\frac{1}{2} (S_i^+ S_j^- + S_i^- S_j^+) + \Delta S_i^z S_j^z \right], \quad (1)$$

where $J_{ij} = J_{1,2}$ for the axial and diagonal bonds (Fig. 1). Δ denotes the exchange anisotropy which is assumed to be the same for both types of bonds. Our results show that the plaquette state in the isotropic SSM persists for small values of exchange anisotropy but develops instability towards AFM ordering at sufficiently strong anisotropies—both Ising-like ($\Delta > 1$) and XY -like ($\Delta < 1$).

II. METHOD

We use a mean-field approach based on Schwinger boson (SB) mapping to elucidate the nature of the plaquette phases. To define the plaquette basis, we choose a square without the diagonal bond as the unit cell. There are two ways to choose such a unit cell, and they are related by the reflection operator about any diagonal bond under a periodic boundary condition. By choosing a specific arrangement as shown in Fig. 1 we have explicitly broken this reflection symmetry at the mean-field level. The spins within a plaquette interact with each other via the axial interaction J_1 , whereas interaction between neighboring plaquettes is mediated by both J_1 and J_2 . The Hilbert space of a single plaquette is composed of the sum of four $S = 1/2$ spins— $h_{\text{PL}} \in \{S_1 \oplus S_2 \oplus S_3 \oplus S_4\}$ and is spanned by a pair of singlets $\{|s_1\rangle, |s_2\rangle\}$, three triplets $\{|t_\alpha\rangle, |l_\alpha\rangle, |r_\alpha\rangle\}$, and one quintuplet $\{|q_\alpha\rangle, |q_1\rangle, |q_2\rangle\}$ with $\alpha \in \{x, y, z\}$. They are the 16 eigenstates of the isotropic Heisenberg interaction on the square. The Brillouin zone in this representation is essentially the Brillouin zone of a square lattice with empty squares as the lattice points and a as the lattice constant.

We introduce 16 SB operators each of which creates an above eigenstate from a fictitious vacuum, e.g., $\hat{s}_1^\dagger |\emptyset\rangle = |s_1\rangle$ with the hard-core boson constraint,

$$\sum_{n=1,2} (\hat{s}_n^\dagger \hat{s}_n + \hat{q}_n^\dagger \hat{q}_n) + \sum_{\alpha} (\hat{l}_\alpha^\dagger \hat{l}_\alpha + \hat{l}_\alpha^\dagger \hat{l}_\alpha + \hat{r}_\alpha^\dagger \hat{r}_\alpha + \hat{q}_\alpha^\dagger \hat{q}_\alpha) = 1. \quad (2)$$

FIG. 1. (Color online) The lattice geometry of the SSM and the phase diagram of the original (isotropic) SSM. In this paper, the unit cell is taken to be the square without the J_2 bond. The numbers label the lattice sites within the unit cell. The lattice constant a is taken to be twice the edge of a unit cell.

In terms of these Schwinger bosons, we can obtain a reducible representation of the spin operators S_n^α , $\alpha \in \{x, y, z\}$ and $n \in \{1-4\}$ as shown in Eq. (7) (with $\phi = 0$).

