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Exotic Phases in Frustrated Quantum Systems

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Abstract

In this work, we study the extended Shastry-Sutherland model which is a Quantum spin system with geometrical frustration. It is originated from the compound $\text{SrCu}_2(\text{BO}_3)_2$ which exhibits nontrivial magnetization plateaus in the external magnetic field. In the first part, we use a generalized spin wave theory (spin wave theory in plaquette representation) to investigate the intermediate phase (extremely frustrated range) of the Shastry-Sutherland model and the associated quasiparticle dispersions. We confirm the existence of this plaquette singlet state by explicitly calculating its energy with quantum corrections from second order perturbation theory. We also propose a more general plaquette valence-bond-solid (PVBS) phase when the anisotropy is turned on. The quasiparticle dispersion changes qualitatively when it passes a critical line within the PVBS phase. The gap splits from $\mathbf{k} = (0,0)$ to four degenerate points which may imply a crossover to the resonating valence bond state.

In the second part, we study the magnetization plateaus and supersolid phases in the extended Shastry-Sutherland model which is expected to be the effective model of the rare earth tetraborides family. Analytical (spiral plaquette representation) and numerical (Stochastic Series Expansion QMC) methods are applied to the Ising and Ising-like XXZ models, respectively. Results from the two methods are qualitatively consistent. Expected plateaus $1/2$ and $1/3$ have been observed in the phase diagram and a few new plateaus including $5/9$ and $2/9$ have been observed in a narrow regime of the phase diagram.

Besides, we also study the Spin-1 Heisenberg model on a bipartite lattice to test the consistency of the QMC and spin wave method. The excellent quantitative and qualitative agreement guarantees the efficiency of the lowest order spin wave theory.

Chapter 1

Introduction

The study of strongly correlated quantum systems is one of the most active research areas in condensed matter physics. Various physical properties of many materials in experiments could be explained and even predicted by effective bosonic or spin models. The physical interactions in these models can be very simple at the microscopic level, however, they can lead to the emergence of very complex phenomena at the macroscopic level. This includes different ground states, which is our main interest in this thesis, different excitations and different classes of phase transitions. The miscellaneous properties in a strongly correlated system is a consequence of the interplay among the interactions, lattice geometry and external potentials. For example, the Bose-Hubbard model only has nearest neighbour hopping and onsite interactions. However, at integer filling, the ground state could be either Mott insulator or superfluid[1], which are completely different. This is a result of competition between two simple interactions. In the strong hopping limit, the ground state is a superfluid while a Mott insulator in the other limit.

1.1 Lattice geometry and frustration

Among the effective factors of a system, the topology of the system (model) plays a very important role. Depending on the objective of the investigation, a material can be mapped onto models with different topologies, even though the interactions in each model could be similar, say nearest neighbor interaction. For example, if we are going to

study the magnetic properties of a material, we only have to consider those magnetically contributing atoms. They can be mapped to a lattice of spins and the lattice geometry is determined by the arrangement of these atoms, neglecting the geometry of the rest. What's more, the lattice of the model doesn't have to preserve the real geometry of the atoms. Instead, the geometry could always be reduced to some well known regular lattices through conformal transformation, for example, a parallelogram could be reduced to a square lattice. The actual geometry could be reflected in the axial anisotropy of the interactions in the model. As a result, different materials could be mapped to models with the same geometry and actually be classified according to the effective lattice. Hence, a complete study of certain lattice with all possible interaction strength is of great importance for a large class of materials. There are many well known and extensively studied lattice systems: square (cubic) lattice, triangular lattice, honeycomb lattice and so on. Among all of them, a lattice with the so-called geometrical frustration is one of the most complicated systems, because of their high sensitivity to the relative changes in the interaction strength and external potential.

As the word itself implies, in a geometrically frustrated system, local interactions cannot be simultaneously optimized in any configuration. This could be illustrated by a canonical example of three classical antiferromagnetically interacting spins located on the three corners of a triangle, as shown in Fig.1.1. Two adjacent spins prefer anti-alignment to lower the energy. However, this could not be achieved for all the three bonds. There is always a bond with two parallel spins. The lowest energy level is doubly degenerate.

When the frustration extends to the whole lattice, the situation becomes much more complicated. Firstly, there exists intensive competition among various long range orderings. Simultaneous ordering becomes possible, for example the supersolid phase near the plateaus in the extended Shastry-Sutherland model (Chapter 6). Secondly, quantum fluctuation is enhanced due to the frustration and may change qualitatively for different parameters even with the same ordering, for example, the excitation dispersion of the plaquette state in Shastry-Sutherland model changes its location of the gap when the parameters change (Chapter 5). Last but not least, some ordering may not even be obvious.

Figure 1.1: Three classical antiferromagnetically interacting spins. The two spins enclosed have maximum interacting energy.

They could occur when the system is in the extensive frustrated regime, for example, the plaquette state in the Shastry-Sutherland model, which appears as an intermediate phase when neither of the two interactions dominates obviously (Chapter 5). As a result, a frustrated system has an extremely rich phase diagram. A small change in the parameters could drive phase transitions from one ordering to another. The fundamental motivation lies in understanding the mechanism behind the emergence of these complex phenomena. Understanding and, even more importantly, controlling the emergent complexity in these systems is a major challenge confronting the community of condensed matter physicists.

1.2 Shastry-Sutherland model and $\text{SrCu}_2(\text{BO}_3)_2$ compound

The Shastry-Sutherland model is a paradigmatic model to study frustrated system. This model was invented in 1981 by Shastry and Sutherland after whom the model is named[2]. It is a two dimensional spin system where $S = 1/2$ moments are arranged in a square lattice. There are two kinds of interactions in this model. One is the usual axial antiferromagnetic (AFM) Heisenberg interaction and the other AFM interaction is on alternating diagonal bonds, as shown in Figure 1.2.

Figure 1.2: The lattice configuration of the Shastry-Sutherland model with axial interaction J_1 and diagonal interaction J_2 .

The Hamiltonian of the system is given by

$$\hat{H} = J_1 \sum_{\langle i,j \rangle} \hat{S}_i \cdot \hat{S}_j + J_2 \sum_{[i,j]} \hat{S}_i \cdot \hat{S}_j \quad (1.1)$$

where $\langle i, j \rangle$ is the nearest neighbour bond and $[i, j]$ is the diagonal bond. $\hat{S} = (\hat{S}^x, \hat{S}^y, \hat{S}^z)$ are spin operators.

When $J_2 = 0$, the system reduces to the well known spin-1/2 Heisenberg model on a square lattice, the ground state of which is known as the Néel state with magnetic long range order. This ordering survives for some small but finite values of J_2 . In the other limit, when $J_1 = 0$, the Hamiltonian consists of isolated dimers on the J_2 bonds. The ground state is simply a direct product of singlets on each J_2 bond, which we call dimer singlet ground state. It is remarkable that this dimer singlet state is an exact eigen state even with finite J_1 . The axial interactions cancel out because of the frustration. Shastry and Sutherland showed that the dimer singlet state remained an exact ground state without any quantum fluctuation for $J_1/J_2 < 0.5$. The critical point was obtained by comparison of the mean field ground state energy between the AFM ordered state and the dimer singlet state. The accuracy of this critical point was improved by Koga and Kawakami [3] using a series expansion calculation and they extended the stability of the dimers singlet ground state to $J_1/J_2 < 0.68$.

Ten years later, in 1991, the $\text{SrCu}_2(\text{BO}_3)_2$ compound was discovered by Smith and Keszler [4]. $\text{SrCu}_2(\text{BO}_3)_2$ has a tetragonal crystal structure with layered CuBO_3 and Sr

planes. The lattice constants are $a = 8.995\text{\AA}$ and $c = 6.649\text{\AA}$ at room temperature. A CuBO_3 layer is shown in Figure 1.3a. The Cu^{2+} ion carries a spin with $S = 1/2$. In the plane, there is one nearest neighbour and four next nearest neighbours for each Cu^{2+} ion*. Two pairs Cu^{2+} ions are connected through the O atoms. At room temperature, the distance of the nearest neighbour is $2.905\text{\AA}$ and $5.132\text{\AA}$ for the next nearest neighbour. The unit cell consists of two types of dimers, the two orthogonal dimers shown in Figure 1.3b. Each type of dimer lies in a plane and the two planes are shifted slightly from each other. However, there exists a critical temperature $T_s = 395\text{K}$ above which all the dimers are in the same plane. Thus, the CuBO_3 layer becomes a mirror plane. This is very important for Dzyaloshinsky-Moriya interactions which may exist for next-nearest neighbours but not the for nearest neighbours since its middle point is an inversion center.

Because the bridge angle of the Cu-O-Cu chain is about 102.42° at room temperature, we could assume the nearest neighbour interaction J is antiferromagnetic, that is AFM interaction within the dimer. However, J alone is not able to explain the experimental observations of this compound, including magnetic plateaus and a very different spin gap. Therefore, a frustrated interaction, AFM J' between next nearest neighbour Cu^{2+} ions, becomes essential. Figure 1.3b shows the lattice structure of the Cu^{2+} ions with the above two interactions. This lattice is topologically equivalent to the Shastry-Sutherland model. The conformal transformation maps the nearest neighbour interaction J in the compound into the alternating diagonal (next nearest neighbour) interactions J_2 and maps the next nearest neighbour interaction J' into the axial (nearest neighbour) interaction J_1 .

By fitting the experimental result of the temperature dependent magnetic susceptibility, the two interactions were obtained as $J_1 = 54\text{K}$ and $J_2 = 85\text{K}$, that is $J_1/J_2 \approx 0.635$ which is in the dimer singlet ground state regime. This spin gapped ground state was confirmed experimentally from the magnetization curve. Therefore, the magnetic properties of the $\text{SrCu}_2(\text{BO}_3)_2$ compound are best described by the Shastry-Sutherland model.

When external magnetic field is applied, the compound shows a sequence of magnetic plateaus at $1/n$ ($n = 2, \dots, 9$) and $2/9$ [5, 6, 7, 8]. This could be particularly explained

*The nearest neighbour and next nearest neighbour have reversed meaning to those in Shastry-Sutherland model.

Figure 1.3: (a) The crystal structure of CuBO_3 : the purple dots represent Cu atoms, the big open circles represent O atoms and small open circles represent B atoms. The unit cell is enclosed by the dotted line. (b) The lattice of Cu^{2+} extracted from the material. (c) The lattice of Cu^{2+} after conformal transformation.

by a hard-core boson model based on the Shastry-Sutherland model[9, 10]. The dimer singlet ground state is treated as an artificial vacuum. We consider the excitation from a singlet to a triplet as creation of a boson on that dimer. Since each dimer could only be in one of the four states (one singlet and three triplet), this is a hard-core boson problem. Momoi and Totsuka [9, 10] derived an effective Hamiltonian using third order perturbation theory. However, they could only produce the $1/2$ and $1/3$ plateaus while the $1/4$ and $1/8$ plateaus were missing. This indicates that interactions from third order approximation is not sufficient enough to explain these two plateaus. Exact diagonalization in finite lattice supported this argument. In a 16-site cluster, both $J_1/J_2 = 0.4$ and $J_1/J_2 = 0.635$ were performed and the $1/8$ plateau was only obtained for $J_1/J_2 = 0.635$.

As a frustrated system, another interesting phenomenon in Shastry-Sutherland model is the novel ground state in different parameter ranges. As discussed above, there exists a phase transition between the AFM long range order and the dimer singlet disordered state. However, in 1996, Mila[11] proposed the helical order as a possible intermediate phase between the two phases with range $0.6 < J_1/J_2 < 0.9$. In 2000, Koga and Kawakami[12] proposed a plaquette singlet state as the intermediate phase with range $0.677(2) < J_1/J_2 < 0.86$. Motivated by the two works, several studies based on a diverse array of approaches have variously predicted a number of different intermediate phases. However, the general consensus has leaned towards an intermediate plaquette singlet phase over a narrow range of J_1/J_2 . In Chapter 5, we apply a plaquette representation

to show explicitly the stability of this plaquette singlet ground state and we also study the dispersions of the excitations. Moreover, we extend our study on this intermediate phase to the anisotropic Shastry-Sutherland model, partially motivated by the rare earth tetraborates as discuss in the next section.

1.3 Rare earth tetraborides and the extended Shastry-Sutherland model

Recently, there have been several new additions to the Shastry-Sutherland family of compounds[13, 14]. These include a complete family of rare earth tetraborides, RB_4 , ($\text{R} = \text{Tm}, \text{Tb}, \text{Dy}, \text{Ho}$ and Er). While the compounds have been known for a long time, their relevance to the Shastry-Sutherland model has only recently been realized. The crystal structure of the compounds consists of weakly coupled layers. Within each layer, the magnetic moment carrying R^{4+} ions form a Shastry-Sutherland lattice. Consequently, the magnetic properties of these materials are expected to be described by generalizations of the Shastry-Sutherland model with varying parameters - such as the magnitude of the spin moment and interaction strengths and possibly additional interactions. Unlike $\text{SrCu}_2(\text{BO}_3)_2$, the rare-earth tetraborides are metallic and have magnetic ground states ranging from Ising-like order for TmB_4 (where the magnetic moments are aligned perpendicular to the plane of the lattice) to XY-like order for TbB_4 (where the magnetic moments are aligned in the lattice plane). The field-induced properties of these compounds are still largely unknown.

In the rare earth tetraborides, the R^{4+} ions carry large magnetic moments that interact via isotropic antiferromagnetic Heisenberg interactions. The strong crystal fields in these compounds produce large single-ion anisotropies at the ions. For example, the Tm^{4+} ions in TmB_4 carry a spin $S = 6$ and a single-ion anisotropy $D \approx 100\text{K}$ where the near-neighbor bare spin exchange is $J \approx 2.2\text{K}$ [14]. The single-ion anisotropy splits the energy levels of the individual spins into doublets separated by gaps $\sim D$. The thermal excitation to higher levels is heavily suppressed at low temperatures ($T \ll J$) and large

$D(D \gg J)$. The properties of the system are well described by an effective $S = 1/2$ model comprising of the lowest doublet. The effective exchange interactions are obtained as a perturbative expansion in D/J : $J_{eff} \sim (D/J)^{2S}$ where S is the magnetic moment of the original spins. The Ising term remains the same in this derivation of the effective mode. This results in a highly anisotropic Heisenberg interactions in the effective model. Significantly, the effective exchange becomes ferromagnetic interaction, as a result, the frustration in the exchange interaction is removed in the effective low energy model although frustration remains in the Ising interaction. This is crucial for computation as it alleviates the negative sign problem normally encountered in the QMC simulations of frustrated quantum systems.

Recent studies[15, 16] on the Ising Shastry-Sutherland model shows a plateau sequence $0 - 1/3 - 1$ while experimentally the compound TmB_4 which is in the Ising-like compound shows a $1/2$ plateau instead of the $1/3$ plateau. This implies that the bare Shastry-Sutherland model with simple anisotropy is not sufficient to explain the RB_4 compounds. Therefore, further interactions should be considered, which could arise as a result of the RKKY interactions between the itinerant electrons and the localized moments. The Hamiltonian is given by

$$\hat{H} = \sum_{n=1}^4 \sum_{\langle i,j \rangle_n} \sum_{\alpha=x,y,z} J_n^\alpha \hat{S}_i^\alpha \hat{S}_j^\alpha \quad (1.2)$$

$\langle i,j \rangle_n$ represents the four interacting bonds, as shown in Figure 1.4.

Figure 1.4: The lattice of the extended Shastry-Sutherland model

In Chapter 6, we explore the possible plateaus from the extended Shastry-Sutherland model in both Ising limit and Ising-like XXZ model. We also investigate possible super-solid phases near the plateaus in the XXZ case.

Chapter 2

Spin Wave Theory

Spin wave analysis is an efficient method to investigate the ground state phase diagram and the low-lying collective excitations of a spin system[17, 18, 19, 20]. In a spin- s system, the local Hilbert space of spin degree of freedom can be mapped to a Bosonic system with certain constraints such that the dimensions of the two Hilbert spaces are the same, $2s+1$. Two of such mappings are well known as the Holstein-Primakoff representation and the Schwinger boson representation.

2.1 Holstein-Primakoff Representation

In Holstein-Primakoff representation[21], the state $|-s\rangle$ is taken as the “vacuum” state $|0\rangle$ while the state $|-s+n\rangle$ is “created” by adding n bosons to the vacuum state. To make sure the dimension of this local Hilbert space is the same as the original, a non-holonomic constraint has to be imposed to the local bosonic system: $0 \leq n \leq 2s$, where n is the number of local bosons. In this representation, the spin operators can be expressed by the bosonic operator $b^\dagger$ and b :

$$S^+ = \hbar\sqrt{2s}\sqrt{1 - \frac{b^\dagger b}{2s}}b, \quad (2.1)$$

$$S^- = \hbar\sqrt{2s}b^\dagger\sqrt{1 - \frac{b^\dagger b}{2s}}, \quad (2.2)$$

$$S_z = \hbar(s - b^\dagger b). \quad (2.3)$$

Generally, the square root is expanded as a Taylor series of $\frac{b^\dagger b}{2s}$. For large s , it is sufficient to expand the square root up to first (lowest) order in $\frac{b^\dagger b}{2s}$. For this reason, this method is also known as linear spin wave analysis. This approximation can also be applied in the Schwinger boson representation even for small s which is more common in practice. Besides, the non-holonomic constraint can sometimes cause difficulty in determining the saddle point of the free energy. In Schwinger boson representation, we can get rid of this difficulty with a holonomic constraint. These are the reasons why we used Schwinger Boson Representation in the spin wave analysis.

2.2 Schwinger Boson Representation

Instead of a single bosonic operator, we introduce $2s + 1$ flavors bosons in Schwinger boson representation such that each spin state is represented by one flavored boson:

$$|s, m\rangle = b_m^\dagger |\emptyset\rangle \quad (2.4)$$

where $|\emptyset\rangle$ is the “vacuum”. The spin operators are then given by

$$\hat{S}^\mu = \sum_{m,n} S_{mn}^\mu \hat{b}_m^\dagger \hat{b}_n \quad (2.5)$$

where

$$S_{mn}^\mu = \langle m | \hat{S}^\mu | n \rangle \quad (2.6)$$

is the matrix element. $\hat{S}^\mu$ can be $\hat{S}^z$, $\hat{S}^\pm$ or even $(\hat{S}^z)^2$. For simplicity, we have thrown away the s in $|s, m\rangle$ in the above and the following expressions. The $(2s + 1)^2$ operators $\hat{b}_m^\dagger \hat{b}_n$ with the holonomic constraint

$$\sum_m \hat{b}_m^\dagger \hat{b}_m = 1 \quad (2.7)$$

form the generators of the $SU(2s + 1)$ group. One should realize that the states $|m\rangle$ are not restricted to the eigenstates of $\hat{S}^z$. This is another advantage of the Schwinger boson

representation. Suppose the ground state $|gs\rangle$ is given by some linear combination of the eigenstates of $\hat{S}^z$. We can perform a unitary transformation to the spin operators and the Hamiltonian operator such that:

$$\hat{b}_0^\dagger |\emptyset\rangle = |gs\rangle. \quad (2.8)$$

The rest of the bosons represent excited states that are orthogonal to the ground state. This provides a good opportunity for us to apply the variational method. We can assume the ground state in terms of the original basis with some free parameters. Those parameters can be determined by the saddle point condition of the free energy.

In a lattice system, we assume the Hilbert space of the whole system is a direct product of the local Hilbert spaces of each lattice site. This makes the mean field character of the approach explicit. Schwinger bosons $\{\hat{b}_{im}^\dagger, \hat{b}_{im}\}$ are then introduced to every local Hilbert space labeled by lattice site i , where they obey the commutation relation:

$$[\hat{b}_{im}, \hat{b}_{jn}^\dagger] = \delta_{ij}\delta_{mn}. \quad (2.9)$$

The holonomic constraint holds locally:

$$\sum_m \hat{b}_{im}^\dagger \hat{b}_{im} = 1, \quad \text{for all lattice sites } i. \quad (2.10)$$

In practice, there are two ways to get rid of the constraints. The first method is the expansion of the square root as applied in the Holstein-Primakoff representation. The other one is the Lagrange multiplier method where chemical potential μ_i is introduced to the Hamiltonian to enforce the local constraint. I will show both approaches separately in the next two sections.

2.3 Spin Wave Analysis with Holstein-Primakoff Approximation

We illustrate this method by applying it to the anisotropic Spin-1 Heisenberg model with single-ion anisotropy[22].

2.3.1 Uniform linear Holstein-Primakoff approximation

The Hamiltonian of the Heisenberg model with single-ion anisotropy $D > 0$ and external field h along z -direction is given by:

$$\hat{H} = J \sum_{\langle i,j \rangle} \left[\frac{1}{2} \left(\hat{S}_i^+ \hat{S}_j^- + \hat{S}_j^+ \hat{S}_i^- \right) + \Delta \hat{S}_i^z \hat{S}_j^z \right] + D \sum_i \left(\hat{S}_i^z \right)^2 - h \sum_i \hat{S}_i^z \quad (2.11)$$

where $\langle i, j \rangle$ indicates nearest neighbours. The ground state of the antiferromagnetic phase breaks the translational symmetry implying two inequivalent sublattices. We can apply a sub-lattice rotation of π along $\hat{S}^z$ for lattice site B such that $\hat{S}_j^\pm \rightarrow -\hat{S}_j^\pm$ for all $j \in B$. The Hamiltonian becomes:

$$\hat{H} = J \sum_{\langle i,j \rangle} \left[-\frac{1}{2} \left(\hat{S}_i^+ \hat{S}_j^- + \hat{S}_j^+ \hat{S}_i^- \right) + \Delta \hat{S}_i^z \hat{S}_j^z \right] + D \sum_i \left(\hat{S}_i^z \right)^2 - h \sum_i \hat{S}_i^z. \quad (2.12)$$

The rotated antiferromagnetic ground state becomes uniform over lattice. The QPM ground state is invariant upon this sublattice rotation. The ground state and excitation energy are invariant as well. Now we introduce three flavors of bosons on each local Hilbert space such that:

$$\hat{b}_{i0}^\dagger |\emptyset\rangle = |0\rangle_i, \quad \hat{b}_{i1}^\dagger |\emptyset\rangle = |1\rangle_i, \quad \text{and} \quad \hat{b}_{i2}^\dagger |\emptyset\rangle = |-1\rangle_i, \quad (2.13)$$

where $|0\rangle_i$, $|1\rangle_i$ and $|-1\rangle_i$ are eigenstates of $\hat{S}_i^z$ at lattice site i . We can define a vector of bosonic operators as:

$$b_i = (\hat{b}_{i0}, \hat{b}_{i1}, \hat{b}_{i2})^T \quad \text{and} \quad b_i^\dagger = (\hat{b}_{i0}^\dagger, \hat{b}_{i1}^\dagger, \hat{b}_{i2}^\dagger). \quad (2.14)$$

The Hamiltonian can then be expressed as:

$$\begin{aligned} \hat{H} = & J \sum_{\langle i,j \rangle} \left[-\frac{1}{2} \left(b_i^\dagger S^+ b_j b_j^\dagger S^- b_j + b_j^\dagger S^+ b_j b_i^\dagger S^- b_i \right) + \Delta b_i^\dagger S^z b_i b_j^\dagger S^z b_j \right] \\ & + D \sum_i (1 - b_i^\dagger A b_i) - h \sum_i b_i^\dagger S^z b_i. \end{aligned} \quad (2.15)$$

where the matrices are given by*:

$$S^- = \sqrt{2} \begin{pmatrix} 0 & 1 & 0 \\ 0 & 0 & 0 \\ 1 & 0 & 0 \end{pmatrix}, \quad S^+ = \sqrt{2} \begin{pmatrix} 0 & 0 & 1 \\ 1 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \quad S^z = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & -1 \end{pmatrix}, \quad A = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}. \quad (2.16)$$

We have used the holonomic constraint $\hat{b}_{i0}^\dagger \hat{b}_{i0} = 1 - \hat{b}_{i1}^\dagger \hat{b}_{i1} - \hat{b}_{i2}^\dagger \hat{b}_{i2}$ in the single-ion anisotropy term.

In this approach, the ground state $|gs\rangle$ consists of condensation of one of the bosons:

$$|gs\rangle = \prod_i \hat{a}_{i0}^\dagger |\emptyset\rangle. \quad (2.17)$$

$\hat{a}_{i0}^\dagger$ along with the other two orthogonal modes $\hat{a}_{i1}^\dagger, \hat{a}_{i2}^\dagger$ can be obtained by applying a global unitary transformation $\mathcal{U}$ to the original vector of bosons:

$$\alpha_i = \mathcal{U} b_i \quad \text{and} \quad \alpha_i^\dagger = b_i^\dagger \mathcal{U}^\dagger, \quad (2.18)$$

where α_i and $\alpha_i^\dagger$ are the transformed vectors of bosons:

$$\alpha_i = (\hat{a}_{i0}, \hat{a}_{i1}, \hat{a}_{i2})^T \quad \text{and} \quad \alpha_i^\dagger = (\hat{a}_{i0}^\dagger, \hat{a}_{i1}^\dagger, \hat{a}_{i2}^\dagger). \quad (2.19)$$

This corresponds to choosing a quantization axis along the direction of the order parameter. The Hamiltonian in terms of the new Schwinger bosons is given by:

*We have set $\hbar = 1$ for simplicity.

$$\begin{aligned} \hat{H} = & J \sum_{\langle i,j \rangle} \left[-\frac{1}{2} \left(\alpha_i^\dagger \tilde{S}^+ \alpha_i \alpha_j^\dagger \tilde{S}^- \alpha_j + \alpha_j^\dagger \tilde{S}^+ \alpha_j \alpha_i^\dagger \tilde{S}^- \alpha_i \right) + \Delta \alpha_i^\dagger \tilde{S}^z \alpha_i \alpha_j^\dagger \tilde{S}^z \alpha_j \right] \\ & + D \sum_i (1 - \alpha_i^\dagger \tilde{A} \alpha_i) - h \sum_i \alpha_i^\dagger \tilde{S}^z \alpha_i. \end{aligned} \quad (2.20)$$

$\tilde{S}^\mu$ and $\tilde{A}$ are the matrices in equation (2.16) after unitary transformation:

$$\tilde{S}^\mu = \mathcal{U} S^\mu \mathcal{U}^\dagger \quad \text{and} \quad \tilde{A} = \mathcal{U} A \mathcal{U}^\dagger. \quad (2.21)$$

The symmetry properties between the original operators are preserved:

$$\tilde{S}^z = (\tilde{S}^z)^\dagger, \quad \tilde{S}^+ = (\tilde{S}^-)^\dagger \quad \text{and} \quad \tilde{A} = \tilde{A}^\dagger. \quad (2.22)$$

Since we have assumed that the local Hilbert space is condensate in $\hat{a}_{i0}^\dagger |\emptyset\rangle$, we can apply Holstein-Primakoff approximation to the holonomic local constraint such that:

$$\hat{a}_{i0}^\dagger \approx \hat{a}_{i0} \approx \sqrt{1 - \hat{a}_{i1}^\dagger \hat{a}_{i1} - \hat{a}_{i2}^\dagger \hat{a}_{i2}} \approx 1 - \frac{1}{2}(\hat{a}_{i1}^\dagger \hat{a}_{i1} + \hat{a}_{i2}^\dagger \hat{a}_{i2}). \quad (2.23)$$

The spin operators $\alpha_i^\dagger \tilde{S}^\mu \alpha_i$ can be approximated up to bilinear terms as:

$$\alpha_i^\dagger \tilde{S}^\mu \alpha_i \approx \tilde{S}_{00}^\mu + (\tilde{S}_{m0}^\mu \hat{a}_{im}^\dagger + \tilde{S}_{0m}^\mu \hat{a}_{im}) + (\tilde{S}_{mn}^\mu - \tilde{S}_{00}^\mu \delta_{mn}) \hat{a}_{im}^\dagger \hat{a}_{in}, \quad (2.24)$$

where $m, n \in \{1, 2\}$. Substitute this into the Hamiltonian, after some algebra, and we can obtain an approximate Hamiltonian up to bilinear terms:

$$\hat{H} \approx H_0 + \hat{H}_1 + \hat{H}_2 \quad (2.25)$$

where

$$H_0 = N \left[-z J \tilde{S}_{00}^+ \tilde{S}_{00}^- + (z J \Delta \tilde{S}_{00}^z - h) \tilde{S}_{00}^z + D(1 - \tilde{A}_{00}) \right], \quad (2.26)$$

$$\begin{aligned}
\hat{H}_1 &= \left[-zJ(\tilde{S}_{00}^+ \tilde{S}_{m0}^- + \tilde{S}_{00}^- \tilde{S}_{m0}^+) + (2zJ\Delta\tilde{S}_{00}^z - h)\tilde{S}_{m0}^z - D\tilde{A}_{m0} \right] \sum_i \hat{a}_{im}^\dagger \\
&\quad + \left[-zJ(\tilde{S}_{00}^+ \tilde{S}_{0m}^- + \tilde{S}_{00}^- \tilde{S}_{0m}^+) + (2zJ\Delta\tilde{S}_{00}^z - h)\tilde{S}_{0m}^z - D\tilde{A}_{0m} \right] \sum_i \hat{a}_{im}, \quad (2.27) \\
\hat{H}_2 &= \lambda_{mn} \sum_i \hat{a}_{im}^\dagger \hat{a}_{in} + \sum_{\langle i,j \rangle} (t_{mn} \hat{a}_{im}^\dagger \hat{a}_{jn} + t_{mn}^* \hat{a}_{im} \hat{a}_{jn}^\dagger) + \sum_{\langle i,j \rangle} (\Delta_{mn} \hat{a}_{im}^\dagger \hat{a}_{jn}^\dagger + \Delta_{mn}^* \hat{a}_{im} \hat{a}_{jn}). \quad (2.28)
\end{aligned}$$

z is the dimension of the system and N is the number of lattice sites. $\tilde{S}_{mn}^\mu$ and $\tilde{A}_{mn}$ are matrices elements. Einstein summation convention has been used here. m and n are summed over 1 and 2. The matrix elements in $\hat{H}_2$ are given by

$$\begin{aligned}
\lambda_{mn} &= -zJ \left[\tilde{S}_{00}^+ (\tilde{S}_{mn}^- - \tilde{S}_{00}^- \delta_{mn}) + \tilde{S}_{00}^- (\tilde{S}_{mn}^+ - \tilde{S}_{00}^+ \delta_{mn}) \right] \\
&\quad + (2zJ\Delta\tilde{S}_{00}^z - h)(\tilde{S}_{mn}^z - \tilde{S}_{00}^z \delta_{mn}) - D(\tilde{A}_{mn} - \tilde{A}_{00} \delta_{mn}) \quad (2.29)
\end{aligned}$$

$$t_{mn} = -\frac{J}{2}(\tilde{S}_{m0}^+ \tilde{S}_{0n}^- + \tilde{S}_{m0}^- \tilde{S}_{0n}^+) + J\Delta\tilde{S}_{m0}^z \tilde{S}_{0n}^z \quad (2.30)$$

$$\Delta_{mn} = -\frac{J}{2}(\tilde{S}_{m0}^+ \tilde{S}_{n0}^- + \tilde{S}_{m0}^- \tilde{S}_{n0}^+) + J\Delta\tilde{S}_{m0}^z \tilde{S}_{n0}^z. \quad (2.31)$$

The symmetry properties are obvious:

$$\lambda_{mn} = \lambda_{nm}^*, \quad t_{mn} = t_{nm}^* \quad \text{and} \quad \Delta_{mn} = \Delta_{nm}. \quad (2.32)$$

H_0 is the classical ground state energy. The parameters in the unitary transformation $\mathcal{U}$ can be obtained by minimizing the ground state energy. $\hat{H}_1$ is linear in the bosonic operators which vanishes at the saddle point. The bilinear term $\hat{H}_2$ is the spin wave Hamiltonian. We investigated its different behavior for different ground states.

It is convenient to study the spin wave Hamiltonian in the momentum space. The Fourier transform of the bosonic operators are given by:

$$\hat{a}_{\mathbf{k}m} = \frac{1}{\sqrt{N}} \sum_i e^{-i\mathbf{k} \cdot \mathbf{r}_i} \hat{a}_{im} \quad \text{and} \quad \hat{a}_{\mathbf{k}m}^\dagger = \frac{1}{\sqrt{N}} \sum_i e^{i\mathbf{k} \cdot \mathbf{r}_i} \hat{a}_{im}^\dagger, \quad (2.33)$$

where the Fourier components obey the usual bosonic commutation relation:

$$[\hat{a}_{\mathbf{k}m}, \hat{a}_{\mathbf{k}'n}^\dagger] = \delta_{\mathbf{k}\mathbf{k}'} \delta_{mn}. \quad (2.34)$$

The inverse Fourier transform is given by:

$$\hat{a}_{im} = \frac{1}{\sqrt{N}} \sum_{\mathbf{k}} e^{i\mathbf{k} \cdot \mathbf{r}_i} \hat{a}_{\mathbf{k}m} \quad \text{and} \quad \hat{a}_{im}^\dagger = \frac{1}{\sqrt{N}} \sum_i e^{-i\mathbf{k} \cdot \mathbf{r}_i} \hat{a}_{\mathbf{k}m}^\dagger. \quad (2.35)$$

After a straightforward substitution into the Hamiltonian, equation (2.28), we arrive at:

$$\mathcal{H} = \lambda_{mn} \sum_{\mathbf{k}} \hat{a}_{\mathbf{k}m}^\dagger \hat{a}_{\mathbf{k}n} + \frac{1}{2} \sum_{\mathbf{k}} f(\mathbf{k}) \left(t_{mn} \hat{a}_{\mathbf{k}m}^\dagger \hat{a}_{\mathbf{k}n} + t_{mn}^* \hat{a}_{-\mathbf{k}m} \hat{a}_{-\mathbf{k}n}^\dagger + \Delta_{mn} \hat{a}_{\mathbf{k}m}^\dagger \hat{a}_{-\mathbf{k}n}^\dagger + \Delta_{mn}^* \hat{a}_{-\mathbf{k}m} \hat{a}_{\mathbf{k}n} \right) \quad (2.36)$$

where

$$f(\mathbf{k}) = \sum_{\mathbf{w}} e^{i\mathbf{k} \cdot \mathbf{w}}. \quad (2.37)$$

$\mathbf{w}$ are vectors connecting two nearest neighbours. In a regular bipartite lattice, for example, in a square or cubic lattice which is what we considered, there are $2z$ numbers of $\mathbf{w}$ and the summation over all $\mathbf{w}$ is real. That is

$$f(\mathbf{k}) = \sum_{\mathbf{w}} \cos(\mathbf{k} \cdot \mathbf{w}). \quad (2.38)$$

Using the symmetry property of t_{mn} as shown in equation (2.32) and the fact that the domain of $\mathbf{k}$ is symmetric about $\mathbf{k} = \mathbf{0}$, we have:

$$\mathcal{H} = \frac{1}{2} \sum_{\mathbf{k}} \left(\varepsilon_{mn} \hat{a}_{\mathbf{k}m}^\dagger \hat{a}_{\mathbf{k}n} + \varepsilon_{mn}^* \hat{a}_{-\mathbf{k}m} \hat{a}_{-\mathbf{k}n}^\dagger + \gamma_{mn} \hat{a}_{\mathbf{k}m}^\dagger \hat{a}_{-\mathbf{k}n}^\dagger + \gamma_{mn}^* \hat{a}_{-\mathbf{k}m} \hat{a}_{\mathbf{k}n} \right) - \frac{1}{2} \sum_{\mathbf{k}} \lambda_{mm}. \quad (2.39)$$

The last term comes from the commutation relation of the bosonic operators. The matrices elements ε_{mn} and γ_{mn} are given by:

$$\varepsilon_{mn} = \lambda_{mn} + f(\mathbf{k}) t_{mn} \quad \text{and} \quad \gamma_{mn} = f(\mathbf{k}) \Delta_{mn}. \quad (2.40)$$

From equation (2.32), we can see that $\varepsilon_{mn} = \varepsilon_{nm}^*$ and $\gamma_{mn} = \gamma_{nm}$.

As before, we define a vector of bosonic operators:

$$\alpha_{\mathbf{k}} = (\hat{a}_{\mathbf{k}1}, \hat{a}_{\mathbf{k}2}, \hat{a}_{-\mathbf{k}1}^\dagger, \hat{a}_{-\mathbf{k}2}^\dagger)^T \quad \text{and} \quad \alpha_{\mathbf{k}}^\dagger = (\hat{a}_{\mathbf{k}1}^\dagger, \hat{a}_{\mathbf{k}2}^\dagger, \hat{a}_{-\mathbf{k}1}, \hat{a}_{-\mathbf{k}2}). \quad (2.41)$$

Define two 2×2 matrices $\mathcal{E}_{\mathbf{k}}$ and $\Gamma_{\mathbf{k}}$ such that:

$$(\mathcal{E}_{\mathbf{k}})_{mn} = \varepsilon_{mn} \quad \text{and} \quad (\Gamma_{\mathbf{k}})_{mn} = \gamma_{mn}. \quad (2.42)$$

Notice that $\mathcal{E}_{\mathbf{k}}$ is Hermitian and $\Gamma_{\mathbf{k}}$ is symmetric. The spin wave Hamiltonian can then be simplified as:

$$\mathcal{H} = \frac{1}{2} \sum_{\mathbf{k}} \alpha_{\mathbf{k}}^\dagger \mathcal{D}_{\mathbf{k}} \alpha_{\mathbf{k}} - \frac{1}{2} \sum_{\mathbf{k}} \lambda_{mm} \quad (2.43)$$

where the *grand dynamical matrix* $\mathcal{D}_{\mathbf{k}}$ is a 4×4 matrix:

$$\mathcal{D}_{\mathbf{k}} = \begin{pmatrix} \mathcal{E}_{\mathbf{k}} & \Gamma_{\mathbf{k}} \\ \Gamma_{\mathbf{k}}^* & \mathcal{E}_{\mathbf{k}}^* \end{pmatrix}. \quad (2.44)$$

The Hermiticity of $\mathcal{D}_{\mathbf{k}}$ is guaranteed by the properties of $\mathcal{E}_{\mathbf{k}}$ and $\Gamma_{\mathbf{k}}$. After Bogoliubov transformation, $\mathcal{H}$ assumes a diagonal form as:

$$\mathcal{H} = \sum_{\mathbf{k}} \omega_{\mathbf{k}m} \left(\hat{c}_{\mathbf{k}m}^\dagger \hat{c}_{\mathbf{k}m} + \frac{1}{2} \right) - \frac{1}{2} \sum_{\mathbf{k}} \lambda_{mm} \quad (2.45)$$

where the elementary exciations are given by:

$$\mathcal{C}_{\mathbf{k}} = P \alpha_{\mathbf{k}} \quad \text{and} \quad \mathcal{C}_{\mathbf{k}}^\dagger = \alpha_{\mathbf{k}}^\dagger P^\dagger \quad (2.46)$$

with $\mathcal{C}_{\mathbf{k}}(\mathcal{C}_{\mathbf{k}}^\dagger)$ defined as usual:

$$\mathcal{C}_{\mathbf{k}} = (\hat{c}_{\mathbf{k}1}, \hat{c}_{\mathbf{k}2}, \hat{c}_{-\mathbf{k}1}^\dagger, \hat{c}_{-\mathbf{k}2}^\dagger)^T \quad \text{and} \quad \mathcal{C}_{\mathbf{k}}^\dagger = (\hat{c}_{\mathbf{k}1}^\dagger, \hat{c}_{\mathbf{k}2}^\dagger, \hat{c}_{-\mathbf{k}1}, \hat{c}_{-\mathbf{k}2}). \quad (2.47)$$

P is a para-unitary matrix such that:

$$P^\dagger \hat{\delta} P = \hat{\delta} \quad \text{with} \quad \hat{\delta} = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & -1 & 0 \\ 0 & 0 & 0 & -1 \end{pmatrix}. \quad (2.48)$$

And $P^{-1} = \hat{\delta} P^\dagger \hat{\delta}$ diagonalizes $\mathcal{D}_{\mathbf{k}}$ as:

$$(P^{-1})^\dagger \mathcal{D}_{\mathbf{k}} P^{-1} = \begin{pmatrix} \omega_{\mathbf{k}1} & 0 & 0 & 0 \\ 0 & \omega_{\mathbf{k}2} & 0 & 0 \\ 0 & 0 & \omega_{\mathbf{k}1} & 0 \\ 0 & 0 & 0 & \omega_{\mathbf{k}2} \end{pmatrix}. \quad (2.49)$$

In the case where $\mathcal{D}_{\mathbf{k}}$ is real, this procedure can be simplified and $\omega_{\mathbf{k}m}^2$ are the eigenvalues of

$$\Omega_{\mathbf{k}} = (\mathcal{E}_{\mathbf{k}} \mp \Gamma_{\mathbf{k}}) (\mathcal{E}_{\mathbf{k}} \pm \Gamma_{\mathbf{k}}). \quad (2.50)$$

Bogoliubov transformation of a general bilinear bosonic Hamiltonian is shown in Section 2.6.

2.3.2 Linear Holstein-Primakoff approximation of two sub-lattices

In this subsection, we consider the Holstein-Primakoff approximation in two sub-lattices. The procedure is a generalization of the method described in the previous section and is applicable to spatially modulated ground state phases. Now we have two sets of matrices in equation (2.16) labeled by A and B . And the unitary transformations on different sub-lattices are generally different. After some algebra, the Hamiltonian can be approximated up to bilinear terms in the bosonic operators, $\hat{a}_{im}^\dagger (\hat{a}_{im})$ and $\hat{b}_{jm}^\dagger (\hat{b}_{jm})$ for $i \in A$ and $j \in B$, respectively:

$$H \approx H_0 + \mathcal{H}, \quad (2.51)$$

with the ground state energy[†]:

$$H_0 = N \left\{ z \left[\frac{1}{2} (S_{00}^{A+} S_{00}^{B-} + S_{00}^{A-} S_{00}^{B+}) + \Delta S_{00}^{Az} S_{00}^{Bz} \right] + \left[D - \frac{1}{2} D (A_{00} + B_{00}) - \frac{1}{2} h (S_{00}^{Az} + S_{00}^{Bz}) \right] \right\} \quad (2.52)$$

and the spin wave Hamiltonian:

$$\mathcal{H} = \sum_{i \in A} \lambda_{mn}^A \hat{a}_{im}^\dagger \hat{a}_{in} + \sum_{j \in B} \lambda_{mn}^B \hat{b}_{jm}^\dagger \hat{b}_{jn} + \sum_{\langle i, j \rangle} \left(t_{mn} \hat{a}_{im}^\dagger \hat{b}_{jn} + t_{mn}^* \hat{a}_{im} \hat{b}_{jn}^\dagger + w_{mn} \hat{a}_{im}^\dagger \hat{b}_{jn}^\dagger + w_{mn}^* \hat{a}_{im} \hat{b}_{jn} \right) \quad (2.53)$$

where the matrices λ^A , λ^B , T and W are given by:

$$\begin{aligned} \lambda_{mn}^A = & z \left[S_{00}^{B-} (S_{mn}^{A+} - S_{00}^{A+} \delta_{mn}) + S_{00}^{B+} (S_{mn}^{A-} - S_{00}^{A-} \delta_{mn}) \right] \\ & + (S_{mn}^{Az} - S_{00}^{Az} \delta_{mn}) (2\Delta z J S_{00}^{Bz} - h) - D (A_{mn} - A_{00} \delta_{mn}), \end{aligned} \quad (2.54)$$

$$\begin{aligned} \lambda_{mn}^B = & z \left[S_{00}^{A-} (S_{mn}^{B+} - S_{00}^{B+} \delta_{mn}) + S_{00}^{A+} (S_{mn}^{B-} - S_{00}^{B-} \delta_{mn}) \right] \\ & + (S_{mn}^{Bz} - S_{00}^{Bz} \delta_{mn}) (2\Delta z J S_{00}^{Az} - h) - D (B_{mn} - B_{00} \delta_{mn}), \end{aligned} \quad (2.55)$$

$$t_{mn} = \frac{1}{2} (S_{m0}^{A+} S_{0n}^{B-} + S_{m0}^{A-} S_{0n}^{B+}) + \Delta S_{m0}^{Az} S_{0n}^{Bz}, \quad (2.56)$$

$$w_{mn} = \frac{1}{2} (S_{m0}^{A+} S_{n0}^{B-} + S_{m0}^{A-} S_{n0}^{B+}) + \Delta S_{m0}^{Az} S_{n0}^{Bz}. \quad (2.57)$$

After Fourier transform, the Hamiltonian can be simplified into a matrix form as:

$$\mathcal{H} = \frac{1}{2} \sum_{\mathbf{k}} u_{\mathbf{k}}^\dagger \mathcal{D}_{\mathbf{k}} u_{\mathbf{k}} - \frac{1}{2} \sum_{\mathbf{k}m} (\lambda_{mm}^A + \lambda_{mm}^B), \quad (2.58)$$

where as before, the vectors are defined as:

$$u_{\mathbf{k}} = \begin{pmatrix} \hat{a}_{\mathbf{k}1} & \hat{a}_{\mathbf{k}2} & \hat{b}_{\mathbf{k}1} & \hat{b}_{\mathbf{k}2} & \hat{a}_{-\mathbf{k}1}^\dagger & \hat{a}_{-\mathbf{k}2}^\dagger & \hat{b}_{-\mathbf{k}1}^\dagger & \hat{b}_{-\mathbf{k}2}^\dagger \end{pmatrix}, \quad (2.59)$$

[†] J has been set to 1.

and the grand dynamical matrix is given by:

$$\mathcal{D}_{\mathbf{k}} = \begin{pmatrix} \lambda^A & f(\mathbf{k})T & 0 & f(\mathbf{k})W \\ f(\mathbf{k})T^\dagger & \lambda^B & f(\mathbf{k})W^\dagger & 0 \\ 0 & f(\mathbf{k})W^* & (\lambda^A)^T & f(\mathbf{k})T^* \\ f(\mathbf{k})W^\dagger & 0 & f(\mathbf{k})T^T & (\lambda^B)^T \end{pmatrix}. \quad (2.60)$$

Similar to last subsection, applying the Bogoliubov transformation, we can obtain the dispersion relations.

2.4 Spin wave theory with Lagrange multiplier

In this section, we demonstrate the second method to implement the hardcore constraint, viz., the Lagrange multiplier (LM) method. Unlike the HP method where the unitary transformation carries a variational parameter, LM method ususally is more efficient the exact classical ground state is known. That is to say we are not able to determine the ground state phases through minimization as in HP method. However, if we could guess the classical ground state correctly, LM method yields a more accurate dispersion relations and more accurate estimates of energy gaps. This advantage allows us to obtain a more accurate phase boundary by investigating at the stability of the dispersions. The comparison of these two methods is shown in Chapter 4.

