Frustrated Magnetism in Strongly Correlated Electron Systems

by

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ABSTRACT

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A deep understanding of magnetism is essential for its application in magnetic semiconductors, spintronic devices and unconventional superconductors. In this work, we study magnetic structures and their corresponding excitations in several strongly correlated electron systems, where exotic orderings can be induced as a result of magnetic frustration and quantum fluctuations. We show that emergent spin textures can arise close to a magnetic field-induced quantum critical point, when the single magnon excitations have several degenerate non-coplanar minima. In this case, quantum fluctuations can lift such degeneracy and lead to the crystallization of the magnetic vortex strings.

Magnetic frustration also plays an important role in Fe-based superconductors. We analyze the spin excitations in the ordered as well as paramagnetic phase of these materials, and find that higher order spin exchanges are essential for understanding the inelastic neutron scattering experiments (INS). The presence of such higher order spin interactions has far-reaching consequences, potentially resulting in more exotic phases, such as the multipolar orders. In particular, we find propensity to ferro-quadrupolar order, which we propose as a candidate for the ground state of the iron selenide FeSe. We find that the calculated spin excitations in this quadrupolar state closely resemble the results of recent INS measurements.
In addition to electron spins, orbital physics also plays a prominent role in Fe-based superconductors. We study the interplay between spin and orbital degrees of freedom and show that the so-called nematic order can be naturally understood as the decoupling of the two transitions, when orbital ordering preempts long-range magnetic spin order. Our results reveal that magnetic frustration plays an important role in several strongly correlated electron systems, and elucidating its consequences is crucial for the understanding and potential application of these materials.
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Preface

This thesis is based on the research conducted in Prof. Andriy H. Nevidomskyy’s group. Part of the work had been performed in collaboration with Prof. Qimiao Si’s group at Rice University and Prof. Cristian D. Batista’s group at Los Alamos National Laboratory. The main results of this thesis can be found in the following publications:


Chapter 1

Introduction

Life is not always without frustration, same as in magnets. In real life, there are always conflicts of choices and politics, under which not every interest can be satisfied. By analogy, in certain situations the competing interactions in magnets are also frustrated: among the many magnetic interactions, sometimes it is impossible to satisfy any given one (by minimizing the fraction of the energy corresponding to this interaction), without sacrificing the others.

While it is difficult to judge frustration in human life, the frustration of magnetic type is always interesting. As we shall see in this thesis, magnetic frustration is able to create exotic states of matter, which host nontrivial static and dynamical properties. In fact, the magnetic structures induced by frustration are not only conceptually important for the understanding of physics, but also practically important because of their (potential) applications in reducing human-life frustration. It is then the motivation of this thesis to study various types of frustration-induced magnetic phenomena in various solid state systems.

1.1 Magnetic Interactions

It is unavoidable to talk about magnetic interactions before talking about magnetic frustration. The commonly seen magnetic interactions mainly fall into two types: ferromagnetic and antiferromagnetic exchanges. In both cases we can write down the
spin exchange between the two sites $i$ and $j$ as follows:

\[ H_{ex} = J S_i \cdot S_j \]  \hspace{1cm} (1.1)

where the sign of $J$ determines the type of exchange of the spins $S_i$ and $S_j$:

- $J < 0$ (ferromagnetic), the two spins favor aligning in the same direction.
- $J > 0$ (anti-ferromagnetic), the two spins favor aligning in the opposite direction.

The origin of such spin exchange varies from material to material. In the following we will look at several most common types:

- Direct exchange mechanism,
- Kinetic exchange mechanism,
- Super-exchange mechanism,
- Double exchange mechanism,
- other mechanisms.

1.1.1 Direct exchange

The electron behavior is strongly affected by the Coulomb repulsion. Consider two electrons in two orbitals (each occupying one orbital), then we can write down the electron wave functions as Slater Determinants. For antisymmetric orbital wave functions, the corresponding spin wave functions of the two electrons then must be symmetric. Due to the Coulomb repulsion, it can be shown that symmetric spin wave function is always favored when the two orbitals are orthogonal, inducing a ferromagnetic direct exchange (also known as “exchange hole” effect) [1].
This is actually nothing but the Hund’s first rule, which favors largest total spin for the ground state of electrons in given single-electron orbitals [2].

Typically, antiferromagnetic exchanges are found more frequently in nature than ferromagnetic exchanges. Indeed, for electrons occupying non-orthogonal orbitals, it is possible to change the interaction from ferromagnetic to antiferromagnetic (when the orthogonality of orbitals becomes poor) [1]. And if we loosen the infinite onsite repulsion constraint, there is also possibility of kinetic exchanges and super-exchanges described in the following sections, which are indeed commonly found to be antiferromagnetic.