For the anisotropic model, the Hamiltonian of the unit cell can be written as

$$\tilde{H}_p = H_p + J_1(\Delta - 1)(S_1^z + S_3^z)(S_2^z + S_4^z). \quad (3)$$

$$\begin{aligned} S_n^\alpha = & \left\{ (-1)^n \left[\left(\frac{1}{\sqrt{6}} \cos \phi + \frac{1}{2\sqrt{3}} \cos \theta_\alpha \sin \phi \right) \hat{t}_\alpha^\dagger \hat{s}_0 + \left(\frac{1}{\sqrt{6}} \sin \phi - \frac{1}{2\sqrt{3}} \cos \theta_\alpha \cos \phi \right) \hat{t}_\alpha^\dagger \hat{q}_0 - \frac{1}{2\sqrt{3}} \sin \theta_\alpha \hat{t}_\alpha^\dagger \hat{q}_2 \right] \right. \\ & - \cos \frac{n\pi}{2} \left[\left(\frac{1}{2\sqrt{3}} \cos \phi - \frac{1}{\sqrt{6}} \cos \theta_\alpha \sin \phi \right) \hat{t}_\alpha^\dagger \hat{s}_0 + \left(\frac{1}{2\sqrt{3}} \sin \phi + \frac{1}{\sqrt{6}} \cos \theta_\alpha \cos \phi \right) \hat{t}_\alpha^\dagger \hat{q}_0 + \frac{1}{2} \hat{r}_\alpha^\dagger \hat{s}_2 + \frac{1}{\sqrt{6}} \sin \theta_\alpha \hat{t}_\alpha^\dagger \hat{q}_2 \right] \\ & + \sin \frac{n\pi}{2} \left[\left(\frac{1}{2\sqrt{3}} \cos \phi - \frac{1}{\sqrt{6}} \cos \theta_\alpha \sin \phi \right) \hat{r}_\alpha^\dagger \hat{s}_0 + \left(\frac{1}{2\sqrt{3}} \sin \phi + \frac{1}{\sqrt{6}} \cos \theta_\alpha \cos \phi \right) \hat{r}_\alpha^\dagger \hat{q}_0 + \frac{1}{2} \hat{t}_\alpha^\dagger \hat{s}_2 + \frac{1}{\sqrt{6}} \sin \theta_\alpha \hat{r}_\alpha^\dagger \hat{q}_2 \right] \\ & + \frac{1}{2} \sin \theta_\alpha \cos \phi \hat{q}_\alpha^\dagger \hat{q}_0 - \frac{1}{2} \sin \theta_\alpha \sin \phi \hat{q}_\alpha^\dagger \hat{s}_0 - \frac{1}{2} \cos \theta_\alpha \hat{q}_\alpha^\dagger \hat{q}_\alpha + \text{H.c.} \Big\} \\ & + \frac{i}{4} \epsilon^{\alpha\beta\gamma} \left[-\hat{t}_\beta^\dagger \hat{t}_\gamma + \hat{q}_\beta^\dagger \hat{q}_\gamma - 2 \sin^2 \frac{n\pi}{2} \hat{t}_\beta^\dagger \hat{t}_\gamma - 2 \cos^2 \frac{n\pi}{2} \hat{r}_\beta^\dagger \hat{r}_\gamma + \sqrt{2} \sin \frac{n\pi}{2} (\hat{r}_\beta^\dagger \hat{t}_\gamma + \hat{t}_\beta^\dagger \hat{r}_\gamma) + \sqrt{2} \cos \frac{n\pi}{2} (\hat{t}_\beta^\dagger \hat{t}_\gamma + \hat{t}_\beta^\dagger \hat{t}_\gamma) \right] \\ & + \frac{i}{4} I^{\alpha\beta\gamma} \left[(-1)^n (\hat{t}_\beta^\dagger \hat{q}_\gamma - \hat{q}_\beta^\dagger \hat{t}_\gamma) - \sqrt{2} \sin \frac{n\pi}{2} (\hat{r}_\beta^\dagger \hat{q}_\gamma - \hat{q}_\beta^\dagger \hat{r}_\gamma) + \sqrt{2} \cos \frac{n\pi}{2} (\hat{t}_\beta^\dagger \hat{q}_\gamma - \hat{q}_\beta^\dagger \hat{t}_\gamma) \right], \end{aligned} \quad (7)$$

where

$$\theta_x = \frac{2}{3}\pi, \quad \theta_y = \frac{4}{3}\pi, \quad \theta_z = 0 \quad \text{and} \quad I^{\alpha\beta\gamma} = |\epsilon^{\alpha\beta\gamma}|. \quad (8)$$

$\epsilon^{\alpha\beta\gamma}$ is the antisymmetric tensor. Einstein summation convention has been applied for repeated indices except for α .

At the isotropic point, the plaquette singlet $|s_1\rangle$ with $\Delta = 1$ is the intermediate ground state between the dimer singlet and the AFM ordering. The explicit configuration of the plaquette singlet is given in Appendix A. When the anisotropy is turned on, we expect the plaquette singlet ground state to be adiabatically connected to the generalized plaquette valence-bond-solid phase described by $|s_0\rangle$ (4), at least for values of Δ close to the isotropic point. Therefore, we assume the mean-field ground state to be a condensation of the generalized plaquette state $|s_0\rangle$. Applying the Holstein-Primakoff approximation [18], up to cubic terms in the bosonic operators, the Hamiltonian can be

H_p is the isotropic Heisenberg interaction. Because the last term does not commute with H_p , the Hamiltonian is no longer diagonal in the above basis. This implies that the total spin does not conserve in general and is no longer a quantum number. But the azimuthal quantum number is still valid since $[\tilde{H}_p, S^z] = 0$. Using the SB representation of the spin operators (7) (with $\phi = 0$) and the constraint (2), we find that only $|s_1\rangle$ and $|q_1\rangle$ are coupled in the anisotropic Hamiltonian. A straightforward diagonalization gives

$$|s_0\rangle = \cos \phi |s_1\rangle - \sin \phi |q_1\rangle, \quad (4)$$

$$|q_0\rangle = \sin \phi |s_1\rangle + \cos \phi |q_1\rangle, \quad (5)$$

with

$$\tan \phi = \frac{2\sqrt{2}(\Delta - 1)}{\Delta + 8 + 3\sqrt{\Delta^2 + 8}}. \quad (6)$$

The corresponding new energies are given in Appendix A.