As usual, we introduce two types of bosons for the different unitary transformations:

$$[\hat{a}_{im}, \hat{a}_{i'n'}^\dagger] = \delta_{ii'}\delta_{mm'} \quad \text{for } i, i' \in A; \quad (2.61)$$

$$[\hat{b}_{jm}, \hat{b}_{j'm'}^\dagger] = \delta_{jj'}\delta_{mm'} \quad \text{for } j, j' \in B. \quad (2.62)$$

That is the above bosonic operators are already in the transformed basis. The grand

Hamiltonian can be written in bosonic language with Lagrange multiplier μ_i as:

$$\tilde{\mathcal{H}} = \sum_{\langle i,j \rangle} \left[\frac{1}{2} \left(S_{mn}^{A+} S_{m'n'}^{B-} \hat{a}_{im}^\dagger \hat{a}_{in} \hat{b}_{jm}^\dagger \hat{b}_{jn'} + S_{mn}^{A-} S_{m'n'}^{B+} \hat{a}_{im}^\dagger \hat{a}_{in} \hat{b}_{jm}^\dagger \hat{b}_{jn'} \right) + \Delta S_{mn}^{Az} S_{m'n'}^{Bz} \hat{a}_{im}^\dagger \hat{a}_{in} \hat{b}_{jm}^\dagger \hat{b}_{jn'} \right] \quad (2.63)$$

$$+ \sum_{i \in A} A_{mn} \hat{a}_{im}^\dagger \hat{a}_{in} + \sum_{j \in B} B_{mn} \hat{b}_{jm}^\dagger \hat{b}_{jn} - h \sum_{i \in A} S_{mn}^{Az} \hat{a}_{im}^\dagger \hat{a}_{in} - h \sum_{j \in B} S_{mn}^{Bz} \hat{b}_{jm}^\dagger \hat{b}_{jn} \quad (2.64)$$

$$- \sum_{i \in A} \mu_i (1 - \hat{a}_{im}^\dagger \hat{a}_{im}) - \sum_{j \in B} \mu_j (1 - \hat{b}_{jm}^\dagger \hat{b}_{jm}). \quad (2.65)$$

For simplicity, we have set the exchange parameter $J = 1$. The matrices A and B characterize the single-ion anisotropy:

$$D(\hat{S}_i^z)^2 = \begin{cases} A_{mn} \hat{a}_{im}^\dagger \hat{a}_{in} & \text{for } i \in A, \\ B_{mn} \hat{b}_{jm}^\dagger \hat{b}_{jn} & \text{for } i \in B. \end{cases} \quad (2.66)$$

At the mean field level, we postulate the local states are condensed in $\hat{a}_{i0}^\dagger$ and $\hat{b}_{j0}^\dagger$ in lattice sites A and B , respectively. The operators can then be approximated with the corresponding condensate fraction:

$$\hat{a}_{i0}^\dagger \approx \hat{a}_{i0} \approx \langle \hat{a}_{i0}^\dagger \rangle \approx \langle \hat{a}_{i0} \rangle \approx a \quad \text{and} \quad \hat{b}_{j0}^\dagger \approx \hat{b}_{j0} \approx \langle \hat{b}_{j0}^\dagger \rangle \approx \langle \hat{b}_{j0} \rangle \approx b \quad (2.67)$$

where a and b are two complex numbers that can be determined from the saddle point conditions. Further more, we assume the chemical potentials or Lagrange multipliers are the same for the same sub-lattice sites. Hence, after some algebra, the Hamiltonian can be expressed as:

$$\begin{aligned} \tilde{\mathcal{H}} = E_0 + \sum_{\langle i,j \rangle} & \left(T_{mn} \hat{a}_{im}^\dagger \hat{b}_{jn} + T_{mn}^* \hat{a}_{im} \hat{b}_{jn}^\dagger + W_{mn} \hat{a}_{im}^\dagger \hat{b}_{jn}^\dagger + W_{mn}^* \hat{a}_{im} \hat{b}_{jn} \right) \\ & + \sum_{i \in A} \lambda_{mn}^A \hat{a}_{im}^\dagger \hat{a}_{in} + \sum_{j \in B} \lambda_{mn}^B \hat{b}_{jm}^\dagger \hat{b}_{jn} \end{aligned} \quad (2.68)$$

where the classical ground state energy E_0 is:

$$E_0 = a^2 b^2 z N \left[\frac{1}{2} (S_{00}^{A+} S_{00}^{B-} + S_{00}^{A-} S_{00}^{B+}) + \Delta S_{00}^{Az} S_{00}^{Bz} \right] + \frac{N}{2} [a^2 A_{00} - a^2 h S_{00}^{Az} - \mu_A (1 - a^2)] + \frac{N}{2} [b^2 B_{00} - b^2 h S_{00}^{Bz} - \mu_B (1 - b^2)]. \quad (2.69)$$

The matrices λ^A , λ^B , T and W are given by:

$$\lambda_{mn}^A = b^2 [z (S_{mn}^{A+} S_{00}^{B-} + S_{mn}^{A-} S_{00}^{B+}) + 2\Delta z S_{mn}^{Az} S_{00}^{Bz}] + A_{mn} - h S_{mn}^{Az} + \mu_A \delta_{mn}, \quad (2.70)$$

$$\lambda_{mn}^B = a^2 [z (S_{mn}^{B+} S_{00}^{A-} + S_{mn}^{B-} S_{00}^{A+}) + 2\Delta z S_{mn}^{Bz} S_{00}^{Az}] + B_{mn} - h S_{mn}^{Bz} + \mu_B \delta_{mn}, \quad (2.71)$$

$$T_{mn} = ab \left[\frac{1}{2} (S_{m0}^{A+} S_{0n}^{B-} + S_{m0}^{A-} S_{0n}^{B+}) + \Delta S_{m0}^{Az} S_{0n}^{Bz} \right], \quad (2.72)$$

$$W_{mn} = ab \left[\frac{1}{2} (S_{m0}^{A+} S_{n0}^{B-} + S_{m0}^{A-} S_{n0}^{B+}) + \Delta S_{m0}^{Az} S_{n0}^{Bz} \right]. \quad (2.73)$$

Notice that λ^A and λ^B are hermitian. As usual, the Fourier counterparts of the bosonic operators are defined as:

$$\hat{a}_{\mathbf{k}m}^\dagger = \sqrt{\frac{2}{N}} \sum_{i \in A} \hat{a}_{im}^\dagger e^{i\mathbf{k} \cdot \mathbf{r}_i} \quad \text{and} \quad \hat{a}_{\mathbf{k}} = \sqrt{\frac{2}{N}} \sum_{i \in A} \hat{a}_{im} e^{-i\mathbf{k} \cdot \mathbf{r}_i}. \quad (2.74)$$

$$\hat{b}_{\mathbf{k}m}^\dagger = \sqrt{\frac{2}{N}} \sum_{j \in B} \hat{b}_{jm}^\dagger e^{i\mathbf{k} \cdot \mathbf{r}_j} \quad \text{and} \quad \hat{b}_{\mathbf{k}} = \sqrt{\frac{2}{N}} \sum_{j \in B} \hat{b}_{jm} e^{-i\mathbf{k} \cdot \mathbf{r}_j}. \quad (2.75)$$

It should be emphasized that $\mathbf{k}$ only takes values within half of the first Brillouin zone.

The Hamiltonian in the Fourier space is then given by:

$$\begin{aligned} \tilde{\mathcal{H}} = E_0 + \sum_{\mathbf{k}} \lambda_{mn}^A \hat{a}_{\mathbf{k}m}^\dagger \hat{a}_{\mathbf{k}n} + \sum_{\mathbf{k}} \lambda_{mn}^B \hat{b}_{\mathbf{k}m}^\dagger \hat{b}_{\mathbf{k}n} \\ + \sum_{\mathbf{k}} f(\mathbf{k}) \left(T_{mn} \hat{a}_{\mathbf{k}m}^\dagger \hat{b}_{\mathbf{k}n} + T_{mn}^* \hat{a}_{\mathbf{k}m} \hat{b}_{\mathbf{k}n}^\dagger + W_{mn} \hat{a}_{\mathbf{k}m}^\dagger \hat{b}_{-\mathbf{k}n}^\dagger + W_{mn}^* \hat{a}_{-\mathbf{k}m} \hat{b}_{\mathbf{k}n} \right). \end{aligned} \quad (2.76)$$

Here $f(\mathbf{k})$ has the same meaning as before except a factor of $\frac{1}{2}$:

$$f(\mathbf{k}) = \sum_{\mathbf{w}} e^{i\mathbf{k} \cdot \mathbf{w}}. \quad (2.77)$$

Define the vector of bosonic operators as:

$$u_{\mathbf{k}} = \begin{pmatrix} \hat{a}_{\mathbf{k}1} & \hat{a}_{\mathbf{k}2} & \hat{b}_{\mathbf{k}1} & \hat{b}_{\mathbf{k}2} & \hat{a}_{-\mathbf{k}1}^\dagger & \hat{a}_{-\mathbf{k}2}^\dagger & \hat{b}_{-\mathbf{k}1}^\dagger & \hat{b}_{-\mathbf{k}2}^\dagger \end{pmatrix}^T, \quad (2.78)$$

$$u_{\mathbf{k}}^\dagger = \begin{pmatrix} \hat{a}_{\mathbf{k}1}^\dagger & \hat{a}_{\mathbf{k}2}^\dagger & \hat{b}_{\mathbf{k}1}^\dagger & \hat{b}_{\mathbf{k}2}^\dagger & \hat{a}_{-\mathbf{k}1} & \hat{a}_{-\mathbf{k}2} & \hat{b}_{-\mathbf{k}1} & \hat{b}_{-\mathbf{k}2} \end{pmatrix}. \quad (2.79)$$

The Hamiltonian can be expressed in a matrix form as:

$$\tilde{\mathcal{H}} = E_0 + \frac{1}{2} \sum_{\mathbf{k}} u_{\mathbf{k}}^\dagger \mathcal{D}_{\mathbf{k}} u_{\mathbf{k}} - \frac{1}{2} \sum_{\mathbf{k}} (\lambda_{mm}^A + \lambda_{mm}^B) \quad (2.80)$$

where the grand dynamical matrix is:

$$\mathcal{D}_{\mathbf{k}} = \begin{pmatrix} \mathcal{E}_{\mathbf{k}} & \Gamma_{\mathbf{k}} \\ \Gamma_{\mathbf{k}}^* & \mathcal{E}_{\mathbf{k}}^* \end{pmatrix} \quad \text{with} \quad \mathcal{E}_{\mathbf{k}} = \begin{pmatrix} \lambda^A & T f(\mathbf{k}) \\ T^\dagger f(\mathbf{k}) & \lambda^B \end{pmatrix}, \quad \Gamma_{\mathbf{k}} = \begin{pmatrix} 0 & W f(\mathbf{k}) \\ W^\dagger f(\mathbf{k}) & 0 \end{pmatrix}. \quad (2.81)$$

This can be diagonalized through Bogoliubov transformation:

$$\tilde{\mathcal{H}} = E_0 + \sum_{\mathbf{k}m} \left(\varepsilon_{\mathbf{k}m}^{(1)} \hat{\alpha}_{\mathbf{k}m}^\dagger \hat{\alpha}_{\mathbf{k}m} + \varepsilon_{\mathbf{k}m}^{(2)} \hat{\beta}_{\mathbf{k}m}^\dagger \hat{\beta}_{\mathbf{k}m} \right) + \frac{1}{2} \sum_{\mathbf{k}m} \left(\varepsilon_{\mathbf{k}m}^{(1)} + \varepsilon_{\mathbf{k}m}^{(2)} - \lambda_{mm}^A - \lambda_{mm}^B \right). \quad (2.82)$$

Minimization of the grand Hamiltonian with respect to the introduced parameters gives the saddle point equations that determine the values of the parameters.

$$\left\langle \frac{\partial \tilde{\mathcal{H}}}{\partial a} \right\rangle = \left\langle \frac{\partial \tilde{\mathcal{H}}}{\partial b} \right\rangle = \left\langle \frac{\partial \tilde{\mathcal{H}}}{\partial \mu_A} \right\rangle = \left\langle \frac{\partial \tilde{\mathcal{H}}}{\partial \mu_B} \right\rangle = 0. \quad (2.83)$$

Solving the above equations gives the optimized parameter sets $\{a, b, \mu_A, \mu_B\}$. Finite values of the condensate fractions a and b imply the robustness of the classical ground state.

2.5 Generalization of spin wave theory

We have demonstrated different versions of spin wave theory in the last few sections. They differ in the implementation of the hardcore boson constraint. However, they are similar

in one aspect. They all work in the minimal local Hilbert space, viz., a single spin. In this case, we have the most classical ground state assumption with the strongest quantum fluctuations. Even in the two-sublattice framework, the entanglement within the unit cell, consisting of two spins, was not considered either, which is essentially the same as a single spin unit cell framework. This is not very problematic for systems with bipartite lattice. However, in frustrated systems, this method easily fails completely because of the strong quantum fluctuation. Therefore, in this section we present a generalized spin wave theory where we take the unit cell containing more than one spins as a basic entity.

We consider a general d -dimensional lattice with Hamiltonian

$$\hat{H} = \sum_{i,j} J_{ij}^{\alpha} \hat{S}_i^{\alpha} \hat{S}_j^{\alpha} \quad (2.84)$$

where $\hat{S}_i^{\alpha}$ are spin- S operators. Suppose we can decompose the lattice into a collection of identical cells with m_s spins within each cell. The local Hilbert space of this cell has dimension $m_h = (2S + 1)^{m_s}$. We could diagonalize this local Hilbert space:

$$\hat{H}_c = \sum_{m=0}^{m_h-1} \varepsilon_m \hat{s}_m^{\dagger} \hat{s}_m \quad (2.85)$$

where $\hat{s}_m$ is the Schwinger boson of the cell and ε_m is the eigen energy of the local Hamiltonian which describes the interactions within the cell only. For example, we consider a cell with two $S = 1/2$ Spins with isotropic Heisenberg interaction. The local Hilbert space could be diagonalized to a singlet and a triplet.

The diagonalization should not be too troublesome provided that dimension of the local Hilbert space is not too large. We have applied this to a 16 dimensional local Hilbert space to investigate the plaquette singlet state in the Shastry-Sutherland model, as discussed in Chapter 5.

When the unit cell is considered as the basic block, the position vector $\mathbf{r}_i$ now denotes the position of the cell. It could be the center of the cell or any points in the cell. There are m_s spins in a unit cell and we label the spin operators at each point as $\hat{S}_{n,i}^{\alpha}$ where i denotes the position and n is the n^{th} spin in the cell. In terms of these cells, the

Hamiltonian can be expressed as

$$\hat{H} = \sum_{\langle i,j \rangle} \sum_{m,n} J_{ij,mn}^{\alpha} \hat{S}_{m,i}^{\alpha} \hat{S}_{n,j}^{\alpha} + \sum_i \hat{H}_{c,i} \quad (2.86)$$

where $\langle i,j \rangle$ sums over all interacting cells. Usually, the lattice of the cells is bipartite. The spin operators could be expressed in terms of the Schwinger bosons of the cell, which is a reducible representation.

$$\hat{S}_n^{\alpha} = S_{n,uv}^{\alpha} \hat{s}_u^{\dagger} \hat{s}_v \quad \text{with} \quad S_{n,uv}^{\alpha} = \langle s_u | \hat{S}_n^{\alpha} | s_v \rangle. \quad (2.87)$$

The matrices S_n^{α} are hermitian.

At the mean field level, if we assume the classical ground state consists of $|s_0\rangle$ in each cell, either Holstein-Primakoff (HP) or Lagrangian Multiplier (LM) method could be applied. Here we take the HP method for example. In the HP approximation, we have as usual

$$\hat{s}_0 \approx \hat{s}_0^{\dagger} \approx 1 - \frac{1}{2} \sum_{m=1}^{m_h-1} \hat{s}_m^{\dagger} \hat{s}_m \quad (2.88)$$

and the spin operator can be approximated up to bilinear terms as

$$\hat{S}_n^{\alpha} \approx S_{n,00}^{\alpha} + S_{n,u0}^{\alpha} \hat{s}_u^{\dagger} + S_{n,0u}^{\alpha} \hat{s}_u + (S_{n,uv}^{\alpha} - \delta_{uv} S_{n,00}^{\alpha}) \hat{s}_u^{\dagger} \hat{s}_v. \quad (2.89)$$

Keeping only constant and bilinear terms, the interaction between two spin operators becomes

$$\begin{aligned} \hat{S}_{m,i}^{\alpha} \hat{S}_{n,j}^{\alpha} \approx & S_{m,00}^{\alpha} S_{n,00}^{\alpha} + S_{m,u0}^{\alpha} S_{n,0v}^{\alpha} \hat{s}_{u,i}^{\dagger} \hat{s}_{v,j} + S_{m,0u}^{\alpha} S_{n,v0}^{\alpha} \hat{s}_{v,j}^{\dagger} \hat{s}_{u,i} \\ & + S_{n,00}^{\alpha} (S_{m,uv}^{\alpha} - \delta_{uv} S_{m,00}^{\alpha}) \hat{s}_{u,i}^{\dagger} \hat{s}_{v,i} + S_{m,00}^{\alpha} (S_{n,uv}^{\alpha} - \delta_{uv} S_{n,00}^{\alpha}) \hat{s}_{u,j}^{\dagger} \hat{s}_{v,j}. \end{aligned} \quad (2.90)$$

Substitute this into the Hamiltonian, we are able to get

$$\hat{H} = E_0 + \hat{H}_1 + \hat{H}_2 \quad (2.91)$$

where the constant E_0 is the classical ground state energy:

$$E_0 = \sum_{\langle i,j \rangle} \sum_{m,n} J_{ij,mn}^\alpha S_{m,00}^\alpha S_{n,00}^\alpha. \quad (2.92)$$

$\hat{H}_1$ is an effective on-cell Hamiltonian and $\hat{H}_2$ describes the interaction between different cells.

$$\hat{H}_1 = \sum_i \left\{ \sum_{\delta} (J_{\delta,m(\delta)n(\delta)}^\alpha + J_{\delta,n(\delta)m(\delta)}^\alpha) (S_{m(\delta),uv}^\alpha - \delta_{uv} S_{m(\delta),00}^\alpha) S_{n(\delta),00}^\alpha + \varepsilon_u \delta_{uv} \right\} \hat{s}_{u,i}^\dagger \hat{s}_{v,i} \quad (2.93)$$

$$\hat{H}_2 = \sum_i \sum_{\delta} J_{\delta,m(\delta)n(\delta)}^\alpha \left(S_{m(\delta),u0}^\alpha S_{n(\delta),0v}^\alpha \hat{s}_{u,i}^\dagger \hat{s}_{v,j} + S_{m(\delta),u0}^\alpha S_{n(\delta),v0}^\alpha \hat{s}_{u,i}^\dagger \hat{s}_{v,i}^\dagger + h.c. \right) \quad (2.94)$$

where $\delta = \mathbf{r}_j - \mathbf{r}_i$ is the vector connecting cell i and j and it always lies in the first quadrant. Such convention avoids double counting. $m(\delta)$ and $n(\delta)$ denote the two spin operators that δ connects. Einstein summation convention has been applied throughout for $m(\delta)$, $n(\delta)$, u , v and α .

The Fourier Transform of the cell Schwinger bosons is :

$$\begin{aligned} \hat{s}_{u,\mathbf{k}}^\dagger &= \frac{1}{\sqrt{N_c}} \sum_i e^{i\mathbf{k} \cdot \mathbf{r}_i} \hat{s}_{u,i}^\dagger \quad \text{and} \quad \hat{s}_{u,i}^\dagger = \frac{1}{\sqrt{N_c}} \sum_{\mathbf{k}} e^{-i\mathbf{k} \cdot \mathbf{r}_i} \hat{s}_{u,\mathbf{k}}^\dagger \\ \hat{s}_{u,\mathbf{k}} &= \frac{1}{\sqrt{N_c}} \sum_i e^{-i\mathbf{k} \cdot \mathbf{r}_i} \hat{s}_{u,i} \quad \text{and} \quad \hat{s}_{u,i} = \frac{1}{\sqrt{N_c}} \sum_{\mathbf{k}} e^{i\mathbf{k} \cdot \mathbf{r}_i} \hat{s}_{u,\mathbf{k}}. \end{aligned} \quad (2.95)$$

where $N_c = N/m_s$ is the number of cells in the lattice and N is the number of lattice sites.

In the momentum space, the Hamiltonian becomes

$$\hat{H}_1 = \sum_{\mathbf{k}} \left\{ \sum_{\delta} (J_{\delta,m(\delta)n(\delta)}^\alpha + J_{\delta,n(\delta)m(\delta)}^\alpha) (S_{m(\delta),uv}^\alpha - \delta_{uv} S_{m(\delta),00}^\alpha) S_{n(\delta),00}^\alpha + \varepsilon_u \delta_{uv} \right\} \hat{s}_{u,\mathbf{k}}^\dagger \hat{s}_{v,\mathbf{k}} \quad (2.96)$$

$$\hat{H}_2 = \sum_{\mathbf{k}} \sum_{\delta} J_{\delta,m(\delta)n(\delta)}^\alpha \left(S_{m(\delta),u0}^\alpha S_{n(\delta),0v}^\alpha e^{i\mathbf{k} \cdot \delta} \hat{s}_{u,\mathbf{k}}^\dagger \hat{s}_{v,\mathbf{k}} + S_{m(\delta),u0}^\alpha S_{n(\delta),v0}^\alpha e^{i\mathbf{k} \cdot \delta} \hat{s}_{u,\mathbf{k}}^\dagger \hat{s}_{v,-\mathbf{k}}^\dagger + h.c. \right). \quad (2.97)$$

To express the Hamiltonian in a more compact matrix form, we define the following

arrays:

$$a_{\mathbf{k}} = \begin{pmatrix} \hat{s}_{1,\mathbf{k}} & \hat{s}_{2,\mathbf{k}} & \cdots & \hat{s}_{m_h-1,\mathbf{k}} & \hat{s}_{1,-\mathbf{k}}^\dagger & \hat{s}_{2,-\mathbf{k}}^\dagger & \cdots & \hat{s}_{m_h-1,-\mathbf{k}}^\dagger \end{pmatrix}^T, \quad (2.98)$$

$$a_{\mathbf{k}}^\dagger = \begin{pmatrix} \hat{s}_{1,\mathbf{k}}^\dagger & \hat{s}_{2,\mathbf{k}}^\dagger & \cdots & \hat{s}_{m_h-1,\mathbf{k}}^\dagger & \hat{s}_{1,-\mathbf{k}} & \hat{s}_{2,-\mathbf{k}} & \cdots & \hat{s}_{m_h-1,-\mathbf{k}} \end{pmatrix}. \quad (2.99)$$

The Hamiltonian can be written in the matrix form as

$$\hat{H} = E_0 + \frac{1}{2} \sum_{\mathbf{k}} \left(a_{\mathbf{k}}^\dagger \Omega_{\mathbf{k}} a_{\mathbf{k}} - \text{Tr}\{A_{\mathbf{k}}\} \right) \quad (2.100)$$

where the grand dynamical matrix $\Omega_{\mathbf{k}}$ is given by

$$\Omega_{\mathbf{k}} = \begin{pmatrix} A_{\mathbf{k}} & B_{\mathbf{k}} \\ B_{-\mathbf{k}}^* & A_{-\mathbf{k}}^* \end{pmatrix} \quad (2.101)$$

$$\begin{aligned} A_{\mathbf{k},uv} = & \varepsilon_u \delta_{uv} + \sum_{\delta} \left(J_{\delta,m(\delta)n(\delta)}^\alpha + J_{\delta,n(\delta)m(\delta)}^\alpha \right) (S_{m(\delta),uv}^\alpha - \delta_{uv} S_{m(\delta),00}^\alpha) S_{n(\delta),00}^\alpha \\ & + \sum_{\delta} J_{\delta,m(\delta)n(\delta)}^\alpha (S_{m(\delta),u0}^\alpha S_{n(\delta),0v}^\alpha e^{i\mathbf{k}\cdot\delta} + S_{n(\delta),u0}^\alpha S_{m(\delta),0v}^\alpha e^{-i\mathbf{k}\cdot\delta}) \end{aligned} \quad (2.102)$$

$$B_{\mathbf{k}} = \sum_{\delta} \left(S_{m(\delta),u0}^\alpha S_{n(\delta),v0}^\alpha e^{i\mathbf{k}\cdot\delta} + S_{m(\delta),v0}^\alpha S_{n(\delta),u0}^\alpha e^{-i\mathbf{k}\cdot\delta} \right). \quad (2.103)$$

The Hamiltonian could be diagonalized using the Bogoliubov transformation:

$$\hat{H} = E_{GS} + \sum_{\mathbf{k}} \sum_{n=1}^{m_h-1} \lambda_{n,\mathbf{k}} \hat{\gamma}_{n,\mathbf{k}}^\dagger \hat{\gamma}_{n,\mathbf{k}} \quad (2.104)$$

where $\lambda_{n,\mathbf{k}}$ are the dispersion relations and $\hat{\gamma}_{n,\mathbf{k}}$ are the quasi-particles. E_{GS} is the ground state energy with quantum corrections:

$$E_{GS} = E_0 + \frac{1}{2} \sum_{\mathbf{k}} \left(\sum_n \lambda_{n,\mathbf{k}} - \text{Tr}\{A_{\mathbf{k}}\} \right). \quad (2.105)$$

We have described a generalization of the spin wave theory to take more quantum fluctuation into account by using larger cells. An explicit application to the Shastry-Sutherland model is shown in Chapter 5. We would like to point out that the above procedure works well for cells containing a single spin too. Actually, it works for a general lattice in any dimension.

2.6 Bogoliubov transformation

In this section, we illustrate the details of the Bogoliubov transformation which is essential in diagonalization of bilinear bosonic Hamiltonian. We have applied it throughout the spin wave calculations.

Usually in bosonic systems, we use an array of bosonic operators to express everything in matrix form.

$$\alpha_k = \begin{pmatrix} a_{1,k} & a_{2,k} & \dots & a_{n,k} & a_{1,-k}^\dagger & a_{2,-k}^\dagger & \dots & a_{n,-k}^\dagger \end{pmatrix}^T, \quad (2.106)$$

$$\alpha_k^\dagger = \begin{pmatrix} a_{1,k}^\dagger & a_{2,k}^\dagger & \dots & a_{n,k}^\dagger & a_{1,-k} & a_{2,-k} & \dots & a_{n,-k} \end{pmatrix}. \quad (2.107)$$

The commutation relation becomes $[\alpha_{i,k}, \alpha_{j,k'}^\dagger] = \hat{\delta}_{ij} \delta_{kk'}$. $\hat{\delta}_{ij}$ is a metric:

$$\hat{\delta} = \begin{pmatrix} I_n & 0 \\ 0 & -I_n \end{pmatrix} \quad (2.108)$$

where I_n an $n \times n$ identity matrix. The bilinear terms in the Hamiltonian is then written as $\sum_k \alpha_k^\dagger H_k \alpha_k$ where H_k has a block matrix form as:

$$H_k = \begin{pmatrix} A_k & B_k \\ B_{-k}^* & A_{-k}^* \end{pmatrix} \quad (2.109)$$

where the submatrices generally have the symmetry:

$$A_k = A_{-k}^* \quad \text{and} \quad B_k = B_{-k}^* \quad (2.110)$$

so that we can write H_k as

$$H_k = \begin{pmatrix} A_k & B_k \\ B_k & A_k \end{pmatrix}. \quad (2.111)$$

The purpose of Bogoliubov transformation is to look for the quasi-particles $\gamma_k = P_k^{-1} \alpha_k$ such that

$$\alpha_k^\dagger H_k \alpha_k = \gamma_k^\dagger P_k^\dagger H_k P_k \gamma_k = \gamma_k^\dagger E_k \gamma_k = \sum_{i=1}^{2n} \varepsilon_{i,k} c_{i,k}^\dagger c_{i,k} \quad (2.112)$$

where $c_{i,k}$ is the Schwinger boson of the quasi-particle and γ_k is the array similar to α_k :

$$\gamma_k = \begin{pmatrix} c_{1,k} & c_{2,k} & \dots & c_{n,k} & c_{1,-k}^\dagger & c_{2,-k}^\dagger & \dots & c_{n,-k}^\dagger \end{pmatrix}^T \quad (2.113)$$

E_k is a diagonal matrix containing the excitation energies $\varepsilon_{i,k}$.

$$E_k = P_k^\dagger H_k P_k. \quad (2.114)$$

The quasi-particles are bosons therefore they should obey the bosonic commutator relation.

$$\hat{\delta}_{ij} \delta_{kk'} = [\alpha_{i,k}, \alpha_{j,k'}^\dagger] = P_{k,im} P_{k',jn}^* [\gamma_{m,k}, \gamma_{n,k'}^\dagger] = \hat{\delta}_{mn} \delta_{kk'} P_{k,im} P_{k',jn}^* = \left(P_k \hat{\delta} P_k^\dagger \right)_{ij} \delta_{kk'}. \quad (2.115)$$

This implies that P_k is a para-unitary matrix instead of a unitary matrix:

$$P_k \hat{\delta} P_k^\dagger = \hat{\delta} \quad \Rightarrow \quad P_k^{-1} = \hat{\delta} P_k^\dagger \hat{\delta} \quad \text{and} \quad P_k^\dagger = \hat{\delta} P_k^{-1} \hat{\delta} \quad (2.116)$$

We express P_k in a block matrix form as $P_k = \begin{pmatrix} P_{1,k} & P_{2,k} \\ P_{3,k} & P_{4,k} \end{pmatrix}$. The four matrices have the

same property as A_k and B_k :

$$P_{i,-k}^* = P_{i,k} \quad \text{for } i = 1, 2, 3, 4. \quad (2.117)$$

We can show that $P_{1,k} = P_{4,k}$ and $P_{2,k} = P_{3,k}$. For $1 \leq i \leq n$, we have

$$a_{i,k} = \alpha_{i,k} = P_{k,ij} \gamma_{j,k} = P_{1,k,ij} c_{j,k} + P_{2,k,ij} c_{j,-k}^\dagger, \quad (2.118)$$

$$a_{i,-k}^\dagger = \alpha_{i+n,k} = P_{3,k,ij} c_{j,k} + P_{4,k,ij} c_{j,-k}^\dagger. \quad (2.119)$$

It is easy to see from the above equations that $P_{1,k} = P_{4,-k}^*$ and $P_{2,k} = P_{3,-k}^*$. With the property in Eq.2.117, we have shown that the matrix P_k can be written as

$$P_k = \begin{pmatrix} U_k & V_k \\ V_k & U_k \end{pmatrix} \quad \text{with} \quad U_{-k}^* = U_k \quad \text{and} \quad V_{-k}^* = V_k \quad (2.120)$$

Besides, Equation 2.116 implies two further properties of these two matrices:

$$U_k^\dagger U_k - V_k^\dagger V_k = I_n \quad \text{and} \quad U_k^\dagger V_k = V_k^\dagger U_k. \quad (2.121)$$

To obtain the energy and the exact expression of P_k , we can transform Equation 2.114 into:

$$H_k P_k = \hat{\delta} P_k \hat{\delta} E_k = \hat{\delta} P_k \Lambda_k \quad \text{with} \quad \Lambda_k = \hat{\delta} E_k = \text{diag}\{\varepsilon_{1,k}, \dots, \varepsilon_{n,k}, -\varepsilon_{1,k}, \dots, -\varepsilon_{n,k}\}. \quad (2.122)$$

We let $u_{i,k}$ and $v_{i,k}$ be the i^{th} column of U_k and V_k , respectively. $\lambda_{i,k}$ is the i^{th} element of Λ_k . We can show that

$$\begin{pmatrix} A_k & B_k \\ B_k & A_k \end{pmatrix} \begin{pmatrix} u_{i,k} \\ v_{i,k} \end{pmatrix} = \lambda_{i,k} \begin{pmatrix} u_{i,k} \\ -v_{i,k} \end{pmatrix} \quad \text{and} \quad \begin{pmatrix} A_k & B_k \\ B_k & A_k \end{pmatrix} \begin{pmatrix} v_{i,k} \\ u_{i,k} \end{pmatrix} = -\lambda_{i,k} \begin{pmatrix} v_{i,k} \\ -u_{i,k} \end{pmatrix}. \quad (2.123)$$

The above relation allows us to focus on the upper sector of Λ_k , that is, we only need to

calculate $\lambda_{i,k} = \varepsilon_{i,k}$. From the above equation, we can extract the following

$$A_k u_{i,k} + B_k v_{i,k} = \lambda_{i,k} \quad \text{and} \quad B_k u_{i,k} + A_k v_{i,k} = -\lambda_{i,k} v_{i,k}. \quad (2.124)$$

Taking the difference and summation of the above equations give

$$(A_k + B_k)(u_{i,k} + v_{i,k}) = \lambda_{i,k}(u_{i,k} - v_{i,k}), \quad (2.125)$$

$$(A_k - B_k)(u_{i,k} - v_{i,k}) = \lambda_{i,k}(u_{i,k} + v_{i,k}). \quad (2.126)$$

Define

$$w_{i,k} = u_{i,k} - v_{i,k} \quad (2.127)$$

and we multiply Eq.2.126 with $(A_k + B_k)$ to get an eigen value problem:

$$(A_k + B_k)(A_k - B_k)w_{i,k} = \lambda_{i,k}^2 w_{i,k}. \quad (2.128)$$

After solving the eigen value problem, we can obtain the expression of $u_{i,k}$ and $v_{i,k}$ through Eq.2.125.

$$u_{i,k} = \frac{1}{2} \left(\frac{1}{\varepsilon_{i,k}} Q_k + I_n \right) w_{i,k} \quad \text{and} \quad v_{i,k} = \frac{1}{2} \left(\frac{1}{\varepsilon_{i,k}} Q_k - I_n \right) w_{i,k}. \quad (2.129)$$

where $Q_k = A_k - B_k$. To satisfy Equation 2.121, we have to divide $u_{i,k}$ and $v_{i,k}$ by a normalization constant which is given by

$$|m_{i,k}|^2 = u_{i,k}^\dagger u_{i,k} - v_{i,k}^\dagger v_{i,k} = \frac{1}{\varepsilon_{i,k}} w_{i,k}^\dagger Q_k w_{i,k}. \quad (2.130)$$

Finally, we have obtained the P_k matrix with

$$u_{i,k} = \frac{1}{2} \left(\frac{1}{\varepsilon_{i,k}} w_{i,k}^\dagger Q_k w_{i,k} \right)^{-1/2} \left(\frac{1}{\varepsilon_{i,k}} Q_k + I_n \right) w_{i,k}, \quad (2.131)$$

$$v_{i,k} = \frac{1}{2} \left(\frac{1}{\varepsilon_{i,k}} w_{i,k}^\dagger Q_k w_{i,k} \right)^{-1/2} \left(\frac{1}{\varepsilon_{i,k}} Q_k - I_n \right) w_{i,k}. \quad (2.132)$$

Chapter 3

Stochastic Series Expansion

Quantum Monte Carlo (QMC) is a very accurate numerical method for simulating Quantum manybody systems and solving the multi-dimensional integrals that arise from the problem in one way or another. There is a large class of algorithms in QMC, each of which is particularly efficient in certain types of systems. Stochastic Series Expansion (SSE) method is one of the very efficient algorithm for Quantum spin and bosonic systems[23, 24, 25, 26, 27]. Our objective with SSE is to evaluate the partition function of a system and perform various measurements. It is based on the Taylor expansion of the density operator and there are two large classes: classical and quantum SSE.

3.1 Classical SSE

We first look at the application of SSE method in classical statistical mechanics. Consider a system of particles and denote the set $\{\alpha\}$ as all possible configurations(states) of the system. The energy of the system in a particular state α is denoted as $E(\alpha)$. In thermal equilibrium, the probability of the system to be in a particular state α is given by the Boltzmann distribution:

$$P(\alpha) = \frac{1}{Z} e^{-\beta E(\alpha)} \quad (3.1)$$

where Z is the partition function defined as

$$Z = \sum_{\{\alpha\}} e^{-\beta E(\alpha)} \quad (3.2)$$

and $\beta = \frac{1}{k_B T}$ is the inverse temperature.

In Boltzmann statistics, the thermal expectation value of a variable f is

$$\langle f \rangle = \frac{1}{Z} \sum_{\{\alpha\}} f(\alpha) e^{-\beta E(\alpha)}. \quad (3.3)$$

Most of the time, it is difficult to calculate both equation (3.2) and (3.3) analytically. We may evaluate the expectation value using Monte Carlo method, where the configurations are importance sampled using Metropolis algorithm according to the Boltzmann distribution

$$\begin{aligned} P(\alpha) &= \frac{1}{Z} W(\alpha), \\ W(\alpha) &= e^{-\beta E(\alpha)}. \end{aligned} \quad (3.4)$$

The expectation value $\langle f \rangle$ can be simply obtained by taking the average of $f(\alpha)$ over the sampled configuration $\alpha(i)$, $i = 1, 2, \dots, N$.

$$\langle f \rangle = \frac{1}{N} \sum_{i=1}^N f(\alpha(i)) \quad (3.5)$$

where N is the number of configurations sampled. When N is very large, the result approaches the exact value.

Now suppose we cannot evaluate the exponential function in equation (3.4).^{*} We can

^{*}In quantum statistical mechanics, it is generally impossible to evaluate the exponential of an operator unless it is diagonal.

Taylor expand the exponential function in equation (3.1) as

$$\langle f \rangle = \frac{1}{Z} \sum_{\{\alpha\}} \sum_{n=0}^{\infty} f(\alpha) \frac{(-\beta E(\alpha))^n}{n!}, \quad (3.6)$$

$$Z = \sum_{\{\alpha\}} \sum_{n=0}^{\infty} \frac{(-\beta E(\alpha))^n}{n!}. \quad (3.7)$$

Equation (3.7) generally converges for any β , since the energy is always finite for most of the systems we are interested in. With equation (3.6) and (3.7), we are now working in an expanded configuration space (α, n) . Now the weight of a certain configuration (α, n) in the expanded space is given by

$$W(\alpha, n) = \frac{(-\beta E(\alpha))^n}{n!}. \quad (3.8)$$

However, we can only use equation (3.8) to do importance sampling provided that it is always non-negative (or the energy is always negative), which is normally not the case. Luckily, we can always subtract a positive constant ε from the energy without changing the physics. From equation (3.1) and (3.2), we can see that doing so is just multiplying the numerator and the denominator by e^ε in equation (3.1). Equation (3.8) then becomes

$$W(\alpha, n) = \frac{\beta^n (\varepsilon - E(\alpha))^n}{n!}. \quad (3.9)$$

With equation (3.8), we are able to write expectation values of thermodynamic observables as weighted averages. For simplicity, we denote $H(\alpha) = \varepsilon - E(\alpha)$. Then equation (3.6), (3.7) and (3.9) become

$$\langle f \rangle = \frac{1}{Z} \sum_{\{\alpha, n\}} f(\alpha) W(\alpha, n), \quad (3.10)$$

$$Z = \sum_{\{\alpha, n\}} W(\alpha, n), \quad (3.11)$$

$$W(\alpha, n) = \frac{\beta^n H(\alpha)^n}{n!}. \quad (3.12)$$

The expectation value of the modified energy of the system is

$$\begin{aligned}
 \langle H \rangle &= \frac{1}{Z} \sum_{\{\alpha\}} \sum_{n=0}^{\infty} H(\alpha) \frac{\beta^n H(\alpha)^n}{n!} \\
 &= \frac{1}{Z\beta} \sum_{\{\alpha\}} \sum_{n=0}^{\infty} (n+1) \frac{\beta^{n+1} H(\alpha)^{n+1}}{(n+1)!} \\
 &= \frac{1}{\beta} \frac{1}{Z} \sum_{\{\alpha\}} \sum_{n=0}^{\infty} n \frac{\beta^n H(\alpha)^n}{n!} \\
 &= \frac{\langle n \rangle}{\beta}.
 \end{aligned} \tag{3.13}$$

The expectation value of the energy (with the additive constant) of the system is then

$$\langle E \rangle = \varepsilon - \frac{\langle n \rangle}{\beta}. \tag{3.14}$$

Equation (3.13) shows remarkable results from the stochastic series expansion (SSE) method. It is worth mentioning again that n is the order in the Taylor expansion and the average value of n is sampled over $\{\alpha\}$ as well as the expansion of $e^{-\beta H(\alpha)}$. Besides, there is a potential danger about the convergency of $\langle n \rangle$. Equation (3.14) shows that $\langle n \rangle$ is convergent provided the average energy of the system is convergent and the temperature is nonzero. Hence, the SSE method works for finite temperatures. We generally consider systems at a low temperature around which the phase transition occurs, which we are actually interested in. In such a case, the expectation value of the energy is generally finite. Therefore, $\langle n \rangle$ is finite. The energy of the system is proportional to the system size N . From equation (3.14) we see that $\langle n \rangle$ is proportional to $\frac{N}{T}$. Therefore, n follows a distribution with average value proportional to $\frac{N}{T}$.

If the temperature of the system is zero, the system is in ground state. But we can still apply SSE method. We actually can not simulate the system in the thermodynamic limit. We will take the system size to be finite. The finite size affects the energy spectrum. In thermodynamic limit, the energy spectrum for a gapless ground state is continuous. In a system of finite size, however, there are gaps between energy levels. In the simulation of such systems, if the temperature is chosen lower than the finite-size gap, the contribu-

tion from the higher energy states will be negligible. In this manner, the system at zero temperature can be simulated by a finite size system at a finite low temperature using SSE method, followed by careful finite size and finite temperature extrapolation[28, 29].

3.2 Quantum SSE

In quantum statistical mechanics, a density matrix $\hat{\rho}$ is used to describe the probability distribution of quantum states

$$\hat{\rho} = \frac{1}{Z} e^{-\beta \hat{H}}, \quad (3.15)$$

$$Z = \text{Tr}\{e^{-\beta \hat{H}}\} = \sum_{\alpha} \langle \alpha | e^{-\beta \hat{H}} | \alpha \rangle \quad (3.16)$$

where $\hat{H}$ is the Hamiltonian of the system and $\text{Tr}\{\hat{A}\}$ is the trace of the operator $\hat{A}$. $\{\alpha\}$ is the collection of the quantum states in the Hilbert space of interest. The expectation value of some operator $\hat{A}$ is then

$$\langle \hat{A} \rangle = \text{Tr}\{\hat{A} \hat{\rho}\} = \frac{1}{Z} \text{Tr}\{\hat{A} e^{-\beta \hat{H}}\}. \quad (3.17)$$

3.2.1 Operator sequence and truncation

The difficulty of the above equations lies in the evaluation of the exponential function of a Hamiltonian. If we are able to find out the eigenstates of the Hamiltonian, the Hamiltonian can be diagonalized and the exponential can be simply calculated, which actually reduces to the classical SSE method. However, the quantum states that we choose with obvious physical meaning, e.g., the z -component of the spin or the number of particles at each lattice site, are generally not the eigenstates of the Hamiltonian. In terms of those bases, the Hamiltonian consists of non-commuting diagonal and off-diagonal operators.

The usual way to deal with exponential of operators or matrices is to perform a Taylor

expansion,

$$e^{-\beta\hat{H}} = \sum_{n=0}^{\infty} \frac{\beta^n}{n!} (-\hat{H})^n. \quad (3.18)$$

Suppose the Hamiltonian can be decomposed as

$$\hat{H} = - \sum_a \hat{H}_a \quad (3.19)$$

where $\hat{H}_a$, $a = 1, 2, \dots$, are operators such that $\hat{H}_a |\alpha\rangle \propto |\beta\rangle$, where both $|\alpha\rangle$ and $|\beta\rangle$ are the chosen basis states. That is each $\hat{H}_a$ maps a basis ket to another basis ket. The great advantage of such decomposition will soon be obvious. Substitute it into equation (3.18) and we will have

$$e^{-\beta\hat{H}} = \sum_{n=0}^{\infty} \frac{\beta^n}{n!} \sum_{\{\hat{H}_a\}} \prod_{p=1}^n \hat{H}_{a(p)} \quad (3.20)$$

where $\{\hat{H}_a\}$ contains all possible sequences of operators $\hat{H}_a$ with size n , that is, there are n operators in each sequence. If we allow the size of the sequences to change, then the summation over the power n can be included in the summation over different sequences of operators. Equation (3.20) then becomes

$$e^{-\beta\hat{H}} = \hat{I} + \sum_{\{\hat{H}_a\}} \prod_{p=1}^n \frac{\beta^n}{n!} \hat{H}_{a(p)}. \quad (3.21)$$

$\hat{I}$ is the identity operator corresponding to $n = 0$. Equation (3.21) is not practical in computation since we have to sum over infinite terms which is impossible on a computer. Practically, we can truncate the Taylor expansion at some cut-off L , which is chosen by the computer itself during the computation. After the truncation, we see that the maximum length of the operator sequence is L . To make the calculation even easier, we insert $(L - n)$ identity operators into the operator sequences with size n in each operator sequence $\hat{H}_a$ so that the all the operator sequences have the same length L . For each operator sequence with size n , there are $\binom{L}{L-n}$ possible ways to insert the identity operators. If we sum over all possible augmented operator sequences with size L , the contribution to the summation from each of the original operator sequence will

be overcounted. Hence, we have to divide it by $\binom{L}{L-n}$ that the operator sequences containing n decomposed Hamiltonian operators. If we denote the identity operator as:

$$\hat{H}_0 = \hat{I}, \quad (3.22)$$

equation (3.21) becomes:

$$e^{-\beta\hat{H}} = \sum_{\{\hat{H}_a\}} \frac{\beta^n (L-n)!}{L!} \prod_{p=1}^L \hat{H}_{a(p)}. \quad (3.23)$$

The partition function is then evaluated as:

$$Z = \sum_{\alpha} \sum_{\{\hat{H}_a\}} \frac{\beta^n (L-n)!}{L!} \langle \alpha | \prod_{p=1}^L \hat{H}_{a(p)} | \alpha \rangle. \quad (3.24)$$

And the thermal average of an observable is:

$$\langle \hat{A} \rangle = \frac{1}{Z} \sum_{\alpha} \sum_{\{\hat{H}_a\}} \frac{\beta^n (L-n)!}{L!} \langle \alpha | \hat{A} \prod_{p=1}^L \hat{H}_{a(p)} | \alpha \rangle. \quad (3.25)$$

Similar to the classical statistical mechanics case, equation (3.24) and (3.25) can be evaluated by Monte Carlo simulation. Instead of the Hilbert space which is the original sample space, now we have an augmented sample space, the direct product of the Hilbert space $\{\alpha\}$ and the operator sequence $\{\hat{H}_a\}$: $\{\alpha, \{\hat{H}_a\}\}$. The elements $\{\alpha, \{\hat{H}_a\}\}$ are sampled according to their weight in the summation. This also requires that all the terms in the summation must be non-negative. This can be achieved for sign-problem free Hamiltonians by using a similar method as what we did in classical case. We can modify the diagonal operators by adding or subtracting some constants to make the operators positive definite without changing the physics. As to the off-diagonal operators, if the lattice system is bipartite, they have to appear in pairs, therefore, the sign does not matter here. However, if the system has non-bipartite lattice system, the off-diagonal interactions have to be negative-definite to avoid frustration. Otherwise, we have the notorious sign problem in Quantum Monte Carlo method.

Explicitly, we can define an auxiliary operator $\hat{H}' = C - \hat{H} = \sum_a \hat{H}'_a$. The constant C is chosen such that all $\hat{H}'_a$ is positive-definite. The partition function becomes:

$$Z = e^{-\beta C} \sum_{\alpha} \sum_{\{\hat{H}_a\}} \frac{\beta^n (L-n)!}{L!} \langle \alpha | \prod_{p=1}^L \hat{H}'_{a(p)} | \alpha \rangle = e^{-\beta C} Z', \quad (3.26)$$

where Z' is the partition function of $-\hat{H}'$. The thermal average of an operator $\hat{A}$ now becomes:

$$\langle \hat{A} \rangle = \frac{1}{Z'} \sum_{\alpha} \sum_{\{\hat{H}_a\}} \frac{\beta^n (L-n)!}{L!} \langle \alpha | \hat{A} \prod_{p=1}^L \hat{H}'_{a(p)} | \alpha \rangle. \quad (3.27)$$

We see that thermal average with respect to the original Hamiltonian is the same as the one generated by the auxiliary $-\hat{H}'$. As a result, in the rest of the thesis, we will use the auxiliary operator instead and its decomposition will just be labeled as $\hat{H}_a$ instead of $\hat{H}'_a$. The thermal average of $\hat{H}'$ is:

$$\langle \hat{H}' \rangle = \frac{1}{Z'} \sum_{n=0} \frac{\beta^n \hat{H}'^{n+1}}{n!} = \frac{1}{Z'} \sum_{n=0} \frac{n}{\beta} \frac{\beta^n}{\hat{H}'} n! = \frac{\langle n \rangle}{\beta}, \quad (3.28)$$

where $\langle n \rangle$ is the thermal average of the length of the operators. In practice, this average is also evaluated in the truncated sequence. Thus, it is the thermal average number of the non-identity operators. The energy of the system is then given by:

$$E = \langle \hat{H} \rangle = C - \frac{\langle n \rangle}{\beta}. \quad (3.29)$$

3.2.2 Determination of the truncation

The truncation length L plays a very important role in SSE. The method described above is based on the assumption that there exists such a truncation length L , that is, we are in fact able to find an appropriate truncation so that Eq.3.23 and 3.24 converge to the real values, Eq.3.16 and 3.17. Besides, we should also have a control on the scale of this truncation. It makes no sense in practice if the truncation length is far too large. Fortunately, we have solutions to all the above problems.