1.1.2 Kinetic exchange

The simplest example of kinetic exchange can be realized in the Hubbard Model [3]:

\[
\mathcal{H} = -t \sum_{\langle i,j \rangle} \sum_{\sigma=\uparrow,\downarrow} (c_i^\dagger c_j + h.c.) + U \sum_i n_{i\uparrow} n_{i\downarrow} \tag{1.2}
\]

where the first term describes the kinetic energy of electrons, while the second term describes the repulsion of electrons which prevents them from occupying the same site.

In the case of strong repulsion \(U/t \gg 1\), the double occupancy of electrons on the same site has a large energy penalty. However, if the two spins on neighboring sites have opposite direction, the exchange of the two can lower the energy of order \(-4t^2/U\), which can be verified by the Schrieffer-Wolff transformation [4]:

\[
\mathcal{H}_{\text{eff}} = e^{-S} \mathcal{H} e^{S} \\
= \mathcal{H} - [S, \mathcal{H}] + \frac{1}{2} [S, [S, \mathcal{H}]] - \frac{1}{3!} [S, [S, [S, \mathcal{H}]]] + \ldots \tag{1.3}
\]

where the operator \(S\) is chose in a way such that the first order term does not connect the two Hubbard subband, which reduces the effective Hamiltonian to the \(t - J\)
model \[1,5\]:

$$
\mathcal{H}_{tJ} = -t \sum_{(i,j)} \sum_{\sigma} \left[ \tilde{c}^\dagger_{i\alpha} c_{j\sigma} + h.c. \right] + \frac{4t^2}{U} \sum_{(i,j)} \left( S_i \cdot S_j - \frac{n_in_j}{4} \right) + \ldots, \quad (1.4)
$$

where

$$
S_i = \frac{1}{2} c^\dagger_{i\alpha} \sigma_{\alpha\beta} \tilde{c}_{i\beta} \quad (1.5)
$$
is the spin operator, and

$$
\tilde{c}_{i\alpha}^\dagger = (1 - n_{i\sigma}) c_{i\sigma} \quad (1.6)
$$
is the fermionic operator projected to the subspace where double-occupancy is not allowed.

We see that an effective antiferromagnetic spin exchange is induced in this large-$U$ limit of the Hubbard Model.

### 1.1.3 Super-exchange

The kinetic exchange previously mentioned requires direct hopping of electrons between different sites. However, in typical transition metal compounds, the electrons on $d$-orbital cations are far separated by anions. Kramers and Anderson showed that \[6,7\], with indirect exchanges through the anion orbital, an effective antiferromagnetic exchange should also arise.

The simplest example of the super-exchange contains one $p_\sigma$ anion orbital sandwiched by two $d_{3z^2-r^2}$ cation orbitals, where the anion orbital is filled with two electrons and the cation orbitals are half-filled (see Fig. 1.1). We can see that the energy can be lowered through a fourth order process, which exchanges the two cation spins and leads to an antiferromagnetic exchange \[6,7,1\].

The super-exchange interactions in real materials goes beyond the above simple analysis, which severely depends on the filling and bonding angles of the cations and
anions. For example, the effective exchange can be even ferromagnetic if the bonding angle is close to 90° [8].

1.1.4 Double exchange

Ferromagnetic exchanges can also arise when there is orbital degeneracy. One famous example is the double exchange [9]: If there are more than one orbitals per site, then the electron hopping events are strongly influenced by Hund’s coupling:

$$\mathcal{H} = -t \sum_{\langle i,j \rangle} \sum_{\sigma} (c_{i\sigma}^\dagger c_{j\sigma} + h.c.) - J_H \sum_i S_i \cdot s_i,$$

(1.7)

where $s_i$ is the spin of the itinerant electrons, while $S_i$ is the effective spin formed by the rest relatively localized orbitals. This model is often called double exchange model or Kondo Lattice model.

Comparing to the previous exchange mechanisms, the double exchange is more complicated:

- The itinerant electron will mediate Ruderman-Kittel-Kasuya-Yosida (RKKY) interactions among the local spins [10][11][12].

- Conversely, the electron band structure will be modified in the local spin background.
In the limit of strong Hund’s coupling, Kondo singlets can even form on each site, reducing the itinerant carriers [13].

In the case of large Hund’s coupling (but not yet forming Kondo singlets), the itinerant electron spins become parallel to the local spins. If the background local spins order ferromagnetically (which is common for large Hund’s coupling), then it also leads to an effective ferromagnetic interaction for the itinerant electrons.