In terms of the new SBs, the spin operators are given by

written as

$$H = H_0 + H_{\text{SW}} + H_I + H_3. \quad (9)$$

H_0 is the classical ground-state energy,

$$\frac{H_0}{N} = -\frac{1}{4}\mu = -\frac{1}{8}J_1(\Delta + \sqrt{\Delta^2 + 8}). \quad (10)$$

N is the number of lattice sites. μ is introduced for convenience. H_I describes three isolated excitations, that is with flat dispersions up to bilinear terms,

$$\begin{aligned} H_I = & J_1 \sqrt{\Delta^2 + 8} \sum_i \hat{q}_{0,i}^\dagger \hat{q}_{0,i} + (\Delta J_1 + \mu) \sum_i \hat{q}_{2i}^\dagger \hat{q}_{2i} \\ & + \mu \sum_i \hat{s}_{2,i}^\dagger \hat{s}_{2,i}. \end{aligned} \quad (11)$$

H_3 contains cubic terms in the bosonic operators. H_{SW} describes the spin-wave excitations of the 12 flavors of bosons.

They can be decoupled into the three “directions:” x , y , and z . We define in the momentum space,

$$b_{\alpha,\mathbf{k}} = (\hat{t}_{\alpha,\mathbf{k}} \hat{t}_{\alpha,\mathbf{k}} \hat{r}_{\alpha,\mathbf{k}} \hat{q}_{\alpha,\mathbf{k}} \hat{t}_{\alpha,-\mathbf{k}}^\dagger \hat{r}_{\alpha,-\mathbf{k}}^\dagger \hat{q}_{\alpha,-\mathbf{k}}^\dagger)^T. \quad (12)$$

After Fourier transformation, the spin-wave Hamiltonian is given by

$$H_{\text{SW}} = \frac{1}{2} \sum_{\mathbf{k},\alpha} b_{\alpha,\mathbf{k}}^\dagger \Omega_{\mathbf{k}}^\alpha b_{\alpha,\mathbf{k}} - \frac{3}{2} N \mu, \quad (13)$$

where

$$\Omega_{\mathbf{k}}^\alpha = \begin{pmatrix} Q^\alpha + R_{\mathbf{k}}^\alpha & R_{\mathbf{k}}^\alpha \\ R_{\mathbf{k}}^\alpha & Q^\alpha + R_{\mathbf{k}}^\alpha \end{pmatrix}. \quad (14)$$

The matrices Q^α and $R_{\mathbf{k}}^\alpha$ are given in Appendix B.

Equation (13) is diagonalized through Bogoliubov transformation [19],

$$H_{\text{SW}} = \sum_{\mathbf{k},\alpha,n} \varepsilon_{\alpha,\mathbf{k}}^{(n)} \gamma_{\alpha,\mathbf{k}}^{(n)\dagger} \gamma_{\alpha,\mathbf{k}}^{(n)} + \frac{1}{2} \sum_{\alpha,\mathbf{k},n} \varepsilon_{\alpha,\mathbf{k}}^{(n)} - \frac{3}{2} N \mu. \quad (15)$$

$n \in \{1-4\}$ labels the four branches of excitations in each direction α . $\varepsilon_{\alpha,\mathbf{k}}^{(n)}$'s are the excitation energies or the dispersions of the quasiparticles $\gamma_{\alpha,\mathbf{k}}^{(n)}$ (Appendix C). The constant term in Eq. (15) is the energy contribution to the ground state from lowest-order quantum fluctuation described by the spin waves. The quasiparticles do not have well-defined total spin numbers except at the isotropic point. It should be noted that in the isotropic limit (the canonical Shastry-Sutherland model), the $S = 2$ SBs $\hat{q}_{\alpha,\mathbf{k}}^\dagger (\hat{q}_{\alpha,\mathbf{k}})$ are decoupled from the remaining $S = 1$ SBs. As such the $S = 2$ states were neglected in previous studies of the plaquette phase [15]. However, they contribute to a more accurate ground-state energy through second-order perturbation theory as we demonstrate below. While this does not affect the estimation of the phase boundary, the $S = 2$ quasiparticles have a profound influence on the nature of the low-lying excitations, as we demonstrate below.