Without the truncation, the partition function is calculated from equation (3.18) as:

$$Z = \sum_{n=0}^{\infty} \frac{(-\beta)^n}{n!} \sum_{\alpha} \langle \alpha | \hat{H}^n | \alpha \rangle \quad (3.30)$$

We can consider the above equation as a series. Let's denote the terms in the summation over n in equation (3.30) as a_n and the ratio between two successive terms as r_n . Then we can apply the ratio test of convergence:

$$r = \lim_{n \rightarrow \infty} |r_n| = \lim_{n \rightarrow \infty} \left| \frac{a_{n+1}}{a_n} \right| = \lim_{n \rightarrow \infty} \left| \frac{\frac{(-\beta)^{n+1}}{(n+1)!} \sum_{\alpha} \langle \alpha | \hat{H}^{n+1} | \alpha \rangle}{\frac{\beta^n}{n!} \sum_{\alpha} \langle \alpha | \hat{H}^n | \alpha \rangle} \right| = \lim_{n \rightarrow \infty} \frac{\beta}{n+1} \left| \frac{\sum_{\alpha} \langle \alpha | \hat{H}^{n+1} | \alpha \rangle}{\sum_{\alpha} \langle \alpha | \hat{H}^n | \alpha \rangle} \right|. \quad (3.31)$$

$\left| \sum_{\alpha} \langle \alpha | \hat{H}^n | \alpha \rangle \right|$ is of the order of $\Gamma |E|^n$, where E is the energy of the system and Γ is the number of states, i.e., the number of $|\alpha\rangle$ we have to sum over. Then equation (3.31) becomes

$$r = \lim_{n \rightarrow \infty} \frac{\beta |E|}{n+1}. \quad (3.32)$$

We can see that $r = 0$ so long as the energy of the system is finite and the temperature is nonzero. Hence, we have shown that equation (3.30) converges. For a convergent series, the summand tends to zero. Hence, we can truncate the series at a_L such that the remainder is exponentially small and makes negligible contribution to the summation. In the language of Monte Carlo simulation, this means that the configuration corresponding to the remainder will never be sampled during the life time of the simulator or even of the universe.

To determine the order of L , we should use equation (3.32). For a convergent series, when n is large enough, the ratio between two successive terms will approximately follow equation (3.32), i.e., they start to converge to the limit. Then we have for sufficiently large L :

$$|a_{L+1}| \simeq \frac{\beta |E|}{L+1} |a_L|. \quad (3.33)$$

We can also approximate the $(L + r)^{\text{th}}$ term as:

$$|a_{L+r}| \simeq \frac{|\beta E|^r}{(L+1)(L+2)\cdots(L+r)} |a_L|. \quad (3.34)$$

We choose $L' = L + r$ and change equation (3.34) into an inequality as:

$$|a_{L'}| \leq \left| \frac{\beta E}{L} \right|^{L'-L} |a_L|. \quad (3.35)$$

Inequality (3.35) shows that the summands in the series start to decay exponentially from the L^{th} term provided that $\left| \frac{\beta E}{L} \right| < 1$. A weak condition for this is that $L = k\beta|E|$, where $k < 1$. Actually, equation (3.34) and inequality (3.35) also shows the method to look for the truncation value computationally. Suppose L is not good enough, then we can try L' from equation (3.34). If L' doesn't satisfy inequality (3.35), then we continue the procedure. Otherwise, we have already determined the truncation value. To implement this procedure in the algorithm, we choose the increment $r = qL$. Then the inequality becomes:

$$|a_{L+qL}| \leq \left| \frac{\beta E}{L} \right|^{qL} |a_L|. \quad (3.36)$$

With the above inequality, we are able to determine a truncation value L such that the remainder of the series is exponentially small and completely negligible. And the order of L can be determined immediately from this inequality:

$$L > \beta|E|. \quad (3.37)$$

We can conclude that in the stochastic series expansion, we can truncate the series to the L^{th} order with exponentially small and negligible error, where L is proportional to $\beta|E|$. Since the total energy is an extensive quantity, i.e., $E \propto N$ (the size of the system), we have $L \sim \beta N$, which is exactly what is observed empirically.

3.2.3 Updating procedures

There are two main updating procedures in Quantum SSE method. One is called **diagonal update** during which the number of identity operators are changed by exchanging the identity operator with diagonal operators. The other is called **off-diagonal update** during which we change the types of the non-identity operators and the state $|\alpha\rangle$ while preserving the number of identity operators. The diagonal update is trivial while in the off-diagonal, the loop update method is applied so that we can update a bunch of operators at the same time. This will be described in detail when we apply to a specific model, Sec.3.3. In this section, we will describe the updating procedures in a non-specific manner and we will introduce diagrams for visualization.

We define the **propagated state**: $|\alpha(l)\rangle$ as

$$|\alpha(l)\rangle = N_l \prod_{i=1}^l \hat{H}_{a(i)} |\alpha\rangle \quad (3.38)$$

where N_l is the normalization constant. We denote $|\alpha\rangle = |\alpha(0)\rangle$. Equation 3.24 can then be written as:

$$Z = \sum_{\alpha} \sum_{\{\hat{H}_a\}} \frac{\beta^n (L-n)!}{L!} \prod_{l=1}^L \langle \alpha(l) | \hat{H}_{a(l)} | \alpha(l-1) \rangle. \quad (3.39)$$

Only terms with $|\alpha(L)\rangle = |\alpha(0)\rangle$ have non-zero contribution. Hence, we can use a closed diagram to represent each element in the sample space, as shown in Fig.3.1.

Each small solid circle represents a propagated state $|\alpha(l)\rangle$ and the link connecting two circles $|\alpha(l-1)\rangle$ and $|\alpha(l)\rangle$ represents the operator $\hat{H}_{a(l)}$ in $\langle \alpha(l) | \hat{H}_{a(l)} | \alpha(l-1) \rangle$. There are exactly L circles in this close loop with $|\alpha(L)\rangle$ and $|\alpha(0)\rangle$ being represented by the same circle. The weight of this configuration is just the summand in Eq.3.39, to which we give a new notation:

$$W(\alpha, \{a\}) = \frac{\beta^n (L-n)!}{L!} \prod_{l=1}^L \langle \alpha(l) | \hat{H}_{a(l)} | \alpha(l-1) \rangle \quad (3.40)$$

We have used $\{a\}$ to represent the operator sequence $\{\hat{H}_a\}$.

The updating procedure is then to change one configuration to another according to

Figure 3.1: The diagram of a general configuration in the sample space. The dotted line represents part of the sequence that is not labeled explicitly.

their relative weights. For simplicity, we call the non-identity operator **H-operator**. In diagonal update, the circles remain the same. Starting from the first link $\hat{H}_{a(1)}$, if the l^{th} link is an identity operator, we try to replace it with some diagonal H-operator $\tilde{H}_{a(l)}$ according to the probability of acceptance:

$$P = \min \left(\frac{BW(\alpha, \{a'\})}{W(\alpha, \{a\})}, 1 \right) = \min \left(\frac{B\beta}{L-n} \langle \alpha(l) | \tilde{H}_{a(l)} | \alpha(l-1) \rangle, 1 \right) \quad (3.41)$$

where B is the number of possible diagonal operators. Generally, the diagonal operators only differ in the lattice position where they operate on. Hence, B is generally the number of positions that we can put a diagonal operator. n is the number of H-operator in the original configuration before update.

If the l^{th} is a diagonal H-operator, we try to replace it with an identity operator with the acceptance probability:

$$P = \min \left(\frac{L-n-1}{B\beta \langle \alpha(l) | \hat{H}_{a(l)} | \alpha(l-1) \rangle}, 1 \right). \quad (3.42)$$

After each diagonal, a few off-diagonal update will be performed when the H-operators and $|\alpha\rangle$ change. In terms of the diagram, we start by changing one of the link to another

type. After this change, the sequence is no longer closed, as shown in Fig.3.2a.

Starting from this new link, for example $\hat{H}'_{a(2)}$ in Fig.3.2b, a new sequence is formed. However, it has to come back at some point because the loop should be closed. Therefore, a new loop is created based on the initial loop, the uniform blue loop in Fig.3.2b. It is possible that the new sequence (the outer part in the Fig.3.2b) comes back after it passes the initial state $|\alpha(0)\rangle$. In this situation, the initial state is updated as well. In practice, the new sequence could go back and forth in both directions. An efficient off-diagonal update, **loop update**, will be discussed in detail in Sec.3.3.

3.2.4 Measurement of a single operator

Measurement is the ultimate purpose of a computational method. In SSE, the expectation values of both diagonal and off-diagonal operators can be calculated very efficiently[24, 28]. For example, the average number of the H-operators gives the energy of the system, Eq.3.29. A diagonal operator does not change the state it operates on, so its measurement could be performed during the diagonal update. Usually the off-diagonal operator could be expressed in terms of the off-diagonal H-operator, therefore its measurement could also be performed during the diagonal update. To those that could not be expressed in terms

of the H-operators, their measurement could be done during the off-diagonal update, for example equal time correlation function, which will be discussed in next subsection.

As shown in Eq.3.27, the thermal average of an operator $\hat{A}$ could be expressed in terms of the propagated state as:

$$\langle \hat{A} \rangle = \frac{1}{Z} \sum_{\alpha} \sum_{\{\hat{H}_a\}} \frac{\beta^n (L-n)!}{L!} \langle \tilde{\alpha}(L) | \hat{A} | \alpha(L) \rangle \prod_{l=1}^L \langle \alpha(l) | \hat{H}_{a(l)} | \alpha(l-1) \rangle \quad (3.43)$$

where $|\tilde{\alpha}(L)\rangle = |\alpha(0)\rangle$.

Diagonal operators

We first consider the case when $\hat{A}$ is a diagonal operator such that $\hat{A} |\alpha(l)\rangle = a_l |\alpha(l)\rangle$.

The thermal average simply becomes:

$$\langle \hat{A} \rangle = \frac{1}{Z} \sum_{\alpha} \sum_{\{\hat{H}_a\}} \frac{\beta^n (L-n)!}{L!} a_L \prod_{l=1}^L \langle \alpha(l) | \hat{H}_{a(l)} | \alpha(l-1) \rangle = \frac{1}{Z} \sum_{\alpha} \sum_{\{\hat{H}_a\}} a_L W(\alpha, \{a\}) \quad (3.44)$$

We define:

$$A(1, \alpha(0), \{a\}) = \langle \alpha(L) | \hat{A} | \alpha(L) \rangle \prod_{l=1}^L \langle \alpha(l) | \hat{H}_{a(l)} | \alpha(l-1) \rangle \quad (3.45)$$

and a cyclic permutation operator $\hat{P}$ such that:

$$\begin{aligned} \hat{P} A(1, \alpha(0), \{a\}) &= \langle \alpha(1) | \hat{H}_{a(1)} | \alpha(0) \rangle \langle \alpha(0) | \hat{A} | \alpha(L) \rangle \prod_{l=2}^L \langle \alpha(l) | \hat{H}_{a(l)} | \alpha(l-1) \rangle \\ &= A(2, \alpha(1), \hat{P}\{a\}) = A(1, \alpha(0), \{a\}). \end{aligned} \quad (3.46)$$

Similarly, we have

$$A(1, \alpha(0), \{a\}) = \hat{P}^m A(1, \alpha(0), \{a\}) = A(m+1, \alpha(m), \hat{P}^m \{a\}) \quad (3.47)$$

where $A(m+1, \alpha(m), \hat{P}^m \{a\})$ means that the operator sequence now is $\hat{P}\{a\}$ with initial state $|\alpha(m)\rangle$ and the measurement of operator $\hat{A}$ is performed at the $(m+1)^{\text{th}}$ position

from left. Hence we have:

$$A(1, \alpha(0), \{a\}) = \frac{1}{M} \sum_{i=1}^M A(m_i + 1, \alpha(m_i), \hat{P}^{m_i} \{a\}) \quad (3.48)$$

where $m_i \in [0, L]$ is an integer. The thermal average now becomes:

$$\langle \hat{A} \rangle = \frac{1}{Z} \sum_{\alpha} \sum_{\{\hat{H}_a\}} \frac{\beta^n (L-n)!}{L!} \frac{1}{M} \sum_{i=1}^M A(m_i + 1, \alpha(m_i), \hat{P}^{m_i} \{a\}). \quad (3.49)$$

Since $\hat{P}^{m_i} \{a\}$ is also an element of $\hat{H}_a$ which is summed over and the same to $|\alpha(m_i)\rangle$, the above equation becomes:

$$\langle \hat{A} \rangle = \frac{1}{Z} \sum_{\alpha} \sum_{\{\hat{H}_a\}} \frac{\beta^n (L-n)!}{L!} \frac{1}{M} \sum_{i=1}^M A(m_i + 1, \alpha, \{a\}). \quad (3.50)$$

Now the above equation has the following meaning. For each initial state $|\alpha\rangle$ and operator sequence $\hat{H}_a$, we can perform the measurement of $\hat{A}$ at any propagated level $|\alpha(m_i)\rangle$ and then take the average. In this manner, we could obtain a more accurate measurement because we are able to perform measurement M times as large as the original one, Eq.3.43, within the same Monte Carlo step. Taking M to be the maximum L , we have

$$\langle \hat{A} \rangle = \frac{1}{Z} \sum_{\alpha} \sum_{\{\hat{H}_a\}} \frac{\beta^n (L-n)!}{L!} \frac{1}{L} \sum_{m=1}^L A(m, \alpha, \{a\}). \quad (3.51)$$

where we have neglected the measurement at the initial level $|\alpha(0)\rangle$ since it has been counted in the final level $|\alpha(L)\rangle$.

H-operators

Now we consider the case when $\hat{A}$ is one of the H-operators. We denoted it as $\hat{H}_b$. It would be more convenient if we start with Equation 3.17:

$$\langle \hat{H}_b \rangle = \frac{1}{Z} \sum_{\alpha} \sum_{\{\hat{H}_a\}} \frac{\beta^n}{n!} \langle \alpha(n+1) | \hat{H}_b | \alpha(n) \rangle \prod_{l=1}^n \langle \alpha(l) | \hat{H}_{a(l)} | \alpha(l-1) \rangle \quad (3.52)$$

where $|\alpha(n+1)\rangle = |\alpha(0)\rangle$. In the above summation, the operator sequence consists of pure H-operators with varying length n . Since $\hat{H}_b$ is also an H-operator, the above sequence can be considered as an augmented sequence which ends with $\hat{H}_b$.

$$\langle \hat{H}_b \rangle = \frac{1}{\beta} \frac{1}{Z} \sum_{\alpha} \sum_{\hat{H}_b \{\hat{H}_a\}} n \frac{\beta^n}{n!} \prod_{l=1}^n \langle \alpha(l) | \hat{H}_{a(l)} | \alpha(l-1) \rangle \quad (3.53)$$

where it is summed over the left coset $\hat{H}_b \{\hat{H}_a\}$. Since n , the length of $\hat{H}_b \{\hat{H}_a\}$, is also an dummy index, we have replaced $n+1$ with n in the above equation. We can extend the summation domain from $\hat{H}_b \{\hat{H}_a\}$ to a more general $\{\hat{H}_a\}$ with varying length n and an delta function $\delta_{a(n),b}$ is introduced to make sure only sequences with last element $\hat{H}_{a(n)} = \hat{H}_b$ contribute. We again define

$$A(\alpha(0), \{\hat{H}_a\}) = \prod_{l=1}^n \langle \alpha(l) | \hat{H}_{a(l)} | \alpha(l-1) \rangle. \quad (3.54)$$

Hence we have

$$\langle \hat{H}_b \rangle = \frac{1}{\beta} \frac{1}{Z} \sum_{\alpha} \sum_{\{\hat{H}_a\}} n \frac{\beta^n}{n!} A(\alpha(0), \{\hat{H}_a\}) \delta_{a(n),b}. \quad (3.55)$$

As before, we can define an cyclic permutation operator $\hat{P}$ such that

$$\hat{P} A(\alpha(0), \{\hat{H}_a\}) \delta_{a(n),b} = A(\alpha(1), \hat{P} \{\hat{H}_a\}) \delta_{a(n-1),b} = A(\alpha(0), \{\hat{H}_a\}) \delta_{a(n),b} \quad (3.56)$$

and similarly

$$\hat{P}^m A(\alpha(0), \{\hat{H}_a\}) \delta_{a(n),b} = A(\alpha(m), \hat{P}^m \{\hat{H}_a\}) \delta_{a(n-m),b} = A(\alpha(0), \{\hat{H}_a\}) \delta_{a(n),b} \quad (3.57)$$

where $A(\alpha(m), \hat{P}^m \{\hat{H}_a\}) \delta_{a(n-m),b}$ means the initial state becomes $|\alpha(m)\rangle$ with sequence $\hat{P}^m \{\hat{H}_a\}$ and the $\hat{H}_b$ now appears as the $(n-m)^{\text{th}}$ operator in the new sequence. Hence we have

$$\langle \hat{H}_b \rangle = \frac{1}{\beta} \frac{1}{Z} \sum_{\alpha} \sum_{\{\hat{H}_a\}} \frac{\beta^n}{n!} \sum_{m=0}^{n-1} A(\alpha(m), \hat{P}^m \{\hat{H}_a\}) \delta_{a(n-m),b} \quad (3.58)$$

Observing the above equation, we realize that $\frac{\beta^n}{n!} A(\alpha(m), \hat{P}^m \{\hat{H}_a\})$ is just the weight of the configuration $\{\alpha(m), \hat{P}^m \{\hat{H}_a\}\}$ which is an element being summed. So we could drop the index m in both $\alpha(m)$ and the operator sequence. This actually means that so long as the sequence contains an $\hat{H}_b$ operator, it could always be counted by some cyclic permutation even though it is not at the final position. So we have:

$$\langle \hat{H}_b \rangle = \frac{1}{\beta} \frac{1}{Z} \sum_{\alpha} \sum_{\{\hat{H}_a\}} \frac{\beta^n}{n!} \sum_{m=0}^{n-1} A(\alpha, \{\hat{H}_a\}) \delta_{a(n-m),b} = \frac{\langle n_b \rangle}{\beta} \quad (3.59)$$

where n_b is the number of $\hat{H}_b$ operator in the operator sequence. The last equality is true because the summation over the delta function simply counts the number of $\hat{H}_b$ appears in the operator sequence $\{\hat{H}_a\}$.

The thermal average $\langle \hat{H} \rangle$, Eq.3.28, could also be obtained in this manner.

$$\langle \hat{H} \rangle = \sum_a \langle \hat{H}_a \rangle = \sum_a \frac{\langle n_a \rangle}{\beta} = \frac{\langle \sum_a n_a \rangle}{\beta} = \frac{\langle n \rangle}{\beta}. \quad (3.60)$$

The last equality is true because the total number of all different H-operators is simply the length of the operator sequence.

Off-diagonal operators

Finally, we consider the case when operator $\hat{A}$ is neither diagonal nor an H-operator. Its thermal average in terms of the non-truncated sequence is again given by

$$\langle \hat{A} \rangle = \frac{1}{Z} \sum_{\alpha} \sum_{\{\hat{H}_a\}} \frac{\beta^n}{n!} \langle \alpha | \hat{A} \prod_{l=1}^n \hat{H}_{a(l)} | \alpha \rangle. \quad (3.61)$$

As before, we can use the property of the trace. We define the cyclic permutation $\hat{P}$ such that

$$\hat{P} \left(\hat{A} \prod_{l=1}^n \hat{H}_{a(l)} \right) = \prod_{l=1}^n \hat{H}_{a(l)} \hat{A} \quad \text{and} \quad \hat{P}^2 \left(\hat{A} \prod_{l=1}^n \hat{H}_{a(l)} \right) = \prod_{l=1}^{n-1} \hat{H}_{a(l)} \hat{A} \hat{H}_{a(n)} \quad (3.62)$$

that is it shifts $\hat{A}$ to the left. The thermal average becomes:

$$\begin{aligned} \langle \hat{A} \rangle &= \frac{1}{Z} \sum_{\alpha} \sum_{\{\hat{H}_a\}} \frac{\beta^n}{n!} \frac{1}{n} \sum_{m=1}^n \langle \alpha | \hat{P}^m \left(\hat{A} \prod_{l=1}^n \hat{H}_{a(l)} \right) | \alpha \rangle \\ &= \frac{1}{Z} \sum_{\alpha} \sum_{\{\hat{H}_a\}} \frac{\beta^n}{n!} \frac{1}{n} \sum_{m=1}^n \langle \alpha | \hat{H}_{a(n)} \hat{H}_{a(n-1)} \cdots \hat{H}_{a(m)} \hat{A} \hat{H}_{a(m-1)} \cdots \hat{H}_{a(1)} | \alpha \rangle \end{aligned} \quad (3.63)$$

We define

$$A(\alpha(l_1 - 1), \{\hat{H}_a\}_{l_1}^{l_2}) = \langle \alpha(l_2) | \hat{H}_{a(l_2)} | \alpha(l_2 - 1) \rangle \cdots \langle \alpha(l_1) | \hat{H}_{a(l_1)} | \alpha(l_1 - 1) \rangle. \quad (3.64)$$

The matrix element in Eq.3.63 can be expressed as:

$$\begin{aligned} \langle \alpha | \hat{P}^m \left(\hat{A} \prod_{l=1}^n \hat{H}_{a(l)} \right) | \alpha \rangle &= \langle \tilde{\alpha}(n) | \hat{H}_{a(n)} | \tilde{\alpha}(n-1) \rangle \cdots \langle \tilde{\alpha}(m) | \hat{H}_{a(m)} | \tilde{\alpha}(m-1) \rangle \\ &\quad \cdot \langle \tilde{\alpha}(m-1) | \hat{A} | \alpha(m-1) \rangle \langle \alpha(m-1) | \hat{H}_{a(m-1)} | \alpha(m-2) \rangle \cdots \\ &\quad \cdot \langle \alpha(1) | \hat{H}_{a(1)} | \alpha(0) \rangle \\ &= A(\tilde{\alpha}(m-1), \{\hat{H}_a\}_m^n) A(\alpha(0), \{\hat{H}_a\}_1^{m-1}) \langle \tilde{\alpha}(m-1) | \hat{A} | \alpha(m-1) \rangle \\ &= A(\alpha(0), \{\hat{H}_a\}_1^n) \frac{A(\tilde{\alpha}(m-1), \{\hat{H}_a\}_m^n)}{A(\alpha(m-1), \{\hat{H}_a\}_m^n)} \langle \tilde{\alpha}(m-1) | \hat{A} | \alpha(m-1) \rangle. \end{aligned} \quad (3.65)$$

In the last step, we have used the fact that

$$A(\alpha(0), \{\hat{H}_a\}_1^n) = A(\alpha(0), \{\hat{H}_a\}_1^{m-1}) \cdot A(\alpha(m-1), \{\hat{H}_a\}_m^n). \quad (3.66)$$

The thermal average then becomes:

$$\langle \hat{A} \rangle = \frac{1}{Z} \sum_{\alpha} \sum_{\{\hat{H}_a\}} \frac{\beta^n}{n!} \frac{1}{n} \sum_{m=1}^n A(\alpha(0), \{\hat{H}_a\}_1^n) \frac{A(\tilde{\alpha}(m-1), \{\hat{H}_a\}_m^n)}{A(\alpha(m-1), \{\hat{H}_a\}_m^n)} \langle \tilde{\alpha}(m-1) | \hat{A} | \alpha(m-1) \rangle. \quad (3.67)$$

Realize that the weight of configuration $(|\alpha\rangle, \{\hat{H}_a\})$, $W(\alpha, \{\hat{H}_a\})$, is simply $\frac{\beta^n}{n!} A(\alpha(0), \{\hat{H}_a\}_1^n)$.

We have

$$\langle \hat{A} \rangle = \frac{1}{Z} \sum_{\alpha} \sum_{\{\hat{H}_a\}} \frac{1}{n} \sum_{m=1}^n W(\alpha, \{\hat{H}_a\}) \frac{A(\tilde{\alpha}(m-1), \{\hat{H}_a\}_m^n)}{A(\alpha(m-1), \{\hat{H}_a\}_m^n)} \langle \tilde{\alpha}(m-1) | \hat{A} | \alpha(m-1) \rangle. \quad (3.68)$$

The ratio in the above equation is simply the relative weight of the two partial sequence.

Hence, $W(\alpha, \{\hat{H}_a\}) \frac{A(\tilde{\alpha}(m-1), \{\hat{H}_a\}_m^n)}{A(\alpha(m-1), \{\hat{H}_a\}_m^n)}$ could be considered as the transition probability.

If the off-diagonal update is initiated by the $\hat{A}$ operator at certain level m with probability $\frac{1}{n}$, then the measurement could be performed during the off-diagonal update and we just have to record $\langle \tilde{\alpha}(m) | \hat{A} | \alpha(m) \rangle$. The average of this measurement at any random level during each off-diagonal update gives the thermal average of $\hat{A}$.

3.2.5 Correlation functions

The correlation function of two operators $\hat{A}_1$ and $\hat{A}_2$ is defined as:

$$\langle \hat{A}_2(\tau) \hat{A}_1(0) \rangle = \langle e^{\tau \hat{H}} \hat{A}_2 e^{-\tau \hat{H}} \hat{A}_1 \rangle. \quad (3.69)$$

Here $\hat{H}$ is the original Hamiltonian. However, as we discussed before, we would use an effective Hamiltonian $(C - \hat{H})$ so that the weight of a configuration is always non-negative.

In terms of the effective Hamiltonian $\hat{H}$, the correlation function becomes:

$$\langle \hat{A}_2(\tau) \hat{A}_1(0) \rangle = \langle e^{-\tau \hat{H}} \hat{A}_2 e^{\tau \hat{H}} \hat{A}_1 \rangle = \frac{1}{Z} \langle e^{(\beta - \tau) \hat{H}} \hat{A}_2 e^{\tau \hat{H}} \hat{A}_1 \rangle \quad (3.70)$$

Taylor expansion is performed simultaneously on the two exponents, then we have:

$$\begin{aligned} \langle \hat{A}_2(\tau) \hat{A}_1(0) \rangle &= \frac{1}{Z} \sum_{\alpha} \sum_{m_1=0}^{\infty} \sum_{m_2=0}^{\infty} \frac{(\beta - \tau)^{m_1} \tau^{m_2}}{m_1! m_2!} \langle \alpha | \hat{H}^{m_1} \hat{A}_2 \hat{H}^{m_2} \hat{A}_1 | \alpha \rangle \\ &= \frac{1}{Z} \sum_{\alpha} \sum_{\{\hat{H}_a\}} \sum_{m=0}^n \frac{(\beta - \tau)^{n-m} \tau^m}{(n-m)! m!} \langle \alpha | \prod_{l=m+1}^n \hat{H}_{a(l)} \hat{A}_2 \prod_{l=1}^m \hat{H}_{a(l)} \hat{A}_1 | \alpha \rangle \end{aligned} \quad (3.71)$$

Diagonal operators

We first consider the case when both $\hat{A}_1$ and $\hat{A}_2$ are diagonal operators. We define:

$$A(l, m) = \langle \alpha(l+m) | \hat{A}_2 | \alpha(l+m) \rangle \langle \alpha(l) | \hat{A}_1 | \alpha(l) \rangle = a_2(l+m)a_1(l), \quad (3.72)$$

$$\bar{A}(m) = \frac{1}{n} \sum_{l=1}^n \langle \alpha(l+m) | \hat{A}_2 | \alpha(l+m) \rangle \langle \alpha(l) | \hat{A}_1 | \alpha(l) \rangle = \frac{1}{n} \sum_{l=1}^n a_2(l+m)a_1(l) \quad (3.73)$$

with $a_{1/2}(l) = \langle \alpha(l) | \hat{A}_{1/2} | \alpha(l) \rangle$ which is periodic $a_{1/2}(l+n) = a_{1/2}(l)$. The correlation function can then be written as

$$\langle \hat{A}_2(\tau) \hat{A}_1(0) \rangle = \frac{1}{Z} \sum_{\alpha} \sum_{\{\hat{H}_a\}} \sum_{m=0}^n B_n(\tau, m) W(\alpha, \{\hat{H}_a\}) A(0, m) \quad (3.74)$$

where $W(\alpha, \{\hat{H}_a\})$ is the weight of the configuration $(\alpha, \{\hat{H}_a\})$ with non-truncated sequences and $B_n(\tau, m)$ is a binomial distribution:

$$B_n(\tau, m) = (1-r)^{n-m} r^m \binom{n}{m} \quad \text{with} \quad r = \frac{\tau}{\beta}. \quad (3.75)$$

Using the property of the trace as before, we can average over all cyclic permutation of the operator sequence in Eq.3.71 and we will have

$$\sum_{\alpha} \sum_{\{\hat{H}_a\}} W(\alpha, \{\hat{H}_a\}) A(0, m) = \sum_{\alpha} \sum_{\{\hat{H}_a\}} W(\alpha, \{\hat{H}_a\}) \bar{A}(m). \quad (3.76)$$

The correlation function becomes:

$$\langle \hat{A}_2(\tau) \hat{A}_1(0) \rangle = \frac{1}{Z} \sum_{\alpha} \sum_{\{\hat{H}_a\}} W(\alpha, \{\hat{H}_a\}) \left(\sum_{m=0}^n B_n(\tau, m) \bar{A}(m) \right). \quad (3.77)$$

Now the weight $W(\alpha, \{\hat{H}_a\})$ has the form as in Equation 3.40.

The above equation implies that the correlation function is simply the thermal average of $\sum_{m=0}^n B_n(\tau, m) \bar{A}(m)$. The generalized susceptibility can be obtained straight forwardly

by integrating over τ :

$$\chi(\beta) = \int_0^\beta \langle \hat{A}_2(\tau) \hat{A}_1(0) \rangle d\tau. \quad (3.78)$$

The integration over the binomial distribution $B_n(\tau, m)$ is given by

$$\int_0^\beta B_n(\tau, m) d\tau = \beta \binom{n}{m} \int_0^\beta (1-r)^{n-m} r^m d\tau = \frac{\beta}{n+1}. \quad (3.79)$$

Hence, the susceptibility could be given by:

$$\begin{aligned} \chi(\beta) &= \frac{1}{Z} \sum_{\alpha} \sum_{\{\hat{H}_a\}} W(\alpha, \{\hat{H}_a\}) \frac{\beta}{n(n+1)} \left\{ \sum_{l=1}^n a_2(l) a_1(l) + \left(\sum_{l=1}^n a_2(l) \right) \left(\sum_{l=1}^n a_1(l) \right) \right\} \\ &= \left\langle \frac{\beta}{n(n+1)} \sum_{l=1}^n a_2(l) a_1(l) \right\rangle + \left\langle \frac{\beta}{n(n+1)} \left(\sum_{l=1}^n a_2(l) \right) \left(\sum_{l=1}^n a_1(l) \right) \right\rangle. \end{aligned} \quad (3.80)$$

If we work with sequence of fixed length L , we can either record $a_{1/2}(l)$ only at levels with H-operators or use the following:

$$\chi(\beta) = \left\langle \frac{\beta}{L(n+1)} \sum_{l=1}^L a_2(l) a_1(l) \right\rangle + \left\langle \frac{\beta}{L(n+1)} \left(\sum_{l=1}^L a_2(l) \right) \left(\sum_{l=1}^L a_1(l) \right) \right\rangle \quad (3.81)$$

where $(n+1)$ remains unchanged because it comes from the integration 3.79.

Off-diagonal operators

Now we consider the case when $\hat{A}_{1/2}$ are off-diagonal. Specifically, we only consider those operators that could initiate the off-diagonal update in SSE, for example, annihilation and creation operators in bosonic systems or spin ladder operators in spin systems. As before, we can define

$$a_{1/2}(l) = \langle \alpha'(l) | \hat{A}_{1/2} | \alpha(l) \rangle \quad \text{with} \quad a_{1/2}(l+n) = a_{1/2}(l), \quad (3.82)$$

$$D(m) = \frac{1}{n} \sum_{l=1}^n a_2(l+m) a_1(l), \quad (3.83)$$

$$A(\alpha(l_1-1), \{\hat{H}_a\}_{l_1}^{l_2}) = \langle \alpha(l_2) | \hat{H}_{a(l_2)} | \alpha(l_2-1) \rangle \cdots \langle \alpha(l_1) | \hat{H}_{a(l_1)} | \alpha(l_1-1) \rangle. \quad (3.84)$$

The correlation function then becomes

$$\begin{aligned} \langle \hat{A}_2(\tau) \hat{A}_1(0) \rangle &= \frac{1}{Z} \sum_{\alpha} \sum_{\{\hat{H}_a\}} \sum_{m=0}^n \sum_{l=1}^n \frac{1}{n} B_n(\tau, m) A(\alpha, \{\hat{H}_a\}_1^l) A(\alpha'(l), \{\hat{H}_a\}_{l+1}^{l+m}) \\ &\quad \cdot A(\alpha''(l+m), \{\hat{H}_a\}_{l+m+1}^n) a_2(l+m) a_1(l) \end{aligned} \quad (3.85)$$

where we have again used the property of the trace. As we have discussed in the single off-diagonal operator, this could actually be written in terms of the transition probability between two configurations.

$$\begin{aligned} \langle \hat{A}_2(\tau) \hat{A}_1(0) \rangle &= \frac{1}{Z} \sum_{\alpha} \sum_{\{\hat{H}_a\}} \sum_{m=0}^n \sum_{l=1}^n \frac{1}{n} B_n(\tau, m) W(\alpha, \{\hat{H}_a\}) a_2(l+m) a_1(l) \\ &\quad \cdot \frac{A(\alpha'(l), \{\hat{H}_a\}_{l+1}^{l+m}) A(\alpha''(l+m), \{\hat{H}_a\}_{l+m+1}^n)}{A(\alpha(l), \{\hat{H}_a\}_{l+1}^{l+m}) A(\alpha(l+m), \{\hat{H}_a\}_{l+m+1}^n)}. \end{aligned} \quad (3.86)$$

The above equation provides a way to calculate the correlation function. In each off-diagonal update step, we can randomly select a level with probability $\frac{1}{n}$ and then update the configuration according to the transition probability in the above equation. During the updating, we can record $B_n(\tau, m) a_2(l+m) a_1(l)$ whenever they appear at the corresponding levels. Hence, the correlation function is given by the average:

$$\langle \hat{A}_2(\tau) \hat{A}_1(0) \rangle = \left\langle \sum_{m=0}^n B_n(\tau, m) a_2(l+m) a_1(l) \right\rangle_T. \quad (3.87)$$

The subscript T means the average is taken during the transition procedures or off-diagonal updates. There are at least three ways to evaluate the above summation[24]. The most efficient way is to split the summation into different intervals where $a_2(l+m) a_1(l)$ is the same in each interval. For each interval $I = [I_1, I_2]$, we define

$$G(\tau, I) = \left(\sum_{m(I)} B_n(\tau, m(I)) \right) a_2(I_2) a_1(I_1) \quad (3.88)$$

where $m(I)$ is the level differences that the interval I passes. For example, if $I = [2, 4]$, then $m(I) \in \{0, 1, 2\}$. The correlation function then becomes:

$$\langle \hat{A}_2(\tau) \hat{A}_1(0) \rangle = \left\langle \sum_I G(\tau, I) \right\rangle_T. \quad (3.89)$$

In practice, we actually work with sequences of fixed length L . Because the insertion of the identity operators does not affect the binomial factor, it still depends on the number of H-operators in the sequence. Equation 3.88 and 3.89 remains the same except that $m(I)$ now only counts the position difference in the H-operators, viz., it counts the level difference in the reduced sequence (removing all the identity operators).

3.3 Anisotropic Spin-1 Heisenberg model

In this section, we demonstrate the details of the updating procedures in SSE by applying it to the anisotropic Spin-1 Heisenberg model with single-ion anisotropy[22]. The results of this calculation will be shown in Chapter 4 where we compare the results obtained from SSE and spin wave calculation.

3.3.1 Decomposition of Hamiltonian

The Spin-1 Heisenberg model with single-ion anisotropy is described by the Hamiltonian:

$$\hat{H} = J \sum_{\langle i, j \rangle} \left[\frac{1}{2} \left(\hat{S}_i^+ \hat{S}_j^- + \hat{S}_j^+ \hat{S}_i^- \right) + \Delta \hat{S}_i^z \hat{S}_j^z \right] + D \sum_i \left(\hat{S}_i^z \right)^2 - h \sum_i \hat{S}_i^z \quad (3.90)$$

$\langle i, j \rangle$ means nearest neighbours. Δ is the spin anisotropy and D is the single-ion anisotropy coming from the strong crystal field. h is the external magnetic field. We only consider bipartite lattices up to 3 dimensions, that is, a line, a square lattice and a cubic lattice. In such a lattice, we can perform a spin rotation about $\hat{S}_y$ direction in one sublattice. Such transformation will reverse the sign of the spin ladder operators $\hat{S}^\pm$ in that sublattice.

We can then rewrite the Hamiltonian in the bond basis as:

$$\hat{H} = \sum_b \left\{ -\frac{J}{2}(\hat{S}_{b_1}^- \hat{S}_{b_2}^- + \hat{S}_{b_1}^- \hat{S}_{b_2}^+) + \Delta J \hat{S}_{b_1}^z \hat{S}_{b_2}^z + \frac{D}{2d} [(\hat{S}_{b_1}^z)^2 + (\hat{S}_{b_2}^z)^2] - \frac{h}{2d}(\hat{S}_{b_1}^z + \hat{S}_{b_2}^z) \right\} \quad (3.91)$$

where d is the dimension of the lattice. b_1 and b_2 are the two lattice points connected by the bond b . The basis we use here consists of eigen states of the $\hat{S}^z$ operator: $\hat{S}^z |s\rangle = s |s\rangle$ with $s \in \{0, \pm 1\}$. The Hamiltonian can be decomposed into:

$$\hat{H} = dNC - \sum_{a=1}^3 \sum_b \hat{H}_{a,b}, \quad (3.92)$$

$$\hat{H}_{1,b} = C - \left\{ \Delta J \hat{S}_{b_1}^z \hat{S}_{b_2}^z + \frac{D}{2d} [(\hat{S}_{b_1}^z)^2 + (\hat{S}_{b_2}^z)^2] - \frac{h}{2d}(\hat{S}_{b_1}^z + \hat{S}_{b_2}^z) \right\}, \quad (3.93)$$

$$\hat{H}_{2,b} = \frac{J}{2} \hat{S}_{b_1}^+ \hat{S}_{b_2}^-, \quad \hat{H}_{3,b} = \frac{J}{2} \hat{S}_{b_1}^- \hat{S}_{b_2}^+. \quad (3.94)$$

C is a constant chosen such that the diagonal operator $\hat{H}_{1,b}$ is positive definite. The minimum value of the diagonal operator is $\langle -1, -1 | \hat{H}_{1,b} | -1, -1 \rangle$. Thus C could be chosen as

$$C = \Delta J + \frac{D}{d} + \frac{h}{d} + \epsilon \quad (3.95)$$

where ϵ is a small positive constant. A nonzero ϵ could reduce the bounce probability in the off-diagonal update, however, the length of the operator sequence L increases as ϵ [27].[†] No general rule could decide the magnitude of ϵ .

To work with sequence with fixed length L , we introduce the identity operator denoted as

$$\hat{H}_{0,0} = \hat{I}. \quad (3.96)$$

The weight of a configuration $\{\alpha, \{\hat{H}_{a,b}\}\}$ is then given by (3.40):

$$W(\alpha, \{a\}) = \frac{\beta^n (L-n)!}{L!} \prod_{l=1}^L \langle \alpha(l) | \hat{H}_{a(l),b(l)} | \alpha(l-1) \rangle. \quad (3.97)$$

[†]Here bounce means that the updated state comes back to its original state immediate after the update.

We call $\langle \alpha(l) | \hat{H}_{a(l),b(l)} | \alpha(l-1) \rangle$ the weight of the **vertex** that consists of the H-operator $\hat{H}_{a(l),b(l)}$ and the two propagated states it connects. From the property of the H-operator, only the following vertices have non-zero contribution.

$$\langle s_1, s_2 | \hat{H}_{1,b} | s_1, s_2 \rangle = C - \left(\Delta J s_1 s_2 + \frac{D}{2d} (s_1^2 + s_2^2) - \frac{h}{2d} (s_1 + s_2) \right) \text{ for all } s_{1/2}, \quad (3.98)$$

$$\langle s_1 + 1, s_2 - 1 | \hat{H}_{2,b} | s_1, s_2 \rangle = J \quad \text{for } s_1 \in \{-1, 0\} \text{ and } s_2 \in \{0, 1\}, \quad (3.99)$$

$$\langle s_1 - 1, s_2 + 1 | \hat{H}_{3,b} | s_1, s_2 \rangle = J \quad \text{for } s_1 \in \{0, 1\} \text{ and } s_2 \in \{-1, 0\}. \quad (3.100)$$

3.3.2 Construct the lattice

We should implement the lattice configuration in computational language. There are $N = L_1 L_2 \cdots L_d$ lattice points in a d -dimensional bipartite lattice with size L_α in each direction α ($\alpha = 1, 2, \dots, d$). The position of the i^{th} lattice point is represented by a d -dimensional vector:

$$\mathbf{r}_i = (x_{1,i}, x_{2,i}, \dots, x_{d,i}) \quad \text{with } x_{\alpha,i} \in \{0, 1, 2, \dots, L_\alpha - 1\}. \quad (3.101)$$

The lattice satisfies the periodic boundary condition along each direction. We can store the position in terms of i instead of $\mathbf{r}_i$ to reduce the memory use:

$$i(\mathbf{r}_i) = 1 + x_{1,i} + x_{2,i} L_1 + x_{3,i} L_2 L_1 + \cdots + x_{\alpha,i} L_{\alpha-1} \cdots L_1. \quad (3.102)$$

And each component $x_{\alpha,i}$ can be expressed as

$$x_{\alpha,i} = \left\lfloor \frac{i(\mathbf{r}_i) - 1}{L_0 L_1 L_2 \cdots L_{\alpha-1}} \right\rfloor \mod L_\alpha \quad (3.103)$$

where $\lfloor x \rfloor$ is the floor function. For convenience, we have also defined $L_0 = 1$ in the above equation.

There are in all $B = dN$ number of bonds in a d -dimensional bipartite lattice system. We can assign a value b to a bond starting from lattice site i and pointing to the positive

α direction:

$$b = (\alpha - 1)N + i \quad \text{if the bond is along } \alpha \text{ direction.} \quad (3.104)$$

Hence, given a bond number b , we can obtain the direction of the bond by

$$\alpha(b) = \lfloor \frac{b}{N} \rfloor + 1. \quad (3.105)$$

The two lattice sites connected by bond b can be labeled as $i(b, 1)$ and $i(b, 2)$ with $i(b, 1) < i(b, 2)$ and they can be obtained as

$$i(b, 1) = b \mod N, \quad (3.106)$$

$$i(b, 2) = i + L_0 L_1 L_2 \cdots L_{\alpha-1} \quad \text{where } \alpha \text{ is the direction of the bond.} \quad (3.107)$$

We have constructed the lattice system and all the information could be extracted from the bond number b using the above formulas. Construction of the lattice system is usually done at the beginning of the computation.

After the lattice is constructed, we can load the spin states on the lattice. We assign each lattice site i a spin variable $sp(i) \in \{-1, 0, 1\}$. The two spin states on each bond b are then given by $sp(i(b, 1))$ and $sp(i(b, 2))$.

3.3.3 Diagonal update

During each Monte Carlo step, the diagonal update is performed once followed by many off-diagonal update. As described in the previous section, the diagonal update changes the number of H-operators in the sequence. At the very beginning of the computation when the operator sequence is initialized, we set the truncation value L to a small number, for example 10. The operator sequence consists only identity operators at the beginning. The spin configurations $sp(i)$ is initiated randomly. The operator sequence length L will be updated as described in Sec.3.2.2 during each Monte Carlo step and finally become stable at certain large value after many Monte Carlo steps. By this time, the operator sequence and the spin configuration could be considered completely random and are not

affected by the initial setting. The above procedure is called **equilibration**. When the system is in equilibrium, we can perform measurement during the Monte Carlo steps.

There are three kinds of operators as shown in Equation 3.93 and 3.94. We can store the operator at level l , $\hat{H}_{a(l),b(l)}$, into an array of integers:

$$OPS(l) = \begin{cases} 3b(l) + a(l) - 1 & \text{for H-operators,} \\ 0 & \text{for identity operator.} \end{cases} \quad (3.108)$$

so that the type $a(l)$ and the bond number $b(l)$ of an H-operator can be extracted easily:

$$a(l) = (OPS(l) \bmod 3) + 1 \quad \text{and} \quad b(l) = \lfloor \frac{OPS(l)}{3} \rfloor. \quad (3.109)$$

Since there is at most one H-operator between two propagated states, we can label the propagated state explicitly with the bond number as

$$|\alpha_{b(l)}\rangle = |sp(i(b, 1)), sp(i(b, 2))\rangle. \quad (3.110)$$

During each diagonal update, we start from $l = 1$. If $OPS(l) = 0$ which means there is no H-operator at level l , we can insert a diagonal operator at bond b with the probability:

$$P(OPS(l) = 0 \rightarrow OPS(l) = 3b) = \min \left(\frac{B\beta}{L-n} \langle \alpha_{b(l)} | \hat{H}_{1,b} | \alpha_{b(l-1)} \rangle, 1 \right) \quad (3.111)$$

where n is the number of H-operator in the sequence before insertion, as explained in the previous section. On the hand, if $OPS(l)$ is not zero, we can obtain its type and bond number through Equation 3.109. If it is a diagonal operator, we try to remove it with probability

$$P(OPS(l) = 3b \rightarrow OPS(l) = 0) = \min \left(\frac{L-n-1}{B\beta \langle \alpha_{(l)} | \hat{H}_{1,b} | \alpha_{b(l-1)} \rangle}, 1 \right). \quad (3.112)$$

If it is an off-diagonal operator, we change the spin configurations accordingly so that we don't have to store the spin configurations of all the propagated states.

3.3.4 Loop update

After each diagonal update, many off-diagonal update steps are performed. Because the configuration of the sample element is a closed string, Fig.3.1, this allows us to change many H-operators at a time. And these H-operator actually form a closed loop in the extended lattice with the propagation level as one extra dimension, as we will see in the following.

There are three types of H-operators and we denote them with arrow diagrams: $\hat{H}_{1,b} : \leftrightarrow$, $\hat{H}_{2,b} : \leftarrow$ and $\hat{H}_{3,b} : \rightarrow$. For illustration purpose, we consider a one dimensional lattice with bond number $B = 5$ and operator sequence length $L = 6$. An example of the configuration is given below.

$sp(1)$	$sp(2)$	$sp(3)$	$sp(4)$	$sp(5)$	$sp(6)$		l	$OPS(l)$
1	-1	0	1	1	0	$ \alpha(0)\rangle$		
$\longleftrightarrow$							1	3
1	-1	0	1	1	0	$ \alpha(1)\rangle$		
		$\longleftarrow$					2	10
1	-1	1	0	1	0	$ \alpha(2)\rangle$		
			$\longleftrightarrow$				3	12
1	-1	1	0	1	0	$ \alpha(3)\rangle$		
$\longleftrightarrow$							4	3
1	-1	1	0	1	0	$ \alpha(4)\rangle$		
				$\longleftrightarrow$			5	15
1	-1	1	0	1	0	$ \alpha(5)\rangle$		
		$\longrightarrow$					6	11
1	-1	0	1	1	0	$ \alpha(6)\rangle$		

From the above diagram, we can see that the configuration is a distribution of the three vertices defined previously:

$$\begin{array}{ccc}
 \begin{array}{c} s_1 \quad s_2 \\ \longleftarrow \\ s_1 + 1 \quad s_2 - 1 \end{array} &
 \begin{array}{c} s_1 \quad s_2 \\ \longleftrightarrow \\ s_1 \quad s_2 \end{array} &
 \begin{array}{c} s_1 \quad s_2 \\ \longrightarrow \\ s_1 - 1 \quad s_2 + 1 \end{array}
 \end{array}$$

The weight of the configuration is simply the product of the weights of the vertices. Thus we can just store the information of each vertex instead of spin configurations in all levels. Along the propagation worldline, some local spins remain the same for a few levels. We

could replace them with straight lines or links between two vertices, as shown below.