1.1.5 Other mechanisms

There are other types of exchange mechanisms beyond the above mentioned simple cases. For example, in the presence of spin-orbit coupling, Dzyaloshinskii-Moriya exchanges can be induced [14, 15]:

\[ \mathcal{H} = D_{ij} \cdot (\mathbf{S}_i \times \mathbf{S}_j). \]  

Higher order interactions should in principle also arise, for example the biquadratic term [1] (such terms only have nontrivial effect on spin \( S \geq 1 \)):

\[ \mathcal{H} = K (\mathbf{S}_i \cdot \mathbf{S}_j)^2, \]  

and the ring-exchange term [1]:

\[ \mathcal{H} = \sum_{ijkl} [(\mathbf{S}_i \cdot \mathbf{S}_j)(\mathbf{S}_k \cdot \mathbf{S}_l) + (\mathbf{S}_j \cdot \mathbf{S}_k)(\mathbf{S}_l \cdot \mathbf{S}_i) - (\mathbf{S}_i \cdot \mathbf{S}_k)(\mathbf{S}_j \cdot \mathbf{S}_l)]. \]  

1.2 Magnetic Frustration

The ground states of magnets are reached by minimizing the various types of exchange interaction energies that exist in the material (or in the theoretical model). Such minimization procedure can be frustrated when there are several equal-energy
configurations. The most famous example is the Ising model on the triangular lattice, with antiferromagnetic nearest neighbor interactions \[16\]. In Fig. 1.2, one of the bonds on the triangle has minimized its energy by having two anti-parallel spins; however, the other two bonds cannot be simultaneously satisfied, no matter how we arrange the third spin. As a result, the ground state of the antiferromagnetic Ising triangular lattice has a macroscopic degeneracy, with an extensive entropy \( \approx 0.3k_B N \) \[16\].

Figure 1.2 : Frustration on antiferromagnetic Ising triangle.

Such geometric frustration can also be realized by competing interactions on a square lattice, with antiferromagnetic nearest neighbor exchange \( J_1 \) and next nearest neighbor \( J_2 \) (see Fig. 1.3). The Néel order (all nearest neighbor spins being anti-parallel) is the preferred ground when there is only \( J_1 \) present. However, the diagonal bonds are frustrated in this case, since the parallel spins on the diagonal get an energy penalty from \( J_2 \). In fact, for the Fe-based superconductors \[17\], \( J_2 \) is relatively large \[18\], in which case the collinear (columnar) antiferromagnetic order is realized \[19\] (see Fig. 1.4).

We can characterize the strength of magnetic frustrations by introducing an empirical parameter \( f = |\Theta_{CW}|/T_c \) \[20, 21\], where \( T_c \) is the magnetic ordering temperature and \( \Theta_{CW} \) is the Curie-Weiss temperature, defined through the magnetic susceptibility
Figure 1.3: Competing Heisenberg interactions (first and second nearest neighbor) on square lattice. The arrows show the Néel order when $J_2/J_1$ is relatively small.

Figure 1.4: Colinear antiferromagnetic order on square lattice, which is realized when $J_2/J_1$ is relatively large.
at high temperature \([22]\):

\[
\chi \approx \frac{C}{T - \Theta_{CW}}. 
\] (1.11)

Typically, a positive (negative) \(\Theta_{CW}\) corresponds to ferromagnetic (antiferromagnetic) ordering tendency. If \(f \gg 1\), then there is a large window \(T_c < T < |\Theta_{CW}|\), where magnetic moments are not ordered (paramagnetic), while short-range order has developed (ordering tendency). The magnets in such a window are defined as the “spin liquid” (cooperative paramagnet), in analogy to the normal liquid \([21]\).

In the extreme case, the magnetic moments do not order even at \(T = 0\), leading to the exotic phase “quantum spin liquid” \([21, 23, 24]\). There are known theoretical examples of quantum spin liquids in the Kitaev model on honeycomb lattice \([25]\), and in the spin-\(\frac{1}{2}\) Heisenberg Model on Kagome lattice \([26]\). Experimentally, neutron scattering experiments on herbertsmithite \(\text{ZnCu}_3(\text{OD})_6\text{Cl}_2\) found a continuum of spin excitations, indicative of the fractionalized excitations characteristic of the quantum spin liquids \([27]\).

Note that the quantum spin liquid is not the only fate of degeneracy. Indeed, there are only very few examples which are known to be spin liquids. And in fact, in the seminal papers by Villain \([28, 29]\), he showed that such degeneracy can actually be lifted due to thermal and/or quantum fluctuations, leading to an ordered ground state. More interestingly, the symmetry breaking of the degeneracy (most of the time corresponding to a discrete symmetry) can be separated from the continuous spin-rotational symmetry breaking, giving two phase transitions at different temperatures \([28, 29]\). Such a situation is known to be realized in the \(J_1 - J_2\) Heisenberg model on square lattice, where there are two degenerate ground states in the large \(J_2/J_1\) case, corresponding to the antiferromagnetic ordering tendency with the ordering wave-vectors \((\pi, 0)\) and \((0, \pi)\) respectively. Quantum fluctuations can lift such
degeneracy, leading to either \((\pi, 0)\) or \((0, \pi)\) magnetic phase \([19]\), as observed in most Fe-based superconductors \([30]\). Furthermore, both theoretically and experimentally, the separation of the discrete symmetry breaking (Ising nematic transition) and continuous symmetry breaking (magnetic transition) is indeed observed in the iron-based family of superconductors \([19, 30]\). Detailed description of the nematic transition in these compounds is the subject of Chapter 5 of this thesis.