To get a more accurate ground-state energy, we include the contribution of the cubic terms H_3 through second-order perturbation theory. It is possible to write H_3 in terms of the 15 quasiparticle operators,

$$H_3 = \sum_{\mathbf{k}_1,\mathbf{k}_2} \sum_{a,b,c} \Gamma^{abc}(\mathbf{k}_1,\mathbf{k}_2, -\mathbf{k}_1 - \mathbf{k}_2) \hat{\gamma}_{a,\mathbf{k}_1}^\dagger \hat{\gamma}_{b,\mathbf{k}_2}^\dagger \hat{\gamma}_{c,-\mathbf{k}_1-\mathbf{k}_2}^\dagger \\ + \Gamma_c^{ab}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_1 + \mathbf{k}_2) \hat{\gamma}_{a,\mathbf{k}_1}^\dagger \hat{\gamma}_{b,\mathbf{k}_2}^\dagger \hat{\gamma}_{c,\mathbf{k}_1+\mathbf{k}_2}^\dagger + \text{H.c.} \quad (16)$$

We have extended the quasiparticle operators to include the the 3 isolated excitations (11) and the 12 dispersive quasiparticles (15). The indices α and n are absorbed in the new set of indices $\{a,b,c\}$. The nonzero contribution to the ground-state energy is given by

$$E_0^{(2)} = - \sum_{\mathbf{k}_1,\mathbf{k}_2} \sum_{\{a,b,c\}} |\bar{\Gamma}^{abc}(\mathbf{k}_1,\mathbf{k}_2, -\mathbf{k}_1 - \mathbf{k}_2)|^2 \\ \times (\varepsilon_{a,\mathbf{k}_1} + \varepsilon_{b,\mathbf{k}_2} + \varepsilon_{c,-\mathbf{k}_1-\mathbf{k}_2})^{-1}. \quad (17)$$

$\bar{\Gamma}^{abc}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3)$ are the symmetrized coefficients,

$$\bar{\Gamma}^{abc}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3) = 3! S_3 \Gamma^{abc}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3), \quad (18)$$

where S_3 is a symmetrization operator that sums over all the permutations of the tuple $\{abc\}$ simultaneously with the

FIG. 2. (Color online) The energy of the plaquette state at $\Delta = 1$ with and without the cubic correction. The inset shows the absolute values of the correction.

associated momenta of the coefficients $\Gamma^{abc}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3)$. The summation in Eq. (17) is carried out over unordered index combinations $\{a,b,c\}$. Combining the contributions from H_{SW} and H_3 , the ground-state energy of the plaquette state assumes the form

$$\frac{E_{\text{PL}}}{N} = -\frac{1}{4} \mu + \frac{1}{2N} \sum_{\mathbf{k},\alpha,n} (\varepsilon_{\alpha,\mathbf{k}}^{(n)} - \mu) + \frac{E_0^{(2)}}{N}. \quad (19)$$

Figure 2 shows the energy of the plaquette state at $\Delta = 1$. The large quantum correction from the cubic term is obvious, which implies its essentiality.

III. RESULTS

For large J_2/J_1 , the ground state retains its dimer character. As J_2 increases, the J_2 bond connecting two adjacent plaquettes will dissolve the generalized plaquette singlet and form a dimer singlet. Hence, in the large J_2/J_1 limit, the dimer singlet state remains the ground state even with the existence of the spin anisotropy. Using the standard dimer basis, it is straightforward to obtain the exact energy (per lattice site) of the dimer singlet as a function of Δ and J_2 ,

$$E_D = -\frac{1}{8}(2 + \Delta)J_2. \quad (20)$$

A straightforward comparison between the two energies gives the critical point of the first-order phase transition between the plaquette and the dimer phases. The boundary separating the two ground states is shown by the solid black line in Fig. 3. The generalized plaquette ground state extends to (slightly) larger J_2/J_1 in the presence of exchange anisotropy.