The above configuration is called the linked vertices. The links between difference vertices are stored in an array.

When we start loop update, we randomly pick up a vertex and try to change the spin at one of its corners. This results in four possible changes as shown in Fig.3.2. The

Figure 3.2: The upper left corner is chosen to be the starting point. The changes are similar starting from different corners.

outgoing arrow means that the spin at that corner has been changed accordingly and this change will be carried forward to the vertex that it connects through the vertex link.

The choice of the exit corner is chosen according to their relative weights.

An example of the loop update is shown above. The lower left corner of the first vertex is chosen as the starting point and the exit is chosen to be the lower right corner. After the change, the vertex changes from a diagonal H-operator to an off-diagonal operator. However, the link is broken now. Hence, this change has to be carried forward to the upper right corner of $OPS(4)$. Similarly, an exit will be chosen according to the relative weights. This new exit will carry the change to the next vertex and the procedure is repeated until it comes back to the starting corner when the loop is closed, as shown below.

The loop update procedure is performed repeatedly a few times before we go into another diagonal updates.

3.4 Generalized Shastry-Sutherland model

In this section, we implement the SSE method to the generalized Shastry-Sutherland model. We will focus on the Hamiltonian decomposition and construction of the lattice since this is not a bipartite lattice. The updating procedures are more or less the same as in the Spin-1 model, thus we will not discuss them here.

The Hamiltonian is given by

$$\hat{H} = \sum_{i,j} \left[-\frac{1}{2} J_{ij} (\hat{S}_i^+ \hat{S}_j^- + \hat{S}_i^- \hat{S}_j^+) + \Delta J_{ij} \hat{S}_i^z \hat{S}_j^z \right] - h \sum_i \hat{S}_i^z. \quad (3.113)$$

There are four types of bonds as illustrated in Introduction.

$$\hat{H} = \sum_{b_i} \left\{ -\frac{J_i}{2} (\hat{S}_{b_i1}^+ \hat{S}_{b_i2}^- + \hat{S}_{b_i1}^- \hat{S}_{b_i2}^+) + \Delta J_i \hat{S}_{b_i1}^z \hat{S}_{b_i2}^z \right\} - h \sum_{b_2} (\hat{S}_{b_21}^z + \hat{S}_{b_22}^z) \quad (3.114)$$

We can decompose the Hamiltonian in the following way

$$\hat{H}_{1,b_i} = C_i - \Delta J_i \hat{S}_{b_i1}^z \hat{S}_{b_i2}^z \quad \text{for } i = 1, 3, 4, \quad (3.115)$$

$$\hat{H}_{1,b_2} = C_2 - \Delta J_i \hat{S}_{b_21}^z \hat{S}_{b_22}^z + h(\hat{S}_{b_21}^z + \hat{S}_{b_22}^z), \quad (3.116)$$

$$\hat{H}_{2,b_i} = \frac{1}{2} J_i (\hat{S}_{b_i1}^+ \hat{S}_{b_i2}^- + \hat{S}_{b_i1}^- \hat{S}_{b_i2}^+), \quad \text{for } i = 1, 2, 3, 4. \quad (3.117)$$

It is straight forward to count the number of different bonds in a square lattice with lattice site number $N = L_x L_y$:

$$B_1 = 2N, \quad B_2 = \frac{1}{2}N, \quad B_3 = N \quad \text{and} \quad B_4 = 2N. \quad (3.118)$$

Both L_x and L_y are even numbers and periodic boundary conditions are applied. The Hamiltonian has the the following decomposition:

$$\hat{H} = \sum_{i=1}^4 B_i C_i - \sum_{a=1}^3 \sum_{b_i} \hat{H}_{a,b_i} \quad (3.119)$$

The constants C_i are given by

$$C_i = \frac{1}{4}\Delta J_i + \epsilon_i \quad \text{for } i = 1, 3, 4 \quad \text{and} \quad C_2 = \frac{1}{4}\Delta J_2 + h + \epsilon_2. \quad (3.120)$$

Now we implement the lattice structures. The J_1 bond is the same as the Spin-1 model, so we have

$$b_1 = (\alpha - 1)N + i \quad \text{if the bond is along } \alpha \text{ direction.} \quad (3.121)$$

$$\alpha(b_1) = \lfloor \frac{b_1}{N} \rfloor + 1, \quad (3.122)$$

$$i(b_1, 1) = b_1 \mod N, \quad i(b_1, 2) = \begin{cases} i + 1 & \text{if } \alpha \text{ is along } x \text{ direction} \\ i + L_x & \text{if } \alpha \text{ is along } y \text{ direction.} \end{cases} \quad (3.123)$$

For J_2 bond, we define the bond numbers in the following manner:

$$b_2 = i, \quad \text{for } i = (2m - 1) + 2(n - 1)L_x \quad (3.124)$$

$$b_2 = i + \frac{N}{4}, \quad \text{for } i = (2m - 1) + (2n - 1)L_x \quad (3.125)$$

where $m = 1, 2, \dots, \frac{L_x}{2}$ and $n = 0, 1, \dots, \frac{L_x}{2} - 1$. The type of the J_2 bond and the two lattice sites it connects could be extracted from b_2 as:

$$\alpha(b_2) = \lfloor \frac{4b_2}{N} \rfloor + 1, \quad (3.126)$$

$$i(b_2, 1) = b_2 \mod \frac{N}{4}, \quad (3.127)$$

$$i(b_2, 2) = \begin{cases} i + L_x + 1 & \text{if } \alpha = 1, \text{ type A } J_2 \text{ bond,} \\ i + L_x - 1 & \text{if } \alpha = 2, \text{ type B } J_2 \text{ bond.} \end{cases} \quad (3.128)$$

There are two types of J_3 bonds as well. We assign a value to the b_3 according to:

$$b_3 = i, \quad \text{for odd } i, \quad (3.129)$$

$$b_3 = i + \frac{N}{2}, \quad \text{for even } i. \quad (3.130)$$

$$\alpha(b_3) = \lfloor \frac{2b_3}{N} \rfloor + 1, \quad (3.131)$$

$$i(b_1, 1) = b_3 \mod \frac{N}{2}, \quad i(b_3, 2) = \begin{cases} i + L_x + 1 & \text{for odd } i, \\ i + L - x - 1 & \text{for even } i. \end{cases} \quad (3.132)$$

J_4 bonds are similar to the J_2 bonds which can be constructed as:

$$b_4 = (\alpha - 1)N + i \quad \text{if the bond is along } \alpha \text{ direction.} \quad (3.133)$$

$$\alpha(b_4) = \lfloor \frac{b_4}{N} \rfloor + 1, \quad (3.134)$$

$$i(b_4, 1) = b_4 \mod N, \quad i(b_4, 2) = \begin{cases} i + 2 & \text{if } \alpha \text{ is along } x \text{ direction} \\ i + 2L_x & \text{if } \alpha \text{ is along } y \text{ direction.} \end{cases} \quad (3.135)$$

After the lattice structure is initiated at the beginning, the updating procedures can then be performed as we did in Spin-1 Heisenberg model. For simplicity, the spin numbers are stored in an integer array $sp(i) = 2s_i$. We note that only the following vertices have contributions:

$$\langle s_1, s_2 | \hat{H}_{1,b_i} | s_1, s_2 \rangle = C_i - s_1 s_2 \Delta J_i \quad \text{for } i = 1, 3, 4, \quad (3.136)$$

$$\langle s_1, s_2 | \hat{H}_{1,b_2} | s_1, s_2 \rangle = C_2 - s_1 s_2 \Delta J_2 + h(s_1 + s_2), \quad (3.137)$$

$$\left\langle \frac{1}{2}, -\frac{1}{2} \middle| \hat{H}_{2,b_i} \middle| -\frac{1}{2}, \frac{1}{2} \right\rangle = \frac{1}{2} J_i \quad \text{and} \quad \left\langle -\frac{1}{2}, \frac{1}{2} \middle| \hat{H}_{2,b_i} \middle| \frac{1}{2}, -\frac{1}{2} \right\rangle = \frac{1}{2} J_i \quad \text{for all } i. \quad (3.138)$$

Chapter 4

Anisotropic Spin-One Magnets

Using Quantum Monte Carlo method, we can perform ideal computational experiments and obtain very accurate numerical results. And analytical methods provide a tool to understand the real physical configurations and the mechanism behind the phenomena observed numerically. Almost all analytical methods come with some approximations in one way or another. If the approximation is simply the guess of the ground state, comparison between the results from the two methods will give a complete picture of the model. Unfortunately, in most of the cases, other approximations have to be adopted so that the analytical methods are able to produce practical results. The consistency between numerical and analytical results implies the applicability and stability of the analytical method. Therefore, it is very important to know the applicability of the analytical method under various approximations so that we are able to determine a suitable analytical approach. In this chapter, we use spin wave method and Stochastic Series Expansion QMC method to explore the phase diagram and investigate the magnetic excitations of an anisotropic spin-one Heisenberg model[22].

4.1 The spin-one model

Recently, there has been a renewed interest in the study of magnetic-field-induced quantum phase transitions in spin-one magnets with strong single-ion and exchange anisotropies[30, 31, 32, 33, 34, 35, 36, 37]. The discovery of $S = 1$ compounds, such as Y_2BaNiO_5 or the

organometallic frameworks $[\text{Ni}(\text{C}_2\text{H}_8\text{N}_2)_2(\text{NO}_2)]\text{ClO}_4$ (NENP), $[\text{Ni}(\text{C}_2\text{H}_8\text{N}_2)_2\text{Ni}(\text{CN}_4)]$ (NENC), and $[\text{NiCl}_2-4\text{SC}(\text{NH}_2)_2]$ (DTN), fueled experimental and theoretical studies of the role of dimensionality and singlet ion anisotropy[30, 33, 34, 35, 36, 37, 38, 39, 40, 41]. In most of the known $S = 1$ magnets, the ubiquitous Heisenberg exchange is complemented by single-ion anisotropy. The interplay between these interactions with external magnetic field and lattice geometry can result in a rich variety of quantum phases and phenomena, including the Haldane phase of quasi-one-dimensional (1D) systems[42], field-induced Bose Einstein condensation (BEC) of magnetic states[30, 31, 32, 33, 34, 35, 36, 37] and field-induced ferronematic ordering[43]. Interest in $S = 1$ Heisenberg antiferromagnets with uniaxial exchange and single-ion anisotropies has gained additional impetus recently after it was shown to exhibit the spin analog of the elusive supersolid phase on a lattice over a finite range of magnetic fields[44, 45, 46].

In contrast to its classical counterpart ($S \rightarrow \infty$), $S = 1$ systems become quantum paramagnets (QPM) for sufficiently strong easy-plane single-ion anisotropy. In other words, the order does not survive at zero temperature $T = 0$ because the dominant anisotropy term $D \sum_{\mathbf{r}} (S_{\mathbf{r}}^z)^2$ ($D > 0$) forces each spin to be predominantly in the non-magnetic $|S_{\mathbf{r}}^z = 0\rangle$ state: $\langle S_{\mathbf{r}}^z = 0 | S_{\mathbf{r}}^\alpha | S_{\mathbf{r}}^z = 0 \rangle = 0$ for $\alpha = \{x, y, z\}$. The application of a magnetic field h along the z -axis reduces the spin gap linearly in h since the field couples to a conserved quantity (total magnetization along the z -axis). The gap is closed at a quantum critical point (QCP) where the bottom of the $S^z = 1$ branch of magnetic excitations touches zero. This QCP belongs to the BEC universality class, and the gapless mode of low-energy $S^z = 1$ excitations remains quadratic for small momenta $\omega \propto k^2$ because the Zeeman term commutes with the rest of the Hamiltonian. Since the dynamical exponent is $z = 2$, the effective dimension is $d + 2$ and the upper critical dimension is $d_c = 2$. This and analogous field-driven transitions have been widely studied experimentally to demonstrate BEC-related phenomena in many quantum magnets[30, 34, 47, 48, 49]. One of these magnets is the metal-organic framework DTN that we mentioned above.

The starting point of any theoretical study of a magnetic- field-induced phase transition in a QPM is to determine the Hamiltonian parameters, that is, the exchange

constants and the amplitude of the different anisotropies. The simplest way of extracting these parameters is to fit the branches of magnetic excitations that are measured with inelastic neutron scattering (INS). The reliability of this procedure is normally limited by the accuracy of the approach that is used to compute the dispersion relation of magnetic excitations. Numerical methods such as quantum Monte Carlo (QMC) and density matrix renormalization group (DMRG) are very accurate, but they can only be applied under special circumstances. While the DMRG method[50] has evolved to the extent that dynamical properties such as the frequency and momentum dependence of the magnetic structure factor can be computed very accurately[51], its application is restricted to quasi-one-dimensional magnets such as HPIP-CuBr₄. On the other hand, QMC methods can only be applied to systems that have no frustration in the exchange interaction, i.e., that are free of the infamous sign problem. Consequently, it is necessary to find simple analytical approaches that are accurate enough to quantitatively reproduce the quantum phase diagram and the dispersion of magnetic excitations.

As introduced in the very beginning, one of the purposes of this chapter is to test different analytical approaches against the results of accurate QMC simulations of a spin-one Heisenberg Hamiltonian with easy-plane single-ion anisotropy. The model is defined either on a square or on a cubic lattice to avoid frustration and make the QMC method applicable. Aside from being relevant for describing real quantum magnets, such as DTN, this model provides one of the simplest realizations of quantum paramagnetism and is ideal for testing methods that can be naturally extended to more complex systems.

The generic $S = 1$ Heisenberg model with uniaxial single-ion anisotropy on an isotropic hypercubic lattice is given by the Hamiltonian

$$\hat{H} = J \sum_{\langle i,j \rangle} \hat{S}_i^\alpha \hat{S}_j^\alpha + \sum_i (D \hat{S}_i^{z^2} - h \hat{S}_i^z) \quad (4.1)$$

where repeated index α is summed over $\{x, y, z\}$. $\langle i, j \rangle$ means nearest neighbour. D is the strength of the single-ion anisotropy and h is the external magnetic field.

The (D, h) quantum phase diagram of this Hamiltonian is well known from mean

field analysis[52, 53, 54], series expansion studies[55] and numerical simulations[56]. The D term splits the local spin states into $|S^z = 0\rangle$ and $|S^z = \pm 1\rangle$ doublet. As we explained above, the ground state is a quantum paramagnet for large $D \gg 1$, i.e., it has no long-range magnetic order and there is a finite-energy gap to spin excitations. At finite magnetic fields, the Zeeman term lowers the energy of the $|S^z = 1\rangle$ state until the gap closes at a critical field h_c . A canted antiferromagnetic (CAFM) phase appears right above h_c : the spins acquire a uniform longitudinal component and an antiferromagnetically ordered transverse component that spontaneously breaks the $U(1)$ symmetry of global spin rotations along the z axis. The CAFM phase can also be described as a condensation of bosonic particles. The particle density $n_{\mathbf{r}}$ is related to the local magnetization along the symmetry axis $n_{\mathbf{r}} = S_{\mathbf{r}}^z + 1$. Therefore, the magnetic field acts as a chemical potential in the bosonic description. For $h > h_c$, the system is populated by a finite density of bosons that condense in the single-particle state with momentum Q with $Q_\alpha = \pi$, ($\alpha = \{x, y, z\}$). The longitudinal magnetization (density of bosons) increases with field and saturates at the fully polarized (FP) state ($S_{\mathbf{r}}^z = 1 \forall \mathbf{r}$) above the saturation field h_s . The FP state corresponds to a bosonic Mott insulator in the language of Bose gases. There exists a critical value of the single-ion anisotropy D_c , below which the CAFM phase extends down to zero field. The nature of the QPM-CAFM quantum phase transition changes between $h = 0$ and $h \neq 0$. The transition belongs to the BEC universality class for $h \neq 0$, while it belongs to the $O(2)$ universality class for $h = 0$.

The details of the spin wave approach have been demonstrated in Chapter 2, thus we will focus on the results from different approximations, Holstein-Primakoff (HP) and Lagrangian multiplier (LM) method, and comparison with the QMC method.

4.2 QPM phase and the fully polarized phase

4.2.1 Holstein-Primakoff approximation

At the mean field level, the paramagnetic (QPM) state

$$|\psi_{QPM}\rangle = \prod_i |S_i^z = 0\rangle = \prod_i \hat{b}_{i0}^\dagger |\emptyset\rangle \quad (4.2)$$

that is the global unitary transformation in (2.18) is the trivial identity matrix. Under Holstein-Primakoff approximation, the quasiparticle dispersion becomes particularly simple in the QPM phase:

$$\omega_{\mathbf{k},\pm} = \sqrt{D^2 + 2Df(\mathbf{k})} \pm h, \quad f(\mathbf{k}) = -2J \sum_{\alpha} \cos k_{\alpha}. \quad (4.3)$$

Both branches have the same dispersion at zero field, as expected from time-reversal symmetry. A finite magnetic field h splits the branches linearly in h without changing the dispersion. This is a consequence of the fact that the external field couples to the total magnetization which is a conserved quantity. Both branches have a minimum at the AFM wave vector $\mathbf{k} = (0,0)$ (after sub-lattice rotation) that determines the size of the gap. The dispersion is quadratic near $\mathbf{k} = (0,0)$ except for the critical point ($D_c = 4dJ$, $h = 0$) that separates the QPM phase from the CAFM phase at $h = 0$. The field induced QCP then belongs to the BEC universality class in dimension $d + 2$, where d is the dimension of the lattice. By expanding around $\mathbf{k} = (0,0)$, we obtain

$$\omega_{\mathbf{k},\pm} \approx Jk^2 \sqrt{D/(D - D_c)} + \sqrt{D(D - D_c)} \pm h. \quad (4.4)$$

It is clear from this expression that the effective mass of the magnetic excitations vanishes for $D \rightarrow D_c$: $m^* \propto \sqrt{D - D_c}$. This is indeed the expected behavior if we keep in mind that the dispersion must be linear at the critical point ($D_c = 4dJ$, $h = 0$). $z = 1$ for the O(2) QCP as we discussed previously. The dispersions at various points are shown in Figure 4.3.

The QPM ground state remains stable for

$$D \geq D_c = 4dJ, \quad h < h_c = \sqrt{D(D - D_c)}, \quad (4.5)$$

as shown in Figure 4.4

When the field is strong enough, the ground state becomes fully polarized. The mean field ground state is exact in this case.

$$|\psi_{FP}\rangle = \prod_i |S_i^z = 1\rangle = \prod_i \hat{b}_{i1}^\dagger |\emptyset\rangle. \quad (4.6)$$

The saturation field can be obtained easily from the closing of the gap in (4.3):

$$h_s = D + 4dJ. \quad (4.7)$$

The energy of the system is proportional to the applied field as expected. The two branches of magnetic excitations above the saturated state are given by

$$\omega_{\mathbf{k},1} = h - D - 2dJ + f(\mathbf{k}), \quad \omega_{\mathbf{k},2} = 2h. \quad (4.8)$$

The flat branch $\omega_{\mathbf{k},2}$ describes the approximated spectrum of two-magnon bound states that appear above a critical value of the single-ion anisotropy.

4.2.2 Lagrangian multiplier method

While the HP approach gives the correct qualitative picture in $d = 3$, it is still far from being quantitatively accurate in $d = 3$ or 2, as we see in the Figure 4.3. This shortcoming can be a serious problem for comparisons against the experimental data. In particular, the Hamiltonian parameters for quantum paramagnets are normally extracted from fits of the quasiparticle dispersions that are measured with INS. The accuracy of the obtained Hamiltonian parameters depends on the accuracy of the approach that is used for computing the dispersions $\omega_{\mathbf{k}}$. Moreover, for quantum paramagnets such as DTN

which have low critical fields $h_c \ll h_s$, the HP approach normally predicts AFM ordering at $h = 0$. Therefore, it is necessary to modify the HP approach in order to obtain a quantitatively accurate description of the low-field paramagnetic ground state and the low-energy excitations.

Lagrangian multiplier method provides an accurate modification. Following the section , we obtain the modified quasiparticle dispersion:

$$\omega_{\mathbf{k},\pm} = \sqrt{\mu^2 + 2\mu s^2 f(\mathbf{k})} \pm h \quad (4.9)$$

where s is the condensate fraction of the $|S^z = 0\rangle$ state and the μ is the chemical potential. Comparing (4.3) and (4.9), we see that Lagrangian multiplier is a renormalization of the single-ion anisotropy and exchange parameters. The chemical potential and condensate fraction can be obtained from (2.83). Explicitly, they are solutions to the following equations:

$$\begin{aligned} D &= \mu \left(1 + \frac{1}{N} \sum_{\mathbf{k}} \frac{f(\mathbf{k})}{\sqrt{\mu^2 + 2s^2 \mu f(\mathbf{k})}} \right), \\ s^2 &= 2 - \frac{1}{N} \sum_{\mathbf{k}} \frac{(\mu + s^2 f(\mathbf{k}))}{\sqrt{\mu^2 + 2s^2 \mu f(\mathbf{k})}}. \end{aligned} \quad (4.10)$$

The stability conditions (4.5) are replaced by

$$\mu \geq \mu_c = 4ds^2 J, \quad h \leq h_c = \sqrt{\mu(\mu - \mu_c)}. \quad (4.11)$$

The behaviour of the dispersion and the phase diagram are shown in Figure 4.3 and 4.4, respectively.

4.2.3 QMC method

We have used two different QMC methods, the standard stochastic series expansion (SSE) with loop updates and a modified directed loop world-line QMC developed in Ref. [25], to study the ground-state and finite-temperature properties of the Hamiltonian (4.1). Since

both methods are unbiased and exact within the statistical error, we refer to them as QMC collectively in this chapter. On the dense parameter grids (temperature for thermal transitions and magnetic field or single-ion anisotropy for ground-state transitions) needed to study the critical region in detail, the statistics of the QMC results can be significantly improved by the use of a parallel tempering scheme. The implementation of tempering schemes in the context of the SSE method has been discussed in detail previously. Ordinarily, the SSE would suffer from the negative sign problem for the AFM Heisenberg interaction. However, the sublattice rotation discussed in Chapter 3 maps the XY part of the Heisenberg interaction into a ferromagnetic exchange term, thus alleviating the sign problem. This transformation maps the AFM ordering vector from $\mathbf{Q} = (\pi, \pi)$ to $\mathbf{Q} = (0, 0)$ in the new basis.

Spin stiffness and finite-size scaling

We compute the spin stiffness ρ_s , defined as the response to a twist in the boundary conditions. The transition to CAFM is efficiently investigated by studying the scaling properties of the spin stiffness ρ_s . For simulations that sample multiple winding-number sectors, the stiffness can be related to the fluctuations of the winding number in the updates and can be estimated readily with great accuracy. For the isotropic systems that are primarily considered in this chapter, the estimates of the stiffness along all the axes are equal within statistical fluctuations.

Along with the spin stiffness, we calculate the square of the order parameters characterizing the different ground states as well as standard thermodynamic observables such as energy and magnetization, and the zz component of the nematic tensor component $Q_{\mathbf{r}}^{zz} = \langle (S_{\mathbf{r}}^z)^2 - 2/3 \rangle$ that is induced by the single-ion anisotropy term. The transverse component of the imaginary-time-dependent spin structure function

$$S^{+-}(\mathbf{q}, \tau) = \frac{1}{N} \sum_{ij} e^{-i\mathbf{q} \cdot (\mathbf{r}_i - \mathbf{r}_j)} \langle S_i^+(\tau) S_j^-(0) \rangle \quad (4.12)$$

provides valuable information about the nature of the ground state. The static spin

structure factor ($\tau = 0$) measures the off-diagonal long-range ordering in the XY plane. Its value at the AFM ordering wave vector $S^{+-}(\mathbf{Q})$ is equal to the square of the XY AFM order parameter divided by N . In the bosonic language, $S^{+-}(\mathbf{Q}, 0)/N$ is the condensate fraction of the BEC. On the other hand, the imaginary-time dependence of $S^{+-}(\mathbf{q}, \tau)$ can be used to estimate the spin gap. In the world-line Monte Carlo method with discontinuities, such as the worm and the directed-loop algorithms, the correlation function 4.12 is obtained by counting the number of events in which two discontinuities created by S^+ and S^- exist in the configuration imaginary-time phase space, with the S^+ and S^- discontinuities located at $(\mathbf{r}_i, \tau)$ and $(\mathbf{r}_j, 0)$, respectively. In the SSE method, we evaluate the correlation function during the construction of the operator loops[24].

The continuous phase transition from the QPM phase to the CAFM phase is marked by the closing of the spin gap. To determine the transition point, we use the finite-size scaling properties of the spin stiffness ρ_s . The finite-size scaling analysis at the critical point predicts that

$$\rho_s(L, \beta, D) \sim L^{2-d-z} Y_{\rho_s}(\beta/L^z, (D - D_c)L^{1/\nu}) \quad (4.13)$$

below the upper critical dimension, i.e., $d + z \leq 4$, where L is the linear dimension of the system, z is the dynamic critical exponent and Y_{ρ_s} is the scaling function. $z = 1$ for QPTs belonging to the O(2) universality class and $z = 2$ for BEC QCPs. Since the effective dimension of the BEC-QCP in $d = 3$, $\mathcal{D} = 3 + 2$, is above the upper critical dimension $\mathcal{D}_c = 4$, we need to apply a modified finite-size scaling[29]

$$\rho_s(L, \beta, h) \sim L^{-(d+z)/2} Y_{\rho_s}(\beta/L^z, (h - h_c)L^{(d+z)/2}). \quad (4.14)$$

The scale invariance at the critical point provides a powerful and widely used tool to simultaneously determine the position of the critical point and verify the value of z . On a plot of $\rho_s L^{d+z-2}$ or $\rho_s L^{(d+z)/2}$ as a function of the driving parameters D or h , the curves for different system sizes will cross at the critical point provided the correct value of z is used.

Figure 4.1 shows the scaling of the stiffness close to the critical point for the QPM-CAFm transition at $h = 0$ driven by varying the single-ion anisotropy D . From field-theoretic arguments, the transition is expected to belong to the $O(2)$ universality class for which $z = 1$. Indeed, the curves were found to exhibit a unique crossing point only for $z = 1$. For a square lattice (top panel), we obtain a critical $D_c = 5.63$, in agreement with previous results[56], whereas the transition occurs at $D_c = 10.02$ on a cubic lattice (bottom panel). Further confirmation of the $O(2)$ universality class of the transition is shown in the inset panels where on a plot of $\rho_s L^{d+z-2}$ versus $(D - D_c)L^{1/\nu}$, the data for different system sizes collapse onto a single curve with our estimated D_c and known critical exponents for the $O(2)$ universality class in $d + 1$ dimensions.

Figure 4.1: Finite-size scaling plots of spin stiffness ρ_s . The four system sizes of the square lattices (upper panel) $L \times L$ are 8×8 (red), 10×10 (blue), 12×12 (black) and 18×18 (purple). The five system sizes of the cubic lattices (lower panel) $L \times L \times L$ are $4 \times 4 \times 4$ (red), $6 \times 6 \times 6$ (green), $8 \times 8 \times 8$ (black), $10 \times 10 \times 10$ (purple) and $12 \times 12 \times 12$ (green). The temperatures are taken to be $T = 1/4L$ in square lattice and $1/2L$ in the cubic lattice. The boundary conditions are periodic.

Figure 4.2 shows the modified finite-size scaling plots of the QPM to CAFm transition for $D > D_c$ as the field h is varied. The transition is expected to belong to the BEC universality class and scale invariance for the stiffness at the critical point is found for

$z = 2$ in accordance with field-theoretic predictions. Thus, the analysis of the stiffness data at the quantum critical points shows that the QPM-CAFm transition belongs to the $O(2)$ universality class for $h = 0$, but changes to BEC universality class for $h \neq 0$.

Figure 4.2: Determination of the critical field through finite-size scaling with $z = 2$ that confirms the BEC universality class of the field-induced quantum critical points.

Dispersion in QMC

The phase boundary between QPM and CAFm phases is also determined by the value of the single-magnon excitation gap Δ_s . Since the Zeeman term commutes with the rest of the Hamiltonian, the spin gap of the QPM phase changes linearly in the magnetic field and vanishes at the critical field $h_c = \Delta_s(h = 0)$. The quasiparticle dispersion and the gap Δ_s can be extracted from the QMC results by analyzing the imaginary-time Green's function

$$G_{\mathbf{k}}^{xx}(\tau) = \frac{1}{L} \sum_{\mathbf{r}} \langle S_{\mathbf{r}}^x(\tau) S_0^x(0) \rangle e^{i\mathbf{k} \cdot \mathbf{r}} \quad (4.15)$$

The quasiparticle dispersion is computed by fitting the QMC data of $G_{\mathbf{k}}^{xx}(\tau)$ with the function

$$f(\tau) = A \left(e^{-\omega\tau} + e^{-\omega(\beta-\tau)} \right), \quad (4.16)$$

where A and ω are fitting parameters. In particular, the parameter ω corresponds to the

Figure 4.3: The dispersions at (a) $D = 8J$ in 2D, (b) $D = D_c$ in 2D, and (c) $D = 12J$ in 3D. In 2D, $D_c = 8, 5.71$ and 5.625 for the HP, LM and QMC approaches, respectively.

magnetic excitation energy for each momentum $\mathbf{k}$. Figure 4.3 shows that the fit is nearly perfect for the $G_{\mathbf{k}}^{xx}(\tau)$ curve that is obtained in the QPM phase. The estimated phase boundary is $h/J = 4.2726(3)$ for $D/J = 12$, $d = 3$ and $L = 12$. This estimate is fully consistent with the modified finite-size scaling analysis (see Fig. 4.2). Since finite-size effects are very small deep inside the QPM state (far from critical point), the field-induced phase boundary can be estimated very precisely with $L = 12$. Figure 4.3 shows the comparison between the quasiparticle dispersions obtained from the QMC results and the analytical expressions (4.3) and (4.9) that we derived in the previous section using the Holstein-Primakoff (HP) and the Lagrange multiplier (LM) approaches. The quantitative agreement with the numerical result is much better for the LM approach

that reproduces not only the value of the spin gap and the overall dispersion inside the QPM phase, but also the spin velocity at the O(2) QCP $D = D_c(h = 0)$.

4.3 CAFM phase

To describe the CAFM phase, we have to use the general expression for the condensed boson with non-trivial transformation matrix. In particular, we use

$$\hat{a}_{i0}^\dagger = \hat{b}_{i0}^\dagger \cos \theta + \hat{b}_{i1}^\dagger \sin \theta \cos \phi + \hat{b}_{i2}^\dagger \sin \theta \sin \phi. \quad (4.17)$$

The other bosonic operators are obtained by orthogonalization. The parameters θ and ϕ are determined by the minimization of the mean field energy. In the absence of any applied field, the AFM ordered phase is invariant under the product of translation by one lattice parameter and a time-reversal transformation. This symmetry implies that $\phi = \frac{\pi}{4}$, i.e., the local moments have equal weights in the $S^z = \pm 1$ states. By minimizing the mean field energy as a function of the remaining variational parameter θ , we obtain

$$\sin^2 \theta = \frac{1}{2} - \frac{D}{16dJ}. \quad (4.18)$$

The dispersion relation consists of two nondegenerate branches that

$$\begin{aligned} \omega_{\mathbf{k},1} &= \sqrt{D_c^2 - D^2 \frac{f(\mathbf{k})}{2z}}, \\ \omega_{\mathbf{k},2} &= \frac{1}{2} \sqrt{\left[(D + D_c) - (D + D_c) \frac{f(\mathbf{k})}{2z} \right] \left[(D + D_c) - (D - D_c) \frac{f(\mathbf{k})}{2z} \right]}. \end{aligned} \quad (4.19)$$

In the low energy limit ($k \rightarrow 0$), they are approximately given by

$$\begin{aligned} \omega_{\mathbf{k},1} &\approx \sqrt{D_c^2 - D^2} + \frac{D^2}{4d\sqrt{D_c^2 - D^2}} k^2, \\ \omega_{\mathbf{k},2} &\approx \sqrt{J(D_c + D)} k \end{aligned} \quad (4.20)$$

Unfortunately, the modified approach based on the inclusion of a Lagrange multiplier

that we introduced in the previous section does not work well inside the ordered phase. Both branches become gapped inside the ordered phase, i.e., the approach misses the Goldstone mode associated with the spontaneous breaking of the $U(1)$ symmetry of global spin rotations along the z axis.

As we explained above, the magnetic-field-induced quantum phase transition from the QPM to the CAFM phase is qualitatively different from the transition between the same two phases that is induced by a change of D at $h = 0$. Equation (4.3) shows that the effect of increasing h from zero at a fixed $D > D_c$ is to reduce the gap $\Delta_s = \omega_{\mathbf{k}=0,-} = \sqrt{D^2 - 4dJD} - h$ linearly in h . The dispersion does not change because h couples to $m_z = \sum_r S_r^z / N$ that is a conserved quantity ($m_z = 1$ for the spin excitations that have dispersion $\omega_{\mathbf{k},-}$). Therefore, the quasiparticle dispersion remains quadratic at the field-induced QCP $h = h_c = \sqrt{D^2 - 4dJD}$, i.e., the dynamical exponent is $z = 2$. The field-induced QCP then belongs to the BEC universality class in dimension $d + 2$. On the other hand, if the single-ion anisotropy is continuously decreased at zero field, the two branches remain degenerate and the gap vanishes at $D = D_c(h = 0)$. The low-energy dispersion becomes linear at the QPM-CAFM phase boundary $\omega_{\mathbf{k}} \approx \sqrt{2DJ}k$ for small $\mathbf{k}$. As it is clear from Equation (4.20), the degeneracy between the two branches at $h = 0$ is lifted inside the CAFM phase. One of the branches, $\omega_{\mathbf{k},2}$, remains gapless with a linear dispersion at low energy (corresponding to the Goldstone mode of the ordered CAFM state), whereas the other mode develops a gap to the lowest excitation.

4.4 Phase diagrams

4.4.1 Quantum phase diagram at zero temperature

The Quantum phase diagrams obtained with different methods, i.e., linear HP approximation, the LM approach and QMC simulations, are shown in Figure 4.4. As it is expected from the comparisons between the quasi-particle dispersions obtained with different methods in the QPM phase (see Fig.4.3), the LM method produces a much better quantitative agreement with the QMC results than the linear HP approximation.

Figure 4.4: Quantum phase diagram in (a) 2D and (b) 3D. The solid line, dashed line and points between QPM and CAFM are the results obtained from the LM, HP and QMC approaches, respectively. For QMC approach, we use the modified finite-size scaling.

Figure 4.5 shows the evolution of some observables that characterize the ground-state phases as the applied field is varied for three representative values of the single-ion anisotropy. For $D > D_c$, the ground state evolves from a QPM phase at low fields ($h < h_c$) to a CAFM phase at intermediate fields ($h_c < h < h_s$) to a fully polarized phase at large fields. The uniform magnetization m_z increases monotonically with the applied field. The zz nematic order parameter Q_r^{zz} also increases monotonically but from a negative to a positive value. Right above $h = h_c$, the magnetization m_z increases with finite slope, but this slope vanishes at the O(2) QCP where $h_c(D_c) = 0$. This result is consistent with the mean field theory described in the previous section which predicts that $m_z \propto [h - h_c(D)]$ for finite $h_c(D)$ and small enough $h - h_c(D)$, while $m_z \propto h_z^3$ for $h_c = 0$ and small enough h_z .

Figure 4.5: The evolution of various characteristic observables with external magnetic field at three representative values of D as the ground state goes through the field-driven quantum phase transitions discussed in the text. The data are for a finite cubic lattice of dimension $16 \times 16 \times 16$.

The stiffness and transverse structure factor decrease monotonically with increasing h for $D \ll D_c$. However, it is clear that the field dependence must be nonmonotonic for

$D \gg D_c$ because a finite critical field is required to induce the transition from the QPM to the ordered XY phase. When the system is in the QPM phase, a critical field $h_c(D)$ is required to induce a finite amplitude of the XY order parameter, i.e., the mean field state of each spin becomes a linear combination of the states $|0\rangle_r$ and $|1\rangle_r$ for $h > h_c$. There is an optimal value of the magnetic field $h_m(D)$ for which the weight of these two states is roughly the same, leading to maxima of the order parameter (XY component of the local moment) and the spin stiffness, as it is shown in Fig. 4.5. Finally, ρ_s and $S^{+-}(\mathbf{Q})$ vanish again at sufficiently strong applied field $h \gg h_s(D)$ because the ground state evolves to the fully polarized phase with $m_z = 1$, and $Q^{zz} = \frac{1}{3}$. The exact boundary between the CAFM and the FP phases is given by Eq. 4.7. A simple continuity argument shows that the nonmonotonic field dependence of ρ_s and $S^{+-}(\mathbf{Q})$ should persist for $D \leq D_c$ as it is clear from Fig. 4.5. The ordering temperature should also exhibit a similar nonmonotonic field dependence, as we will see in the next section. This observation can be used to detect quantum magnets that exhibit magnetic ordering at $h = 0$, but are near the QCP, i.e., close to becoming quantum paramagnets.

4.4.2 Phase diagram at finite temperature

For three-dimensional systems, the CAFM phase survives up to a finite temperature $T_c(D, h)$ above which the system becomes a paramagnet via a second-order classical phase transition that belongs to the $O(2)$ universality class in dimension d . The second-order transition is replaced by a Berezinskii-Kosterlitz-Thouless phase transition at $T = T_{BKT}$ when the system is two dimensional. In this case, only quasi-long-range ordering survives at finite temperatures $T \leq T_{BKT}$. Figure 4.6 shows the field dependence of the critical temperature T_c for some representative values of D . T_c is determined by exploiting the scale invariance of the spin stiffness at the critical point with the finite-size scaling

$$\rho_s(L, T) \sim L^{2-d} Y_{\rho_s}[(T - T_c)L^{1/\nu}]. \quad (4.21)$$

Figure 4.6: The critical temperatures of the thermal phase transition into different ground states.

The thermal transition out of the CAFM phase is driven by phase fluctuations of the order parameter and belongs to the $d = 3$ $O(2)$ universality class ($\nu \approx 0.67$). At small values of D , the system is dominated by the Heisenberg AFM interaction and $T_c(h)$ decreases monotonically as a function of increasing h to $T_c(h_s) = 0$ at the QMP-FP boundary. As D increases, the spins acquire a significant $S^z = 0$ (nematic) component and the resultant decrease in the local magnetization leads to a suppression of the critical temperature. As we explained in the previous section, the applied field increases the magnitude of the local moments for $D \leq D_c$ and this effect leads to an accompanying increase in $T_c(h)$. At higher values of the applied field, the spins acquire an increasing (ferromagnetic) component along the field direction, while the AFM-ordered component decreases beyond the optimal field $h_m(D)$. Consequently, the critical temperature starts decreasing monotonically to $T_c(h) = 0$ for $h > h_m(D)$. For $D > D_c$, the system is in a QPM ground state at low fields (with the local spins being predominantly in the $S^z = 0$ state) and $T_c = 0$. A sufficiently strong external field induces a transition to the CAFM

phase with $T_c \propto (h - h_c)^{2/3}$ for small enough $h - h_c$. The transition temperature increases initially as the magnitude of the local moments increases and eventually decreases as the moments acquire a dominant ferromagnetic component parallel to the applied field, going to $T_c = 0$ at $h = h_s$.

4.5 Summary

In summary, we have investigated the quantum phase diagram and the nature of the quantum phase transitions in the $S = 1$ Heisenberg model with easy-plane single-ion anisotropy and an external magnetic field. By using a generalized spin-wave approach, we showed that the low-energy quasi-particle dispersion is qualitatively different at the phase boundary depending on the presence or absence of an external field. This difference is reflected in the universality class of the underlying QCP and has direct consequences on the low-temperature behavior. The nature of the QPM-CAFm transition in the presence and absence of an external field is directly confirmed by using large-scale QMC simulations and finite-size scaling.

We have used two different analytical approaches to describe the QPM. By comparing the results of both approaches against our QMC results, we have found important quantitative differences in the region near the $O(2)$ QCP that signals the transition to the CAFm phase. By “quantitative differences” we are not referring to the already known critical behaviors predicted by both approaches, but to the phase boundary $D_c(h)$ and the dispersion of the low-energy quasiparticle excitations. To make a clear distinction between these two different aspects of the problem, we will discuss the critical behavior in the first place. It is clear that both analytical treatments reproduce the correct critical behavior for $d = 3$ up to logarithmic corrections because $d_c \geq 3$ for the QCPs [$O(2)$ and BEC] that appear in the quantum phase diagram. The situation is different for $d = 2$ because the upper critical dimension of the $O(2)$ QCP is $d_c = 3$. We note that the approach based on the inclusion of the Lagrange multiplier and the saddle-point approximation becomes exact in the large $\mathcal{N} \rightarrow \infty$ limit ($\mathcal{N}$ is the number of components

of the order parameter of the broken symmetry state, i.e., $\mathcal{N} = 2$ for the case under consideration). Since $\nu = 1/(d1)$ for $\mathcal{N} \rightarrow \infty$, the LM approach leads to a spin gap that closes linearly in $(D - D_c)$ for $d = 2$. In contrast, the HP approach produces the expected mean field exponent $\nu = 1/2$. Naturally, neither of these approaches can reproduce the correct value of the exponent ν [$\nu \approx 0.67$ for the O(2) QCP in dimension $\mathcal{D} = 2 + 1$] because $2 < d_c$. However, the LM approach can be systematically improved by including higher-order corrections in $1/\mathcal{N}$.

Since the limitations of the LM and HP approaches for describing the critical behavior of the O(2) QCP are already known, we have focused on the overall quantitative agreement for the phase boundary $D_c(h)$ and the dispersion of the low-energy quasiparticle excitations in comparison with the numerical results. The very good agreement between the LM and QMC results is rather surprising if we consider that it holds true even for $d = 2$. Indeed, a similar treatment has been successfully applied to the quasi-one-dimensional organic quantum magnet known as DTN. In this compound, the $S = 1$ moments are provided by Ni^{2+} ions which are arranged in a tetragonal lattice. The magnetic properties are well described by the Hamiltonian with parameters $D = 8.9K$, $J_c = 2.2K$, and $J_a = J_b = 0.18K$, where J_α denotes the strength of the Heisenberg exchange interaction along the different crystal axes. Once again, the introduction of a Lagrange multiplier to enforce the constraint leads to a critical field value of 2T, which is in very good agreement with the result of QMC simulations and with the experiments. In contrast, the linear HP approach incorrectly predicts that this compound should be magnetically ordered in absence of the applied magnetic field. We note that the phase boundary obtained with the LM approach for $d = 2$ remains quantitatively more accurate near the O(2) QCP even when the next (second-) order corrections in $1/S$ are included in the HP approach. Our results then indicate that introducing a Lagrange multiplier for describing the low-energy physics of quantum paramagnets improves considerably the estimation of the spin gap and the quasiparticle dispersion. This improvement is particularly important for quantum paramagnets that have a small spin gap and consequently are close to the QCP that signals the onset of magnetic ordering. Since the Hamiltonian

parameters are typically extracted from fits of the quasiparticle dispersion measured with INS, it is crucial to have a reliable approach for computing such dispersion. The QMC method can only be applied to Hamiltonians that are free of the sign problem. However, the analytical approach is always applicable.

Chapter 5

Plaquette Valence Bond Solid

When the original lattice model was invented by Shastry and Sutherland[2], they showed an exact non-magnetic eigen state that consists of dimer singlets along each diagonal J_2 bond, as shown in Figure. 5.1.

Figure 5.1: The dimer singlet state – a direct product of singlets along each J_2 bond:
 $\frac{1}{\sqrt{2}} (|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle)$

It is interesting that interactions between singlets through J_1 cancel out exactly because of the special geometry of the lattice. Consequently, the energy of the dimer singlet state only depends on J_2 and is given by $E_D = -\frac{3}{8}J_2$ per lattice site. It is then obvious that when J_1 is small, the dimer singlet becomes the exact ground state of the spin system. This is a non-magnetic disordered ground state without long range spin ordering. On the other hand, when J_1 is much larger than J_2 , the spin system is very close to the Heisenberg model on a square lattice, the ground state of which is magnetic and has long range spin ordering. Because of the very different behavior in the two limit, a Quantum

phase transition has been expected at certain value of J_2/J_1 . A simple calculation using variational method gives an estimate of this critical point.

The Hamiltonian (1.1) could be considered as a collection of triangles with local Hamiltonian $H_t = J_1 \vec{S}_1 \cdot (\vec{S}_2 + \vec{S}_3) + \frac{1}{2} J_2 \vec{S}_2 \cdot \vec{S}_3^*$. The lowest energy per triangle is $-\frac{3}{8} J_2$ for $J_2 > 2J_1$ (dimer singlet) and $\left(\frac{1}{8} J_2 - J_1\right)$ for $J_2 < 2J_1$ (AFM). Since the energy of the AFM phase is obtained from the variational method, it is higher than the actual ground state energy. Simple mean field theory compares the energies of the two states. The critical point of the transition between the dimer singlet and the AFM state should be $J_2/J_1 < 2$ ($J_1/J_2 > 0.5$). In 1999, Ueda[57] and Weihong[58] obtained the critical point $J_2/J_1 \approx 0.69$ using perturbation theory and series expansion method, respectively. However, the nature of the transition was not clear yet. Ueda claimed that it should be either a weak first order or a continuous phase transition.

In addition to these two states, Mila proposed a helical order as an intermediate phase between the dimer singlet and the AFM ordering[11]. He used Schwinger Boson Mean Field Theory (SBMF) to determine the range of the helical order as $1.1 < J_2/J_1 < 1.65$. On the other hand, using series expansion method, Koga and Kawakami proposed a plaquette valence-bond-solid state where plaquette singlets are formed on alternative empty squares with spin gap[12] as the ground state in the parameter range $1.16 < J_2/J_1 < 1.48$. Motivated by these works, several authors investigated the intermediate phase using diverse techniques[59, 60, 61, 62, 63]. Ueda gave a detailed review about this model in 2003[64]. Many numerical methods have been used, including series expansion and perturbation theory. The general consensus has leaned towards a plaquette singlet state as the intermediate phase. Especially in 2013, Mila verified the existence of the plaquette singlet using tensor network study[63], which is considered a highly accurate numerical tool. They obtained the range of the plaquette singlet state as $0.675 < J_1/J_2 < 0.765$.

Even though the existence of the plaquette singlet is reliably established by many numerical methods, it remains far from resolved whether there is an intermediate phase

*The 1/2 comes from the fact that the diagonal J_2 bond has been counted twice.

and what is the nature of this intermediate phase. The low energy excitations (quasi-particles) and their stability remains largely unexplored. Only in 2008, Sigrist approached this phase using spin wave theory with quadrumerized Schwinger bosons[61]. However, they truncated the local Hilbert space into the four lowest states. As a result, they were not able to get an accurate ground state energy because a large part of quantum fluctuations had been neglected. We found that these quantum fluctuations were crucial to both the behavior of the quasi-particle dispersions and the ground state energy.

Furthermore, we also investigated a similar plaquette singlet state in the anisotropic case. Anisotropy occurs when there is large crystal field in the lattice, which exists in the rare-earth tetraboride. We found a more general plaquette valence bond solid which is the ground state over finite range of parameters.

5.1 Schwinger Bosons in plaquette representation

We take the central square without J_2 bond in FIG.5.1 as the unit cell. We label the four corners in clockwise manner starting from the upper-left corner, as shown in FIG.5.2.

Figure 5.2: The unit cell with labels at the four corners. The position of the unit cell is labeled by the center of the square. We take right and up as the positive x and y direction, respectively. There are two ways to choose the unit cells and the correlation between these two choices will become important when J_2 becomes stronger. In this thesis, we choose the left one as our unit cell.