1.3 Description of Magnetic States

1.3.1 Spin Structures

In an analogue of water, we can classify magnets also into three states:

- Gas (fully disordered paramagnets);
- Liquid (spin liquid, paramagnets with short-range correlations);
- Solid (magnetically ordered states with long-range correlations).

Now we should ask the question: How do we identity these different types of states?

Theoretically, the different magnetic states are described by their static spin structure factor:

\[
S(q) = \langle S_q \cdot S_{-q} \rangle = \frac{1}{N} \sum_{i,j} e^{i q \cdot (r_i - r_j)} \langle S_i \cdot S_j \rangle,
\]

where \(N\) is total number of lattice sites.

For the fully disordered paramagnets (gases), since there is no correlation between spins, \(S(q)\) is totally flat.
In the simplest example of a ferromagnetically ordered state, since all the spins are parallel, Eq. (1.12) yields

\[ S(q)_{FM} = \frac{1}{N} \sum_{i,j} e^{iq \cdot (r_i - r_j)} = N\delta_{q,0}. \]  

(1.13)

More generally, all the magnetically ordered states (solids) are signatured by Bragg peaks in \( S(q) \). And depending on the number of Bragg peaks in the first Brillouin zone, we classify these states as single-\( Q \) (one Bragg peak) or multi-\( Q \) (more than one Bragg peaks) states. In Chapter 2, we will see that multi-\( Q \) ordering can give rise to exotic vortex structure, even with three-dimensional modulation when these ordering wavevectors are non-coplanar.

The structure factor \( S(q) \) in the spin liquid states has no Bragg peaks, since there is no long range correlation. However, since short range order has started developing, \( S(q) \) is not featureless. As we see from Chapter 3, even slightly above the ordering temperature, there are still dispersive feature in the spin structure factor. And the broadening of the Bragg peaks typically corresponds to \( 1/\lambda \), where \( \lambda \) is the spin correlation length.

Note that the reverse is not always true: if \( S(q) \) has no Bragg peaks, we should not jump to the conclusion that it is a spin liquid. In fact, there are non-magnetic states, which has no magnetization \( \langle S_i \rangle = 0 \), but still breaks some type of continuous symmetry. We shall see in Chapter 4 that, in the spin multipolar ordered states, spin rotational symmetry can be spontaneously broken, while no Bragg peaks in the spin (dipolar) structure factor \( S(q) \) are found.
1.3.2 Spin Excitations

Besides the ground state property, the magnetic states are also characterized by their dynamic properties.

Assuming the ground state of the system is $|\Psi\rangle_0$, then the simplest excitation is the single spin flip (single-magnon):

$$|\Psi\rangle_{\text{ex}} = \sum_i \alpha_i S_i^+ |\Psi\rangle_0,$$

where $\alpha_i$ is the linear combination coefficients for the magnon excitation.

Since such single-magnon excitation is essentially a bosonic degree of freedom, Matsubara and Matsuda introduced the exact mapping from spin-$\frac{1}{2}$ operators to hard-core bosons \[31\]:

$$S_i^+ = b_i, \quad S_i^- = b_i^\dagger, \quad S_i^z = \frac{1}{2} - b_i^\dagger b_i.$$  \[1.15\]

In such a description, the spin-$\frac{1}{2}$ Heisenberg Model can be described by dispersive single-magnon excitations with density-density interactions.

Further generalization of spin-$S$ Hamiltonian can be realized by the Holstein-Primakoff bosons \[32\]:

$$S_i^+ = \left(\sqrt{2S - b_i^\dagger b_i}\right) b_i, \quad S_i^- = b_i^\dagger \left(\sqrt{2S - b_i^\dagger b_i}\right), \quad S_i^z = S - b_i^\dagger b_i.$$  \[1.16\]

To describe the excitations, we can expand the square root in the Holstein-Primakoff representation:

$$\sqrt{2S - b_i^\dagger b_i} = \sqrt{2S} \left(1 - \frac{b_i^\dagger b_i}{4S} - \frac{b_i^\dagger b_i b_i^\dagger b_i}{32S^2} \ldots\right),$$  \[1.17\]

which describes the spin fluctuations around the $z$-direction. In such expansion the corresponding Hamiltonian up to quadratic order ("spin wave" Hamiltonian) describes the spin excitations in the large-$S$ limit. Then in such magnon language,
the magnetic ordering can be described by the Bose-Einstein Condensation (BEC) of single-magnons \[33\].