The same approach cannot be used to determine the boundary between the plaquette phase and the AFM phase at small J_2/J_1 . Instead we study the lowest branch dispersion of the quasiparticles at various values of $x = J_2/J_1$ and Δ . We restrict ourselves to the spin-wave Hamiltonian (15) for the rest of the paper. The generalized plaquette ground state has gapped excitations. The gap reduces towards the AFM phase (small $x = J_2/J_1$), and the lowest excitation becomes gapless at the phase boundary where the system undergoes a continuous

FIG. 3. (Color online) The phase range of the plaquette state. The AFM-plaquette line is exact on the left of the critical line obtained from Eq. (21). The five critical points $(J_2/J_1, \Delta)$ are as follows: $E_1 = (1.25, 1)$, $E_2 = (1.514, 1.3)$, $E_3 = (1.650, 0.33)$, $\Delta_1 = (1.450, 1.21)$, and $\Delta_2 = (1.448, 0.63)$. The four points (a)–(d) mark the corresponding locations of the spectrum in Fig. 5. The inset shows the gap of the plaquette singlet phase in the isotropic case.

phase transition from the plaquette ground state to the AFM ground state. The excitation becomes unstable within the AFM phase. The stability condition gives the phase boundary between the plaquette state and the AFM state, the blue line in Fig. 3. We observe that the plaquette ground state extends only over a finite range of Δ . A sufficiently strong anisotropy—both Ising- and XY-like—drives a transition to the AFM state. The AFM-PL phase boundary ends at two critical points— $E_2 = (1.51, 1.3)$ and $E_3 = (1.65, 0.33)$ —beyond which there is a direct AFM to dimer transition. For $\Delta = 1$, the boundary of the plaquette phase is found to be $1.25 \leq J_2/J_1 \leq 1.46$, in good agreement with the results from more rigorous tensor network calculations [17].

The most interesting outcome of the present study is the nature of the low-lying excitations in the plaquette phase. The lowest excitation is gapped with a quadratic dispersion at the minimum. The gapped nature is shown for $\Delta = 1$ in the inset of Fig. 3. As one approaches the AFM-plaquette boundary, the dispersion at the minimum changes to linear at the boundary along with the vanishing of the gap along expected lines. The unexpected finding is the change in the position of the minimum. Over a majority of the plaquette phase, the minimum is at $\mathbf{k} = (0, 0)$, consistent with previous studies. However, there exists a separatrix, shown in red in Fig. 3, at which the location of the minimum gap splits from $\mathbf{k} = (0, 0)$ into four points. The fourfold degenerate points are closed under the Z_4 symmetry group generated by the transformation $(k_x, k_y) \rightarrow (-k_y, k_x)$. As shown in Fig. 4 with $\Delta = 0.63$, the locations of the minima move away from $(0, 0)$ as J_2/J_1 increases in the PL2 region. The lowest excitations at four different points in the phase space have been shown in Fig. 5. This qualitative change in the nature of the lowest excitation branch effectively splits the plaquette phase into two regions—PL1 and PL2. The boundary separating the two regions meets the AFM phase boundary at two points

FIG. 4. (Color online) The minimum gap location vector $\mathbf{k}_0$ increases logarithmically in magnitude. The inset shows the exact locations of the minimum gaps at different values of J_2/J_1 .

($x_1 = 1.450, \Delta_1 = 1.21$) and ($x_2 = 1.448, \Delta_2 = 0.63$). The boundary between the AFM and the PL1 can be expressed in a closed form

$$\begin{aligned} \frac{J_2}{J_1} &= 2 - \frac{3(\mu - J_1)}{4J_1 e_z^2} \quad \text{for } 0.64 \leq \Delta \leq 1, \\ \frac{J_2}{J_1} &= 2 - \frac{3(\mu - \Delta J_1)}{4\Delta J_1 e_z^2} \quad \text{for } 1 \leq \Delta \leq 1.22. \end{aligned} \quad (21)$$

The Δ -dependent e_α is given in Appendix B. The boundary between the PL2 and the AFM phases cannot be expressed in a closed form and is instead computed by numerically solving for the parameters where the (fourfold degenerate) gap to the lowest excitation vanishes. The nature of the low-lying excitation in PL2 is contrary to previous calculations of low-energy excitations [16] in the Shastry-Sutherland model where the minimum of the lowest excitation (in the limit $\Delta = 1$) was always found to be at $\mathbf{k} = (0, 0)$. The discrepancy is explained by noting that only the $\hat{t}_\alpha$ branch was considered in the calculation of the excitation spectrum in Ref. [16], whereas we retain all the SBs in our calculations. The qualitative change in the character of the excitation underscores the fact that the low-energy physics has significant contributions from the additional SBs. The boundary separating the plaquette valence-bond phases with distinct low-energy excitation is determined by studying the nature of the dispersion of the lowest excitation branch at $\mathbf{k} = (0, 0)$ and determining the parameter values (J_2/J_1 and Δ) where it changes from a minimum to a (local) maximum (Appendix C). It is interesting to note that our estimate of the boundary between the plaquette valence-bond phase and the dimer phase at the isotropic point, viz., $J_2/J_1 = 1.456$ agrees well with that from the tensor network calculations, although the nature of the lowest excitation branch that drives this transition (by getting soft at the phase boundary) is distinct from previous results. It is worth mentioning that the boundary between PL1 and PL2 is not smooth at $\Delta = 1$ as a consequence of the different natures of the excitations in the Ising-like and XY-like regions.