The Hamiltonian of this square can be written as:

$$\hat{H}_p = J_1(\vec{S}_1 + \vec{S}_3) \cdot (\vec{S}_2 + \vec{S}_4) = \frac{1}{2}J_1(S^2 - S_{13}^2 - S_{24}^2). \quad (5.1)$$

S is the total spin number of the square. S_{13} (S_{24}) is the total spin number of the two

spins at corner 1 and 3 (2 and 4). The local Hilbert space can be described by the three Quantum numbers: $|S, S_{13}, S_{24}\rangle$ with possible degeneracy. There are in all 16 states with 5 energy levels (one quintuplet, three triplets and two singlets):

$$\begin{aligned}
E_{s_1} &= -2J_1, \quad |s_1\rangle = |0, 1, 1\rangle, \\
E_{t_\alpha} &= -J_1, \quad |t_\alpha\rangle = |1, 1, 1\rangle, \\
E_{s_2} &= 0, \quad |s_2\rangle = |0, 0, 0\rangle, \\
E_{l_\alpha} &= 0, \quad |l_\alpha\rangle = |1, 1, 0\rangle, \\
E_{r_\alpha} &= 0, \quad |r_\alpha\rangle = |1, 0, 1\rangle \\
E_{q_\alpha} &= E_{q_1} = E_{q_2} = J_1, \quad |q_\alpha\rangle = |2, 1, 1\rangle, |q_1\rangle = |2, 1, 1\rangle, |q_2\rangle = |2, 1, 1\rangle \quad (5.2)
\end{aligned}$$

$\alpha = x, y$ or z . For $|t_\alpha\rangle$, $|l_\alpha\rangle$ and $|r_\alpha\rangle$, we have $\hat{S}^\alpha |\beta\rangle = i\epsilon^{\alpha\beta\gamma} |\gamma\rangle$. And for $|q_\alpha\rangle$, we have $\hat{S}^\alpha |q_\beta\rangle = I^{\alpha\beta\gamma} |q_\gamma\rangle$. $\epsilon^{\alpha\beta\gamma}$ and $I^{\alpha\beta\gamma}$ are the antisymmetric tensor and the symmetric tensor, respectively. $\hat{S}^\alpha$ are the spin operators. More explicitly, these basis states can be expressed in terms of $|m_1, m_2, m_3, m_4\rangle$ where m_i is the eigenvalue of S^z at the i^{th} corner.

$$\begin{aligned}
|s_1\rangle &= \frac{1}{2\sqrt{3}} (|\uparrow\uparrow\downarrow\downarrow\rangle + |\downarrow\downarrow\uparrow\uparrow\rangle + |\uparrow\downarrow\downarrow\uparrow\rangle + |\downarrow\uparrow\uparrow\downarrow\rangle - 2|\uparrow\downarrow\uparrow\downarrow\rangle - 2|\downarrow\uparrow\downarrow\uparrow\rangle) \\
|s_2\rangle &= \frac{1}{2} (|\uparrow\uparrow\downarrow\downarrow\rangle + |\downarrow\downarrow\uparrow\uparrow\rangle - |\uparrow\downarrow\downarrow\uparrow\rangle - |\downarrow\uparrow\uparrow\downarrow\rangle) \\
|t_x\rangle &= \frac{1}{2\sqrt{2}} (|\uparrow\downarrow\downarrow\downarrow\rangle + |\downarrow\downarrow\uparrow\downarrow\rangle + |\uparrow\uparrow\downarrow\uparrow\rangle + |\downarrow\uparrow\uparrow\uparrow\rangle - |\uparrow\downarrow\downarrow\downarrow\rangle - |\uparrow\downarrow\uparrow\uparrow\rangle - |\downarrow\downarrow\uparrow\uparrow\rangle - |\uparrow\uparrow\downarrow\downarrow\rangle) \\
|t_y\rangle &= \frac{i}{2\sqrt{2}} (|\uparrow\uparrow\uparrow\downarrow\rangle + |\uparrow\downarrow\downarrow\downarrow\rangle + |\uparrow\downarrow\uparrow\uparrow\rangle + |\downarrow\downarrow\uparrow\downarrow\rangle - |\uparrow\uparrow\downarrow\uparrow\rangle - |\downarrow\uparrow\uparrow\uparrow\rangle - |\uparrow\downarrow\downarrow\downarrow\rangle - |\downarrow\downarrow\uparrow\uparrow\rangle) \\
|t_z\rangle &= \frac{1}{\sqrt{2}} (|\uparrow\downarrow\uparrow\downarrow\rangle - |\downarrow\uparrow\downarrow\uparrow\rangle), |q_z\rangle = \frac{1}{\sqrt{2}} (|\downarrow\downarrow\downarrow\downarrow\rangle - |\uparrow\uparrow\uparrow\uparrow\rangle), |q_2\rangle = \frac{1}{\sqrt{2}} (|\uparrow\uparrow\uparrow\uparrow\rangle + |\downarrow\downarrow\downarrow\downarrow\rangle) \\
|l_x\rangle &= \frac{1}{2} (|\downarrow\uparrow\downarrow\downarrow\rangle + |\uparrow\downarrow\uparrow\uparrow\rangle - |\uparrow\uparrow\uparrow\downarrow\rangle - |\downarrow\downarrow\downarrow\uparrow\rangle), |r_x\rangle = \frac{1}{2} (|\uparrow\downarrow\downarrow\downarrow\rangle + |\downarrow\uparrow\uparrow\uparrow\rangle - |\downarrow\downarrow\uparrow\downarrow\rangle - |\uparrow\uparrow\downarrow\uparrow\rangle) \\
|l_y\rangle &= \frac{i}{2} (|\uparrow\uparrow\uparrow\downarrow\rangle - |\downarrow\downarrow\downarrow\uparrow\rangle + |\downarrow\uparrow\downarrow\downarrow\rangle - |\uparrow\downarrow\uparrow\uparrow\rangle), |r_y\rangle = \frac{i}{2} (|\uparrow\uparrow\downarrow\uparrow\rangle + |\uparrow\downarrow\downarrow\downarrow\rangle - |\downarrow\uparrow\uparrow\uparrow\rangle - |\downarrow\downarrow\uparrow\downarrow\rangle) \\
|l_z\rangle &= \frac{1}{2} (|\uparrow\uparrow\downarrow\downarrow\rangle - |\downarrow\downarrow\uparrow\uparrow\rangle + |\downarrow\uparrow\uparrow\downarrow\rangle - |\uparrow\downarrow\downarrow\uparrow\rangle), |r_z\rangle = \frac{1}{2} (|\uparrow\uparrow\downarrow\downarrow\rangle - |\downarrow\downarrow\uparrow\uparrow\rangle + |\uparrow\downarrow\downarrow\uparrow\rangle - |\downarrow\uparrow\uparrow\downarrow\rangle) \\
|q_x\rangle &= \frac{1}{2\sqrt{2}} (|\uparrow\uparrow\uparrow\downarrow\rangle + |\uparrow\uparrow\downarrow\uparrow\rangle + |\uparrow\downarrow\uparrow\uparrow\rangle + |\downarrow\uparrow\uparrow\uparrow\rangle + |\downarrow\downarrow\downarrow\uparrow\rangle + |\downarrow\downarrow\uparrow\downarrow\rangle + |\uparrow\downarrow\downarrow\downarrow\rangle + |\uparrow\downarrow\downarrow\uparrow\rangle) \\
|q_y\rangle &= \frac{i}{2\sqrt{2}} (|\uparrow\uparrow\uparrow\downarrow\rangle + |\uparrow\uparrow\downarrow\uparrow\rangle + |\uparrow\downarrow\uparrow\uparrow\rangle + |\downarrow\uparrow\uparrow\uparrow\rangle - |\downarrow\downarrow\downarrow\uparrow\rangle - |\downarrow\downarrow\uparrow\downarrow\rangle - |\uparrow\downarrow\downarrow\downarrow\rangle - |\uparrow\downarrow\downarrow\uparrow\rangle) \\
|q_1\rangle &= \frac{1}{\sqrt{6}} (|\uparrow\uparrow\downarrow\downarrow\rangle + |\downarrow\downarrow\uparrow\uparrow\rangle + |\uparrow\downarrow\downarrow\uparrow\rangle + |\downarrow\uparrow\uparrow\downarrow\rangle + |\uparrow\downarrow\uparrow\downarrow\rangle + |\downarrow\uparrow\downarrow\uparrow\rangle)
\end{aligned}$$

We denote these 16 states as the plaquette basis and introduce 16 Schwinger Bosons to represent them. The position of a plaquette state is represented by its center. Each of these states is created from a vacuum $|\emptyset\rangle$:

$$\hat{s}_{1,2}^\dagger |\emptyset\rangle = |s_{1,2}\rangle, \hat{t}_\alpha^\dagger |\emptyset\rangle = |t_\alpha\rangle, \hat{l}_\alpha^\dagger |\emptyset\rangle = |l_\alpha\rangle, \hat{r}_\alpha^\dagger |\emptyset\rangle = |r_\alpha\rangle, \hat{q}_\alpha^\dagger |\emptyset\rangle = |q_\alpha\rangle, \hat{q}_{1,2}^\dagger |\emptyset\rangle = |q_{1,2}\rangle \quad (5.3)$$

They obey the single occupancy constraint:

$$\sum_{m=1,2} (\hat{s}_m^\dagger \hat{s}_m + \hat{q}_m^\dagger \hat{q}_m) + \sum_{\alpha=x,y,z} (\hat{t}_\alpha^\dagger \hat{t}_\alpha + \hat{l}_\alpha^\dagger \hat{l}_\alpha + \hat{r}_\alpha^\dagger \hat{r}_\alpha + \hat{q}_\alpha^\dagger \hat{q}_\alpha) = 1 \quad (5.4)$$

for each square. They also satisfy the bosonic commutation relation. In terms of this basis state, the spin operators at each corner have an irreducible form as:

$$\begin{aligned} S_n^\alpha = & \left[(-1)^n \left(\frac{1}{\sqrt{6}} \hat{t}_\alpha^\dagger \hat{s}_1 - \frac{1}{2\sqrt{3}} \cos \theta_\alpha \hat{q}_1^\dagger \hat{t}_\alpha - \frac{1}{2\sqrt{3}} \sin \theta_\alpha \hat{q}_2^\dagger \hat{t}_\alpha \right) \right. \\ & - \cos \frac{n\pi}{2} \left(\frac{1}{2\sqrt{3}} \hat{l}_\alpha^\dagger \hat{s}_1 + \frac{1}{2} \hat{r}_\alpha^\dagger \hat{s}_2 + \frac{1}{\sqrt{6}} \cos \theta_\alpha \hat{q}_1^\dagger \hat{l}_\alpha + \frac{1}{\sqrt{6}} \sin \theta_\alpha \hat{q}_2^\dagger \hat{l}_\alpha \right) \\ & + \sin \frac{n\pi}{2} \left(\frac{1}{2\sqrt{3}} \hat{r}_\alpha^\dagger \hat{s}_1 + \frac{1}{2} \hat{l}_\alpha^\dagger \hat{s}_2 + \frac{1}{\sqrt{6}} \cos \theta_\alpha \hat{q}_1^\dagger \hat{r}_\alpha + \frac{1}{\sqrt{6}} \sin \theta_\alpha \hat{q}_2^\dagger \hat{r}_\alpha \right) \\ & \left. + \frac{1}{2} \sin \theta_\alpha \hat{q}_1^\dagger \hat{q}_\alpha - \frac{1}{2} \cos \theta_\alpha \hat{q}_2^\dagger \hat{q}_\alpha + h.c. \right] \\ & + \frac{i}{4} \epsilon^{\alpha\beta\gamma} \left[-\hat{t}_\beta^\dagger \hat{t}_\gamma + \hat{q}_\beta^\dagger \hat{q}_\gamma - 2 \sin^2 \frac{n\pi}{2} \hat{l}_\beta^\dagger \hat{l}_\gamma - 2 \cos^2 \frac{n\pi}{2} \hat{r}_\beta^\dagger \hat{r}_\gamma \right. \\ & \quad \left. + \sqrt{2} \sin \frac{n\pi}{2} (\hat{r}_\beta^\dagger \hat{t}_\gamma + \hat{t}_\beta^\dagger \hat{r}_\gamma) + \sqrt{2} \cos \frac{n\pi}{2} (\hat{l}_\beta^\dagger \hat{t}_\gamma + \hat{t}_\beta^\dagger \hat{l}_\gamma) \right] \\ & + \frac{i}{4} I^{\alpha\beta\gamma} \left[(-1)^n (\hat{t}_\beta^\dagger \hat{q}_\gamma - \hat{q}_\beta^\dagger \hat{t}_\gamma) - \sqrt{2} \sin \frac{n\pi}{2} (\hat{r}_\beta^\dagger \hat{q}_\gamma - \hat{q}_\beta^\dagger \hat{r}_\gamma) + \sqrt{2} \cos \frac{n\pi}{2} (\hat{l}_\beta^\dagger \hat{q}_\gamma - \hat{q}_\beta^\dagger \hat{l}_\gamma) \right] \end{aligned} \quad (5.5)$$

n represents the n^{th} corner and θ_α is given by:

$$\theta_x = \frac{2}{3}\pi, \quad \theta_y = \frac{4}{3}\pi, \quad \text{and} \quad \theta_z = 0. \quad (5.6)$$

The original Hamiltonian becomes:

$$\begin{aligned} \hat{H} = \sum_i \hat{H}_{p,i} + J_2 \sum_i (\vec{S}_{1,i} \cdot \vec{S}_{3,i-x} + \vec{S}_{2,i} \cdot \vec{S}_{4,i+y}) \\ + J_1 \sum_i (\vec{S}_{2,i} \cdot \vec{S}_{1,i+x} + \vec{S}_{3,i} \cdot \vec{S}_{4,i+x} + \vec{S}_{1,i} \cdot \vec{S}_{4,i+y} + \vec{S}_{2,i} \cdot \vec{S}_{3,i+y}) \end{aligned} \quad (5.7)$$

Here, i labels the position of the center of the square. Using Eq.5.5, the Hamiltonian can be expressed in terms of the Schwinger Bosons. Particularly, the bilinear term $\hat{H}_p$ now assumes a diagonal form as:

$$\hat{H}_p = -2J_1 \hat{s}_1^\dagger \hat{s}_1 - J_1 \hat{t}_\alpha^\dagger \hat{t}_\alpha + J_1 \hat{q}_\alpha^\dagger \hat{q}_\alpha + J_1 (\hat{q}_1^\dagger \hat{q}_1 + \hat{q}_2^\dagger \hat{q}_2) \quad (5.8)$$

Einstein summation convention has been used here and will be used in the whole thesis unless further specified.

In the anisotropic case, the Hamiltonian of the square could be written as:

$$\hat{H}_{ap} = J_1^\alpha (\hat{S}_1^\alpha + \hat{S}_3^\alpha) (\hat{S}_2^\alpha + \hat{S}_4^\alpha) = \hat{H}_p + J_1 (\Delta - 1) (\hat{S}_1^z + \hat{S}_3^z) (\hat{S}_2^z + \hat{S}_4^z) \quad (5.9)$$

where $J_1^\alpha = J_1$ for $\alpha = x, y$ and ΔJ_1 for $\alpha = z$, respectively. The last term in the above equation can be expressed as bilinear terms of the Schwinger bosons using Eq.5.5 and the property of single occupancy. As a result, the anisotropic Hamiltonian in each square becomes:

$$\begin{aligned} \hat{H}_{ap} = J_1 \left[-\frac{2}{3} (\Delta + 2) \hat{s}_1^\dagger \hat{s}_1 - \Delta \hat{t}_z^\dagger \hat{t}_z + \frac{1}{3} (4 - \Delta) \hat{q}_1^\dagger \hat{q}_1 + \Delta (\hat{q}_2^\dagger \hat{q}_2 + \hat{q}_z^\dagger \hat{q}_z) \right. \\ \left. - (\hat{t}_x^\dagger \hat{t}_x + \hat{t}_y^\dagger \hat{t}_y) + (\hat{q}_x^\dagger \hat{q}_x + \hat{q}_y^\dagger \hat{q}_y) + \frac{\sqrt{2}}{3} (\Delta - 1) (\hat{s}_1^\dagger \hat{q}_1 + \hat{q}_1^\dagger \hat{s}_1) \right] \end{aligned} \quad (5.10)$$

And the original anisotropic Hamiltonian becomes:

$$\begin{aligned} \hat{H}_a = \sum_i \hat{H}_{ap,i} + J_2^\alpha \sum_i (\hat{S}_{1,i}^\alpha \hat{S}_{3,i-x}^\alpha + \hat{S}_{2,i}^\alpha \hat{S}_{4,i+y}^\alpha) \\ + J_1^\alpha \sum_i (\hat{S}_{2,i}^\alpha \hat{S}_{1,i+x}^\alpha + \hat{S}_{3,i}^\alpha \hat{S}_{4,i+x}^\alpha + \hat{S}_{1,i}^\alpha \hat{S}_{4,i+y}^\alpha + \hat{S}_{2,i}^\alpha \hat{S}_{3,i+y}^\alpha). \end{aligned} \quad (5.11)$$

In the presence of anisotropy, the $|s_1\rangle$ and $|q_1\rangle$ are coupled to each other while the others remain isolated. A simple diagonalization gives two new diagonal plaquette states:

$$|s_0\rangle = \cos \phi |s_1\rangle - \sin \phi |q_1\rangle, \quad (5.12)$$

$$|q_0\rangle = \sin \phi |s_1\rangle + \cos \phi |q_1\rangle \quad (5.13)$$

where ϕ satisfies:

$$\tan \phi = \frac{2\sqrt{2}(\Delta - 1)}{8 + \Delta + 3\sqrt{\Delta^2 + 8}}. \quad (5.14)$$

These two new plaquette states do not have a definite total spin number because of the coupling between the $S = 0$ and the $S = 2$ states. Hence, $|s_0\rangle$ is no longer a plaquette singlet state in the usual sense. The energy of the two new plaquette states is:

$$E_{s_0} = -\frac{1}{2}J_1(\Delta + \sqrt{\Delta^2 + 8}), \quad (5.15)$$

$$E_{q_0} = \frac{1}{2}J_1(\sqrt{\Delta^2 + 8} - \Delta). \quad (5.16)$$

For any $\Delta \geq 0$, the new plaquette ‘singlet’ state $|s_0\rangle$ always has the lowest energy. The local Hamiltonian becomes diagonal:

$$\begin{aligned} \hat{H}_{ap} = & -\frac{1}{2}J_1(\Delta + \sqrt{\Delta^2 + 8})\hat{s}_0^\dagger\hat{s}_0 + \frac{1}{2}J_1(\sqrt{\Delta^2 + 8} - \Delta)\hat{q}_0^\dagger\hat{q}_0 \\ & - \Delta J_1\hat{t}_z^\dagger\hat{t}_z + \Delta J_1(\hat{q}_2^\dagger\hat{q}_2 + \hat{q}_z^\dagger\hat{q}_z) - J_1(\hat{t}_x^\dagger\hat{t}_x + \hat{t}_y^\dagger\hat{t}_y) + J_1(\hat{q}_x^\dagger\hat{q}_x + \hat{q}_y^\dagger\hat{q}_y) \end{aligned} \quad (5.17)$$

In terms of the new basis including $|s_0\rangle$ and $|q_0\rangle$, the spin operators can be rewritten as:

$$\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 \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{l}_\alpha^\dagger \hat{s}_0 + \left(\frac{1}{2\sqrt{3}} \sin \phi + \frac{1}{\sqrt{6}} \cos \theta_\alpha \cos \phi \right) \hat{l}_\alpha^\dagger \hat{q}_0 \right. \\
& \quad \left. + \frac{1}{2} \hat{r}_\alpha^\dagger \hat{s}_2 + \frac{1}{\sqrt{6}} \sin \theta_\alpha \hat{l}_\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 \right. \\
& \quad \left. + \frac{1}{2} \hat{l}_\alpha^\dagger \hat{s}_2 + \frac{1}{\sqrt{6}} \sin \theta_\alpha \hat{r}_\alpha^\dagger \hat{q}_2 \right] \\
& - \frac{(-1)^n}{2\sqrt{3}} \sin \theta_\alpha \hat{q}_2^\dagger \hat{t}_\alpha + \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}_2^\dagger \hat{q}_\alpha + 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{l}_\beta^\dagger \hat{l}_\gamma - 2 \cos^2 \frac{n\pi}{2} \hat{r}_\beta^\dagger \hat{r}_\gamma \right. \\
& \quad \left. + \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{l}_\beta^\dagger \hat{t}_\gamma + \hat{t}_\beta^\dagger \hat{l}_\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{l}_\beta^\dagger \hat{q}_\gamma - \hat{q}_\beta^\dagger \hat{l}_\gamma) \right]
\end{aligned} \tag{5.18}$$

Starting from this plaquette representation, we can apply the generalized spin wave theory to investigate the behaviour of the excitation dispersion and obtain the stability condition of the plaquette ‘singlet’ state. Because the size of the unit cell (square) is larger than the usual dimer unit cell, more local Quantum fluctuation has been taken into account automatically. Therefore, we will get a more accurate ground state energy in the representation. In fact, after we take into account of a further correction from the cubic terms using perturbation theory, the ground state energy we have obtained is very close to that from the tensor network method[63].

5.2 Dispersion relation of the excitations

At the mean field level, we assume the ground state is a condensate of the $\hat{s}_0$ particles. That is the classical ground state is assumed to be the one with the generalized plaquette state $|s_0\rangle$ formed in every unit cell, shown in Fig.5.2. In the generalized spin wave

language, this means we have the following Holstein-Primakoff approximation for $\hat{s}_0$ and $\hat{s}_0^\dagger$:

$$\hat{s}_0 = \hat{s}_0^\dagger = 1 - \frac{1}{2} \left(\hat{s}_2^\dagger \hat{s}_2 + \hat{q}_0^\dagger \hat{q}_0 + \hat{q}_2^\dagger \hat{q}_2 + \hat{t}_\alpha^\dagger \hat{t}_\alpha + \hat{l}_\alpha^\dagger \hat{l}_\alpha + \hat{r}_\alpha^\dagger \hat{r}_\alpha + \hat{q}_\alpha^\dagger \hat{q}_\alpha \right). \quad (5.19)$$

Under such approximation, the local Hamiltonian of each unit cell $\hat{H}_{ap,i}$ becomes:

$$\begin{aligned} \hat{H}_{ap,i} = & E_0 + J_1 \sqrt{\Delta^2 + 8} \hat{q}_{0,i}^\dagger \hat{q}_{0,i} - (E_0 + J_1^\alpha) \hat{t}_{\alpha,i}^\dagger \hat{t}_{\alpha,i} + (J_1^\alpha - E_0) \hat{q}_{\alpha,i}^\dagger \hat{q}_{\alpha,i} \\ & + (\Delta J_1 - E_0) \hat{q}_{2,i}^\dagger \hat{q}_{2,i} - E_0 (\hat{l}_{\alpha,i}^\dagger \hat{l}_{\alpha,i} + \hat{r}_{\alpha,i}^\dagger \hat{r}_{\alpha,i} + \hat{s}_{2,i}^\dagger \hat{s}_{2,i}) \end{aligned} \quad (5.20)$$

where E_0 is the classical ground state energy per unit cell:

$$E_0 = -\frac{1}{2} J_1 (\Delta + \sqrt{\Delta^2 + 8}). \quad (5.21)$$

This expression is actually exact because it could be obtained from the constraint on the particle number per unit cell, Eq.5.4. By doing so, we have actually taken $\bigotimes_i |s_{0,i}\rangle$ as the new vacuum. In other words, we have adopted the following projection:

$$\hat{f}_i^\dagger |\emptyset\rangle \rightarrow \hat{f}_i^\dagger \hat{s}_{0,i} |s_{0,i}\rangle \iff \hat{f}_i^\dagger \hat{s}_{0,i} \rightarrow \hat{f}_i^\dagger \quad \text{and} \quad \hat{s}_{0,i}^\dagger \hat{f}_i \rightarrow \hat{f}_i \quad (5.22)$$

where $\hat{f}_i$ ($\hat{f}_i^\dagger$) is any of the other 15 flavors Schwinger bosons. Now the vacuum has an energy density of E_0 per unit cell and all the other bosons are considered as excitations from this new vacuum.

The hopping process of the Schwinger bosons comes from the spin interactions between nearest neighbouring unit cells. Observing Eq.5.18, we see that under Holstein-Primakoff approximation the lowest order in the spin operator $\hat{S}_n^\alpha$ is linear in Schwinger bosons. Consequently, the bilinear terms in the effective Hamiltonian are only contributed from the lowest order approximation of the spin operator which is:

$$\hat{S}_n^\alpha = (-1)^n e_\alpha (\hat{t}_\alpha^\dagger + \hat{t}_\alpha) - \cos \frac{n\pi}{2} h_\alpha (\hat{l}_\alpha^\dagger + \hat{l}_\alpha) + \sin \frac{n\pi}{2} h_\alpha (\hat{r}_\alpha^\dagger + \hat{r}_\alpha) - d_\alpha (\hat{q}_\alpha^\dagger + \hat{q}_\alpha) \quad (5.23)$$

where

$$e_\alpha = \frac{1}{\sqrt{6}} \cos \phi + \frac{1}{2\sqrt{3}} \sin \phi \cos \theta_\alpha, \quad (5.24)$$

$$h_\alpha = \frac{1}{2\sqrt{3}} \cos \phi - \frac{1}{\sqrt{6}} \sin \phi \cos \theta_\alpha, \quad (5.25)$$

$$d_\alpha = \frac{1}{2} \sin \phi \sin \theta_\alpha. \quad (5.26)$$

ϕ and θ_α are angles defined in Eq.5.14 and Eq.5.6, respectively. Schwinger bosons of different directions, α , are well separated in the bilinear effective Hamiltonian. Therefore, the Hamiltonian and the excitations can be decoupled into the three directions.

For each direction α , i.e., the repeated index α in the following is not summed over, the hopping process contains terms like:

$$\begin{aligned} \hat{S}_{n,i}^\alpha \hat{S}_{m,j}^\alpha = & \left[(-1)^n e_\alpha \hat{t}_{\alpha,i}^\dagger - \cos \frac{n\pi}{2} h_\alpha \hat{l}_{\alpha,i}^\dagger + \sin \frac{n\pi}{2} h_\alpha \hat{r}_{\alpha,i}^\dagger - d_\alpha \hat{q}_{\alpha,i}^\dagger \right] \\ & \cdot \left[(-1)^m e_\alpha \hat{t}_{\alpha,j}^\dagger - \cos \frac{m\pi}{2} h_\alpha \hat{l}_{\alpha,j}^\dagger + \sin \frac{m\pi}{2} h_\alpha \hat{r}_{\alpha,j}^\dagger - d_\alpha \hat{q}_{\alpha,j}^\dagger \right] \\ & + \left[(-1)^n e_\alpha \hat{t}_{\alpha,i}^\dagger - \cos \frac{n\pi}{2} h_\alpha \hat{l}_{\alpha,i}^\dagger + \sin \frac{n\pi}{2} h_\alpha \hat{r}_{\alpha,i}^\dagger - d_\alpha \hat{q}_{\alpha,i}^\dagger \right] \\ & \cdot \left[(-1)^m e_\alpha \hat{t}_{\alpha,j}^\dagger - \cos \frac{m\pi}{2} h_\alpha \hat{l}_{\alpha,j}^\dagger + \sin \frac{m\pi}{2} h_\alpha \hat{r}_{\alpha,j}^\dagger - d_\alpha \hat{q}_{\alpha,j}^\dagger \right] + h.c. \end{aligned} \quad (5.27)$$

The Fourier transform of the Schwinger bosons $\hat{f}_{\alpha,i}$ ($\hat{f}_{\alpha,i}^\dagger$) is[†]:

$$\hat{f}_{\alpha,\mathbf{k}}^\dagger = \frac{1}{\sqrt{N_p}} \sum_i e^{i\mathbf{k}\cdot\mathbf{r}_i} \hat{f}_{\alpha,i}^\dagger \quad \text{and} \quad \hat{f}_{\alpha,\mathbf{k}} = \frac{1}{\sqrt{N_p}} \sum_i e^{-i\mathbf{k}\cdot\mathbf{r}} \hat{f}_{\alpha,i} \quad (5.28)$$

$$\hat{f}_{\alpha,i}^\dagger = \frac{1}{\sqrt{N_p}} \sum_{\mathbf{k}} e^{-i\mathbf{k}\cdot\mathbf{r}} \hat{f}_{\alpha,\mathbf{k}}^\dagger \quad \text{and} \quad \hat{f}_{\alpha,i} = \frac{1}{\sqrt{N_p}} \sum_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{r}} \hat{f}_{\alpha,\mathbf{k}} \quad (5.29)$$

N_p is the number of the unit cell which is one quarter of the lattice sites N : $N = 4N_p$.

The summation over the real space $\mathbf{r}$ in Eq.5.11 can be transformed into the momentum space $\mathbf{k}$:

$$\sum_i \hat{S}_{n,i}^\alpha \hat{S}_{m,j}^\alpha = \frac{1}{2} \sum_{\mathbf{k}} (T_{nm}^\alpha(\mathbf{k}, \boldsymbol{\delta}) + T_{nm}^\alpha(-\mathbf{k}, \boldsymbol{\delta})) \quad (5.30)$$

[†]As before, $\hat{f}_{\alpha,i}$ represents any of the 15 Schwinger bosons.

where $T_{nm}^\alpha(\mathbf{k})$ is given by:

$$\begin{aligned}
T_{nm}^\alpha(\mathbf{k}, \boldsymbol{\delta}) = & e^{i\mathbf{k} \cdot \boldsymbol{\delta}} \left[(-1)^n e_\alpha \hat{t}_{\alpha, \mathbf{k}}^\dagger - \cos \frac{n\pi}{2} h_\alpha \hat{l}_{\alpha, \mathbf{k}}^\dagger + \sin \frac{n\pi}{2} h_\alpha \hat{r}_{\alpha, \mathbf{k}}^\dagger - d_\alpha \hat{q}_{\alpha, \mathbf{k}}^\dagger \right] \\
& \cdot \left[(-1)^m e_\alpha \hat{t}_{\alpha, -\mathbf{k}}^\dagger - \cos \frac{m\pi}{2} h_\alpha \hat{l}_{\alpha, -\mathbf{k}}^\dagger + \sin \frac{m\pi}{2} h_\alpha \hat{r}_{\alpha, -\mathbf{k}}^\dagger - d_\alpha \hat{q}_{\alpha, -\mathbf{k}}^\dagger \right] \\
& + e^{i\mathbf{k} \cdot \boldsymbol{\delta}} \left[(-1)^n e_\alpha \hat{t}_{\alpha, \mathbf{k}}^\dagger - \cos \frac{n\pi}{2} h_\alpha \hat{l}_{\alpha, \mathbf{k}}^\dagger + \sin \frac{n\pi}{2} h_\alpha \hat{r}_{\alpha, \mathbf{k}}^\dagger - d_\alpha \hat{q}_{\alpha, \mathbf{k}}^\dagger \right] \\
& \cdot \left[(-1)^m e_\alpha \hat{t}_{\alpha, \mathbf{k}}^\dagger - \cos \frac{m\pi}{2} h_\alpha \hat{l}_{\alpha, \mathbf{k}}^\dagger + \sin \frac{m\pi}{2} h_\alpha \hat{r}_{\alpha, \mathbf{k}}^\dagger - d_\alpha \hat{q}_{\alpha, \mathbf{k}}^\dagger \right] + h.c. \quad (5.31)
\end{aligned}$$

where $\boldsymbol{\delta} = \mathbf{r}_j - \mathbf{r}_i$ is the fixed vector pointing to one of the nearest neighbour $\mathbf{r}_j$ of $\mathbf{r}_i$.

We introduce a basis in the momentum space:

$$b_{\alpha, \mathbf{k}}^\dagger = \{\hat{t}_{\alpha, \mathbf{k}}^\dagger, \hat{l}_{\alpha, \mathbf{k}}^\dagger, \hat{r}_{\alpha, \mathbf{k}}^\dagger, \hat{q}_{\alpha, \mathbf{k}}^\dagger, \hat{t}_{\alpha, -\mathbf{k}}, \hat{l}_{\alpha, -\mathbf{k}}, \hat{r}_{\alpha, -\mathbf{k}}, \hat{q}_{\alpha, -\mathbf{k}}\} \quad (5.32)$$

$$b_{\alpha, \mathbf{k}} = \{\hat{t}_{\alpha, \mathbf{k}}, \hat{l}_{\alpha, \mathbf{k}}, \hat{r}_{\alpha, \mathbf{k}}, \hat{q}_{\alpha, \mathbf{k}}, \hat{t}_{\alpha, -\mathbf{k}}^\dagger, \hat{l}_{\alpha, -\mathbf{k}}^\dagger, \hat{r}_{\alpha, -\mathbf{k}}^\dagger, \hat{q}_{\alpha, -\mathbf{k}}^\dagger\}^T \quad (5.33)$$

In this basis, the bilinear terms $T_{nm}^\alpha(\mathbf{k}, \boldsymbol{\delta}) + T_{nm}^\alpha(-\mathbf{k}, \boldsymbol{\delta})$ can be written into a matrix form as:

$$T_{nm}^\alpha(\mathbf{k}, \boldsymbol{\delta}) + T_{nm}^\alpha(-\mathbf{k}, \boldsymbol{\delta}) = \begin{pmatrix} R_{nm}^\alpha(\mathbf{k}, \boldsymbol{\delta}) & R_{nm}^\alpha(\mathbf{k}, \boldsymbol{\delta}) \\ R_{nm}^\alpha(\mathbf{k}, \boldsymbol{\delta}) & R_{nm}^\alpha(\mathbf{k}, \boldsymbol{\delta}) \end{pmatrix} \quad (5.34)$$

where $R_{nm}^\alpha(\mathbf{k}, \boldsymbol{\delta})$ is a 4×4 hermitian matrix. Because of the page limit, we further split the matrix into a 2×2 block matrix form:

$$R_{nm}^\alpha(\mathbf{k}, \boldsymbol{\delta}) = \begin{pmatrix} {}_1R_{nm}^\alpha(\mathbf{k}, \boldsymbol{\delta}) & {}_2R_{nm}^\alpha(\mathbf{k}, \boldsymbol{\delta}) \\ {}_3R_{nm}^\alpha(\mathbf{k}, \boldsymbol{\delta}) & {}_4R_{nm}^\alpha(\mathbf{k}, \boldsymbol{\delta}) \end{pmatrix} \quad (5.35)$$

where

$$\begin{aligned}
{}_1R_{nm}^\alpha(\mathbf{k}, \boldsymbol{\delta}) = & \begin{pmatrix} (-1)^{n+m} 2e_\alpha^2 \cos \mathbf{k} \cdot \boldsymbol{\delta} & -e_\alpha h_\alpha \left((-1)^m \cos \frac{n\pi}{2} e^{-i\mathbf{k} \cdot \boldsymbol{\delta}} + (-1)^n \cos \frac{m\pi}{2} e^{i\mathbf{k} \cdot \boldsymbol{\delta}} \right) \\ -e_\alpha h_\alpha \left((-1)^m \cos \frac{n\pi}{2} e^{i\mathbf{k} \cdot \boldsymbol{\delta}} + (-1)^n \cos \frac{m\pi}{2} e^{-i\mathbf{k} \cdot \boldsymbol{\delta}} \right) & 2 \cos \frac{n\pi}{2} \cos \frac{m\pi}{2} h_\alpha^2 \cos \mathbf{k} \cdot \boldsymbol{\delta} \end{pmatrix} \quad (5.36)
\end{aligned}$$

$$\begin{aligned}
{}_2R_{nm}^\alpha(\mathbf{k}, \boldsymbol{\delta}) = & \begin{pmatrix} e_\alpha h_\alpha \left((-1)^m \sin \frac{n\pi}{2} e^{-i\mathbf{k} \cdot \boldsymbol{\delta}} + (-1)^n \sin \frac{m\pi}{2} e^{i\mathbf{k} \cdot \boldsymbol{\delta}} \right) & -e_\alpha d_\alpha \left((-1)^m e^{-i\mathbf{k} \cdot \boldsymbol{\delta}} + (-1)^n e^{i\mathbf{k} \cdot \boldsymbol{\delta}} \right) \\ -h_\alpha^2 \left(\cos \frac{m\pi}{2} \sin \frac{n\pi}{2} e^{-i\mathbf{k} \cdot \boldsymbol{\delta}} + \cos \frac{n\pi}{2} \sin \frac{m\pi}{2} e^{i\mathbf{k} \cdot \boldsymbol{\delta}} \right) & h_\alpha d_\alpha \left(\cos \frac{m\pi}{2} e^{-i\mathbf{k} \cdot \boldsymbol{\delta}} + \cos \frac{n\pi}{2} e^{i\mathbf{k} \cdot \boldsymbol{\delta}} \right) \end{pmatrix}
\end{aligned} \quad (5.37)$$

$${}_4R_{nm}^\alpha(\mathbf{k}, \boldsymbol{\delta}) = \begin{pmatrix} 2 \sin \frac{n\pi}{2} \sin \frac{m\pi}{2} h_\alpha^2 \cos \mathbf{k} \cdot \boldsymbol{\delta} & -h_\alpha d_\alpha \left(\sin \frac{m\pi}{2} e^{-i\mathbf{k} \cdot \boldsymbol{\delta}} + \sin \frac{n\pi}{2} e^{i\mathbf{k} \cdot \boldsymbol{\delta}} \right) \\ -h_\alpha d_\alpha \left(\sin \frac{m\pi}{2} e^{i\mathbf{k} \cdot \boldsymbol{\delta}} + \sin \frac{n\pi}{2} e^{-i\mathbf{k} \cdot \boldsymbol{\delta}} \right) & 2d_\alpha^2 \cos \mathbf{k} \cdot \boldsymbol{\delta} \end{pmatrix} \quad (5.38)$$

and ${}_3R_{nm}^\alpha(\mathbf{k}, \boldsymbol{\delta}) = {}_2R_{nm}^\alpha(\mathbf{k}, \boldsymbol{\delta})^\dagger$.

Sum over all the pairs in Eq.5.11 and we have:

$$R_{\mathbf{k}}^\alpha = J_2^\alpha (R_{13}^\alpha(\mathbf{k}, -\mathbf{x}) + R_{24}^\alpha(\mathbf{k}, \mathbf{y})) + J_1^\alpha (R_{21}^\alpha(\mathbf{k}, \mathbf{x}) + R_{34}^\alpha(\mathbf{k}, \mathbf{x}) + R_{14}^\alpha(\mathbf{k}, \mathbf{y}) + R_{23}^\alpha(\mathbf{k}, \mathbf{y})) \quad (5.39)$$

which has a matrix form:

$$R_{\mathbf{k}}^\alpha = 2J_1^\alpha \begin{pmatrix} (x-2)e_\alpha^2 f(\mathbf{k}) & -ie_\alpha h_\alpha w_1(\mathbf{k}) & ie_\alpha h_\alpha w_2(\mathbf{k}) & -xe_\alpha d_\alpha g(\mathbf{k}) \\ ie_\alpha h_\alpha w_1(\mathbf{k}) & -xh_\alpha^2 \cos k_x & h_\alpha^2 g(\mathbf{k}) & -ih_\alpha d_\alpha w_3(\mathbf{k}) \\ -ie_\alpha h_\alpha w_2(\mathbf{k}) & h_\alpha^2 g(\mathbf{k}) & -xh_\alpha^2 \cos k_y & ih_\alpha d_\alpha w_4(\mathbf{k}) \\ -xe_\alpha d_\alpha g(\mathbf{k}) & ih_\alpha d_\alpha w_3(\mathbf{k}) & -ih_\alpha d_\alpha w_4(\mathbf{k}) & (x+2)d_\alpha^2 f(\mathbf{k}) \end{pmatrix} \quad (5.40)$$

where

$$f(\mathbf{k}) = \cos k_x + \cos k_y \quad \text{and} \quad g(\mathbf{k}) = \cos k_x - \cos k_y \quad (5.41)$$

$$w_1(\mathbf{k}) = (x-1) \sin k_x - \sin k_y, \quad (5.42)$$

$$w_2(\mathbf{k}) = \sin k_x + (x-1) \sin k_y, \quad (5.43)$$

$$w_3(\mathbf{k}) = (x+1) \sin k_x + \sin k_y, \quad (5.44)$$

$$w_4(\mathbf{k}) = \sin k_x - (x+1) \sin k_y. \quad (5.45)$$

k_x and k_y are the $\hat{x}$ - and $\hat{y}$ -components of the momentum, respectively. We have set

$|\delta| = 1$. We have also defined the ratio between the two interactions: $x = J_2/J_1$. Now we include the diagonal terms $\hat{H}_{ap,i}$ and we obtain the effective Hamiltonian in the momentum space as:

$$\hat{H} = N_p E_0 + \hat{H}_I + \hat{H}_{SW} \quad (5.46)$$

where E_0 is the classical ground state energy given by Eq.5.21. H_I contains the isolated excitations, in other words, excitations with flat dispersions.

$$\hat{H}_I = J_1 \sqrt{\Delta^2 + 8} \sum_{\mathbf{k}} \hat{q}_{0,\mathbf{k}}^\dagger \hat{q}_{0,\mathbf{k}} + (\Delta J_1 - E_0) \sum_{\mathbf{k}} \hat{q}_{2,\mathbf{k}}^\dagger \hat{q}_{2,\mathbf{k}} - E_0 \sum_{\mathbf{k}} \hat{s}_{2,\mathbf{k}}^\dagger \hat{s}_{2,\mathbf{k}} \quad (5.47)$$

$\hat{H}_{SW}$ is the spin wave Hamiltonian which describes the dispersion of the excitations and also provides a Quantum correction to the ground state energy.

$$\hat{H}_{SW} = \frac{1}{2} \sum_{\mathbf{k}, \alpha} b_{\alpha, \mathbf{k}}^\dagger \Omega_{\mathbf{k}}^\alpha b_{\alpha, \mathbf{k}} + 6N_p E_0 \quad (5.48)$$

The grand dynamical matrix $\Omega_{\mathbf{k}}^\alpha$ is an 8×8 matrix:

$$\Omega_{\mathbf{k}}^\alpha = \begin{pmatrix} Q^\alpha + R_{\mathbf{k}}^\alpha & R_{\mathbf{k}}^\alpha \\ R_{\mathbf{k}}^\alpha & Q^\alpha + R_{\mathbf{k}}^\alpha \end{pmatrix} \quad (5.49)$$

where Q^α is a 4×4 diagonal matrix:

$$Q^\alpha = \begin{pmatrix} -J_1^\alpha - E_0 & 0 & 0 & 0 \\ 0 & -E_0 & 0 & 0 \\ 0 & 0 & -E_0 & 0 \\ 0 & 0 & 0 & J_1^\alpha - E_0 \end{pmatrix}. \quad (5.50)$$

The grand dynamical matrix can be diagonalized through Bogoliubov transformation:

$$\hat{H}_{SW} = \sum_{\mathbf{k}, \alpha, n} \varepsilon_{\alpha, \mathbf{k}}^{(n)} \hat{\gamma}_{\alpha, n, \mathbf{k}}^\dagger \hat{\gamma}_{\alpha, n, \mathbf{k}} + \frac{1}{2} \sum_{\alpha, \mathbf{k}, n} \left(\varepsilon_{\alpha, \mathbf{k}}^{(n)} + E_0 \right). \quad (5.51)$$

Here $n \in \{1, 2, 3, 4\}$ represents the four branches dispersion relations $\varepsilon_{\alpha, \mathbf{k}}^{(n)}$ for each direction α . And $\hat{\gamma}_{\alpha, n, \mathbf{k}}^\dagger$ ($\hat{\gamma}_{\alpha, n, \mathbf{k}}$) are the Schwinger bosons for the corresponding quasi-particles (excitations). As discussed in Section 2.6, the explicit form of the dispersion relations can be obtained from the matrix $M_{\mathbf{k}}^\alpha = (2R_{\mathbf{k}}^\alpha + Q^\alpha)Q^\alpha$ as[†].

$$\begin{aligned}\varepsilon_{\alpha, \mathbf{k}}^{(1,2)} &= \left[-\frac{1}{4} \left(\alpha_3 - \sqrt{\alpha_3^2 - 4\alpha_2 + 4y_\alpha} \right) \pm \sqrt{D_-} \right]^{1/2} \\ \varepsilon_{\alpha, \mathbf{k}}^{(3,4)} &= \left[-\frac{1}{4} \left(\alpha_3 + \sqrt{\alpha_3^2 - 4\alpha_2 + 4y_\alpha} \right) \pm \sqrt{D_+} \right]^{1/2}\end{aligned}\quad (5.52)$$

with $D_\pm$ given by

$$D_\pm = \frac{1}{4} \left(\alpha_3 \pm \sqrt{\alpha_3^2 - 4\alpha_2 + 4y_\alpha} \right)^2 - 2 \left(y_\alpha \pm \sqrt{y_\alpha^2 - 4\alpha_0} \right). \quad (5.53)$$

α_i ($i \in \{0, 1, 2, 3\}$) are the coefficients of the characteristic polynomial of matrix $M_{\mathbf{k}}^\alpha$:

$$\alpha_0 = \det \{M_{\mathbf{k}}^\alpha\} \quad (5.54)$$

$$\alpha_1 = -\frac{1}{6} \left(\text{Tr} \{M_{\mathbf{k}}^\alpha\}^3 - 3 \text{Tr} \{(M_{\mathbf{k}}^\alpha)^2\} \text{Tr} \{M_{\mathbf{k}}^\alpha\} + 2 \text{Tr} \{(M_{\mathbf{k}}^\alpha)^3\} \right) \quad (5.55)$$

$$\alpha_2 = \frac{1}{2} \left(\text{Tr} \{M_{\mathbf{k}}^\alpha\}^2 - \text{Tr} \{(M_{\mathbf{k}}^\alpha)^2\} \right) \quad (5.56)$$

$$\alpha_3 = -\text{Tr} \{M_{\mathbf{k}}^\alpha\}. \quad (5.57)$$

And y_α is the real number solution of a cubic equation and is given by:

$$y_\alpha = \frac{1}{3}\alpha_2 - 2\sqrt{-\frac{m_\alpha}{3}} \cos\left(\psi_\alpha + \frac{2n\pi}{3}\right) \quad (5.58)$$

$$\cos 3\psi_\alpha = \frac{1}{2}p_\alpha \left(-\frac{3}{m_\alpha}\right)^{3/2} \quad (5.59)$$

$$p_\alpha = -\frac{2}{27}\alpha_2^3 + \frac{1}{3}\alpha_1\alpha_2\alpha_3 + \frac{8}{3}\alpha_0\alpha_2 - \alpha_1^2 - \alpha_3^2\alpha_0 \quad (5.60)$$

$$m_\alpha = \alpha_1\alpha_3 - 4\alpha_0 - \frac{1}{3}\alpha_2^2 \quad (5.61)$$

where n is any integer such that y_α is a real number.

[†] $A_{\mathbf{k}} = R_{\mathbf{k}}^\alpha + Q_{\mathbf{k}}^\alpha$ and $B_{\mathbf{k}} = Q_{\mathbf{k}}^\alpha$.