There are also other types of exact mappings of the spin operators, for example the Schwinger bosons \[5\]:

\[
S_i^+ = a_i^\dagger b_i, \quad S_i^- = b_i^\dagger a_i, \quad S_i^z = \frac{1}{2}(a_i^\dagger a_i - b_i^\dagger b_i),
\]

subject to the constraint

\[
a_i^\dagger a_i + b_i^\dagger b_i = 2S.
\]

Similarly, the Abrikosov fermions describe the fermionic excitations in the magnets \[23\]:

\[
S_i = \frac{1}{2} f_{i\alpha}^\dagger \sigma_{\alpha\beta} f_{i\beta},
\]

subject to the constraint

\[
\sum_\alpha f_{i\alpha}^\dagger f_{i\alpha} = 1.
\]

In real magnetic systems, there could be other low-lying excitations besides single-magnons. In spins liquids, the single-magnon excitations are gapped out, and indeed Schwinger bosons or Abrikosov fermions are more natural descriptions \[23\]. A more generalized version of representation is the supersymmetry, where spin operators are represented by both bosons and fermions \[34\].

Moreover, when the effective interactions among single-magnons are attractive, then the low-lying excitations can instead be bound states of multi-magnons. Especially, when multi-magnon bound states go through BEC, the system will exhibit exotic orderings, for example the spin multipolar order \[35\].

### 1.4 Outline of the Thesis

This thesis is organized in the following way:
In Chapter 2, we describe the magnetic vortex crystal which can be realized as a ground state in highly frustrated quantum magnets. In the language of hard-core bosons, the magnetic ordering is described by the BEC of single-magnons. Further, the ground state degeneracy in the multi-minimum single-magnon manifold is lifted by quantum fluctuations, determining the unique ground state it belongs to.

In Chapter 3, we focus on the paramagnetic phase close to the collinear antiferromagnetic ordering. The spin dynamics is studied using the Dyson-Maleev and Schwinger boson mean-field calculations. By comparing the theoretical predictions with the inelastic neutron scattering experiments, we found that higher order bi-quadratic spin exchanges are essential for the understanding of Fe-based superconductors.

In Chapter 4, the “non-magnetic” phase in the Fe-based superconductor FeSe is studied within the same model as in Chapter 3. The key difference is that, in a certain parameter regime, we found that this model hosts an exotic phase which is magnetically disordered but with long range quadrupolar order. Spin dynamics in such a quadrupolar phase is studied by the flavor wave theory (which is a generalization of the Schwinger boson method, see Chapter 4 for more details), and our theoretical results are found to be consistent results with the inelastic neutron scattering experiments.

In Chapter 5, we turn to the more realistic model for Fe-based superconductors, which includes both magnetic and orbital degrees of freedom. By using both analytical (random phase approximation) and numerical (variational cluster approximation) approaches, we found that the Ising nematic transition in Fe-based superconductors can be naturally understood as the decoupling of the orbital and magnetic transitions.
Chapter 2

Three Dimensional Crystallization of Vortex Strings in Frustrated Quantum Magnets

2.1 Introduction

Topological spin structures of great potential interest in future applications of spin-electronic techniques [36]. The skyrmion crystals discovered in the $B20$-structure metallic alloys MnSi and Fe$_{1-x}$Co$_x$ [37, 38, 39] and in the Mott insulator Cu$_2$OSeO$_3$ [40, 41] are prominent examples. While the emergence of crystals of topological structures is reminiscent of the Abrikosov vortex lattice of type-II superconductors [42, 43], their origin is completely different in magnets. The basic difference is that magnetic systems are neutral Bose gases [33], while the charged Cooper pairs are coupled to the electromagnetic gauge field. In other words, the orbital coupling to an external field that stabilizes the Abrikosov vortex crystal in type-II superconductors is basically absent in magnets.

Topological spin structures must then be stabilized by other means. A key aspect of magnetic systems is that competing interactions are ubiquitous. A common outcome of this competition is a magnetic susceptibility that is maximized by several low-symmetry wave vectors $Q$ connected by point-group transformations of the underlying material. Topological spin structures can emerge when the effective interaction between the different $Q$ modes favors a multi-$Q$ ordering. This is the case of the $B20$ materials, in which the Dzyaloshinskii-Moriya [14, 15] interaction $D$ that
arises from their noncentrosymmetric nature shifts the susceptibility maximum from \( Q = 0 \) favored by the ferromagnetic exchange \( J \) to a finite vector \( |Q| \sim D/J \) that can have different orientations due to the cubic symmetry of the B20 structure. Thermal fluctuations then play an important role for stabilizing the \( 6\cdot Q \) structure that leads to the hexagonal skyrmion crystals in bulk versions of the B20 materials \[37\]. In contrast, the phase is already stable at the mean field level in 2D thin films \[40\]. In addition to chiral magnets, skyrmion crystals \[44\], soliton crystals \[45\] \[46\], and \( Z_2 \) vortex crystals \[47\] have been theoretically predicted in other classical spin systems. All of these examples correspond to 2D crystals of topological structures, i.e., they are not modulated along the third dimension.

More recently, it was proposed the realization of magnetic vortex crystals in a quantum spin system of weakly coupled triangular layers near a magnetic field-induced quantum critical point (QCP) \[48\]. The basic idea is to use geometric frustration as the source of competing interactions and quantum fluctuations to stabilize the multi-\( Q \) vortex crystal states. This study focuses on a case with six degenerate coplanar \( Q \) vectors that are connected by the \( C_6 \) symmetry transformations of the underlying lattice. Consequently, as in the previous examples, the resulting vortex crystal is not modulated along the third direction.