To summarize, we have used a plaquette basis to obtain the stability range of a generalized plaquette valence-bond

FIG. 5. (Color) The lowest branch dispersion at (a) $\Delta = 1$ and $J_2/J_1 = 1.25$: gapless and linear at $\mathbf{k} = (0,0)$; (b) $\Delta = 0.5$ and $J_2/J_1 = 1.519$: gapless and linear at $\pm(0.2\pi, 0.14\pi)$ and $\pm(0.14\pi, -0.2\pi)$; (c) $\Delta = 1.1$ and $J_2/J_1 = 1.442$: gapped and smooth at $\mathbf{k} = (0,0)$; (d) $\Delta = 0.6$ and $J_2/J_1 = 1.5$: gapped with four minima at $\pm(0.17\pi, 0.12\pi)$ and $\pm(0.12\pi, -0.17\pi)$.

solid as the ground state of the Shastry-Sutherland model with exchange anisotropy. We found that the plaquette state survives for a finite range of anisotropies— $0.33(1) \leq \Delta \leq 1.30(1)$, $1.25 \leq J_2/J_1 \leq 1.650(1)$ —and is unstable towards AFM ordering for both Ising-like ($\Delta > 1$) and XY-like ($\Delta < 1$) anisotropies. At the isotropic point, our result agrees well with those obtained from the variational tensor network method using infinite projected entangled-pair states—a more accurate computational method [17]. However, more insight into the plaquette phase is obtained from the current approach. This is manifested by the surprising finding that the lowest excitation branch changes its nature qualitatively as one approaches the dimer phase while still maintaining the plaquette nature of the ground state.

ACKNOWLEDGMENTS

It is a pleasure to thank C. Batista, K. Wierschem, and F. Mila for valuable discussions. The work was partially supported by Grant No. MOE2011-T2-1-108 from the Ministry of Education, Singapore.

APPENDIX A

The configuration of the condensed new plaquette state $|s_0\rangle$,

$$|s_0\rangle = -\left(\frac{1}{\sqrt{6}}\sin\phi + \frac{1}{\sqrt{3}}\cos\phi\right)\left(\begin{vmatrix}\uparrow & \downarrow \\ \downarrow & \uparrow\end{vmatrix} + \begin{vmatrix}\downarrow & \uparrow \\ \uparrow & \downarrow\end{vmatrix}\right) \\ + \left(\frac{1}{2\sqrt{3}}\cos\phi - \frac{1}{\sqrt{6}}\sin\phi\right)\left(\begin{vmatrix}\uparrow & \uparrow \\ \downarrow & \downarrow\end{vmatrix} + \begin{vmatrix}\downarrow & \downarrow \\ \uparrow & \uparrow\end{vmatrix}\right) \\ + \left(\frac{1}{2\sqrt{3}}\cos\phi - \frac{1}{\sqrt{6}}\sin\phi\right)\left(\begin{vmatrix}\uparrow & \downarrow \\ \uparrow & \downarrow\end{vmatrix} + \begin{vmatrix}\downarrow & \uparrow \\ \downarrow & \uparrow\end{vmatrix}\right).$$

The energies of the anisotropic Heisenberg interaction on a single square,

$$E_{s_0} = -\mu = -\frac{1}{2}J_1(\Delta + \sqrt{\Delta^2 + 8}), \\ E_{t_\alpha} = -J_1^\alpha, \quad E_{q_\alpha} = J_1^\alpha, \quad E_{s_2} = E_{l_\alpha} = E_{r_\alpha} = 0, \\ E_{q_0} = \frac{1}{2}J_1(\sqrt{\Delta^2 + 8} - \Delta), \quad E_{q_2} = \Delta J_1.$$