The quasi-particle Schwinger bosons:

$$\gamma_{\alpha,\mathbf{k}}^\dagger = \{\hat{\gamma}_{\alpha,1,\mathbf{k}}^\dagger, \hat{\gamma}_{\alpha,2,\mathbf{k}}^\dagger, \hat{\gamma}_{\alpha,3,\mathbf{k}}^\dagger, \hat{\gamma}_{\alpha,4,\mathbf{k}}^\dagger, \hat{\gamma}_{\alpha,1,-\mathbf{k}}, \hat{\gamma}_{\alpha,2,-\mathbf{k}}, \hat{\gamma}_{\alpha,3,-\mathbf{k}}, \hat{\gamma}_{\alpha,4,-\mathbf{k}}\}, \quad (5.62)$$

$$\gamma_{\alpha,\mathbf{k}} = \{\hat{\gamma}_{\alpha,1,\mathbf{k}}, \hat{\gamma}_{\alpha,2,\mathbf{k}}, \hat{\gamma}_{\alpha,3,\mathbf{k}}, \hat{\gamma}_{\alpha,4,\mathbf{k}}, \hat{\gamma}_{\alpha,1,-\mathbf{k}}^\dagger, \hat{\gamma}_{\alpha,2,-\mathbf{k}}^\dagger, \hat{\gamma}_{\alpha,3,-\mathbf{k}}^\dagger, \hat{\gamma}_{\alpha,4,-\mathbf{k}}^\dagger\}^T \quad (5.63)$$

are obtained from the transformation:

$$\gamma_{\alpha,\mathbf{k}} = P_{\alpha,\mathbf{k}}^{-1} b_{\alpha,\mathbf{k}} \quad \text{with} \quad P_{\alpha,\mathbf{k}}^{-1} = \hat{\delta} P_{\alpha,\mathbf{k}}^\dagger \hat{\delta}, \quad (5.64)$$

and $P_{\alpha,\mathbf{k}} = \begin{pmatrix} U_{\alpha,\mathbf{k}} & V_{\alpha,\mathbf{k}} \\ V_{\alpha,\mathbf{k}} & U_{\alpha,\mathbf{k}} \end{pmatrix}$. The 4×4 matrices $U_{\alpha,\mathbf{k}}$ and $V_{\alpha,\mathbf{k}}$ contain the following columns $u_{\alpha,\mathbf{k},n}$ and $v_{\alpha,\mathbf{k},n}$, respectively:

$$u_{\alpha,\mathbf{k},n} = \frac{1}{2} \left(\frac{z_{\alpha,\mathbf{k},n}^\dagger Q^\alpha z_{\alpha,\mathbf{k},n}}{\varepsilon_{\alpha,\mathbf{k}}^{(n)}} \right)^{-1/2} \left(\frac{1}{\varepsilon_{\alpha,\mathbf{k}}^{(n)}} Q^\alpha + 1 \right) z_{\alpha,\mathbf{k},n} \quad (5.65)$$

$$v_{\alpha,\mathbf{k},n} = \frac{1}{2} \left(\frac{z_{\alpha,\mathbf{k},n}^\dagger Q^\alpha z_{\alpha,\mathbf{k},n}}{\varepsilon_{\alpha,\mathbf{k}}^{(n)}} \right)^{-1/2} \left(\frac{1}{\varepsilon_{\alpha,\mathbf{k}}^{(n)}} Q^\alpha - 1 \right) z_{\alpha,\mathbf{k},n} \quad (5.66)$$

where $z_{\alpha,\mathbf{k},n}$ is the eigen vector of $M_{\mathbf{k}}^\alpha$ with eigenvalue $\varepsilon_{\alpha,\mathbf{k}}^{(n)}$. The two matrices have the following property:

$$U_{\alpha,\mathbf{k}}^* = U_{\alpha,-\mathbf{k}} \quad \text{and} \quad V_{\alpha,\mathbf{k}}^* = V_{\alpha,-\mathbf{k}}. \quad (5.67)$$

The expression of the quasi-particle Schwinger bosons is essential in the calculation of the further Quantum correction to the ground state energy from the cubic terms, which will be discussed in the next section.

There are in all 15 branches of dispersion relations with 12 coming from the three directions α and 3 flat dispersions. The lowest branch is of great importance because it describes the low energy physics of the quasi-particles and the behaviour of the gap is closely related to the stability of the ground state from which we could determine the phase boundary of this generalized plaquette valence bond solid. The behaviour of the excitation dispersion will be discussed in detail in Section 5.4.

5.3 Quantum correction from the cubic Hamiltonian

The constant term in the diagonalized spin wave Hamiltonian (5.51) is the Quantum fluctuation above the classical ground state, in other words it is the Quantum correction to the classical ground state energy. Therefore, the ground state energy per lattice site of this generalized plaquette valence bond solid is $|s_0\rangle$ is:

$$E_{GS} = \frac{1}{4}E_0 + \frac{1}{2N} \sum_{\alpha, \mathbf{k}, n} \left(\varepsilon_{\alpha, \mathbf{k}}^{(n)} + E_0 \right). \quad (5.68)$$

Because of the large amount of quasi-particles, we have tried to get a more accurate ground state energy by including more quantum fluctuations. The next lowest order beyond the spin wave Hamiltonian is the cubic terms resulting from the Holstein-Primakoff approximation. With this approximation, the bilinear terms in the spin operator (5.18) containing $\hat{s}_0^\dagger$ ($\hat{s}_0$) becomes linear in Schwinger boson operators while the rest remain bilinear form. As a result, $S_{n,i}^\alpha S_{m,j}^\alpha$ contains bilinear terms, which contribute to the spin wave Hamiltonian, and additional cubic terms and quartic terms. After our calculation, we found that the next dominating contribution to the ground state energy came from the cubic terms up to second order perturbation theory.

5.3.1 The cubic Hamiltonian

In the real space, the cubic Hamiltonian contains terms: $\hat{f}_{1,i}^\dagger \hat{f}_{2,j}^\dagger \hat{f}_{3,j}$, $\hat{f}_{1,i} \hat{f}_{2,j}^\dagger \hat{f}_{3,j}$ and their hermitian conjugate. As usual, $\hat{f}_{n,i}^\dagger$ ($\hat{f}_{n,i}$) represents any Schwinger boson operators. After Fourier transform, they become terms in the momentum space:

$$\sum_i \hat{f}_{1,i}^\dagger \hat{f}_{2,j}^\dagger \hat{f}_{3,j} = \frac{1}{\sqrt{N_p}} \sum_{\mathbf{k}_1, \mathbf{k}_2} \hat{f}_{1,\mathbf{k}_1}^\dagger \hat{f}_{2,\mathbf{k}_2}^\dagger \hat{f}_{3,\mathbf{k}_1+\mathbf{k}_2} e^{i\mathbf{k}_1 \cdot \boldsymbol{\delta}}, \quad (5.69)$$

$$\sum_i \hat{f}_{2,i}^\dagger \hat{f}_{3,i}^\dagger \hat{f}_{1,j} = \frac{1}{\sqrt{N_p}} \sum_{\mathbf{k}_1, \mathbf{k}_2} \hat{f}_{2,\mathbf{k}_2}^\dagger \hat{f}_{3,\mathbf{k}_1+\mathbf{k}_2}^\dagger \hat{f}_{1,\mathbf{k}_1} e^{-i\mathbf{k}_1 \cdot \boldsymbol{\delta}}, \quad (5.70)$$

$$\sum_i \hat{f}_{1,i}^\dagger \hat{f}_{2,j}^\dagger \hat{f}_{3,j} = \frac{1}{\sqrt{N_p}} \sum_{\mathbf{k}_1, \mathbf{k}_2} \hat{f}_{1,\mathbf{k}_1}^\dagger \hat{f}_{2,\mathbf{k}_1+\mathbf{k}_2}^\dagger \hat{f}_{3,\mathbf{k}_2} e^{-i\mathbf{k}_1 \cdot \boldsymbol{\delta}}, \quad (5.71)$$

$$\sum_i \hat{f}_{2,i}^\dagger \hat{f}_{3,i}^\dagger \hat{f}_{1,j} = \frac{1}{\sqrt{N_p}} \sum_{\mathbf{k}_1, \mathbf{k}_2} \hat{f}_{2,\mathbf{k}_1+\mathbf{k}_2}^\dagger \hat{f}_{3,\mathbf{k}_2}^\dagger \hat{f}_{1,\mathbf{k}_1} e^{i\mathbf{k}_1 \cdot \boldsymbol{\delta}} \quad (5.72)$$

As before, $\boldsymbol{\delta} = \mathbf{r}_j - \mathbf{r}_i$. We call the cubic terms $\hat{f}_{1,\mathbf{k}_1}^\dagger \hat{f}_{2,\mathbf{k}_2}^\dagger \hat{f}_{3,\mathbf{k}_1+\mathbf{k}_2}$ and $\hat{f}_{1,\mathbf{k}_1} \hat{f}_{2,\mathbf{k}_1+\mathbf{k}_2}^\dagger \hat{f}_{3,\mathbf{k}_2}$ **vertices** and they can be represented by the following diagrams:

Outgoing arrows represent creation operators while ingoing ones represent annihilation operators. Each operator carries a momentum $\mathbf{k}$ when pointing to the right while $-\mathbf{k}$ when pointing the left. The conservation of the momentum is obvious. We realize that the coefficient of the vertex only depends on one momentum even though the vertex as a whole depends on two independent momenta. For simplicity, we let the upper arrow carry the momentum on which the coefficient depends.

The exact coefficient of each vertex can be obtained from Eq.5.11 by grouping all the identical vertices from the bilinear terms $S_{n,i}^\alpha S_{m,j}^\alpha$. For the sake of convenience, we re-index the 15 Schwinger bosons as the following:

Figure 5.3: The left diagram represents $\hat{f}_{1,\mathbf{k}_1}^\dagger \hat{f}_{2,\mathbf{k}_2}^\dagger \hat{f}_{3,\mathbf{k}_1+\mathbf{k}_2}$ and the right $\hat{f}_{1,\mathbf{k}_1} \hat{f}_{2,\mathbf{k}_1+\mathbf{k}_2}^\dagger \hat{f}_{3,\mathbf{k}_2}$. The upper arrow always carries the momentum on which the coefficient of the vertex depends.

$$\begin{aligned}
 \hat{f}_1 &= \hat{t}_x, & \hat{f}_2 &= \hat{l}_x, & \hat{f}_3 &= \hat{r}_x, & \hat{f}_4 &= \hat{q}_x, & \hat{f}_5 &= \hat{t}_y, & \hat{f}_6 &= \hat{l}_y, \\
 \hat{f}_7 &= \hat{r}_y, & \hat{f}_8 &= \hat{q}_y, & \hat{f}_9 &= \hat{t}_z, & \hat{f}_{10} &= \hat{l}_z, & \hat{f}_{11} &= \hat{r}_z, & \hat{f}_{12} &= \hat{q}_z \\
 \hat{f}_{13} &= \hat{s}_2, & \hat{f}_{14} &= \hat{q}_0, & \hat{f}_{15} &= \hat{q}_2
 \end{aligned} \tag{5.73}$$

We define $A_{n,a}^\alpha$ as the coefficient of the linear term $\hat{f}_a$ in the effective spin operator S_n^α (5.18) under Holstein-Primakoff approximation. Here a is an integer representing the flavor of the Schwinger boson operator. From Eq.5.23, we can see that the coefficient of the $\hat{f}_a^\dagger$ is also $A_{n,a}^\alpha$. Similarly, we define $B_{n,ab}^\alpha$ as the coefficient of the bilinear term $\hat{f}_a^\dagger \hat{f}_b$. The coefficients $A_{n,a}^\alpha$ and $B_{n,ab}^\alpha$ are shown in Table.5.1 and 5.2, respectively.

Operator	Coefficient $A_{n,a}^\alpha$
$\hat{t}_\alpha, \hat{t}_\alpha^\dagger$:	$(-1)^n \left(\frac{1}{\sqrt{6}} \cos \phi + \frac{1}{2\sqrt{3}} \sin \phi \cos \theta_\alpha \right)$
$\hat{l}_\alpha, \hat{l}_\alpha^\dagger$:	$-\cos \frac{n\pi}{2} \left(\frac{1}{2\sqrt{3}} \cos \phi - \frac{1}{\sqrt{6}} \sin \phi \cos \theta_\alpha \right)$
$\hat{r}_\alpha, \hat{r}_\alpha^\dagger$:	$\sin \frac{n\pi}{2} \left(\frac{1}{2\sqrt{3}} \cos \phi - \frac{1}{\sqrt{6}} \sin \phi \cos \theta_\alpha \right)$
$\hat{q}_\alpha, \hat{q}_\alpha^\dagger$:	$-\frac{1}{2} \sin \phi \sin \theta_\alpha$

Table 5.1: Coefficients of the linear operators.

It is obvious that the two vertices in Eq.5.69 and 5.70 are the same. Thus, we can combine them into a single vertex $\hat{f}_{a,\mathbf{k}_1}^\dagger \hat{f}_{b,\mathbf{k}_2}^\dagger \hat{f}_{c,\mathbf{k}_3}$, the coefficient of which is denoted by $W_c^{ab}(\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3)$. We have used upper indices for creation operators and lower indices for annihilation operators. The coefficient of $\hat{f}_{a,\mathbf{k}_1} \hat{f}_{b,\mathbf{k}_2}^\dagger \hat{f}_{c,\mathbf{k}_3}$ is then just $W_a^b{}_c(\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3)$. From Eq.5.69-5.72, we realize that both of the coefficients $W_c^{ab}(\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3)$ and $W_a^b{}_c(\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3)$ only depend on $\mathbf{k}_1$. And from the property of the symmetry property of the Hamiltonian, we have $W_a^b{}_c(\mathbf{k}_1, \mathbf{k}_1 + \mathbf{k}_2, \mathbf{k}_2) = W_c^{ab}(-\mathbf{k}_1, \mathbf{k}_2, -\mathbf{k}_1 + \mathbf{k}_2)$.

Operator	Coefficient $B_{n,ab}^\alpha$	Operator	Coefficient $B_{n,ab}^\alpha$
$\hat{t}_\alpha^\dagger \hat{q}_0$:	$(-1)^n \left(\frac{1}{\sqrt{6}} \sin \phi - \frac{1}{2\sqrt{3}} \cos \phi \cos \theta_\alpha \right)$	$\hat{t}_\beta^\dagger \hat{t}_\gamma$:	$-\frac{i}{4} \epsilon^{\alpha\beta\gamma}$
$\hat{l}_\alpha^\dagger \hat{q}_0$:	$-\cos \frac{n\pi}{2} \left(\frac{1}{2\sqrt{3}} \sin \phi + \frac{1}{\sqrt{6}} \cos \phi \cos \theta_\alpha \right)$	$\hat{q}_\beta^\dagger \hat{q}_\gamma$:	$\frac{i}{4} \epsilon^{\alpha\beta\gamma}$
$\hat{r}_\alpha^\dagger \hat{q}_0$:	$\sin \frac{n\pi}{2} \left(\frac{1}{2\sqrt{3}} \sin \phi + \frac{1}{\sqrt{6}} \cos \phi \cos \theta_\alpha \right)$	$\hat{l}_\beta^\dagger \hat{l}_\gamma$:	$-\frac{i}{2} \epsilon^{\alpha\beta\gamma} \sin^2 \frac{n\pi}{2}$
$\hat{q}_\alpha^\dagger \hat{q}_0$:	$\frac{1}{2} \cos \phi \sin \theta_\alpha$	$\hat{r}_\beta^\dagger \hat{r}_\gamma$:	$-\frac{i}{2} \epsilon^{\alpha\beta\gamma} \cos^2 \frac{n\pi}{2}$
$\hat{l}_\alpha^\dagger \hat{s}_2$:	$\frac{1}{2} \sin \frac{n\pi}{2}$	$\hat{t}_\beta^\dagger \hat{r}_\gamma$:	$\frac{1}{2\sqrt{2}} i \epsilon^{\alpha\beta\gamma} \sin \frac{n\pi}{2}$
$\hat{r}_\alpha^\dagger \hat{s}_2$:	$-\frac{1}{2} \cos \frac{n\pi}{2}$	$\hat{t}_\beta^\dagger \hat{l}_\gamma$:	$\frac{1}{2\sqrt{2}} i \epsilon^{\alpha\beta\gamma} \cos \frac{n\pi}{2}$
$\hat{t}_\alpha^\dagger \hat{q}_2$:	$-(-1)^n \frac{1}{2\sqrt{3}} \sin \theta_\alpha$	$\hat{t}_\beta^\dagger \hat{q}_\gamma$:	$(-1)^n \frac{i}{4} I^{\alpha\beta\gamma}$
$\hat{l}_\alpha^\dagger \hat{q}_2$:	$-\frac{1}{\sqrt{6}} \cos \frac{n\pi}{2} \sin \theta_\alpha$	$\hat{r}_\beta^\dagger \hat{q}_\gamma$:	$-\frac{1}{2\sqrt{2}} i I^{\alpha\beta\gamma} \sin \frac{n\pi}{2}$
$\hat{r}_\alpha^\dagger \hat{q}_2$:	$\frac{1}{\sqrt{6}} \sin \frac{n\pi}{2} \sin \theta_\alpha$	$\hat{l}_\beta^\dagger \hat{q}_\gamma$:	$\frac{1}{2\sqrt{2}} i I^{\alpha\beta\gamma} \cos \frac{n\pi}{2}$
$\hat{q}_\alpha^\dagger \hat{q}_2$:	$-\frac{1}{2} \cos \theta_\alpha$		

Table 5.2: Coefficients of the bilinear operators.

The coefficient $W_c^{ab}(\mathbf{k}, \mathbf{k}', \mathbf{k} + \mathbf{k}')$ could be obtained from Eq.5.11:

$$\begin{aligned}
W_c^{ab}(\mathbf{k}, \mathbf{k}', \mathbf{k} + \mathbf{k}') &= e^{ik_x} \left(J_2^\alpha B_{1,bc}^\alpha A_{3,a}^\alpha + J_1^\alpha A_{2,a}^\alpha B_{1,bc}^\alpha + J_1^\alpha A_{3,a}^\alpha B_{4,bc}^\alpha \right) \\
&+ e^{-ik_x} \left(J_2^\alpha A_{1,a}^\alpha B_{3,bc}^\alpha + J_1^\alpha B_{2,bc}^\alpha A_{1,a}^\alpha + J_1^\alpha B_{3,bc}^\alpha A_{4,a}^\alpha \right) \\
&+ e^{ik_y} \left(J_2^\alpha A_{2,a}^\alpha B_{4,bc}^\alpha + J_1^\alpha A_{1,a}^\alpha B_{4,bc}^\alpha + J_1^\alpha A_{2,a}^\alpha B_{3,bc}^\alpha \right) \\
&+ e^{-ik_y} \left(J_2^\alpha B_{2,bc}^\alpha A_{4,a}^\alpha J_1^\alpha B_{1,bc}^\alpha A_{4,a}^\alpha + J_1^\alpha B_{2,bc}^\alpha A_{3,a}^\alpha \right). \quad (5.74)
\end{aligned}$$

α is not summed over here. The linear terms only contain Schwinger bosons of the kinds $\hat{t}_\alpha$, $\hat{l}_\alpha$, $\hat{r}_\alpha$ and $\hat{q}_\alpha$, i.e., $\hat{f}_a$ with $1 \leq a \leq 12$, and their hermitian conjugates. Thus, $W_c^{ab}(\mathbf{k}, \mathbf{k}', \mathbf{k} + \mathbf{k}')$ is zero for $a \in \{13, 14, 15, 16\}$. α is just the direction of $\hat{f}_a^\dagger$ in the corresponding vertex. The cubic Hamiltonian is then given by:

$$\hat{H}_3 = \frac{1}{\sqrt{N_p}} \sum_{\substack{\mathbf{k}, \mathbf{k}' \\ a, b, c}} \left(W_c^{ab}(\mathbf{k}, \mathbf{k}', \mathbf{k} + \mathbf{k}') \hat{f}_{a,\mathbf{k}}^\dagger \hat{f}_{b,\mathbf{k}'}^\dagger \hat{f}_{c,\mathbf{k}+\mathbf{k}'} + W_a^{bc}(\mathbf{k}, \mathbf{k} + \mathbf{k}', \mathbf{k}') \hat{f}_{a,\mathbf{k}} \hat{f}_{b,\mathbf{k}+\mathbf{k}'}^\dagger \hat{f}_{c,\mathbf{k}'} \right) \quad (5.75)$$

Observing the above equation, we find that $\hat{f}_{a,\mathbf{k}}^\dagger \hat{f}_{b,\mathbf{k}'}^\dagger \hat{f}_{c,\mathbf{k}+\mathbf{k}'}$ and $\hat{f}_{b,\mathbf{k}'} \hat{f}_{a,\mathbf{k}} \hat{f}_{c,\mathbf{k}+\mathbf{k}'}$ are actually

the same but they have different coefficients. Hence, we perform a symmetrization so that identical operators have equal coefficients. We define the symmetrized coefficients:

$$\bar{W}_c^{ab}(\mathbf{k}, \mathbf{k}', \mathbf{k} + \mathbf{k}') = \frac{1}{2} [W_c^{ab}(\mathbf{k}, \mathbf{k}', \mathbf{k} + \mathbf{k}') + W_c^{ba}(\mathbf{k}', \mathbf{k}, \mathbf{k} + \mathbf{k}')], \quad (5.76)$$

$$\bar{W}_a^b{}_c(\mathbf{k}, \mathbf{k} + \mathbf{k}', \mathbf{k}') = \frac{1}{2} [W_a^b{}_c(\mathbf{k}, \mathbf{k} + \mathbf{k}', \mathbf{k}') + W_c^b{}_a(\mathbf{k}', \mathbf{k} + \mathbf{k}', \mathbf{k})]. \quad (5.77)$$

The symmetrizer sums over all the permutations of the indices of the same level, upper or lower indices, simultaneously with the associated momenta. We list the properties of $\bar{W}_c^{ab}(\mathbf{k}, \mathbf{k}', \mathbf{k} + \mathbf{k}')$ in the following:

1. $\bar{W}_c^{ab}(\mathbf{k}, \mathbf{k}', \mathbf{k} + \mathbf{k}') = \bar{W}_a^b{}_c(-\mathbf{k}, -\mathbf{k} - \mathbf{k}', -\mathbf{k}')$,
2. $\bar{W}_c^{ab}(\mathbf{k}, \mathbf{k}', \mathbf{k} + \mathbf{k}')^* = \bar{W}_{ba}^c(\mathbf{k} + \mathbf{k}', \mathbf{k}', \mathbf{k})$,
3. $\bar{W}_{ab}^c(\mathbf{k} + \mathbf{k}', \mathbf{k}, \mathbf{k}') = \bar{W}_a^c{}_b(\mathbf{k}, \mathbf{k} + \mathbf{k}', \mathbf{k}')$,
4. $\bar{W}_c^{ab}(\mathbf{k}, \mathbf{k}', \mathbf{k} + \mathbf{k}') = \bar{W}_c^{ba}(\mathbf{k}', \mathbf{k}, \mathbf{k} + \mathbf{k}')$,
5. $\bar{W}_a^c{}_b(\mathbf{k}, \mathbf{k} + \mathbf{k}', \mathbf{k}') = \bar{W}_b^c{}_a(\mathbf{k}', \mathbf{k} + \mathbf{k}', \mathbf{k})$.

The Hamiltonian can also be expressed:

$$\hat{H}_3 = \frac{1}{\sqrt{N_p}} \sum_{\substack{\mathbf{k}, \mathbf{k}' \\ a, b, c}} \left(W_c^{ab}(\mathbf{k}, \mathbf{k}', \mathbf{k} + \mathbf{k}') \hat{f}_{a, \mathbf{k}}^\dagger \hat{f}_{b, \mathbf{k}'}^\dagger \hat{f}_{c, \mathbf{k} + \mathbf{k}'} + h.c \right) \quad (5.78)$$

We calculate the quantum correction from $\hat{H}_3$ using second order perturbation theory. And since the spin wave Hamiltonian has been diagonalized into the quasi-particle Schwinger bosons, the perturbative Hamiltonian $\hat{H}_3$ should be written in the quasi-particle representation.

Using Eq.5.64-5.66, we can transform the perturbative Hamiltonian $\hat{H}_3$ into the quasi-particle basis $\gamma_{\alpha, \mathbf{k}}$. Each vertex in the original basis $\hat{f}_{a, \mathbf{k}}$ will split into 8 vertices formed

by the quasi-particles operators. The perturbative Hamiltonian then becomes:

$$\begin{aligned} \hat{H}_3 = \frac{1}{\sqrt{N_p}} \sum_{\substack{\mathbf{k}, \mathbf{k}' \\ m, n, l}} & \left(\Gamma^{mnl}(\mathbf{k}, \mathbf{k}', -\mathbf{k} - \mathbf{k}') \hat{\gamma}_{m, \mathbf{k}}^\dagger \hat{\gamma}_{n, \mathbf{k}'}^\dagger \hat{\gamma}_{l, -\mathbf{k} - \mathbf{k}'}^\dagger + \Gamma^{mn}_l(\mathbf{k}, \mathbf{k}', \mathbf{k} + \mathbf{k}') \hat{\gamma}_{m, \mathbf{k}}^\dagger \hat{\gamma}_{n, \mathbf{k}'}^\dagger \hat{\gamma}_{l, \mathbf{k} + \mathbf{k}'}^\dagger \right. \\ & \left. + \Gamma_m^{nl}(\mathbf{k} + \mathbf{k}', \mathbf{k}, \mathbf{k}') \hat{\gamma}_{m, \mathbf{k} + \mathbf{k}'} \hat{\gamma}_{n, \mathbf{k}}^\dagger \hat{\gamma}_{l, \mathbf{k}'}^\dagger + \Gamma_n^{ml}(\mathbf{k}, \mathbf{k} + \mathbf{k}', \mathbf{k}') \hat{\gamma}_{m, \mathbf{k}}^\dagger \hat{\gamma}_{n, \mathbf{k} + \mathbf{k}'} \hat{\gamma}_{l, \mathbf{k}'}^\dagger + h.c. \right) \end{aligned} \quad (5.79)$$

The new coefficients are given by:

$$\begin{aligned} \Gamma^{mnl}(\mathbf{k}, \mathbf{k}', -\mathbf{k} - \mathbf{k}') = & \bar{W}_c^{ab}(\mathbf{k}, \mathbf{k}', \mathbf{k} + \mathbf{k}') U_{\mathbf{k}, am}^* U_{\mathbf{k}', bn}^* V_{\mathbf{k} + \mathbf{k}', lc} \\ & + \bar{W}_{ab}{}^c(-\mathbf{k}, -\mathbf{k}', -\mathbf{k} - \mathbf{k}') V_{\mathbf{k}, am}^* V_{\mathbf{k}', bn}^* U_{\mathbf{k} + \mathbf{k}', cl} \end{aligned} \quad (5.80)$$

$$\begin{aligned} \Gamma^{mn}_l(\mathbf{k}, \mathbf{k}', \mathbf{k} + \mathbf{k}') = & \bar{W}_c^{ab}(\mathbf{k}, \mathbf{k}', \mathbf{k} + \mathbf{k}') U_{\mathbf{k}, am}^* U_{\mathbf{k}', bn}^* U_{\mathbf{k} + \mathbf{k}', cl} \\ & + \bar{W}_{ab}{}^c(-\mathbf{k}, -\mathbf{k}', -\mathbf{k} - \mathbf{k}') V_{\mathbf{k}, am}^* V_{\mathbf{k}', bn}^* V_{\mathbf{k} + \mathbf{k}', cl} \end{aligned} \quad (5.81)$$

$$\begin{aligned} \Gamma_m^{nl}(\mathbf{k}, \mathbf{k}', \mathbf{k} - \mathbf{k}') = & \bar{W}_c^{ab}(-\mathbf{k}, \mathbf{k}', -\mathbf{k} + \mathbf{k}') V_{\mathbf{k}, am} U_{\mathbf{k}', bn}^* V_{-\mathbf{k} + \mathbf{k}', cl} \\ & + \bar{W}_{ab}{}^c(\mathbf{k}, -\mathbf{k}', \mathbf{k} - \mathbf{k}') U_{\mathbf{k}, am} V_{\mathbf{k}', bn}^* U_{-\mathbf{k} + \mathbf{k}', cl} \end{aligned} \quad (5.82)$$

$$\begin{aligned} \Gamma_n^{ml}(\mathbf{k}, \mathbf{k}', -\mathbf{k} + \mathbf{k}') = & \bar{W}_c^{ab}(\mathbf{k}, -\mathbf{k}', \mathbf{k} - \mathbf{k}') U_{\mathbf{k}, am}^* V_{\mathbf{k}', bn} V_{\mathbf{k} - \mathbf{k}', cl} \\ & + \bar{W}_{ab}{}^c(-\mathbf{k}, \mathbf{k}', -\mathbf{k} + \mathbf{k}') V_{\mathbf{k}, am}^* U_{\mathbf{k}', bn} U_{\mathbf{k} - \mathbf{k}', cl} \end{aligned} \quad (5.83)$$

The two 15×15 matrices $U_{\mathbf{k}}$ and $V_{\mathbf{k}}$ are block diagonal with $U_{\alpha, \mathbf{k}}$ and $V_{\alpha, \mathbf{k}}$ along the diagonal, respectively.

$$U_{\mathbf{k}} = \begin{pmatrix} U_{x, \mathbf{k}} & \mathbf{0}_4 & \mathbf{0}_4 & \mathbf{0}_3 \\ \mathbf{0}_4 & U_{y, \mathbf{k}} & \mathbf{0}_4 & \mathbf{0}_3 \\ \mathbf{0}_4 & \mathbf{0}_4 & U_{z, \mathbf{k}} & \mathbf{0}_3 \\ \mathbf{0}_3 & \mathbf{0}_3 & \mathbf{0}_3 & \mathbf{I}_3 \end{pmatrix} \quad \text{and} \quad V_{\mathbf{k}} = \begin{pmatrix} V_{x, \mathbf{k}} & \mathbf{0}_4 & \mathbf{0}_4 & \mathbf{0}_3 \\ \mathbf{0}_4 & V_{y, \mathbf{k}} & \mathbf{0}_4 & \mathbf{0}_3 \\ \mathbf{0}_4 & \mathbf{0}_4 & V_{z, \mathbf{k}} & \mathbf{0}_3 \\ \mathbf{0}_3 & \mathbf{0}_3 & \mathbf{0}_3 & \mathbf{I}_3 \end{pmatrix} \quad (5.84)$$

$\mathbf{0}_n$ and $\mathbf{I}_n$ are $n \times n$ zero matrix and identity matrix, respectively.

Again, we realize that some of the coefficients are actually represent the same vertex, therefore, we perform the symmetrization on the new coefficient as well.

$$\begin{aligned}\bar{\Gamma}^{mnl}(\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3) &= \hat{P}_3 \Gamma^{mnl}(\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3) \\ &= \frac{1}{3!} (\Gamma^{mnl}(\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3) + \Gamma^{mln}(\mathbf{k}_1, \mathbf{k}_3, \mathbf{k}_2) + \Gamma^{nml}(\mathbf{k}_2, \mathbf{k}_1, \mathbf{k}_3) \\ &\quad + \Gamma^{nlm}(\mathbf{k}_2, \mathbf{k}_3, \mathbf{k}_1) + \Gamma^{lmn}(\mathbf{k}_3, \mathbf{k}_1, \mathbf{k}_2) + \Gamma^{lnm}(\mathbf{k}_3, \mathbf{k}_2, \mathbf{k}_1))\end{aligned}\quad (5.85)$$

$$\begin{aligned}\bar{\Gamma}^{mn}_l(\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3) &= \hat{P}_2 [\Gamma^{mn}_l(\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3) + \Gamma^{m\ n}_l(\mathbf{k}_1, \mathbf{k}_3, \mathbf{k}_2) + \Gamma_l^{mn}(\mathbf{k}_3, \mathbf{k}_1, \mathbf{k}_2)] \\ &= \frac{1}{2!} (\Gamma^{mn}_l(\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3) + \Gamma^{m\ n}_l(\mathbf{k}_1, \mathbf{k}_3, \mathbf{k}_2) + \Gamma^{nm}_l(\mathbf{k}_2, \mathbf{k}_1, \mathbf{k}_3) \\ &\quad + \Gamma_l^{n\ m}(\mathbf{k}_2, \mathbf{k}_3, \mathbf{k}_1) + \Gamma_l^{mn}(\mathbf{k}_3, \mathbf{k}_1, \mathbf{k}_2) + \Gamma_l^{nm}(\mathbf{k}_3, \mathbf{k}_2, \mathbf{k}_1))\end{aligned}\quad (5.86)$$

$\hat{P}_n$ is the symmetrizer that averages over all the n permutations of the indices of the same level and the associated momenta simultaneously. Besides the symmetry property, the symmetrized coefficients also satisfy:

$$\bar{\Gamma}^{mnl} = (\bar{\Gamma}_{mnl})^* \quad \text{and} \quad \bar{\Gamma}^{mn}_l = (\bar{\Gamma}_{mn}^{\ l})^*. \quad (5.87)$$

The perturbative Hamiltonian now has a more compact and symmetric form with normal ordering:

$$\begin{aligned}\hat{H}_3 &= \frac{1}{\sqrt{N_p}} \sum_{\substack{\mathbf{k}, \mathbf{k}' \\ m, n, l}} \left(\bar{\Gamma}^{mnl}(\mathbf{k}, \mathbf{k}', -\mathbf{k} - \mathbf{k}') \hat{\gamma}_{m, \mathbf{k}}^\dagger \hat{\gamma}_{n, \mathbf{k}'}^\dagger \hat{\gamma}_{l, -\mathbf{k} - \mathbf{k}'}^\dagger \right. \\ &\quad \left. + \bar{\Gamma}^{mn}_l(\mathbf{k}, \mathbf{k}', \mathbf{k} + \mathbf{k}') \hat{\gamma}_{m, \mathbf{k}}^\dagger \hat{\gamma}_{n, \mathbf{k}'}^\dagger \hat{\gamma}_{l, \mathbf{k} + \mathbf{k}'} + h.c. \right)\end{aligned}\quad (5.88)$$

5.3.2 Perturbative correction of $\hat{H}_3$

The effective Hamiltonian including the cubic terms now becomes:

$$\hat{H} = N_p E_0 + \hat{H}_I + \hat{H}_{SW} + \hat{H}_3. \quad (5.89)$$

We consider $\hat{H}_3$ as our perturbative Hamiltonian. Because the first term is only a scalar constant, we will take the unperturbed Hamiltonian to be:

$$\hat{H}_0 = \hat{H}_I + \hat{H}_{SW}. \quad (5.90)$$

By doing so, all the energy levels have been shifted positively by $N_p E_0$. The ground state energy now becomes zero, $E_{gs} = 0$.

It is obvious that the first order perturbation gives zero contribution to the ground state energy. Hence, the lowest order correction comes from the second order perturbation, which is given by

$$E_{gs}^{(2)} = \langle gs | \hat{H}_3 (E_{gs} - \hat{H}_0)^{-1} Q_{gs} \hat{H}_3 | gs \rangle = - \langle gs | \hat{H}_3 \hat{H}_0^{-1} Q_{gs} \hat{H}_3 | gs \rangle \quad (5.91)$$

where Q_{gs} is the projection operator that projects out the ground state, that is to the excited states. Since the ground state $|gs\rangle$ is actually the vacuum state with respect to the 15 Schwinger bosons and the vertices in $\hat{H}_3$ are in normal order, only two types of vertices contribute to the second order perturbation. They are $\hat{\gamma}_{m,\mathbf{k}}^\dagger \hat{\gamma}_{n,\mathbf{k}'}^\dagger \hat{\gamma}_{l,-\mathbf{k}-\mathbf{k}'}^\dagger$ and $\hat{\gamma}_{m,\mathbf{k}} \hat{\gamma}_{n,\mathbf{k}'} \hat{\gamma}_{l,-\mathbf{k}-\mathbf{k}'}$, or in terms of diagram, Fig.5.4. The correction can then be written

Figure 5.4: The only two contributing vertices on the left. The vertex on the right is the contraction of the two vertices.

explicitly as:

$$E_{gs}^{(2)} = \frac{1}{N_p} \sum_{\substack{\mathbf{k}_1, \mathbf{k}'_1 \\ a, b, c}} \sum_{\substack{\mathbf{k}_2, \mathbf{k}'_2 \\ m, n, l}} \langle gs | \hat{\gamma}_{a, \mathbf{k}_1} \hat{\gamma}_{b, \mathbf{k}'_1} \hat{\gamma}_{c, -\mathbf{k}_1 - \mathbf{k}'_1} \hat{\gamma}_{m, \mathbf{k}_2}^\dagger \hat{\gamma}_{n, \mathbf{k}'_2}^\dagger \hat{\gamma}_{l, -\mathbf{k}_2 - \mathbf{k}'_2}^\dagger | gs \rangle (\varepsilon_{m, \mathbf{k}_2} + \varepsilon_{n, \mathbf{k}'_2} + \varepsilon_{l, -\mathbf{k}_2 - \mathbf{k}'_2})^{-1} \\ \cdot \bar{\Gamma}_{abc}(\mathbf{k}_1, \mathbf{k}'_1, -\mathbf{k}_1 - \mathbf{k}'_1) \bar{\Gamma}^{mnl}(\mathbf{k}_2, \mathbf{k}'_2, -\mathbf{k}_2 - \mathbf{k}'_2) \quad (5.92)$$

where $\varepsilon_{m, \mathbf{k}}$ are the energies of the quasi-particles, Eq.5.47 and Eq.5.52. All the 15 excitations have been absorbed into one index. It is obvious from the diagram language that only the vertex on the right in Fig.5.4 has finite value.

Algebraically, using Wick's theorem, all the normal ordering terms become zero. Only the contraction contributes to the correction which is

$$E_{gs}^{(2)} = \frac{3!}{N_p} \sum_{\substack{\mathbf{k}, \mathbf{k}' \\ m, n, l}} \hat{P}_3 [\bar{\Gamma}_{mnl}(\mathbf{k}, \mathbf{k}', -\mathbf{k} - \mathbf{k}')] \bar{\Gamma}^{mnl}(\mathbf{k}, \mathbf{k}', -\mathbf{k} - \mathbf{k}') (\varepsilon_{m, \mathbf{k}} + \varepsilon_{n, \mathbf{k}'} + \varepsilon_{l, -\mathbf{k} - \mathbf{k}'})^{-1} \quad (5.93)$$

where, again, $\hat{P}_3$ is the symmetrizer that averages all the permutation of the index tuple (m, n, l) simultaneously with the associated momenta. Because the coefficients $\bar{\Gamma}_{mnl}(\mathbf{k}, \mathbf{k}', -\mathbf{k} - \mathbf{k}')$ are symmetric under permutation, we have:

$$E_{gs}^{(2)} = \frac{3!}{N_p} \sum_{\substack{\mathbf{k}, \mathbf{k}' \\ m, n, l}} |\bar{\Gamma}^{mnl}(\mathbf{k}, \mathbf{k}', -\mathbf{k} - \mathbf{k}')|^2 (\varepsilon_{m, \mathbf{k}} + \varepsilon_{n, \mathbf{k}'} + \varepsilon_{l, -\mathbf{k} - \mathbf{k}'})^{-1}. \quad (5.94)$$

We have also used Eq.5.87. We can further utilize the symmetric property to simplify the calculation. The summation over the ordered indices tuple (m, n, l) can be reduced to summation over un-ordered tuple $\{m, n, l\}$. Thus, we finally have the quantum correction from the cubic Hamiltonian $\hat{H}_3$ using second order perturbation theory:

$$E_{gs}^{(2)} = \frac{36}{N_p} \sum_{\substack{\mathbf{k}, \mathbf{k}' \\ \{m, n, l\}}} |\bar{\Gamma}^{mnl}(\mathbf{k}, \mathbf{k}', -\mathbf{k} - \mathbf{k}')|^2 (\varepsilon_{m, \mathbf{k}} + \varepsilon_{n, \mathbf{k}'} + \varepsilon_{l, -\mathbf{k} - \mathbf{k}'})^{-1}. \quad (5.95)$$

The ground state energy per lattice site with this correction is then given by:

$$E_{gs} = \frac{1}{4}E_0 + \frac{1}{2N} \sum_{\alpha, \mathbf{k}, n} \left(\varepsilon_{\alpha, \mathbf{k}}^{(n)} + E_0 \right) + \frac{E_{gs}^{(2)}}{N}. \quad (5.96)$$

The ground state energies for the isotropic Hamiltonian ($\Delta = 1$) both with and without

Figure 5.5: Ground state energy of the original Shastry-Sutherland model ($\Delta = 1$). The gap shows the importance of the cubic terms.

correction $E_{gs}^{(2)}$ are shown in Fig.5.5. The contribution from $\hat{H}_3$ is obviously non-trivial. The inset shows the magnitude of the correction $E_{gs}^{(2)}$ which decreases towards large J_2/J_1 . This is actually closely related to the behaviour of the dispersion relations. As we will show in the next section, when J_2 increases, the gaps of the dispersions become larger. The decrease is then obvious from Eq.5.95. We also realize, from the blue curve, that quantum fluctuations from the spin wave Hamiltonian is becoming smaller for large J_2 since the classical ground state energy does not depend on J_2 . This is also explained in the next section.

5.4 Phase diagram

In this section, we show partial phase diagram of the generalized Shastry-Sutherland model in the $(J_2/J_1, \Delta)$ space. There are three competing phases: antiferromagnetic phase (AFM) with long range magnetic ordering, the plaquette valence bond solid (PVBS) and the dimer singlet (DS). We also show an unexpected and interesting phenomenon of the dispersion relations.

5.4.1 Towards AFM phase

The AFM phase is the ground state for small J_2/J_1 . Therefore, when J_2/J_1 decreases in the PVBS phase, there must be a critical point where the PVBS is taken over by the AFM phase. This turns out to be a continuous phase transition. Starting from a point in PVBS, the gap of the dispersion vanishes towards small J_2/J_1 . Besides, the gap becomes smaller as well when Δ is away from 1. In fact, if the minimum gap of the dispersion occurs at $\mathbf{k} = (0, 0)$, it could be expressed in a closed form as:

$$\left\{ \begin{array}{ll} E_g^{XY} = \sqrt{(J_1 + E_0) [(J_1 + E_0) - 8(J_2 - 2J_1)e_x^2]} & \text{for } \Delta < 1, \\ E_g^I = \sqrt{(\Delta J_1 + E_0) [(\Delta J_1 + E_0) - 8\Delta(J_2 - 2J_1)e_z^2]} & \text{for } \Delta \geq 1. \end{array} \right. \quad (5.97)$$

Because $(J_1^\alpha + E_0)$ is always negative, Eq.5.21, it is then obvious that the gap is a monotonic decreasing function of J_2/J_1 . The tendency of the change of the gap with respect to Δ is not obvious from the above equations. A plot of the gap with respect to Δ at a fixed J_2/J_1 helps. Take $J_2/J_1 = 1.4$ as an example, Figure 5.6 shows that, indeed, the gap closes away from isotropic point $\Delta = 1$.

The figure also shows the positions where the gap closes in both Ising- and XY-like regions. From Equation 5.97, we can actually obtain the critical value J_2/J_1 for each Δ

at which the gap vanishes.

$$\begin{cases} \left(\frac{J_2}{J_1}\right)_c^{XY} = 2 + \frac{J_1 + E_0}{8J_1 e_x^2} & \text{for } \Delta < 1, \\ \left(\frac{J_2}{J_1}\right)_c^I = 2 + \frac{\Delta J_1 + E_0}{8\Delta J_1 e_z^2} & \text{for } \Delta \geq 1. \end{cases} \quad (5.98)$$

The above equations for the critical points are only valid provided that the minimum gap of the dispersion relation appears at $\mathbf{k} = (0, 0)$, which is true for a large area in the PVBS phase. Particularly, in the isotropic case ($\Delta = 1$), the critical point is given by 1.25. These two critical lines are shown in Fig.5.8, the phase boundary between the AFM and the PL1. The vanishing of the gap along the boundary implies a continuous phase transition from the PVBS phase to the AFM phase. Both J_2/J_1 and Δ could drive this continuous phase transition. When it is driven by the anisotropy Δ , the transition is similar to the Bose-Einstein condensation (BEC) which is driven by the chemical potential. The Δ plays a similar role as the chemical potential, therefore, this transition may have the same universality class as BEC. On the other hand, when it is driven by J_2/J_1 , it is effectively driven by the transverse interaction which is similar to the XY phase transition. Thus, this transition could belong to the XY universality class.

5.4.2 The two PVBS

We have obtained the phase boundary between AFM and the PVBS in the last subsection by assuming the fact the minimum gap occurs at $\mathbf{k} = (0, 0)$. However, it is clear from Fig.5.7 that the minimum gap does change its location. We see that at some points, there are 4 degenerate points that have the same minimum gap.

We can actually derive the critical line where the minimum gap changes location. When the minimum gap is located at $\mathbf{k} = (0, 0)$, the dispersion relation has local minimum at $\mathbf{k} = (0, 0)$. And when the minimum is no longer at $\mathbf{k} = (0, 0)$, the dispersion has a local maximum at $\mathbf{k} = (0, 0)$ instead. Thus, this change could be detected by second derivative test. Suppose $\varepsilon_{\mathbf{k}}$ is the lowest branch dispersion and its Hessian matrix with

Figure 5.6: The figure shows the square of the gap versus Δ . The two gaps meet at $\Delta = 1$ for any J_2/J_1 . Both Ising-like gap and XY-like gap touch zero at certain values of Δ implying instability of the corresponding quasi-particles.

respect to $\mathbf{k}$ at $\mathbf{k} = (0, 0)$ is:

$$H = \begin{pmatrix} \frac{\partial^2 \varepsilon_{\mathbf{k}}}{\partial k_x^2} & \frac{\partial^2 \varepsilon_{\mathbf{k}}}{\partial k_x \partial k_y} \\ \frac{\partial^2 \varepsilon_{\mathbf{k}}}{\partial k_x \partial k_y} & \frac{\partial^2 \varepsilon_{\mathbf{k}}}{\partial k_y^2} \end{pmatrix}_{\mathbf{k}=(0,0)} \quad (5.99)$$

The second derivative test states:

1. If the determinant of H , $\det\{H\} > 0$, and $\frac{\partial^2 \varepsilon_{\mathbf{k}}}{\partial k_x^2} > 0$, then $\varepsilon_{\mathbf{k}}$ has local minimum at $\mathbf{k} = (0, 0)$;
2. If the determinant $\det\{H\} > 0$ and $\frac{\partial^2 \varepsilon_{\mathbf{k}}}{\partial k_x^2} < 0$, then $\varepsilon_{\mathbf{k}}$ has a local maximum at $\mathbf{k} = (0, 0)$;
3. If $\det\{H\} < 0$, then $\varepsilon_{\mathbf{k}}$ has a saddle point at $\mathbf{k} = (0, 0)$;
4. If $\det\{H\} = 0$, the test is inconclusive.

The derivatives of the dispersion relations can be obtained from Eq.5.52. The critical line is then obtained as shown in the red line in Figure 5.8. The plaquette valence

bond solid phase in the PL1 region has excitation dispersions with minimum gap at $\mathbf{k} = (0,0)$ while in the PL2 region, the excitations above the plaquette valence bond solid have four minima. The four minima are symmetric under Z_4 symmetry group generated by the transformation: $(k_x, k_y) \rightarrow (-k_y, k_x)$. This critical line intersects the phase boundary between AFM and PVBS at two points: $(J_2/J_1 = 1.210, \Delta = 1.450)$ and $(J_2/J_1 = 1.448, \Delta = 0.630)$. We also find that away from these two points along the PVBS-AFM boundary, the dispersion relation becomes linear while remaining gapless. In the PL1 region, it is linear at $\mathbf{k} = (0,0)$ while in the PL2 region, it is linear at the four minima. This phenomenon is shown in the 4 typical dispersion relations in Fig.5.7.

Figure 5.7: 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 $J_2/J_1 = 1.450$: gapless and smooth at $\mathbf{k} = (0,0)$; (d) $\Delta = 0.6$ and $J_2/J_1 = 1.448$: gapped with four minima at $\pm(0.17\pi, 0.12\pi)$ and $\pm(0.12\pi, -0.17\pi)$.

The two different PVBS phases are closely related to the stability of the plaquette state over different quantum fluctuations. There are actually two ways to form the plaquette valence bond solid. Besides the configuration in Fig.5.2, the unit cell can also be chosen on the other empty squares. As a result, we will obtain another plaquette valence bond solid where the plaquette is formed on the these empty square.

When J_2/J_1 is small, the quantum fluctuation that drives the transition between the

two PVBS phases is weak. The dominating quantum fluctuations are those particle-like excitations, viz., the quasi-particles that are created on a unit cell and become dispersive through the strong J_1 and relatively weak J_2 interactions. In other words, the structure of the plaquette or the unit cell is robust against the hopping through J_1 and J_2 . This robustness could also be enhanced by the anisotropy Δ . From the relation between the excitation gap and Δ , we see that both Ising-like and XY-like anisotropy will stabilize the excitation as an entity. That is the plaquette structure will be preserved. This could be seen from Fig.5.8 as well. In a small range of J_2/J_1 , when Δ is away from 1, it is possible to enter the PL1 phase from the PL2 phase.