In this chapter, we demonstrate that a similar mechanism can also stabilize exotic 3D crystals of vortex lines. Unlike the case of the 2D vortex crystals, we are unaware of alternative realizations of 3D vortex crystals. As we explained above, the observation of magnetic skyrmion lattices unveiled the relevance of multi-\( Q \) orderings that produce 2D crystals of topological structures. However, much less effort has been devoted to the 3D crystals that can also arise from multi-\( Q \) orderings. The recent real-space observation of a skyrmion-antiskyrmion cubic lattice in MnGe \[49\] con-
firms the physical relevance of these 3D structures. The key to realize 3D crystals of topological objects is to find regions of stability of multiple noncoplanar-$Q$ orderings. Consequently, we study the body-centered cubi (bcc) and face-centered cubic (fcc) lattices that commonly occur in nature (typical examples are the transition-metal oxides and fluorides \[8\], solid $^3$He \[50\], 3D Wigner crystals \[51\], and the alkali-metal fulleride Cs$_3$C$_{60}$ \[52, 53\]). By extending the exchange interactions up to third nearest neighbors, we produce a single-magnon dispersion with multiple degenerate minima at noncoplanar $Q$ vectors connected by the cubic point group. We compute the optimal single-particle state for condensing the magnons and find that several multi-$Q$ states corresponding to different vortex crystals span sizable regions of the phase diagrams with isotropic exchange. These phases are further stabilized by symmetric exchange anisotropy that arises from, e.g., dipole-dipole interactions or spin-orbit coupling. The resulting spin textures consist of exotic 3D patterns of vortex strings.

2.2 Model

We consider a spin-$\frac{1}{2}$ Heisenberg model on bcc and fcc lattices coupled to a magnetic field:

$$\mathcal{H} = \sum_{\langle ij \rangle} J_{ij} S_i \cdot S_j - \sum_i H \cdot S_i,$$

(2.1)

where $J_{ij}$ are the Heisenberg interactions up to 3rd nearest neighbor $\{J_1, J_2, J_3\}$. In this study, we focus on the external field $H$ applied along the high symmetry [111] direction. The spin-$\frac{1}{2}$ operators can be represented by hard-core bosons \[31\]:

$S^+_i = b_i$, $S^-_i = b^\dagger_i$, $S^z_i = \frac{1}{2} - b^\dagger_i b_i$, where the z axis is along the magnetic field direction. The Hamiltonian is thus transformed into a model for an interacting Bose
gas:

\[ \mathcal{H} = \frac{J_1}{2} \sum_{(ij)} (b_i^\dagger b_j + b_j^\dagger b_i) + \frac{J_2}{2} \sum_{\langle \langle ij \rangle \rangle} (b_i^\dagger b_j + b_j^\dagger b_i) + \frac{J_3}{2} \sum_{\langle \langle \langle ij \rangle \rangle \rangle} (b_i^\dagger b_j + b_j^\dagger b_i) + \frac{U}{2} \sum_i n_i(n_i - 1) + J_1 \sum_{(ij)} n_i n_j + J_2 \sum_{\langle \langle ij \rangle \rangle} n_i n_j + J_3 \sum_{\langle \langle \langle ij \rangle \rangle \rangle} n_i n_j - \left( \frac{z_1 J_1 + z_2 J_2 + z_3 J_3}{2} - H \right) \sum_i n_i \]  

(2.2)

where \( U \) is the on-site hard-core repulsion, which is sent to infinity in the calculation, and \( z_1, z_2, z_3 \) are the coordination numbers of the 1st, 2nd and 3rd nearest neighbors.

Figure 2.1 : The Heisenberg interactions \( J_1, J_2, J_3 \) are defined on the 1st, 2nd and 3rd nearest neighbors. (a) bcc lattice. (b) fcc lattice.

By Fourier transformation \( b_i^\dagger = \frac{1}{\sqrt{N}} \sum_k e^{-i\mathbf{k} \cdot \mathbf{r}_i} b_k^\dagger \), the Hamiltonian is written down in \( \mathbf{k} \)-space:

\[ \mathcal{H} = \sum_k [\epsilon(k) - \epsilon(0) + H] b_k^\dagger b_k + \frac{1}{2N} \sum_{k,k',q} (U + V_q) b_{k+q}^\dagger b_{k-q}^\dagger b_k b_k, \]  

(2.3)

where

\[ \epsilon(k) = \frac{J_1}{2} \sum_{n_1} e^{i\mathbf{k} \cdot \mathbf{r}_{n_1}} + \frac{J_2}{2} \sum_{n_2} e^{i\mathbf{k} \cdot \mathbf{r}_{n_2}} + \frac{J_3}{2} \sum_{n_3} e^{i\mathbf{k} \cdot \mathbf{r}_{n_3}}, \]  