APPENDIX B

The matrix $R_{\mathbf{k}}^\alpha$ is given by

$$R_{\mathbf{k}}^\alpha = J_1^\alpha \begin{pmatrix} -\frac{1}{3}(2-x)e_\alpha^2 f(\mathbf{k}) & -\frac{i}{3}e_\alpha h_\alpha w_1(\mathbf{k}) & \frac{i}{3}e_\alpha h_\alpha w_2(\mathbf{k}) & -\frac{1}{\sqrt{6}}xe_\alpha d_\alpha g(\mathbf{k}) \\ \frac{i}{3}e_\alpha h_\alpha w_1(\mathbf{k}) & -\frac{1}{3}xh_\alpha^2 \cos k_x a & \frac{1}{3}h_\alpha^2 g(\mathbf{k}) & -\frac{i}{\sqrt{6}}h_\alpha d_\alpha w_3(\mathbf{k}) \\ -\frac{i}{3}e_\alpha h_\alpha w_2(\mathbf{k}) & \frac{1}{3}h_\alpha^2 g(\mathbf{k}) & -\frac{1}{3}xh_\alpha^2 \cos k_y a & \frac{i}{\sqrt{6}}h_\alpha d_\alpha w_4(\mathbf{k}) \\ -\frac{1}{\sqrt{6}}xe_\alpha d_\alpha g(\mathbf{k}) & \frac{i}{\sqrt{6}}h_\alpha d_\alpha w_3(\mathbf{k}) & -\frac{i}{\sqrt{6}}h_\alpha d_\alpha w_4(\mathbf{k}) & \frac{1}{2}d_\alpha^2(x+2)f(\mathbf{k}) \end{pmatrix},$$

where $x = \frac{J_z}{J_1}$ and a is twice the lattice constant as shown in Fig. 1. The constants and functions of $\mathbf{k}$ in the above matrix elements are given by

$$\begin{aligned} f(\mathbf{k}) &= \cos k_x a + \cos k_y a, \\ g(\mathbf{k}) &= \cos k_x a - \cos k_y a, \\ w_1(\mathbf{k}) &= (x-1) \sin k_x a - \sin k_y a, \\ w_2(\mathbf{k}) &= \sin k_x a + (x-1) \sin k_y a, \\ w_3(\mathbf{k}) &= (x+1) \sin k_x a + \sin k_y a, \\ w_4(\mathbf{k}) &= \sin k_x a - (x+1) \sin k_y a, \\ e_\alpha &= \cos \phi + \frac{1}{\sqrt{2}} \sin \phi \cos \theta_\alpha, \\ h_\alpha &= \sin \phi \cos \theta_\alpha - \frac{1}{\sqrt{2}} \cos \phi, \\ d_\alpha &= \sin \phi \sin \theta_\alpha. \end{aligned}$$

The matrix Q^α is given by

$$Q^\alpha = \begin{pmatrix} \mu - J_1^\alpha & 0 & 0 & 0 \\ 0 & \mu & 0 & 0 \\ 0 & 0 & \mu & 0 \\ 0 & 0 & 0 & \mu + J_1^\alpha \end{pmatrix}.$$

APPENDIX C

The dispersions $\varepsilon_{\alpha,\mathbf{k}}^{(n)}$ are given by the eigenvalues of the matrix $M_{\mathbf{k}}^\alpha = Q^\alpha(2R_{\mathbf{k}}^\alpha + Q^\alpha)$,

$$\varepsilon_{\alpha,\mathbf{k}}^{(1,2)} = \left[-\frac{1}{4}(\alpha_3 - \sqrt{\alpha_3^2 - 4\alpha_2 + 4y_\alpha}) \pm \sqrt{D_-} \right]^{1/2} \quad (C1)$$

$$\varepsilon_{\alpha,\mathbf{k}}^{(3,4)} = \left[-\frac{1}{4}(\alpha_3 + \sqrt{\alpha_3^2 - 4\alpha_2 + 4y_\alpha}) \pm \sqrt{D_+} \right]^{1/2}, \quad (C2)$$

with

$$D_\pm = \frac{1}{4}(\alpha_3 \pm \sqrt{\alpha_3^2 - 4\alpha_2 + 4y_\alpha})^2 - 2(y_\alpha \pm \sqrt{y_\alpha^2 - 4\alpha_0}). \quad (C3)$$

y_α is a real number given by

$$y_\alpha = \frac{1}{3}\alpha_2 - 2\sqrt{-\frac{m_\alpha}{3}} \cos\left(\psi_\alpha + \frac{2n\pi}{3}\right), \quad (C4)$$

$$\cos 3\psi_\alpha = \frac{1}{2}p_\alpha \left(-\frac{3}{m_\alpha}\right)^{3/2}, \quad (C5)$$

$$p_\alpha = -\frac{2}{27}\alpha_2^3 + \frac{1}{3}\alpha_1\alpha_2\alpha_3 + \frac{8}{3}\alpha_0\alpha_2 - \alpha_1^2 - \alpha_3^2\alpha_0, \quad (C6)$$