As J_2 increases, the quantum fluctuation now prefers to drive the transition between the two configurations, that is the two choices of the unit cell. Unlike the excitations in PL1 which are mediated by the hopping process, the excitations in PL2 are more like a bridge that link the two PVBS which have the same energy. In other words, the structure of the plaquette as a single entity has become unstable. The resonance between the two PVBS through the quantum fluctuation becomes dominating. This also explains why there is less correction to the PVBS energy from quantum fluctuations.

5.4.3 Towards dimer singlet

We have already known that for $\Delta = 1$, when J_2 is strong, the ground state is the dimer singlet phase where a singlet is formed in each J_2 bond. This is also true in the anisotropic case. When J_2 increases further in the PL2 phase, the plaquette valence bond solid is dissolved and the dimer singlet is formed. The ground state energy of the anisotropic dimer singlet phase could be obtained by diagonalizing the anisotropic two-spin interaction. The energy per lattice is given by:

$$E_D = -\frac{1}{8}(2 + \Delta)J_2. \quad (5.100)$$

A comparison between this energy and the energy of the PVBS (5.96) gives a first order phase transition from the PVBS to the dimer singlet phase. The black line in Figure 5.8

is the first order phase boundary between the PVBS (PL2) and the dimer singlet phase. Particularly, the critical point at the isotropic point is given by 1.456(1), which agrees quite well with the tensor network study[63]. This critical line intersects with the PVBS-AFM boundary at two points: $(J_2/J_1 = 1.514, \Delta = 1.3)$ and $(J_2/J_1 = 1.650, \Delta = 0.33)$. Hence we have obtained a generalized plaquette valence bond solid phase which is enclosed by the AFM and the dimer singlet phase. In other words, the PVBS phase is strongly suppressed by the AFM phase in the anisotropic case.

Figure 5.8: The phase range of the plaquette state. The PL1-AFM line is exact on the left of the critical line obtained from Eq.5.98. The five critical points $(J_2/J_1, \Delta)$ are: $E1 = (1.25, 1)$ $E2 = (1.514, 1.3)$, $E3 = (1.650, 0.33)$, $\Delta_1 = (1.21, 1.450)$ and $\Delta_2 = (1.448, 0.63)$.

5.5 Conclusion

We have obtained a phase diagram for the anisotropic Shastry-Sutherland model for a certain parameter range where we have focused on a generalized plaquette valence bond solid. We found that there were two PVBS phases with very different behaviours of excitations. In PL1, small J_2/J_1 , the lowest excitation has minimum at $\mathbf{k} = (0, 0)$ and the dominating quantum fluctuation preserves the plaquette structure. In PL2, large J_2/J_1 , the lowest excitation has four minima and the quantum fluctuation tries to dissolve the

plaquette structure by generating resonance between the two configurations of the PVBS.

In the PVBS region, we found that both anisotropy and strong J_1 will drive the phase transition from the PVBS to the AFM phase. However they could belong to different universality class: BEC universality when driven by the anisotropy and XY universality class when driven by J_1 . Along the PVBS-AFM boundary, the lowest dispersion relation is gapless and tends to be linear away from critical points between PL1 and PL2. As J_2 increases, it drives a first order phase transition from the PL2 PVBS to the dimer singlet phase.

Chapter 6

Plateaus in Extended Shastry-Sutherland Model

One of the most interesting phenomena in the rare-earth tetraborides compounds is the appearance of the magnetization plateaus. There have been several attempts[15, 16, 65, 66, 67, 68] to explain the mechanism and the plateau configurations by numerically studying the extended Shastry-Sutherland model (SSM). Both spin anisotropy Δ and further interactions J_3 have been considered. It has been shown that the Shastry-Sutherland model with pure Ising interactions is not enough to explain the plateau sequences. There is only one $1/3$ plateau in the Ising limit[15, 16]. With the transverse interactions included, there is one more narrow $1/2$ plateau[15]. To produce a complete magnetization plateau sequence, we have to extend the interaction range from J_1 and J_2 in the original SSM to four interactions: J_1 , J_2 , J_3 and J_4 as shown in Fig.1.4. These further interactions are consequences of the RKKY interactions between the itinerant electrons and localized magnetic moments. In this chapter, we explore possible magnetization plateaus in the strong Ising limit, using two complementing approaches. On the one hand, we use SSE QMC to study an effective low energy $S = 1/2$ XXZ model. On the other hand, we develop a Schwinger boson approach to gain better insight into the nature of and the mechanism behind the emergence of the plateaus. We start with a detailed description of the latter.

Figure 6.1: The spiral plaquette contains four dimers.

6.1 Ising limit

6.1.1 Spiral plaquette

We first consider the pure Ising interactions in the extended SSM with four interactions. We found that a spiral plaquette (s-plaquette) was a good candidate to investigate the plateau configurations in the Ising limit. Figure 6.1 shows the configuration of this spiral plaquette which contains 8 lattice sites or 4 dimers. The position of the s-plaquette is represented by the center of the square surrounded by the four dimers. We denote the state of the s-plaquette $|sp_i\rangle$ in terms of the four dimer states:

$$|sp_i\rangle = |a_1, a_2, a_3, a_4\rangle_i \quad (6.1)$$

where $|a_n\rangle_i$ is the state of the n^{th} dimer with the center of the square at $\mathbf{r}_i$. Each dimer state could be represented by three different basis. The first one consists of direct products of the spin states at the two lattice sites connected by the dimer which is more convenient in the strong Ising limit. The other two consist of one singlet and one triplet.

The three bases are shown below.

$$\left\{ \begin{array}{l} |u\rangle = |\uparrow\uparrow\rangle \\ |d\rangle = |\downarrow\downarrow\rangle \\ |l\rangle = |\uparrow\downarrow\rangle \\ |r\rangle = |\downarrow\uparrow\rangle \end{array} \right., \quad \left\{ \begin{array}{l} |t_1\rangle = |u\rangle \\ |t_{-1}\rangle = |d\rangle \\ |t_0\rangle = \frac{1}{\sqrt{2}}(|l\rangle + |r\rangle) \\ |s\rangle = \frac{1}{\sqrt{2}}(|l\rangle - |r\rangle) \end{array} \right. \quad \text{and} \quad \left\{ \begin{array}{l} |t_x\rangle = -\frac{1}{\sqrt{2}}(|u\rangle - |d\rangle) \\ |t_y\rangle = \frac{i}{\sqrt{2}}(|u\rangle + |d\rangle) \\ |t_z\rangle = \frac{1}{\sqrt{2}}(|l\rangle + |r\rangle) \\ |s\rangle = \frac{1}{\sqrt{2}}(|l\rangle - |r\rangle) \end{array} \right. . \quad (6.2)$$

In all bases, the first state in $|s_1, s_2\rangle$ represents the spin state at the corner of the central square while the second state represents the spin state at the corresponding edge of the s-plaquette. The first basis actually consists of eigen states of the Ising interactions $\hat{S}_l^z \hat{S}_r^z$ where $\hat{S}_l^\alpha$ ($\alpha = x, y, z$) denotes the spin operator at the corner of the central square and $\hat{S}_r^\alpha$ denotes the one at the edge. The second basis are eigen states of the Heisenberg interaction $\hat{S}_l^\alpha \hat{S}_r^\alpha$, where Einstein summation convention has been applied as before. In the last basis, the triplet are eigen states of the Spin-1 operator $\hat{T}^\alpha$ with eigenvalue zero.

It is obvious from the figure that there are constraints in this s-plaquette representation. The 3rd (4th) dimer state at $\mathbf{r}_i$ is the same as the 1st (2nd) state at $\mathbf{r}_i + \mathbf{x}$ ($\mathbf{y}$) where $\mathbf{x}$ ($\mathbf{y}$) is the horizontal (vertical) vector connecting two adjacent s-plaquette with one common dimer. However, the edge of dimer 3 will become the corner of the central square in dimer 1. Hence, we define the following convention:

$$|\bar{u}\rangle = |u\rangle, \quad |\bar{d}\rangle = |d\rangle, \quad |\bar{l}\rangle = |r\rangle \quad \text{and} \quad |\bar{r}\rangle = |l\rangle. \quad (6.3)$$

The constraint can be expressed as

$$|a_3\rangle_i = |\bar{a}_1\rangle_{i+x} \quad \text{and} \quad |a_4\rangle_i = |\bar{a}_2\rangle_{i+y}. \quad (6.4)$$

The below equality follows immediately:

$$\hat{S}_{3,l,i}^\alpha = \hat{S}_{1,r,i+x}^\alpha, \quad \hat{S}_{3,r,i}^\alpha = \hat{S}_{1,l,i+x}^\alpha, \quad \hat{S}_{4,r,i}^\alpha = \hat{S}_{2,l,i+y}^\alpha \quad \text{and} \quad \hat{S}_{4,l,i}^\alpha = \hat{S}_{2,r,i+y}^\alpha. \quad (6.5)$$

In this representation, both J_1 and J_2 become internal interactions within the s-plaquette while J_3 and J_4 mediate the interactions between nearest neighbour s-plaquettes. The Hamiltonian is given by:

$$\begin{aligned} \hat{H} = & \sum_{i,n} \frac{1}{2} J_2^\alpha \hat{S}_{n,l,i}^\alpha \hat{S}_{n,r,i}^\alpha - h(\hat{S}_{n,l,i}^z + \hat{S}_{n,r,i}^z) + J_1^\alpha (\hat{S}_{n,l,i}^\alpha + \hat{S}_{n,r,i}^\alpha) \hat{S}_{n+1,l,i}^\alpha \\ & + J_3^\alpha \sum_i \hat{S}_{1,l,i}^\alpha \hat{S}_{1,r,i+x}^\alpha + \hat{S}_{1,l,i}^\alpha \hat{S}_{1,r,i+y}^\alpha + \hat{S}_{2,l,i}^\alpha \hat{S}_{2,r,i+y}^\alpha + \hat{S}_{2,r,i}^\alpha \hat{S}_{2,l,i+x}^\alpha \\ & + J_4^\alpha \sum_{\langle i,j \rangle} \hat{S}_{1,l,i}^\alpha \hat{S}_{1,l,j}^\alpha + \hat{S}_{1,r,i}^\alpha \hat{S}_{1,r,j}^\alpha + \hat{S}_{2,l,i}^\alpha \hat{S}_{2,l,j}^\alpha + \hat{S}_{2,r,i}^\alpha \hat{S}_{2,r,j}^\alpha \end{aligned} \quad (6.6)$$

where $\langle i, j \rangle$ sums over all nearest neighbours and $J_m^\alpha = (\Delta J_m, \Delta J_m, J_m)$ for all m . In the J_2 term above, the spin operator is periodic in n with period 4.

We introduce Schwinger bosons for each state as in the spin wave theory. The spin operators in the n^{th} dimer can be expressed as:

$$\hat{S}_{n,l}^\alpha = \frac{1}{2} (\hat{t}_{n,\alpha}^\dagger \hat{s}_n + \hat{s}_n^\dagger \hat{t}_{n,\alpha}) + \frac{1}{2} \hat{T}_n^\alpha, \quad (6.7)$$

$$\hat{S}_{n,r}^\alpha = -\frac{1}{2} (\hat{t}_{n,\alpha}^\dagger \hat{s}_n + \hat{s}_n^\dagger \hat{t}_{n,\alpha}) + \frac{1}{2} \hat{T}_n^\alpha \quad (6.8)$$

where $\alpha = x, y, z$ and the Spin-1 operator is given by

$$\hat{T}_n^\alpha = -i\epsilon^{\alpha\beta\gamma} \hat{t}_\beta^\dagger \hat{t}_\gamma. \quad (6.9)$$

In the Ising limit, it is more convenient to work in the basis of $\{|u\rangle, |d\rangle, |l\rangle, |r\rangle\}$ since only $\hat{S}_{n,l}^z$ and $\hat{S}_{n,r}^z$ are involved. In terms of this basis, we have

$$\hat{S}_{n,l}^z = \frac{1}{2} (\hat{l}_n^\dagger \hat{l}_n - \hat{r}_n^\dagger \hat{r}_n + \hat{u}_n^\dagger \hat{u}_n + \hat{d}_n^\dagger \hat{d}_n) = \frac{1}{2} (\hat{L}_n - \hat{R}_n + \hat{U}_n - \hat{D}_n), \quad (6.10)$$

$$\hat{S}_{n,r}^z = \frac{1}{2} (-\hat{l}_n^\dagger \hat{l}_n + \hat{r}_n^\dagger \hat{r}_n + \hat{u}_n^\dagger \hat{u}_n + \hat{d}_n^\dagger \hat{d}_n) = \frac{1}{2} (-\hat{L}_n + \hat{R}_n + \hat{U}_n - \hat{D}_n) \quad (6.11)$$

where $\hat{L}_n = \hat{l}_n^\dagger \hat{l}_n$ is the number operator and similarly for the rest. After some algebra,

the Hamiltonian in the Ising limit can be derived as:

$$\begin{aligned}
 \hat{H}_{Ising} = & \sum_{i,n} \left(\frac{1}{8} J_2 - \frac{1}{2} h \right) \hat{U}_{n,i} + \left(\frac{1}{8} J_2 + \frac{1}{2} h \right) \hat{D}_{n,i} - \frac{1}{8} J_2 (\hat{L}_{n,i} + \hat{R}_{n,i}) \\
 & + \frac{1}{2} J_1 \sum_{i,n} (\hat{U}_{n,i} - \hat{D}_{n,i}) (\hat{U}_{n+1,i} + \hat{L}_{n+1,i} - \hat{D}_{n+1,i} - \hat{R}_{n+1,i}) \\
 & + \frac{1}{4} (2J_4 + J_3) \sum_{\langle i,j \rangle} (\hat{U}_{1,i} - \hat{D}_{1,i}) (\hat{U}_{1,j} - \hat{D}_{1,j}) + (\hat{U}_{2,i} - \hat{D}_{2,i}) (\hat{U}_{2,j} - \hat{D}_{2,j}) \\
 & + \frac{1}{4} (2J_4 - J_3) \sum_{\langle i,j \rangle} (\hat{L}_{1,i} - \hat{R}_{1,i}) (\hat{L}_{1,j} - \hat{R}_{1,j}) + (\hat{L}_{2,i} - \hat{R}_{2,i}) (\hat{L}_{2,j} - \hat{R}_{2,j}) \\
 & + \frac{1}{4} J_3 \sum_{\langle i,j \rangle} (\hat{L}_{1,i} - \hat{R}_{1,i}) (\hat{U}_{1,j} - \hat{D}_{1,j}) - (\hat{U}_{1,i} - \hat{D}_{1,i}) (\hat{L}_{1,j} - \hat{R}_{1,j}) \\
 & + \frac{1}{4} J_3 \sum_{\langle i,j \rangle} \eta_{ij} (\hat{U}_{2,i} - \hat{D}_{2,i}) (\hat{L}_{2,j} - \hat{R}_{2,j}) - \eta_{ij} (\hat{L}_{2,i} - \hat{R}_{2,i}) (\hat{U}_{2,j} - \hat{D}_{2,j}) \quad (6.12)
 \end{aligned}$$

where $\eta_{ij} = 1$ if the bond is along $\mathbf{x}$ direction and -1 along $\mathbf{y}$ direction. We take the convention that the vector $\mathbf{r}_j - \mathbf{r}_i$ always points to the positive directions.

It has been well known that the small magnetization plateaus contain very large unit cell. In this s-plateau representation, there is a very simple relation between the magnetization plateau and the minimum size of the unit cell. In a q/p plateau, suppose the unit cell contains N s-plaquettes. Each plaquette has maximum magnetization 4 and takes values from -4 to 4 . Hence we have

$$\frac{q}{p} = \frac{1}{4N} \sum_i S_i^z \quad (6.13)$$

where S_i^z is the magnetization of each s-plaquette. Since the summation on the right hand side is always an integer, we should have that $4N/p$ be an integer as well. Hence N should be integers of the form $pZ/4$ where Z is any integer. For instance, in the $1/2$ plateau ($p = 2$ and $q = 1$), $N = Z/2$, that is $N = 1, 2, 3, \dots$. The minimum unit cell contains only one s-plaquette. In the $1/3$ plateau ($p = 3$ and $q = 1$), $N = 3Z/4 = 3, 6, 9, \dots$. The minimum unit cell contains 3 s-plaquettes. This relation is important in numerical study as well. The system size should be set properly to observe stable plateaus. For example, to observe a $1/3$ plateau, the system should contain at least 3 s-plaquettes.

Properties of the s-plaquette

The choice of the unit cell and some other information about the configuration of the ground state could be inferred from Ising Hamiltonian (6.12). In this thesis, only AFM J_1 and J_2 are considered. (J_1, J_2, h) give the self-energy of each s-plaquette and J_2 prefers $|l\rangle, |r\rangle$ to $|u\rangle, |d\rangle$, as expected. J_1 determines the internal structure of each s-plaquette. AFM J_1 will prefer the neighbouring dimers having different states. We notice that two adjacent $|l\rangle, |r\rangle$ dimers effectively do not interact through J_1 which goes against J_2 , resulting a competition between J_2 and J_1 . This is actually the competition between the dimer state and the Néel state with critical point at $J_2 = 2J_1$. The unit cells of both of these two states contain only one s-plaquette, which will be shown in next section. In the existence of field, the number of s-plaquettes in the unit cell could increase because the constraint (6.4) always flips two dimers in two adjacent s-plaquettes in opposite way. This is indeed the case as we shall see in the $1/3$ plateau.

For fixed J_1 and J_2 , the configurations of the neighbouring s-plaquettes are further determined by J_3 and J_4 . In fact, the number of s-plaquettes in one unit cell is primarily determined by large $|J_3|$ and/or $|J_4|$. Only when J_3 and J_4 are relatively close to zero will it be determined by (J_1, J_2) or all of the four parameters.

The $\frac{1}{4}(2J_4 \pm J_3)$ terms are effective nearest neighbour interactions in a bipartite lattice. Strong (relative to J_1 and J_2) FM J_4 prefers a single s-plaquette unit cell. With FM J_3 , s-plaquette formed by $|u\rangle, |d\rangle$ has lowest energy while for AFM J_3 , $|l\rangle, |r\rangle$ will have lower energy. This single s-plaquette preference actually extends to AFM J_4 as well, provided that J_3 is strong AFM. In this range, it is an effective FM model. On the other hand, in the effective AFM range, the ground state prefers two s-plaquette unit cell of pattern $\begin{smallmatrix} A & B \\ B & A \end{smallmatrix}$. When it is close to the boundary between effective AFM and FM, the interplay among all the four parameters complicates the situation. The system may have two s-plaquettes with pattern $\begin{smallmatrix} A & B \\ A & B \end{smallmatrix}$ or other numbers of s-plaquettes. However, unit cells with more than two s-plaquettes will always have higher energy contributed from (J_3, J_4) and not be preferred by $\frac{1}{4}(2J_4 \pm J_3)$. Hence in this range, it is most likely to be the competition between the (J_1, J_2) pair and (J_3, J_4) pair where we would expect some phase boundary

in the (J_3, J_4) phase diagram purely determined by (J_1, J_2) if any.

Besides (J_1, J_2) , the last two terms in the equation also prefer large unit cells because of the directional dependence. It is not difficult to see that they are non-zero only for unit cells containing at least 3 s-plaquettes. This preference in large unit cell competes with $\frac{1}{4}(2J_4 \pm J_3)$, or more precisely, a competition between AFM J_4 and AFM J_3 , because this directional interaction alone has no chance to lower the energy by increasing the size of the unit cell in the FM J_4 range or FM J_3 range.

To summarise, in the strong FM J_4 range, the ground state prefers unit cells with single s-plaquette. In the extreme frustrated region, strong AFM J_4 and FM J_3 range, it prefers unit cells with two s-plaquettes. In the rest of the parameter space, large unit cell competes with unit cells with single s-plaquette and two s-plaquettes. As a direct consequence, there is more frustration in optimizing the configuration with larger unit cell than the minimum size required, therefore we will always consider unit cell with as small size as possible. With all this in mind, we explore a few plateaus in the following.

6.1.2 Phase diagram in zero field

In the absence of magnetic field, the total magnetization is zero. From Equation 6.13, we can see that the unit cell could contain any number of s-plaquettes. We can divide them into different classes according to the number of the s-plaquettes.

Single s-plaquette unit cell

The smallest unit cell contains only one s-plaquette and we denote the state as $|a_1, a_2, a_3, a_4\rangle$. The constraint (6.4) implies that $|\bar{a}_3\rangle = |a_1\rangle$ and $|\bar{a}_4\rangle = |a_2\rangle$. Because the magnetization is zero, the total spin moment should be zero, that is to say $S^z(a_1) + S^z(a_2) + S^z(a_3) + S^z(a_4) = 0$. Obeying these conditions, there are only 6 such states:

$$|l, l, r, r\rangle, \quad |r, r, l, l\rangle, \quad |l, r, r, l\rangle, \quad |r, l, l, r\rangle, \quad |u, d, u, d\rangle \quad \text{and} \quad |d, u, d, u\rangle. \quad (6.14)$$

We soon realize that some of the above states have the same energy because they are related by a symmetry transformation: 90 degree rotation about the center of the central square. The extended SSM has this 4-fold rotational symmetry. If the unit cell consists of only one s-plaquette, under this transformation, the following states are degenerate:

$$E(|a_1, a_2, a_3, a_4\rangle) = E(|a_2, a_3, a_4, a_1\rangle) = E(|a_3, a_4, a_1, a_2\rangle) = E(|a_4, a_1, a_2, a_3\rangle). \quad (6.15)$$

Thus, there are only two independent configurations in this case: $|l, l, r, r\rangle$ and $|u, d, u, d\rangle$ and their energies are obtained from Equation 6.12:

$$|\psi_1^0\rangle = |l, l, r, r\rangle \quad \text{with} \quad E_1 = -\frac{1}{2}J_2 - J_3 + 2J_4, \quad (6.16)$$

$$|\psi_2^0\rangle = |u, d, u, d\rangle \quad \text{with} \quad E_2 = -2J_1 + \frac{1}{2}J_2 + J_3 + 2J_4. \quad (6.17)$$

Next we consider the two s-plaquettes case. They can be further classified into three groups. First group has the structure $\begin{smallmatrix} S_1 & S_2 \\ S_1 & S_2 \end{smallmatrix}$ while the second group has the structure $\begin{smallmatrix} S_1 & S_2 \\ S_2 & S_1 \end{smallmatrix}$, where S_1 and S_2 mean the two s-plaquettes. The third group $\begin{smallmatrix} S_2 & S_2 \\ S_1 & S_1 \end{smallmatrix}$ is actually connected to the first group by a 90 degree rotation. Hence, the third and the first group are degenerate and we will only consider the first and the second groups.

Two s-plaquettes – unit cell-1

We consider the $\begin{smallmatrix} S_1 & S_2 \\ S_1 & S_2 \end{smallmatrix}$ group first. The two s-plaquette states can be denoted as $|S_1\rangle = |a_1, a_2, a_3, a_4\rangle$ and $|S_2\rangle = |b_1, b_2, b_3, b_4\rangle$. According to the constraint (6.4), we have

$$|b_4\rangle = |\bar{a}_1\rangle, \quad |a_4\rangle = |\bar{a}_2\rangle, \quad |b_4\rangle = |\bar{b}_2\rangle \quad \text{and} \quad |b_1\rangle = |\bar{a}_3\rangle. \quad (6.18)$$

The total magnetic moment is zero, so we have

$$\sum_i S^z(a_i) + \sum_i S^z(b_i) = 0 \Rightarrow S^z(a_1) + S^z(a_2) + S^z(a_3) + S^z(b_2) = 0. \quad (6.19)$$

The last equality is true because $S^z(\bar{a}_i) = S^z(a_i)$ for any $|a_i\rangle$. Therefore, there are only 3 independent variables: (a_1, a_2, a_3, b_2) with the constraint above. Hence we can denote the two s-plaquette states by (a_1, a_2, a_3, b_2) , that is,

$$(a_1, a_2, a_3, b_2)_1 \doteq |a_1, a_2, a_3, \bar{a}_1\rangle_i \otimes |\bar{a}_3, b_2, \bar{a}_1, \bar{b}_2\rangle_{i+x}. \quad (6.20)$$

As in the single s-plaquette case, some of the states are actually degenerate because of the symmetry transformation. The translational transformation maps the state $|S_1\rangle_i |S_2\rangle_{i+x}$ to $|S_2\rangle_i |S_1\rangle_{i+x}$. Explicitly,

$$|a_1, a_2, a_3, \bar{a}_1\rangle_i \otimes |\bar{a}_3, b_2, \bar{a}_1, \bar{b}_2\rangle_{i+x} \Rightarrow |\bar{a}_3, b_2, \bar{a}_1, \bar{b}_2\rangle_i \otimes |a_1, a_2, a_3, \bar{a}_1\rangle_{i+x}, \quad (6.21)$$

or

$$(a_1, a_2, a_3, b_2)_1 \Rightarrow (\bar{a}_3, b_2, \bar{a}_1, a_2)_1. \quad (6.22)$$

The 90 degree rotation maps the state into some state in the third group as explained above. Under 180 degree rotation about the common dimer between the two s-plaquettes, the state is transformed as

$$|a_1, a_2, a_3, \bar{a}_1\rangle_i \otimes |\bar{a}_3, b_2, \bar{a}_1, \bar{b}_2\rangle_{i+x} \Rightarrow |\bar{a}_1, \bar{b}_2, \bar{a}_3, b_2\rangle_i \otimes |a_3, \bar{a}_2, a_1, a_2\rangle_{i+x}, \quad (6.23)$$

or

$$(a_1, a_2, a_3, b_2)_1 \Rightarrow (\bar{a}_1, \bar{b}_2, \bar{a}_3, \bar{a}_2)_1. \quad (6.24)$$

As a result of the above symmetry properties, the below four states are degenerate:

$$E(a_1, a_2, a_3, b_2) = E(\bar{a}_1, \bar{b}_2, \bar{a}_3, \bar{a}_2) = E(\bar{a}_3, b_2, \bar{a}_1, a_2) = E(a_3, \bar{a}_2, a_1, \bar{b}_2). \quad (6.25)$$

We list in Table 6.1 all the states with their energies that are not connected by any of the above symmetry transformations.

Configuration of the state	Energy
$(u, u, d, d)_1$	$\frac{1}{2}J_2$
$(u, d, l, l)_1, (d, u, r, r)_1$	$-J_1 + J_4$
$(u, d, l, r)_1, (u, d, r, l)_1, (d, u, l, r)_1, (d, u, r, l)_1$	$-\frac{1}{2}J_1 + J_4$
$(u, d, l, l)_1, (d, u, r, r)_1$	J_4
$(l, u, l, d)_1, (r, d, r, u)_1$	J_1
$(l, d, l, u)_1, (r, u, r, d)_1$	$-J_1$
$(l, u, r, d)_1, (r, u, l, d)_1, (l, d, r, u)_1, (r, d, l, u)_1$	$-\frac{1}{2}J_3 + J_4$
$(l, l, l, l)_1, (l, l, r, r)_1$	$-\frac{1}{2}J_2 - \frac{1}{2}J_3 + J_4$
$(l, l, l, r)_1$	$-\frac{1}{2}J_2$

Table 6.1: The 20 configurations with their energies.

Two s-plaquettes – unit cell-2

Now we consider the $\frac{S_1}{S_2} \frac{S_2}{S_1}$ case. The state is again denoted by $|S_1\rangle = |a_1, a_2, a_3, a_4\rangle$ and $|S_2\rangle = |b_1, b_2, b_3, b_4\rangle$ and the constraint gives:

$$|b_1\rangle = |\bar{a}_3\rangle, \quad |b_2\rangle = |\bar{a}_4\rangle, \quad |b_3\rangle = |\bar{a}_1\rangle \quad \text{and} \quad |b_4\rangle = |\bar{a}_2\rangle. \quad (6.26)$$

Hence the configurate is completely determined by the first s-plaquette $(a_1, a_2, a_3, a_4)_2$.

Similarly, we also have that the total magnetic moment is zero: $\sum_i S^z(a_i) = 0$.

As in the first group, some of the states here are connected by the translational and rotational transformations. The translational transformation both in $\mathbf{x}$ and $\mathbf{y}$ directions connects the two states:

$$(a_1, a_2, a_3, a_4)_2 \Rightarrow (\bar{a}_3, \bar{a}_4, \bar{a}_1, \bar{a}_2)_2. \quad (6.27)$$

The 90 degree rotations connect the following four states:

$$(a_1, a_2, a_3, a_4)_2 \Rightarrow (a_2, a_3, a_4, a_1)_2 \Rightarrow (a_3, a_4, a_1, a_2)_2 \Rightarrow (a_4, a_1, a_2, a_3)_2. \quad (6.28)$$

There are in all 10 configurations that are not related by any of the symmetry transformations which are listed in Table 6.2.

Configuration of the state	Energy
$(u, u, d, d)_2$	$\frac{1}{2}J_2 - J_3 - 2J_4$
$(u, d, l, l)_2, (d, u, l, l)_2, (u, d, l, r)_2, (d, u, l, r)_2$	$-\frac{1}{2}J_1$
$(u, l, d, l)_2$	$-2J_4$
$(u, l, d, r)_2$	$-J_3$
$(l, l, l, l)_2, (l, r, l, r)_2$	$-\frac{1}{2}J_2 + J_3 - 2J_4$
$(l, l, l, r)_2$	$-\frac{1}{2}J_2$

Table 6.2: The 10 configurations with their energies.

Phase diagram

Comparing all of the energies obtained above, we are able to draw a phase diagram in the (J_3, J_4) plane. It turns out that there are only five possible configurations. These five energy values and their phase ranges are shown in Table 6.3.

$E_1 = -\frac{1}{2}J_2 - J_3 + 2J_4$ $J_3 \geq J_1 - \frac{1}{2}J_2, \quad J_4 \leq \min \left\{ \frac{1}{4}J_2, \frac{1}{2}J_3, \frac{1}{2}J_3 + \frac{1}{4}(J_2 - 2J_1) \right\}$
$E_2 = -2J_1 + \frac{1}{2}J_2 + J_3 + 2J_4$ $J_3 \leq J_1 - \frac{1}{2}J_2, \quad J_4 \leq \min \left\{ \frac{1}{4}(2J_1 - J_2), -\frac{1}{2}J_3 + \frac{1}{4}(2J_1 - J_2) \right\}$
$E_3 = \frac{1}{2}J_2 - (J_3 + 2J_4)$ $J_3 \geq \frac{1}{2}J_2, \quad J_4 \geq \max \left\{ \frac{1}{4}J_2, -\frac{1}{2}J_3 + \frac{1}{4}(2J_1 + J_2) \right\}$
$E_4 = -\frac{1}{2}J_2 + J_3 - 2J_4$ $J_3 \leq \frac{1}{2}J_2, \quad J_4 \geq \max \left\{ \frac{1}{4}(2J_1 - J_2), \frac{1}{2}J_3, \frac{1}{2}J_3 + \frac{1}{4}(2J_1 - J_2) \right\}$
$E_5 = -J_1$ $J_2 \leq 2J_1, 0 \leq J_3 \leq \min \{J_1, J_2\},$ $\frac{1}{2}J_2 - J_1 \leq 2J_4 - J_3 \leq J_1 - \frac{1}{2}J_2$ and $J_1 - \frac{1}{2}J_2 \leq 2J_4 + J_3 \leq J_1 + \frac{1}{2}J_2$

Table 6.3: The energies of the five phases and their corresponding phase range.

The fifth phase E_5 only exists for $J_2 \leq 2J_1$. The phase diagrams for different J_1 and J_2 values are shown in Figure 6.2a-6.2c. The points and the lines in the figures are given

by:

$$\begin{aligned}
 A_1 &= \left(J_1 - \frac{1}{2}J_2, 0 \right), \quad A_2 = \left(J_1, \frac{1}{4}J_2 \right), \quad A_3 = \left(\frac{1}{2}J_2, \frac{1}{2}J_1 \right), \quad A_4 = \left(0, \frac{1}{2}J_1 - \frac{1}{4}J_2 \right), \\
 B_1 &= \left(\frac{1}{2}J_2, \frac{1}{4}J_2 \right), \quad B_2 = \left(J_1 - \frac{1}{2}J_2, \frac{1}{2}J_1 - \frac{1}{4}J_2 \right), \\
 L_1 : J_4 &= \frac{1}{2}J_3 + \frac{1}{4}(J_2 - 2J_1) \quad \text{where} \quad J_1 - \frac{1}{2}J_2 \leq J_3 \leq J_1, \\
 L_2 : J_4 &= -\frac{1}{2}J_3 + \frac{1}{4}(2J_1 + J_2) \quad \text{where} \quad \frac{1}{2}J_2 \leq J_4 \leq J_1, \\
 L_3 : J_4 &= \frac{1}{2}J_3 + \frac{1}{4}(2J_1 - J_2) \quad \text{where} \quad 0 \leq J_3 \leq \frac{1}{2}J_2, \\
 L_4 : J_4 &= -\frac{1}{2}J_3 + \frac{1}{4}(2J_1 - J_2) \quad \text{where} \quad 0 \leq J_3 \leq J_1 - \frac{1}{2}J_2, \\
 L_5 : J_4 &= \frac{1}{2}J_3 \quad \text{where} \quad J_1 - \frac{1}{2}J_2 \leq J_3 \leq \frac{1}{2}J_2
 \end{aligned} \tag{6.29}$$

The representative configurations of each phase are:

$E_1 : l, l, r, r\rangle,$	single s-plaquette
$E_2 : u, d, u, d\rangle,$	single s-plaquette
$E_3 : u, u, d, d\rangle_i \otimes d, d, u, u\rangle_{i+x},$	$\begin{smallmatrix} S_1 & S_2 \\ S_2 & S_1 \end{smallmatrix}$ pattern
$E_4 : l, l, l, l\rangle_i \otimes r, r, r, r\rangle_{i+x}, l, r, l, r\rangle_i \otimes r, l, r, l\rangle_{i+x},$	$\begin{smallmatrix} S_1 & S_2 \\ S_2 & S_1 \end{smallmatrix}$ pattern
$E_5 : l, d, l, d\rangle_i \otimes r, u, r, u\rangle_{i+x}, r, u, r, u\rangle_i \otimes l, d, l, d\rangle_{i+x},$	$\begin{smallmatrix} S_1 & S_2 \\ S_1 & S_2 \end{smallmatrix}$ pattern

The two configurations in E_4 and E_5 are actually connected by reflection about the J_2 bond, respectively. The explicit diagrams are shown in Figure 6.3.

The E_1 phase is in the strong AFM J_3 and FM J_4 or weak AFM J_4 range and the configuration is actually the columnar state. In the E_2 phase, both J_3 and J_4 are at most weak AFM where J_1 dominates, hence, it is actually the Néel AFM state. As explained previously, FM J_4 prefers identical adjacent s-plaquettes, that is single s-plaquette unit cell and this is exactly the case in phase E_1 and E_2 . It is also clear that the choice of the types of the dimer depends on J_3 . The E_3 phase is in the strong AFM J_3 and J_4 range where the J_2 columnar state will be stabilized. As shown in Fig.6.3c, the J_2 columnar state is formed by alternative diagonal stripes of J_2 dimers. This is an effective AFM

 (a) The phase diagram for $J_1 \geq J_2$.

 (b) The phase diagram for $\frac{1}{2}J_2 \leq J_1 \leq J_2$.

 (c) The phase diagram for $J_1 \leq \frac{1}{2}J_2$.

 Figure 6.2: Phase diagram at various values of J_1 and J_2 .

model, as explained before, thus the unit cell contains two s-plaquettes. The E_4 phase is in the range of AFM J_4 and at most weak AFM J_3 which also prefers two s-plaquette unit cell yet with l, r dimers. Thus, we have the J_3 plaquette chess board state, as shown in Fig.6.3d. The E_5 phase only exists when $J_1 > \frac{1}{2}J_2$. Though it has AFM J_3 and J_4 , they are too weak to form E_1 , E_3 and E_4 and too strong to form E_2 . It is also near the boundary between effective AFM and FM, which results in $\frac{S_1}{S_1} \frac{S_2}{S_2}$ pattern. Hence, all the J_3 and J_4 interactions are cancelled out. The E_5 phase is more like an intermediate phase between the other four phases. The area of this phase is shrunk when J_1 approaches $J_2/2$ and eventually disappears when $J_1 < J_2/2$.

Figure 6.3: The blue points represent spin up while the white represent spin down.

6.1.3 $1/2$ plateaus

We explore the possible $1/2$ plateaus in this subsection. From Equation 6.13, it is straightforward to see that the unit cell could contain any integer number of s-plaquettes and the total magnetic moment in the unit cell is $2N$ where N is the number of s-

plaquettes.

Single s-plaquette

In the single s-plaquette case, the state is again denoted as $|a_1, a_2, a_3, a_4\rangle$ with constraint $|a_3\rangle = |\bar{a}_1\rangle$ and $|a_4\rangle = |\bar{a}_2\rangle$. The symmetry, Eq.6.15, holds as well. Hence, there is only one independent state with energy:

$$\left| \psi_1^{1/2} \right\rangle = |u, l, u, r\rangle \quad \text{with} \quad E = 2J_4 - h. \quad (6.30)$$

Two s-plaquette unit cell-1

Now we consider the two s-plaquette unit cell with pattern: $\begin{smallmatrix} S_1 & S_2 \\ S_1 & S_2 \end{smallmatrix}$. The state is again denoted as:

$$(a_1, a_2, a_3, b_2)_1 \doteq |a_1, a_2, a_3, \bar{a}_1\rangle_i \otimes |\bar{a}_3, b_2, \bar{a}_1, \bar{b}_2\rangle_{i+x}. \quad (6.31)$$

Everything is the same as in the zero plateau case including the symmetry in Equation 6.25 except that the total magnetic moment is 2 instead of 0.

$$\sum_i S^z(a_i) + \sum_i S^z(b_i) = 2 \Rightarrow S^z(a_1) + S^z(a_2) + S^z(a_3) + S^z(b_2) = 2. \quad (6.32)$$

There are only 8 independent states that are not connected by the translational and rotational symmetry transformation and are listed in Table 6.4

Configuration of the state	Energy
$(u, u, u, d)_1, (u, u, d, u)_1$	$\frac{1}{2}J_2 + \frac{1}{2}J_3 + J_4 - h$
$(u, u, l, l)_1, (u, u, r, r)_1$	$\frac{1}{2}J_1 + J_4 - h$
$(u, l, u, r)_1, (l, u, l, u)_1$	$\frac{1}{2}J_3 + J_4 - h$
$(l, l, u, u)_1$	$J_4 - h$
$(r, r, u, u)_1$	$J_1 + J_4 - h$

Table 6.4: The 8 configurations with their energies.

Two s-plaquette unit cell-2

The $\begin{smallmatrix} S_1 & S_2 \\ S_2 & S_1 \end{smallmatrix}$ pattern is similar to that in the zero plateau case and the state is denoted by the first s-plaquette state $(a_1, a_2, a_3, a_4)_2$ as well. The symmetry properties, Eq.6.27

and Eq.6.28, are satisfied. It is quite straightforward to show that there are only 4 independent states in this pattern as shown in Table 6.5.

Configuration of the state	Energy
$(u, u, u, d)_2$	$\frac{1}{2}J_2 - h$
$(u, u, l, l)_2, (u, u, l, r)_2$	$\frac{1}{2}J_1 - h$
$(u, l, u, l)_2$	$J_3 - h$

Table 6.5: The 4 configurations with their energies.

Phase diagram

Energy	Phase range
$E_1 = 2J_4 - h$	$J_4 \leq \min\{\frac{1}{2}J_3, 0\}$
$E_2 = J_4 - h$	$J_3 \geq 0, \quad 0 \leq J_4 \leq \min\{J_3, \frac{1}{2}J_1, \frac{1}{2}J_2\}$
$E_3 = J_3 - h$	$J_3 \leq \min\{\frac{1}{2}J_1, \frac{1}{2}J_2\}, \quad J_4 \geq \max\{\frac{1}{2}J_3, J_3\}$
$E_4 = \frac{1}{2}J_1 - h$	$J_1 \leq J_2, \quad J_3 \geq \frac{1}{2}J_1, \quad J_4 \geq \frac{1}{2}J_1$
$E_5 = \frac{1}{2}J_2 - h$	$J_1 \geq J_2, \quad J_3 \geq \frac{1}{2}J_2, \quad J_4 \geq \frac{1}{2}J_2$

Table 6.6: The energies of the 5 phases and their corresponding phase range.

Similar to the zero plateau situation, comparison of the energies of different states will give the phase diagram in the (J_3, J_4) plane. After some algebra, we are able to show that there are only 5 phases which listed with the phase range in Table6.6. The phase diagram is shown in Figure 6.4. E_4 and E_5 phases share the same range but they do not exist simultaneously, as can be seen from the table. When $J_1 < J_2$, E_4 has lower energy while E_5 has lower energy in the other way round.

The points and the lines in the phase diagram are defined as:

$$\begin{aligned}
 A_0 &= (0, 0), \quad A_1 = \left(\frac{1}{2} \min\{J_1, J_2\}, \frac{1}{2} \min\{J_1, J_2\} \right), \\
 L_1 : J_4 &= J_3, \quad L_2 : J_4 = \frac{1}{2}J_3.
 \end{aligned} \tag{6.33}$$

Table 6.7 lists the representative configurations of the 5 half plateaus and the explicit lattice configurations are shown in Figure 6.5.

E_1 is in the FM J_4 range, similar to the columnar phase in zero plateau. This could actually be seen from Fig.6.5a. The down spins are flipped in an alternating order along

Figure 6.4: The phase diagram of the 1/2 plateau

E_1 :	$ u, l, u, r\rangle_i$	Single s-plaquette
E_2 :	$ l, l, l, r\rangle_i \otimes r, u, r, u\rangle_{i+x}$	$\begin{smallmatrix} s_1 & s_2 \\ s_1 & s_2 \end{smallmatrix}$ pattern
E_3 :	$ u, l, u, l\rangle_i \otimes u, r, u, r\rangle_{i+x}$	$\begin{smallmatrix} s_1 & s_2 \\ s_2 & s_1 \end{smallmatrix}$ pattern
E_4 :	$ u, u, l, l\rangle_i \otimes r, r, u, u\rangle_{i+x}$	$\begin{smallmatrix} s_1 & s_2 \\ s_2 & s_1 \end{smallmatrix}$ pattern
E_5 :	$ u, u, u, d\rangle_i \otimes u, d, u, u\rangle_{i+x}$	$\begin{smallmatrix} s_1 & s_2 \\ s_2 & s_1 \end{smallmatrix}$ pattern

Table 6.7: The representative configurations of 1/2 plateaus

the column while the up spin do not change. The J_4 term remains the same. The E_2 phase is in the AFM J_3 and J_4 range which could also be considered as an excitation from the columnar state as shown in Fig.6.5b. The E_3 phase is in the at most weak AFM J_3 range which could be considered as excited from the J_3 plaquette chess board phase in zero plateau. Two diagonal down spins in the plaquette are flipped keeping the J_3 unchanged as shown in Fig.6.5c. Both E_4 and E_5 phases are in the strong AFM J_3 and J_4 range which can be considered as excited from the J_2 dimer columnar state in two different ways. In E_4 , J_2 is larger than J_1 , the dimer prefers $|l\rangle$ and $|r\rangle$, therefore, the spin down dimers are flipped to $|\uparrow\downarrow\rangle$. In E_5 , $J_1 > J_2$, the down spin dimers are flipped to up spin dimers in an alternating manner keeping the J_2 energy unchanged.

Figure 6.5: The blue points represent spin up while the white represent spin down.

So far, we only compare the energy in the same plateau. We will see that when other plateaus are taken into account, some of the half plateaus no long have the lowest energy. We will come back to this and update the phase diagram later.

6.1.4 1/3 plateau

1/3 plateau appears in the original Ising SSM without the extended interactions. We investigated how the long range interaction will affect this plateau in this subsection. From Equation 6.13, we obtain the number of s-plaquettes in a unit cell should be at least 3. The total magnetic moment in the 3 s-plaquettes should be 4, that is

$$\sum_{i=1}^3 \sum_{j=1}^4 S^z(a_{ij}) = 4 \quad (6.34)$$

where $|a_{ij}\rangle$ is the j^{th} dimer in the i^{th} s-plaquette. We consider the following patterns:

$$\begin{array}{ccc} & S_1 & S_2 & S_3 \\ \text{pattern 1: } & S_1 & S_2 & S_3 \quad \text{and} \quad \text{pattern 2: } & S_2 & S_3 & S_1 \\ & & & & S_3 & S_1 & S_2 \end{array}$$

where S_i represents one s-plaquette.

Pattern 1

We consider the stripe pattern first. The states in the unit cell is are denoted as

$$|a_{11}, a_{12}, a_{13}, a_{14}\rangle_i \otimes |a_{21}, a_{22}, a_{23}, a_{24}\rangle_{i+x} \otimes |a_{31}, a_{32}, a_{33}, a_{34}\rangle_{i+2x}. \quad (6.35)$$

The constraint (6.4) and the periodicity result in the following identities:

$$|a_{21}\rangle = |\bar{a}_{13}\rangle, |a_{31}\rangle = |\bar{a}_{23}\rangle, |a_{14}\rangle = |\bar{a}_{12}\rangle, |a_{24}\rangle = |\bar{a}_{22}\rangle, |a_{34}\rangle = |\bar{a}_{32}\rangle, |a_{33}\rangle = |\bar{a}_{11}\rangle. \quad (6.36)$$

Hence, there are only 6 independent variables, we define the following representation:

$$(a_1, a_2, a_3, a_4, a_5, a_6)_1 = |a_1, a_2, a_3, \bar{a}_2\rangle_i \otimes |\bar{a}_3, a_4, a_5, \bar{a}_4\rangle_{i+x} \otimes |\bar{a}_5, a_6, \bar{a}_1, \bar{a}_6\rangle_{i+2x}. \quad (6.37)$$

The constraint on the total magnetic moment requires that

$$\sum_{i=1}^6 S^z(a_i) = 2. \quad (6.38)$$

Besides, the translational operation connects the following three states:

$$(a_1, a_2, a_3, a_4, a_5, a_6)_1 \Rightarrow (\bar{a}_3, a_4, a_5, a_6, \bar{a}_1, a_2)_1 \Rightarrow (\bar{a}_5, a_6, \bar{a}_1, a_2, a_3, a_4)_1. \quad (6.39)$$

This symmetry property helps simplifying the calculation a lot. We can divide the state in the following manner:

$$(a_1, a_2, a_3, a_4, a_5, a_6)_1 \rightarrow [S^z(a_1) + S^z(a_2), S^z(a_3) + S^z(a_4), S^z(a_5) + S^z(a_6)]. \quad (6.40)$$

Each pair could only have magnetic moment from -2 to 2 , thus we only have to consider the following 5 groups:

$$[2, 2, -2], \quad [2, 1, -1], \quad [2, -1, 1], \quad [2, 0, 0] \quad \text{and} \quad [1, 1, 0]. \quad (6.41)$$

Besides, the 180 degree rotation about the center of the s-plaquette has the effect:

$$(a_1, a_2, a_3, a_4, a_5, a_6)_1 \Rightarrow (\bar{a}_1, \bar{a}_6, \bar{a}_5, \bar{a}_4, \bar{a}_3, \bar{a}_2)_1. \quad (6.42)$$

Together with the translational symmetry, there could be as many as 6 states that are topologically equivalent. This simplifies the calculation as well.

[2,2,-2] The first group is the simplest with only one element $(u, u, u, u, d, d)_1$. The energy is given:

$$E = \frac{1}{2}J_2 - \frac{2}{3}h + \frac{2}{3}J_1 + \frac{1}{3}J_3 + \frac{2}{3}J_4. \quad (6.43)$$

[2,1,-1] and [2,-1,1] There in all 32 possible configurations in these two groups. Each configuration has an identical term in the energy: $\frac{1}{2}J_2 - \frac{2}{3}h$. The configurations can be further classify into 8 sets where only 2 of them are independent. Let b_i be states l or

r . We can show that only $(u, u, u, b_1, d, b_1)_1$ and $(u, u, b_1, d, b_2, u)_1$ are not connected to other states by symmetry transformations.

$$(u, u, u, b_1, b_2, d)_1 \rightarrow (u, u, u, d, \bar{b}_2, \bar{b}_1)_1 \in [2, 0, 0]$$

$$(u, u, d, b_1, b_2, u)_1 \rightarrow (u, u, \bar{b}_2, \bar{b}_1, d, u)_1 \in [2, 0, 0]$$

$$(u, u, d, b_1, u, b_2)_1 \rightarrow (u, \bar{b}_2, u, \bar{b}_1, d, u)_1 \in [1, 1, 0]$$

$$(u, u, b_1, u, d, b_2)_1 \rightarrow (b_1, u, u, \bar{b}_2, d, u)_1 \in [1, 1, 0]$$

$$(u, u, b_1, d, u, b_2)_1 \rightarrow (b_1, u, u, \bar{b}_2, u, d)_1 \in [1, 1, 0]$$

$$(u, u, b_1, u, b_2, d)_1 \rightarrow (b_2, u, \bar{b}_1, u, u, d)_1 \in [1, 1, 0]$$

For convenience, we still give the full configurations in Table 6.8 and those connected configurations in $[2, 0, 0]$ and $[1, 1, 0]$ will not be listed.