(2.4)

here \( \mathbf{r}_n \) denote the positions of the neighboring sites. And

\[ V_q = 2\epsilon(q). \]  

(2.5)
To be explicit, for bcc lattice:

\[ \epsilon(k) = 4J_1 \cos \frac{k_x}{2} \cos \frac{k_y}{2} \cos \frac{k_z}{2} + J_2 \left( \cos k_x + \cos k_y + \cos k_z \right) \]

\[ + 2J_3 \left( \cos k_x \cos k_y + \cos k_y \cos k_z + \cos k_z \cos k_x \right). \] (2.6)

For fcc lattice:

\[ \epsilon(k) = 2J_1 \left( \cos \frac{k_x}{2} \cos \frac{k_y}{2} + \cos \frac{k_y}{2} \cos \frac{k_z}{2} + \cos \frac{k_z}{2} \cos \frac{k_x}{2} \right) + J_2 \left( \cos k_x + \cos k_y + \cos k_z \right) \]

\[ + 4J_3 \left( \cos k_x \cos \frac{k_y}{2} \cos \frac{k_z}{2} + \cos k_y \cos \frac{k_z}{2} \cos \frac{k_x}{2} + \cos k_z \cos \frac{k_x}{2} \cos \frac{k_y}{2} \right). \] (2.7)

We define the minimum value of \( \epsilon(k) \) to be \( \epsilon_{\text{min}} \), in this way \( \omega_k \equiv \epsilon(k) - \epsilon_{\text{min}} \) has minimum value equals to zero. The Hamiltonian is rewritten as:

\[ H = \sum_k (\omega_k - \mu) b_k^\dagger b_k + \frac{1}{2N} \sum_{k,k',q} (U + V_q) b_{k+q}^\dagger b_{k'-q} b_k^\dagger b_k, \] (2.8)

where \( \omega_k \) is the single-boson (magnon) dispersion, the chemical potential:

\[ \mu = [\epsilon(0) - \epsilon_{\text{min}}] - H \equiv H_{\text{sat}} - H. \] (2.9)

Because of the frustration, the single magnon dispersion \( \omega_k \) can have multiple degenerate minima at different \( Q \)-vectors. In Fig. 2.2, we compute the number of minima in \( \omega_k \), for both bcc and fcc lattices.

The relative strengths of \( J_1 \), \( J_2 \), and \( J_3 \) determine the number of degenerate minima in the single-magnon dispersion \( \omega_k \). Phases with six and eight minima exist in both the bcc and fcc lattices. A phase with twelve minima also exists in bcc lattice (see Fig. 2.2). For concreteness, we will focus on the region with six degenerate minima, whose positions are denoted by \( \pm Q_n = \pm Q \hat{e}_n \), where \( n = 1, 2, 3 \). The value of \( Q \) is given by \( \cos \frac{Q}{2} = -J_1/(J_2 + 4J_3) \) for the bcc lattice and \( \cos \frac{Q}{2} = -(J_1 + 2J_3)/(J_2 + 4J_3) \)
Figure 2.2 : The single-magnon phase diagrams, with each region denoted either by the number of minima (when there are multiple minima at incommensurate $Q$-vectors), or denoted by the positions of the $Q$-vectors (when $Q$ is commensurate). (a)(b) bcc lattice. (c)(d) fcc lattice.

for the fcc lattice. Correspondingly, the saturation field values are:

$$H_{\text{bcc}}^{\text{sat}} = \frac{2J_1^2}{J_2 + 4J_3} + 4J_1 + 2J_2 + 8J_3,$$  \hspace{1em} (2.10a)  

$$H_{\text{fcc}}^{\text{sat}} = \frac{2(J_1 + 2J_3)^2}{J_2 + 4J_3} + 4J_1 + 2J_2 + 16J_3.$$  \hspace{1em} (2.10b)  

2.3 Multi-Q Condensation and Effective Interactions

The single-magnon dispersion becomes gapless at $H = H_{\text{sat}}$ which signals the phase transition into a Bose-Einstein condensate \cite{54, 55, 56, 57, 58, 59, 60, 61, 48}. In the vicinity of this transition $|H| \lesssim |H_{\text{sat}}|$, the boson density is vanishingly small, and we can use Beliaev’s dilute boson approach \cite{62} to compute the effective boson-boson interactions in the long-wavelength limit. Because this is a controlled expansion
in the small lattice gas parameter (ratio between the scattering length and the average interparticle distance), the result is asymptotically exact in the dilute limit. The next step is to condense the bosons in the most general single-particle state, i.e., to replace the bosonic operators for each wave vector $Q$ by six complex amplitudes: $\langle b_{\pm Q} \rangle / \sqrt{N} = \sqrt{\rho_{\pm Q}} \exp(i\phi_{\pm Q})$. The total energy is the sum of the low-energy terms allowed by translation symmetry, i.e., density-density interactions between bosons in the same ($\Gamma_1$) and different ($\Gamma_2, \Gamma_3$) minima, as well as a $\Gamma_4$ vertex that scatters bosons between two pairs of opposite minima,