$$m_\alpha = \alpha_1\alpha_3 - 4\alpha_0 - \frac{1}{3}\alpha_2^2. \quad (C7)$$

n is any integer such that y_α is real. The $\mathbf{k}$ -dependent parameters α_n can be expressed using the trace and determinant of $M_{\mathbf{k}}^\alpha$,

$$\alpha_0 = \det\{M_{\mathbf{k}}^\alpha\}, \quad (C8)$$

$$\alpha_1 = -\frac{1}{6}[\text{Tr}\{M_{\mathbf{k}}^\alpha\}^3 - 3\text{Tr}\{(M_{\mathbf{k}}^\alpha)^2\}\text{Tr}\{M_{\mathbf{k}}^\alpha\} + 2\text{Tr}\{(M_{\mathbf{k}}^\alpha)^3\}], \quad (C9)$$

$$\alpha_2 = \frac{1}{2}[\text{Tr}\{M_{\mathbf{k}}^\alpha\}^2 - \text{Tr}\{(M_{\mathbf{k}}^\alpha)^2\}], \quad (C10)$$

$$\alpha_3 = -\text{Tr}\{M_{\mathbf{k}}^\alpha\}. \quad (C11)$$

The behavior of the dispersions at $\mathbf{k} = (0,0)$ could be observed by looking at their Hessian matrix,

$$H_\alpha = \begin{pmatrix} \frac{\partial^2}{\partial k_x^2} \varepsilon_{\alpha,\mathbf{k}}^{(n)} & \frac{\partial^2}{\partial k_x \partial k_y} \varepsilon_{\alpha,\mathbf{k}}^{(n)} \\ \frac{\partial^2}{\partial k_x \partial k_y} \varepsilon_{\alpha,\mathbf{k}}^{(n)} & \frac{\partial^2}{\partial k_y^2} \varepsilon_{\alpha,\mathbf{k}}^{(n)} \end{pmatrix}_{\mathbf{k}=(0,0)}. \quad (C12)$$

The critical point at which the minimum of the dispersion is no longer at $\mathbf{k} = (0,0)$ could be determined from the second derivative test.

-
- [1] B. S. Shastry and B. Sutherland, *Physica B+C* **108**, 1069 (1981).
 - [2] T. Momoi and K. Totsuka, *Phys. Rev. B* **62**, 15067 (2000).
 - [3] C. H. Chung, J. B. Marston, and S. Sachdev, *Phys. Rev. B* **64**, 134407 (2001).
 - [4] S. Miyahara and K. Ueda, *J. Phys.: Condens. Matter* **15**, R327 (2003).
 - [5] R. Darradi, J. Richter, and D. J. J. Farnell, *Phys. Rev. B* **72**, 104425 (2005).
 - [6] J. Dorier, K. P. Schmidt, and F. Mila, *Phys. Rev. Lett.* **101**, 250402 (2008).
 - [7] A. Abendschein and S. Capponi, *Phys. Rev. Lett.* **101**, 227201 (2008).
 - [8] J. Lou, T. Suzuki, K. Harada, and N. Kawashima, *arXiv:1212.1999*.
 - [9] R. W. Smith and D. A. Keszler, *J. Solid State Chem.* **93**, 430 (1991).
 - [10] H. Kageyama, K. Yoshimura, R. Stern, N. V. Mushnikov, K. Onizuka, M. Kato, K. Kosuge, C. P. Slichter, T. Goto, and Y. Ueda, *Phys. Rev. Lett.* **82**, 3168 (1999).
 - [11] S. Miyahara and K. Ueda, *Phys. Rev. Lett.* **82**, 3701 (1999).
 - [12] M. Albrecht and F. Mila, *Europhys. Lett.* **34**, 145 (1996).
 - [13] A. Koga and N. Kawakami, *Phys. Rev. Lett.* **84**, 4461 (2000).
 - [14] Y. Takushima, A. Koga, and N. Kawakami, *J. Phys. Soc. Jpn.* **70**, 1369 (2001).
 - [15] W. Zheng, J. Oitmaa, and C. J. Hamer, *Phys. Rev. B* **65**, 014408 (2001).
 - [16] A. Läuchli, S. Wessel, and M. Sigrist, *Phys. Rev. B* **66**, 014401 (2002).
 - [17] P. Corboz and F. Mila, *Phys. Rev. B* **87**, 115144 (2013).
 - [18] T. Holstein and H. Primakoff, *Phys. Rev.* **58**, 1098 (1940).
 - [19] J. Colpa, *Physica A* **93**, 327 (1978).