Energy: $\frac{1}{6}J_2 - \frac{2}{3}h +$	Configurations
$\frac{2}{3}J_4$	$(u, u, u, r, d, l)_1, (u, u, r, d, l, u)_1$
J_4	$(u, u, u, r, r, d)_1, (u, u, d, l, l, u)_1$
$\frac{1}{3}J_1 + J_4$	$(u, u, u, r, l, d)_1, (u, u, l, u, l, d)_1, (u, u, r, u, d, r)_1, (u, u, l, d, u, l)_1,$ $(u, u, d, r, u, r)_1, (u, u, d, r, l, u)_1$
$\frac{2}{3}J_1 + J_4$	$(u, u, u, l, d, l)_1, (u, u, u, r, d, r)_1, (u, u, l, d, l, u)_1, (u, u, r, d, r, u)_1$
$-\frac{1}{3}J_1 + J_4$	$(u, u, r, u, d, l)_1, (u, u, r, u, r, d)_1, (u, u, d, l, u, l)_1, (u, u, r, d, u, l)_1$
$\frac{1}{3}J_1 + \frac{2}{3}J_4$	$(u, u, r, u, l, d)_1, (u, u, d, r, u, l)_1$
$\frac{4}{3}J_1 + \frac{1}{3}J_3 + \frac{2}{3}J_4$	$(u, u, u, l, d, r)_1, (u, u, l, d, r, u)_1$
$\frac{2}{3}J_1 + \frac{1}{3}J_3 + J_4$	$(u, u, u, l, l, d)_1, (u, u, d, r, r, u)_1$
$\frac{1}{3}J_1 + \frac{1}{3}J_3 + J_4$	$(u, u, u, l, r, d)_1, (u, u, l, u, d, r)_1, (u, u, l, d, u, r)_1, (u, u, d, l, r, u)_1$
$-\frac{1}{3}J_1 + \frac{1}{3}J_3 + J_4$	$(u, u, l, u, d, l)_1, (u, u, r, d, u, r)_1$
$-\frac{1}{3}J_1 + \frac{1}{3}J_3 + \frac{2}{3}J_4$	$(u, u, l, u, r, d)_1, (u, u, d, l, u, r)_1$

Table 6.8: The 32 configurations of $[2, 1, -1]$ and $[2, -1, 1]$.

[2,0,0] This group can be further classified into 3 sets according to the J_2 term. The first set contains only u and d states with $\frac{1}{2}J_2 - \frac{2}{3}h$, as shown in Table 6.9. The second set contains 4 u, d states with $\frac{1}{6}J_2 - \frac{2}{3}h$. The topologically independent configurations are shown in Table 6.10. The last set contains 2 u, d states with $-\frac{1}{6}J_2 - \frac{2}{3}h$ and are shown in Table 6.11.

[1,1,0] There are two sets in this group. The first set contains 4 u, d states with $\frac{1}{6}J_2 - \frac{2}{3}h$ and most of the configurations in this set are connected to previous groups. The topologically independent configurations are shown in Table 6.12. The second set contains 2 u, d with $-\frac{1}{6}J_2 - \frac{2}{3}h$. Table 6.13 lists the 28 independent configurations.

Energy: $\frac{1}{2}J_2 - \frac{2}{3}h +$	Configurations
$-\frac{2}{3}J_1 + \frac{2}{3}J_3 + \frac{4}{3}J_4$	$(u, u, u, d, u, d)_1, (u, u, d, u, d, u)_1$
$\frac{2}{3}J_1 + \frac{1}{3}J_3 + \frac{2}{3}J_4$	$(u, u, u, d, d, u)_1$
$-\frac{2}{3}J_1 + \frac{1}{3}J_3 + \frac{2}{3}J_4$	$(u, u, d, u, u, d)_1$

Table 6.9: Four of the configurations in [2,0,0] group set 1

Energy: $\frac{1}{6}J_2 - \frac{2}{3}h +$	Configurations
J_4	$(u, u, d, u, l, r)_1, (u, u, l, r, u, d)_1$
$-\frac{1}{3}J_1 + J_4$	$(u, u, d, u, l, l)_1, (u, u, r, r, u, d)_1$
$-\frac{1}{3}J_1 + \frac{1}{3}J_3 + J_4$	$(u, u, d, u, r, r)_1, (u, u, l, l, u, d)_1$
$-\frac{2}{3}J_1 + \frac{1}{3}J_3 + J_4$	$(u, u, d, u, r, l)_1, (u, u, r, l, u, d)_1$

Table 6.10: Eight of the independent configurations in [2,0,0] group set 2

Energy: $-\frac{1}{6}J_2 - \frac{2}{3}h +$	Configurations
$\frac{1}{3}J_1 - \frac{1}{3}J_3 + \frac{4}{3}J_4$	$(u, u, l, l, l, l)_1, (u, u, r, r, r, r)_1$
$\frac{2}{3}J_1 + J_4$	$(u, u, l, l, l, r)_1, (u, u, l, r, r, r)_1$
$\frac{1}{3}J_1 + J_4$	$(u, u, l, l, r, l)_1, (u, u, r, l, r, r)_1$
$\frac{1}{3}J_1 - \frac{1}{3}J_3 + J_4$	$(u, u, l, r, l, l)_1, (u, u, r, r, l, r)_1$
$\frac{1}{3}J_1 + \frac{2}{3}J_4$	$(u, u, l, r, r, l)_1, (u, u, r, l, l, r)_1$
$-\frac{1}{3}J_3 + J_4$	$(u, u, r, l, l, l)_1, (u, u, r, r, r, l)_1$
$\frac{2}{3}J_1 + \frac{1}{3}J_3 + \frac{2}{3}J_4$	$(u, u, l, l, r, r)_1$
$\frac{2}{3}J_1 - \frac{1}{3}J_3 + \frac{4}{3}J_4$	$(u, u, l, r, l, r)_1$
$-\frac{1}{3}J_3 + \frac{4}{3}J_4$	$(u, u, r, l, r, l)_1$
$-\frac{1}{3}J_3 + \frac{2}{3}J_4$	$(u, u, r, r, l, l)_1$

Table 6.11: 16 of the configurations in [2,0,0] group set 3

Pattern 2

There are actually 2 ways to stack the s-plaquettes in pattern 2: $\begin{smallmatrix} S_1 & S_2 & S_3 \\ S_2 & S_3 & S_1 \\ S_3 & S_1 & S_2 \end{smallmatrix}$ and $\begin{smallmatrix} S_1 & S_2 & S_3 \\ S_3 & S_1 & S_2 \\ S_2 & S_3 & S_1 \end{smallmatrix}$.

However, they are not topologically independent. Instead, they are connected by the 90 degree rotation about the center of the s-plaquette. Thus, we only consider the first case.

Under the periodicity and the constraints, the 3 s-plaquette unit cell could be represented

Energy: $\frac{1}{6}J_2 - \frac{2}{3}h +$	Configurations
$-\frac{2}{3}J_1 + \frac{1}{3}J_3 + \frac{4}{3}J_4$	$(u, l, u, r, u, d)_1, (r, u, r, u, d, u)_1$
$-\frac{2}{3}J_1 + \frac{2}{3}J_3 + \frac{4}{3}J_4$	$(u, r, u, l, u, d)_1, (l, u, l, u, d, u)_1$
$-\frac{2}{3}J_1 + \frac{1}{3}J_3 + \frac{5}{3}J_4$	$(u, l, u, l, u, d)_1, (u, r, u, r, u, d)_1, (l, u, r, u, d, u)_1, (r, u, l, u, d, u)_1$

 Table 6.12: The 8 independent configurations in $[1,1,0]$ group set 1

by:

$$(a_1, a_2, a_3, a_4, a_5, a_6)_2 = |a_1, a_4, a_2, a_5\rangle_i \otimes |\bar{a}_2, a_6, a_3, \bar{a}_4\rangle_{i+x} \otimes |\bar{a}_3, \bar{a}_5, \bar{a}_1, \bar{a}_6\rangle_{i+2x}. \quad (6.44)$$

The reason of labeling in this order will be clear soon. Under translational transformation and 90 degree rotation, the following 6 configurations are topologically equivalent:

$$\begin{aligned} (a_1, a_2, a_3, a_4, a_5, a_6)_2 &\equiv (\bar{a}_2, a_3, \bar{a}_1, a_6, \bar{a}_4, \bar{a}_5)_2 \equiv (\bar{a}_3, \bar{a}_1, a_2, \bar{a}_5, \bar{a}_6, a_4)_2 \\ &\equiv (a_3, \bar{a}_2, a_1, \bar{a}_4, a_6, a_5)_2 \equiv (a_2, a_1, \bar{a}_3, a_5, a_4, \bar{a}_6)_2 \equiv (\bar{a}_1, \bar{a}_3, \bar{a}_2, \bar{a}_6, \bar{a}_5, \bar{a}_4)_2. \end{aligned} \quad (6.45)$$

There is a clear separation between the first and the last three indices. We can then divide the configurations into different groups according to:

$$(a_1, a_2, a_3, a_4, a_5, a_6)_2 \rightarrow [S^z(a_1) + S^z(a_2) + S^z(a_3), S^z(a_4) + S^z(a_5) + S^z(a_6)]. \quad (6.46)$$

Energy: $-\frac{1}{6}J_2 - \frac{2}{3}h +$	Configurations
J_4	$(u, l, u, r, l, r)_1, (l, u, l, u, r, l)_1, (u, l, u, r, r, r)_1, (l, u, l, u, r, r)_1$
$\frac{2}{3}J_4$	$(u, r, r, u, l, l)_1, (u, l, l, u, r, r)_1$
$-\frac{1}{3}J_3 + \frac{5}{3}J_4$	$(u, l, u, l, l, l)_1, (r, u, l, u, l, l)_1, (r, u, l, u, l, r)_1, (u, l, u, l, r, l)_1$
$-\frac{1}{3}J_1 + J_4$	$(u, l, u, r, l, l)_1, (u, r, l, u, l, l)_1, (u, l, u, r, r, l)_1, (u, l, l, u, r, l)_1, (r, u, l, u, r, l)_1, (r, u, l, u, r, r)_1$
$-\frac{1}{3}J_3 + \frac{4}{3}J_4$	$(u, l, l, u, l, l)_1, (u, r, l, u, l, r)_1$
$\frac{1}{3}J_1 - \frac{1}{3}J_3 + J_4$	$(u, l, r, u, l, l)_1, (u, l, l, u, l, r)_1$
$\frac{1}{3}J_3 + J_4$	$(l, u, l, u, l, l)_1, (u, l, u, l, l, r)_1, (l, u, l, u, l, r)_1, (u, l, u, l, r, r)_1$
$\frac{2}{3}J_1 - \frac{1}{3}J_3 + \frac{2}{3}J_4$	$(u, l, r, u, l, r)_1$
$-\frac{2}{3}J_1 + \frac{1}{3}J_3 + \frac{2}{3}J_4$	$(u, r, l, u, r, l)_1$

 Table 6.13: The 26 independent configurations in $[1,1,0]$ group set 2

There are in all five groups:

$$[3, -1], \quad [2, 0], \quad [1, 1], \quad [0, 2] \quad \text{and} \quad [-1, 3]. \quad (6.47)$$

[3,-1],[-1,3] There are two sets in these two groups according to the J_2 term, respectively. The first set contains one single independent configuration:

$$(u, u, u, u, d, d)_2, (u, d, d, u, u, u)_2 \quad \text{with energy} \quad \frac{1}{2}J_2 - \frac{2}{3}h - \frac{2}{3}J_1 + \frac{1}{3}J_3 + \frac{2}{3}J_4. \quad (6.48)$$

The second set contains 2 l, r states with J_2 term: $\frac{1}{6}J_2 - \frac{2}{3}h$. There are only 3 topologically independent configurations which are listed in Table 6.14.

Energy: $\frac{1}{6}J_2 - \frac{2}{3}h +$	Configurations
$-\frac{2}{3}J_1 + \frac{1}{3}J_3 + \frac{2}{3}J_4$	$(u, u, u, d, l, l)_2, (u, u, u, d, r, r)_2$
$-\frac{2}{3}J_1 + \frac{1}{3}J_3 + \frac{4}{3}J_4$	$(u, u, u, d, l, r)_2, (d, l, l, u, u, u)_2$
$-\frac{2}{3}J_1 + \frac{2}{3}J_3 + \frac{2}{3}J_4$	$(d, l, r, u, u, u)_2, (d, r, l, u, u, u)_2$

Table 6.14: The 3 configurations in [3,-1] group set 2

[2,0],[0,2] According to the J_2 term, these two groups can be divided into 2 sets, respectively. The first set with $\frac{1}{6}J_2 - \frac{2}{3}h$ is shown in Table 6.15 while the second set with $-\frac{1}{6}J_2 - \frac{2}{3}h$ is shown in Table 6.16.

Energy: $\frac{1}{6}J_2 - \frac{2}{3}h +$	Configurations
$\frac{1}{3}J_3$	$(u, u, l, u, d, l)_2, (u, u, l, d, u, r)_2$
$-\frac{1}{3}J_3$	$(u, u, l, u, d, r)_2, (u, u, l, d, u, l)_2, (u, u, l, u, r, d)_2, (u, u, l, d, l, u)_2,$ $(u, u, l, r, u, d)_2, (u, u, l, l, d, u)_2$
$\frac{1}{3}J_1 + \frac{1}{3}J_3$	$(u, u, l, u, l, d)_2, (u, u, l, l, u, d)_2$
$-\frac{1}{3}J_1 + \frac{1}{3}J_3$	$(u, u, l, d, r, u)_2, (u, u, l, r, d, u)_2$
$-\frac{1}{3}J_1$	$(u, d, l, u, u, l)_2, (u, d, r, u, u, l)_2, (l, d, u, u, u, l)_2, (r, d, u, u, u, l)_2$
$\frac{1}{3}J_1$	$(d, u, l, u, u, l)_2, (d, u, r, u, u, l)_2, (l, u, d, u, u, l)_2, (r, u, d, u, u, l)_2$
0	$(u, l, d, u, u, l)_2, (u, r, d, u, u, l)_2, (d, l, u, u, u, l)_2, (d, r, u, u, u, l)_2$

Table 6.15: The 24 configurations in set 1 of [2,0] and [0,2]

Energy: $-\frac{1}{6}J_2 - \frac{2}{3}h +$	Configurations
$\frac{1}{3}J_3$	$(u, u, l, l, r, r)_2, (u, u, l, r, l, l)_2, (l, l, l, u, u, l)_2, (l, l, r, u, u, l)_2$ $(l, r, l, u, u, l)_2, (r, l, r, u, u, l)_2, (r, r, l, u, u, l)_2, (r, r, r, u, u, l)_2$
$\frac{1}{3}J_1 + \frac{1}{3}J_3$	$(u, u, l, l, l, l)_2, (u, u, l, l, l, r)_2$
$-\frac{1}{3}J_1 + \frac{1}{3}J_3$	$(u, u, l, r, r, l)_2, (u, u, l, r, r, r)_2$
$-\frac{1}{3}J_3 + \frac{4}{3}J_4$	$(u, u, l, l, r, l)_2, (u, u, l, r, l, r)_2, (l, r, r, u, u, l)_2, (r, l, l, u, u, l)_2$

 Table 6.16: The 16 configurations in set 2 of $[2,0]$ and $[0,2]$

[1,1] As before, according to J_2 term, there are three sets. The first set contains only two independent configurations:

$$(u, u, d, u, u, d)_2 \quad \text{with energy} \quad \frac{1}{2}J_2 - \frac{2}{3}h + \frac{2}{3}J_1 - \frac{1}{3}J_3 - \frac{2}{3}J_4, \quad (6.49)$$

$$(u, u, d, u, d, u)_2 \quad \text{with energy} \quad \frac{1}{2}J_2 - \frac{2}{3}h - \frac{1}{3}J_3 - \frac{2}{3}J_4. \quad (6.50)$$

The second set has 14 independent configurations with $\frac{1}{6}J_2 - \frac{2}{3}h$, as shown in Table 6.17.

And the last set with $-\frac{1}{6}J_2 - \frac{2}{3}h$ is shown in Table 6.18.

Energy: $\frac{1}{6}J_2 - \frac{2}{3}h +$	Configurations
$-\frac{1}{3}J_3$	$(u, u, d, u, r, l)_2, (u, u, d, l, r, u)_2, (l, r, u, u, u, d)_2$
$-\frac{2}{3}J_4$	$(l, l, u, u, u, d)_2, (r, r, u, u, u, d)_2$
$\frac{1}{3}J_1 - \frac{1}{3}J_3$	$(u, l, l, u, u, d)_2, (u, r, r, u, u, d)_2$
$\frac{1}{3}J_1 - \frac{2}{3}J_4$	$(u, l, r, u, u, d)_2, (u, r, l, u, u, d)_2$
$-\frac{1}{3}J_3 - \frac{2}{3}J_4$	$(u, u, d, u, r, r)_2$
$\frac{2}{3}J_1 - \frac{1}{3}J_3$	$(u, u, d, u, l, r)_2$
$\frac{2}{3}J_1 + \frac{1}{3}J_3 - \frac{2}{3}J_4$	$(u, u, d, u, l, l)_2$
$\frac{2}{3}J_1 - \frac{1}{3}J_3 - \frac{2}{3}J_4$	$(u, u, d, l, l, u)_2$
$-\frac{2}{3}J_1 + \frac{1}{3}J_3 - \frac{2}{3}J_4$	$(u, u, d, r, r, u)_2$

 Table 6.17: The 14 configurations in set 2 of $[1,1]$

Phase diagram

We have already computed the energy of all possible configurations in the 3 s-plaquette unit cell. After comparison of these energies, we find that there are in all 11 possible phases. These energies and their corresponding phase range are shown in Table 6.19. We also list the representative configurations of these phase in Table 6.20.

An example phase diagram of the $1/3$ plateau is given in Figure 6.6 where we have set $J_1 = 2$ and $J_2 = 3$. We notice that the $1/3$ plateau appears even in the FM J_3

Energy: $-\frac{1}{6}J_2 - \frac{2}{3}h +$	Configurations
0	$(u, l, r, u, r, l)_2, (u, l, r, r, l, u)_2$
$\frac{1}{3}J_3$	$(u, l, l, u, l, l)_2, (u, l, l, r, r, u)_2$
$-\frac{1}{3}J_3$	$(u, l, l, r, u, l)_2$
$-\frac{2}{3}J_4$	$(u, l, r, r, u, l)_2$
$\frac{1}{3}J_1$	$(u, l, r, u, l, r)_2, (u, l, r, l, u, l)_2, (u, l, r, r, u, r)_2, (u, l, r, l, r, u)_2$
$\frac{1}{3}J_1 - \frac{1}{3}J_3$	$(u, l, l, u, r, r)_2, (u, l, l, l, l, u)_2$
$\frac{2}{3}J_1 + \frac{1}{3}J_3$	$(u, l, l, l, u, r)_2$
$-\frac{1}{3}J_3 + \frac{2}{3}J_4$	$(u, l, l, u, r, l)_2, (u, l, l, r, l, u)_2$
$\frac{2}{3}J_3 - \frac{2}{3}J_4$	$(u, l, r, u, l, l)_2, (u, l, r, r, r, u)_2$
$\frac{1}{3}J_1 - \frac{2}{3}J_4$	$(u, l, r, u, r, r)_2, (u, l, r, l, l, u)_2$
$\frac{1}{3}J_1 - \frac{1}{3}J_3 + \frac{2}{3}J_4$	$(u, l, l, u, l, r)_2, (u, l, l, l, u, l)_2, (u, l, l, r, u, r)_2, (u, l, l, l, r, u)_2$
$\frac{2}{3}J_1 + \frac{2}{3}J_3 - \frac{2}{3}J_4$	$(u, l, r, l, u, r)_2$

Table 6.18: The 14 configurations in set 2 of [1,1]

and J_4 range which seems to be contradictoray to our expectation. When we consider the magnetization sequence, these unexpected $1/3$ plateaus will disappear, as will be discussed in the end of this section.

Figure 6.6: The $1/3$ plateau phase diagram with $J_1 = 2$ and $J_2 = 3$.

6.1.5 Other plateaus

Besides the $1/2$ and $1/3$ plateaus, there exists a ubiquitous $5/9$ plateau in the AFM J_3 and J_4 range. This plateau takes over the $1/2$ plateau in a large area of parameter space. Its appearance in the extreme frustrated region is not a coincidence. This could be explained

$$\begin{aligned}
 E_1 &= -\frac{1}{6}J_2 - \frac{2}{3}h - \frac{2}{3}J_1 + \frac{1}{3}J_3 + \frac{2}{3}J_4 \\
 \max\{-10J_1, -J_2, -\frac{2}{3}(J_1 + J_2)\} &\leq J_3 \leq J_1, \\
 \max\{-2J_1, -\frac{1}{3}J_2, -\frac{1}{2}(J_2 + J_3), \frac{2}{3}(J_3 - J_1)\} &\leq J_4 \leq \{\frac{1}{4}J_2, (J_1 - J_3), \frac{1}{4}(2J_1 \pm J_3)\}
 \end{aligned}$$

$$\begin{aligned}
 E_2 &= -\frac{1}{6}J_2 - \frac{2}{3}h - \frac{1}{3}J_3 + \frac{5}{3}J_4 \\
 J_3 &\geq \frac{1}{2}(2J_1 - J_2), J_4 \leq \min\{0, J_3, \frac{2}{3}(J_3 - J_1)\}
 \end{aligned}$$

$$\begin{aligned}
 E_3 &= -\frac{1}{6}J_2 - \frac{2}{3}h + \frac{1}{3}J_3 + J_4 \quad (J_2 \geq 6J_1) \\
 \max\{4J_1 - J_2, \frac{5}{6}(2J_1 - J_2)\} &\leq J_3 \leq -2J_1, \\
 \max\{J_3, 2J_1 - J_2 - J_3\} &\leq J_4 \leq \min\{-2J_1, \frac{1}{5}J_3\}
 \end{aligned}$$

$$\begin{aligned}
 E_4 &= -\frac{1}{6}J_2 - \frac{2}{3}h - \frac{1}{3}J_3 \\
 J_3 &\geq \max\{\frac{2}{3}J_1, \frac{1}{4}(4J_1 - J_2)\}, \max\{0, J_1 - J_3\} \leq J_4 \leq \min\{\frac{1}{2}J_2, \frac{1}{2}J_3, \frac{1}{2}(2J_3 - 2J_1 + J_2)\}
 \end{aligned}$$

$$\begin{aligned}
 E_5 &= -\frac{1}{6}J_2 - \frac{2}{3}h - \frac{2}{3}J_4 \quad (J_1 < J_2) \\
 \max\{0, 2J_1 - J_2\} &\leq J_3 \leq J_2, J_4 \geq \max\{\frac{1}{2}J_3, \frac{1}{4}(2J_1 - J_3)\}
 \end{aligned}$$

$$\begin{aligned}
 E_6 &= -\frac{1}{6}J_2 - \frac{2}{3}h + \frac{2}{3}J_3 - \frac{2}{3}J_4 \\
 J_3 &\leq \min\{0, J_2 - 2J_1\}, J_4 \geq \max\{\frac{1}{5}J_3, \frac{1}{6}(2J_1 - J_2), \frac{1}{4}(2J_1 - J_2), \frac{1}{4}(2J_1 + J_3)\}
 \end{aligned}$$

$$\begin{aligned}
 E_7 &= \frac{1}{6}J_2 - \frac{2}{3}h - \frac{2}{3}J_1 + \frac{2}{3}J_3 + \frac{4}{3}J_4 \\
 J_3 &\leq \min\{-\frac{1}{3}J_2, \frac{1}{2}(2J_1 - J_2)\}, J_3 \leq J_4 \leq \min\{0, \frac{1}{6}(2J_1 - J_2), -\frac{1}{2}(J_2 + J_3), 2J_1 - J_2 - J_3\}
 \end{aligned}$$

$$\begin{aligned}
 E_8 &= \frac{1}{6}J_2 - \frac{2}{3}h - \frac{2}{3}J_1 + \frac{1}{3}J_3 + \frac{5}{3}J_4 \\
 J_3 &\leq \frac{1}{2}(2J_1 - J_2), J_4 \leq \min\{J_3, -\frac{1}{3}J_2\}
 \end{aligned}$$

$$\begin{aligned}
 E_9 &= \frac{1}{6}J_2 - \frac{2}{3}h - \frac{2}{3}J_1 + \frac{2}{3}J_3 + \frac{2}{3}J_4 \quad (2J_1 \geq J_2) \\
 J_3 &\leq -J_2, 0 \leq J_4 \leq \min\{\frac{1}{4}(2J_1 - J_2), -\frac{1}{4}J_3\}
 \end{aligned}$$

$$\begin{aligned}
 E_{10} &= \frac{1}{6}J_2 - \frac{2}{3}h - \frac{1}{3}J_3 - \frac{2}{3}J_4 \\
 J_3 &\geq \max\{J_1, J_2\}, J_4 \geq \frac{1}{2}J_2
 \end{aligned}$$

$$\begin{aligned}
 E_{11} &= \frac{1}{6}J_2 - \frac{2}{3}h - \frac{2}{3}J_1 + \frac{1}{3}J_3 - \frac{2}{3}J_4 \quad (2J_1 \geq J_2) \\
 J_2 - 2J_1 &\leq J_3 \leq \min\{J_1, 2J_1 - J_2\}, J_4 \geq \max\{\frac{1}{4}J_2, -\frac{1}{4}J_3, -\frac{1}{2}(2J_3 - 2J_1 + J_2)\}
 \end{aligned}$$

 Table 6.19: The 11 phases and their corresponding phase range in the J_3 and J_4 plane.

$E_1:$	$(u, r, l, u, r, l)_1$
$E_2:$	$(u, l, u, l, l, l)_1, (r, u, l, u, l, l)_1, (r, u, l, u, l, r)_1, (u, l, u, l, r, l)_1$
$E_3:$	$(l, u, l, u, l, l)_1, (u, l, u, l, l, r)_1, (l, u, l, u, l, r)_1, (u, l, u, l, r, r)_1$
$E_4:$	$(u, l, l, r, u, l)_2$
$E_5:$	$(u, l, r, r, u, l)_2$
$E_6:$	$(u, l, r, u, l, l)_2, (u, l, r, r, r, u)_2$
$E_7:$	$(u, r, u, l, u, d)_1, (l, u, l, u, d, u)_1$
$E_8:$	$(u, l, u, l, u, d)_1, (u, r, u, r, u, d)_1, (l, u, r, u, d, u)_1, (r, u, l, u, d, u)_1$
$E_9:$	$(d, l, r, u, u, u)_2, (d, r, l, u, u, u)_2$
$E_{10}:$	$(u, u, d, u, r, r)_2$
$E_{11}:$	$(u, u, d, r, r, u)_2$

Table 6.20: The representative configurations of the 11 phases. The subscript 1 and 2 denoting different stacking patterns are defined in Equation 6.42 and 6.44.

as the following. The fully polarised state has no frustration at all, because the system now has maximum energy from the spin-spin interactions. When the field decreases and a spin flip becomes possible, the configuration should simultaneously maximize the uniform magnetization and minimize the spin-spin interaction. This turns out to be a 5/9 plateau as shown in Figure 6.7. The unit cell of the 5/9 plateau consists of 9 s-plaquettes:

$$\begin{matrix} s_1 & s_2 & s_3 \\ s_4 & s_5 & s_6 \\ s_7 & s_8 & s_9 \end{matrix}$$

with

$$\begin{aligned} |s_1\rangle &= |l, u, u, r\rangle, |s_2\rangle = |u, r, u, u\rangle, |s_3\rangle = |u, u, r, l\rangle, \\ |s_4\rangle &= |u, u, r, r\rangle, |s_5\rangle = |l, l, l, l\rangle, |s_6\rangle = |r, u, u, u\rangle, \\ |s_7\rangle &= |r, l, u, u\rangle, |s_8\rangle = |u, u, u, r\rangle, |s_9\rangle = |u, r, l, u\rangle. \end{aligned}$$

The 5/9 plateau has energy:

$$E_{5/9} = -\frac{10}{9}h + \frac{2}{9}J_1 + \frac{1}{18}J_2 + \frac{1}{9}J_3 + \frac{2}{9}J_4. \quad (6.51)$$

From the figure, we can see that, within the interacting range, all the $-1/2$ spins (white dots) are surrounded by $1/2$ spins which minimizes the spin-spin interactions on all of the four different bonds. The 5/9 plateau is very stable even in the existence of quantum fluctuations from the spin anisotropy which we will see later.

In the weak field region, there is another possible 2/9 plateau similar to that observed

in the original Shastry-Sutherland model[65]. Its configuration is shown in Figure 6.8 with energy

$$E_{2/9} = -\frac{4}{9}h - \frac{5}{18}J_2 + \frac{1}{3}J_3 - \frac{4}{9}J_4. \quad (6.52)$$

Finally, we also have a trivial plateau, the fully polarized state with all $1/2$ spins and it has energy:

$$E_{full} = -2h + 2J_1 + \frac{1}{2}J_2 + J_3 + 2J_4. \quad (6.53)$$

Figure 6.7: The $5/9$ plateau: all the $-1/2$ spins have minimum interaction energy with surrounding $1/2$ spins.

Figure 6.8: The configurations of the $2/9$ plateau.

6.1.6 Magnetization sequence

As mentioned previously, the phase diagram of each plateau was obtained by comparing the energy of different configurations. This could result in some artificial plateau con-

configurations which are not stable actually. The stability of a plateau configuration can be obtained from its field range. For a particular plateau configuration, we can compare its energy with higher plateau and lower plateau, which gives an upper bound h_+ and lower bound h_- of the field, respectively. The plateau configuration is stable only when $h_- < h_+$.

For example, in the FM J_3 and AFM J_4 region, there is a possible magnetization sequence $0 - 1/3 - 1/2 - 1$. The 0-plateau is in the E_4 region, Table 6.3, with energy $E_0 = -\frac{1}{2}J_2 + J_3 - 2J_4$. The $1/3$ plateau has configuration E_6 , Table 6.19 with energy $E_{1/3} = -\frac{2}{3}h - \frac{1}{6}J_2 + \frac{2}{3}J_3 - \frac{2}{3}J_4$. The $1/2$ plateau has configuration E_3 , Table 6.6, with energy $E_{1/2} = J_3 - h$. The energy of the fully polarized state is given by Equation 6.53. We could obtain the field range of the $1/3$ plateau as following:

$$\begin{cases} E_{1/3} < E_0 & \rightarrow h > h_- = \frac{1}{2}J_2 - \frac{1}{2}J_3 + 2J_4 \\ E_{1/3} < E_{1/2} & \rightarrow h < h_+ = \frac{1}{2}J_2 + J_3 + 2J_4 \end{cases} \quad (6.54)$$

For a stable configuration, we require:

$$h_- < h_+ \Rightarrow J_3 > 0. \quad (6.55)$$

However, the phase range of E_6 in $1/3$ plateau is $J_3 < \min\{0, J_2 - 2J_1\} \leq 0$. Therefore, E_6 is an unstable $1/3$ plateau and will not appear in the magnetization sequence.

Applying the same procedures to all possible magnetization sequences, we can obtain the correct phase diagram of each plateau, as shown in Table 6.21.

The configurations of the $1/2$ plateaus are the same as before. The 5 representative configurations of the $1/3$ plateau are given in Figure 6.9.

The new phase diagram of the plateaus with different values of J_1 and J_2 are shown in Figure 6.10-6.13. Particularly, the magnetization sequence with $J_1 = J_2 = 1$ is shown in Figure 6.14. The parameters for TmB_4 and ErB_4 are indicated in the figure. The two points SS1 and SS2 denote two supersolid phases appear just below the $1/2$ plateau which will be discussed in next section.

$E_1^{1/3} = -\frac{1}{6}J_2 - \frac{2}{3}h + \frac{2}{3}J_3 - \frac{2}{3}J_4$ $-\frac{1}{2}J_2 \leq J_3 \leq J_1,$ $\max\{-\frac{1}{4}J_2, -2J_1, \frac{1}{2}(J_3 - J_1), -\frac{1}{2}(J_2 + J_3)\} \leq J_4 \leq \min\{\frac{1}{4}J_2, J_1 - J_3, -\frac{1}{4}J_3 + \frac{1}{2}J_1\}$
$E_2^{1/3} = -\frac{1}{6}J_2 - \frac{2}{3}h - \frac{1}{3}J_3$ $J_3 \geq \max\{\frac{2}{3}J_1, \frac{1}{4}(4J_1 - J_2)\}, \max\{0, J_1 - J_3\} \leq J_4 \leq \min\{\frac{1}{2}J_2, \frac{1}{2}J_3, \frac{1}{2}(2J_3 - 2J_1 + J_2)\}$
$E_3^{1/3} = -\frac{1}{6}J_2 - \frac{2}{3}h - \frac{2}{3}J_4$ $J_1 < J_2, \max\{0, 2J_1 - J_2\} \leq J_3 \leq J_2, J_4 \geq \max\{\frac{1}{2}J_3, \frac{1}{4}(2J_1 - J_3)\}$
$E_4^{1/3} = \frac{1}{6}J_2 - \frac{2}{3}h - \frac{1}{3}J_3 - \frac{2}{3}J_4$ $J_3 \geq \max\{J_1, J_2\}, J_4 \geq \frac{1}{2}J_2$
$E_5^{1/3} = \frac{1}{6}J_2 - \frac{2}{3}h - \frac{2}{3}J_1 + \frac{1}{3}J_3 - \frac{2}{3}J_4$ $2J_1 > J_2, \frac{1}{2}J_2 - J_1 \leq J_3 \leq \min\{J_1, 2J_1 - J_2\}, J_4 \geq \max\{\frac{1}{4}J_2, -\frac{1}{4}J_3, \frac{1}{2}(2J_3 - 2J_1 + J_2)\}$
$E_1^{1/2} = 2J_4 - h$ $J_3 \geq -\frac{1}{2}J_2, J_4 \leq \min\{0, \frac{1}{2}J_3\}$
$E_2^{1/2} = J_4 - h$ $J_3 > 0, 0 \leq J_4 \leq \min\{J_3, \frac{1}{3}J_1, \frac{1}{5}(J_1 + J_3), \frac{1}{8}J_3 + \frac{1}{8}(J_1 + \frac{1}{2}J_2), \frac{1}{10}(2J_1 + J_2), \frac{1}{3}J_3 + \frac{1}{6}J_2\}$
$E_3^{1/2} = J_3 - h$ $J_3 \leq \frac{1}{10}J_2, J_4 \geq \max\{-\frac{1}{4}J_2, \frac{1}{2}J_3, \frac{5}{2}J_3\}$
$E_4^{1/2} = \frac{1}{2}J_1 - h$ $J_1 < \frac{1}{4}J_2, J_3 \geq \frac{3}{2}J_1, J_4 \geq \frac{3}{4}J_1$
$E_5^{1/2} = \frac{1}{2}J_2 - h$ $J_1 \geq \frac{5}{2}J_2, J_3 \geq \frac{1}{2}J_2, J_4 \geq \max\{\frac{1}{2}J_2, -\frac{1}{2}J_3 - \frac{1}{2}J_1 + \frac{9}{4}J_2\}$

Table 6.21: Phase range of the stable 1/2 and 1/3 plateaus

Figure 6.9: The five configurations of the 1/3 plateau.

 Figure 6.10: $1/3$ plateau with $J_1 = 1$ and $J_2 = 3$.

 Figure 6.11: $1/3$ plateau with $J_1 = J_2 = 1$.

 Figure 6.12: $1/3$ plateau with $J_1 = 2$ and $J_2 = 1$.

 Figure 6.13: $1/2$ plateau with $J_1 = J_2 = 1$.

 Figure 6.14: Magnetization sequence with $J_1 = J_2 = 1$.

6.2 The XXZ model

The spin anisotropy introduces quantum fluctuations to the spin system. Together with the thermal fluctuation, it could probably melt the plateaus or generate new plateaus in the highly frustrated region, AFM J_3 and J_4 , via the quantum order by disorder mechanism. Near stable plateaus, quantum fluctuations could result in supersolid phase as well. In this section, we focus on the Ising-like XXZ model, $\Delta < 1$.

6.2.1 Weak frustration

In the weak frustrated region, AFM J_3 and FM J_4 or FM J_3 and AFM J_4 , we find that the $1/2$ plateau is stable against the quantum fluctuation. In the AFM J_3 and FM J_4 region, the Ising Hamiltonian predicts a magnetization sequence from columnar state to the $E_1^{1/2}$ $1/2$ plateau without a $1/3$ plateau. This is qualitatively stable against the quantum fluctuation, as shown in Figure 6.15. m_c^2 and m_s^2 are the columnar and stagger magnetization, respectively. Both of the order parameter characterize the spin crystal ordering. The figure clearly shows that the ground state in the low field is indeed a columnar state. m_z and ρ_s are the uniform magnetization and spin stiffness, respectively. Finite spin stiffness indicates the spin superfluid state. When the field increases from zero, the columnar state remains because of the spin gap. The system undergoes a first order phase transition at the first critical field around 1.24, when it becomes spin superfluid. The finite yet small crystal ordering is considered as the finite size effect which decreases to zero as the system sizes becomes larger. Around $h = 2$, the spin system undergoes another phase transition and becomes crystalized with simultaneous superfluid ordering, that is in the supersolid phase (SS1). The FM J_4 stabilizes the crystal ordering while quantum fluctuations through the AFM J_3 and J_1 enhance the superfluid ordering, as can be clearly seen from Figure 6.5a.

Similarly, in the other weakly frustrated region, AFM J_4 and FM J_3 , there exist the magnetization sequence from J_3 chessboard state to the $E_3^{1/2}$ $1/2$ plateau and another supersolid phase (SS2) just below the plateau. The ordering parameters are shown in

Figure 6.15: The $1/2$ plateau and the supersolid phase with columnar magnetic order just below the plateau in the weakly frustrated region. We have set $J_1 = J_2 = 1$.

Figure 6.16. In this case, the FM J_3 stabilizes the chessboard crystal ordering while the J_4 mediates the quantum fluctuations resulting in the superfluid ordering.

One intriguing difference between these two supersolid phase is that while they both emerge from superfluid phases as a half plateau is approached from below, in SS1 this occurs as a first order phase transition while in SS2 this is a continuous phase transition. Comparing the configurations in Figure 6.3d and 6.5c, while the superfluid in SS2 is formed through transverse J_4 interaction, there are also condensation of up spins along the J_3 diagonal chain with all down spins in the J_3 chessboard state. When the density of the condensation increases, the superfluid velocity decreases because the quantum fluctuation is suppressed by the dense up spins. When the condensation saturates, the superfluidity is gone immediately because the condensation becomes frozen in the space and a crystal of $1/2$ plateau is formed completely. The crystal ordering is formed gradually during the saturation procedure, resulting a continuous phase transition. In the SS1 case, there is no obvious competition between the superfluid motion and the formation of the crystal ordering. The superfluid only disappears when the spin system is crystalized again and

Figure 6.16: The $1/2$ plateau and the supersolid phase with chessboard magnetic order just below the plateau in the weakly frustrated region. We have set $J_1 = J_2 = 1$.

this crystalization is a first order phase transition.

6.2.2 Strong frustration

In the extreme frustrated region, AFM J_3 and J_4 , there is strong competition between two s-plaquette unit cell and large sized unit cell. As a result, some other plateaus which could be further stabilized by the spin anisotropy may appear. Though the s-plaquette method is able to calculate all the possible plateaus, when the unit cell becomes large, it becomes not efficient due to the many possible configurations of the plateaus. Thus we use stochastic series expansion QMC method to explore possible plateaus in this extreme frustrated region. The result also serves as a verification of the prediction of the plateaus from the s-plaquette in the Ising limit.

Different relations between J_1 and J_2 could result in very obvious qualitative change in the phase diagram and consequent difference in the plateaus. Hence, we focus on two regimes of the bare Shastry-Sutherland interactions, namely the Néel regime $J_1 > 2J_2$ and the dimer regime $J_1 < 2J_2$. In the first case, we set $5J_1 = 2J_2$ and in the unit of J_2 . In the second case, we set $J_1 = J_2$ and also in the unit of J_2 . The spin anisotropy

is set to be $\Delta = 0.1$ in both cases. To reduce the number of variables, we fix J_4 and the magnetic phase diagram is given in terms of J_3 and the field h . The phase diagram of both cases are shown in Figure 6.17 and 6.18, respectively.

Figure 6.17: The magnetic phase diagram for $J_1 = J_2 = 1$ and $J_4 = 0.2$. The X axis is h , the Y axis is J_3 and the Z axis is the magnetization m_z . The inverse temperature is set to be $\beta = 32$ and the system size is 12×12 . The spin anisotropy is $\Delta = 0.1$.

In the weak FM J_3 regime, there is only one $1/2$ plateau in both cases as we have predicted in the Ising limit. This means the quantum fluctuation is not strong enough to melt the $1/2$ plateau or generate another plateau in this regime. On the other hand, in the extreme frustrated regime, the broad stability of the $5/9$ plateau complements the $1/2$ plateau in the weak FM J_3 regime. This also implies that the $5/9$ plateau is robust against the quantum fluctuations.

Similar to the supersolid phase near the $1/2$ plateau, there could be possible related supersolid phase for magnetizations just below $5/9$ plateau. This putative supersolid phase may even extend down to $1/3$ plateaus. If this is indeed the case, it would be quite interesting as the $1/3$ and $5/9$ plateaus do not share the same ordering wave vectors. It is likely that the putative supersolid phase does not completely span the distance between these plateaus, but rather undergoes a first order phase transition in one case and a continuous phase transition in the other. Further investigation is required to verify this supersolid phase.

Figure 6.18: The magnetic phase diagram for $5J_1 = 2J_2 = 2$ and $J_4 = 0.2$. The X axis is h , the Y axis is J_3 and the Z axis is the magnetization m_z . The inverse temperature is set to be $\beta = 32$ and the system size is 12×12 . The spin anisotropy is $\Delta = 0.1$.

Figure 6.19: The magnetization versus field for $J_1 = J_2 = 1$, $J_3 = 1.8$ and $J_4 = 0.3$. The $2/9$ plateau is obvious while we see a possibly unstable $1/3$ plateau.

From the magnetic phase diagram, we also notice a possible $2/9$ plateau as we have expected in the Ising limit. From the configuration of the $2/9$ plateau, large J_4 is required to stabilize it. Hence, we further investigate the case with $J_4 = 0.3J_2$ and a stable $2/9$ plateau is observed clearly, as shown in Figure 6.19. Besides, we realized that the $1/3$ plateau may become unstable and actually be complemented by the $2/9$ plateau.

6.3 Conclusion

In this chapter, we have used both the spiral plaquette language and the stochastic series expansion Quantum Monte Carlo to explore the possible plateaus and supersolid phases in the extended Shastry-Sutherland model. In the Ising limit, the s-plaquette method is very efficient for plateau configuration and energy in the weakly frustrated regime. It also predicts the possible size of the unit cell therefore suggests possible plateaus in the extremely frustrated regime. The results agree qualitatively well with stochastic series expansion method. The stochastic series expansion is very efficient in the Ising-like XXZ model in both weakly and extremely frustrated regime. We have observed many plateaus in different parameter ranges. Besides the ubiquitous $1/2$ and $1/3$ plateaus, we also found a $5/9$ plateau in the extremely frustrated regime complementing the $1/2$ plateau in the weakly frustrated regime. For large J_4 , we also found a stable $2/9$ plateau possibly complementing the $1/3$ plateau in the strongly frustrated regime. We have also observed different supersolid phases near the plateaus generated by the quantum fluctuations due to the spin anisotropy. Depending on the structures of the plateaus, the phase transition to supersolid could be either first order or continuous.

Chapter 7

General conclusion

Shastry-Sutherland model is a good candidate for studying the interplay between the interaction and geometrical frustration. While the ground states are well known in the two weakly frustrated limits, the ground state in the extremely frustrated regime remains not so clear, even though several numerical studies have suggested a plaquette singlet state. We have used a plaquette representation and a second order perturbation theory to obtain the ground state energy and explicitly showed that the plaquette singlet is indeed the intermediate phase in the strongly frustrated regime. Partially motivated by the relevance between Shastry-Sutherland model and the rare earth tetraborides, we extended our study to the anisotropic case. And we find a generalized plaquette valence bond solid (PVBS) phase with the plaquette singlet as a special one. The PVBS phase is suppressed by the anisotropy, both Ising- and XY-like. In the large J_1/J_2 limit, the ground state develops an antiferromagnetic long range order through a continuous phase transition. In the other limit, the ground state becomes the dimer singlet state through a first order phase transition.

We have also studied the behaviour of the excitations above the generalized PVBS. To capture a more complete dispersion relation, we used the full local Hilbert space without any truncation and we found a critical line where the behaviour of the lowest dispersion changes qualitatively. The dispersion has a single gap at $\mathbf{k} = (0, 0)$ for small J_2/J_1 while on the other side of the critical line, the dispersion has four degenerate gaps. We consider

this as a direct consequence from the qualitative change in the quantum fluctuations which implies a qualitative change in the ground state, since the ground state actually consists of two parts: the classical ground state (PVBS) and quantum fluctuations. We expect the ground state has resonating valence bond order in the latter case.

Magnetization plateau is always an interesting phenomenon in magnetic materials, especially those with geometrical frustration. Depending on the lattice of interaction, many possible plateaus could occur and there even exist many different configurations for the same magnetization plateau. To understand the mechanism behind the magnetization plateaus in the rare earth tetraborides, we studied the extended Shastry-Sutherland model with two further interactions. One of the reason is that previous studies based on the bare Shastry-Sutherland model were not able to reproduce the experimental observations. Another reason lies in the fact that the existence of the itinerant electrons will induce the RKKY interactions with the localized magnetic moments which may result in effective further interactions.

In the Ising limit, we have used the spiral plaquette representation as our analytical method to construct possible magnetization plateaus. Depending on the nature of the interactions (AFM or FM) and the relative strength, each plateau has more than one possible configurations. And we also found regions without any plateaus. Combined the phase diagrams of different plateaus and we obtained various magnetization sequence in the (J_3, J_4) parameter space. These configurations of the plateaus serve as the classical ground state of possible plateaus in the Ising-like XXZ model.

In the Ising-like XXZ model, we used Stochastic Series Expansion Quantum Monte Carlo method as our numerical method to explore possible plateaus in the extremely frustrated regime where our spiral plaquette method may not be efficient and may miss some other possible plateaus. The numerical results are actually consistent qualitatively with the analytical prediction from the Ising limit. Quantum fluctuations due to the transverse interactions shrink the stability of the plateaus. Besides, we also found supersolid phases near some of the plateaus.

The consistency of the numerical method (QMC) and the analytical method (spin

wave theory) have been tested on a Spin-1 anisotropic Heisenberg model. We have obtained the phase diagram in the parameter space of the single-ion anisotropy and external field. The results from both methods agreed very well both quantitatively and qualitatively. Besides, the lowest order excitation dispersions above the QPM phase from spin wave theory agreed well with that obtained from QMC. The results guaranteed the validity and efficiency of both methods, especially the spin wave method which could provide convenient and intuitive idea about the physics of various models.

In conclusion, we have used both analytical and numerical methods to study Quantum spin systems with geometrical frustration. We have observed novel quantum phases and the consequences of the interplay between quantum fluctuations and geometrical frustration.

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