$$
E = \frac{\Gamma_1}{2} \sum_{n, \sigma = \pm} \rho_{\sigma n}^2 + \Gamma_2 \sum_n \rho_{Q_n} \rho_{-Q_n} + \Gamma_3 \sum_{n < m} \rho_{\sigma_1 Q_n} \rho_{\sigma_2 Q_m} \\
+ 2\Gamma_4 \sum_{n < m} \sqrt{\rho_{Q_n} \rho_{-Q_n} \rho_{Q_m} \rho_{-Q_m}} \cos (\Phi_n - \Phi_m) - \mu \rho,
$$

(2.11)

where $\rho = \sum_n (\rho_{Q_n} + \rho_{-Q_n})$ is the total boson density and $\Phi_n = \phi_{Q_n} + \phi_{-Q_n}$. The interaction vertices $\Gamma_1, \ldots, \Gamma_4$ are obtained by summing over the ladder diagrams at zero total frequency.

Figure 2.3: (a)-(f) Schematic momentum-space representations of the multi-$Q$ condensates near the field-induced QCP for the case of six degenerate minima. The arrows representing the phases $\Phi_n$ of the $Q_n$ component of the order parameter [see Eq. (2.11)] are only shown for states in which their relative values are fixed by the interactions or anisotropy. The gray (light) color indicates no correlation among the different phases $\Phi_n$. 
2.4 Phase Diagram Calculation

2.4.1 Solving Bethe-Salpeter Equation

The effective interactions in the dilute limit for hard-core bosons are calculated by the Bethe-Salpeter equation, which is equivalent to summing over all the ladder diagrams (Fig. 2.4).

\[
\Gamma_q(k, k') = U + V_q - \int \frac{d^3 q'}{V_{BZ}} \frac{\Gamma_q(k, k')(U + V_{q-q'})}{\omega_{k+q'} + \omega_{k'-q'}}
\]

(2.12)

where \(V_{BZ}\) is the volume of the 1st BZ.

![Ladder diagrams](image)

Figure 2.4 : Ladder diagrams.

When the magnetic field \(H\) is close to the saturation value \(H_{sat}\), the system is unstable towards BEC at the dispersion minima. In this case we can take the long wavelength limit \(k \rightarrow \pm Q_i\), and calculate the corresponding vertex functions (schematically shown in Fig. 2.5):

\[
\begin{align*}
\Gamma_1 &= \Gamma_0(Q_n, Q_n), \\
\Gamma_2 &= \Gamma_0(Q_n, -Q_n) + \Gamma_{-2Q_n}(Q_n, -Q_n), \\
\Gamma_3 &= \Gamma_0(Q_n, Q_m) + \Gamma_{Q_m-Q_n}(Q_n, Q_m), \\
\Gamma_4 &= \Gamma_{Q_m-Q_n}(Q_n, -Q_n) + \Gamma_{-Q_m-Q_n}(Q_n, -Q_n).
\end{align*}
\]

(2.13)
To solve the Bethe-Salpeter equation, we start from the following ansatz:

\[ \Gamma_q = \langle \Gamma \rangle + \sum_\eta A_\eta V(r_\eta)e^{i\mathbf{q} \cdot \mathbf{r}_\eta}, \]  

(2.14)

where \( r_\eta \) denotes the positions of the 1st, 2nd, and 3rd neighboring sites. The \( \mathbf{k}, \mathbf{k}' \) index in \( \Gamma_q(\mathbf{k}, \mathbf{k}') \) are omitted for simplicity, and \( \langle \Gamma \rangle = \int \frac{d^3q}{V_{\text{BZ}}} \Gamma_q' \). We also assume that \( V_q \) is centro-symmetric, i.e.

\[ \int d^3q V(q) = 0. \]  

(2.15)

By substituting the ansatz into the Bethe-Salpeter equation and taking the hard-core limit, we get the following form of linear equations:

\[ \sum_\eta V(r_\eta)(\tau_1^\eta)^* A_\eta + \tau_0 \langle \Gamma \rangle = 1, \]  

(2.16a)

\[ \sum_\nu (\tau_2^{\eta \nu} V(r_\nu) + \delta_{\eta \nu}) A_\nu + \tau_1^\eta \langle \Gamma \rangle = 1. \]  

(2.16b)

where the integrals are defined as:

\[ \tau_0 = \int \frac{d^3q}{V_{\text{BZ}}} \frac{1}{\omega_{\mathbf{k}+q} + \omega_{\mathbf{k}'-q}}, \]  

(2.17a)

\[ \tau_1^\eta = \int \frac{d^3q}{V_{\text{BZ}}} e^{-i\mathbf{q} \cdot \mathbf{r}_\eta} \frac{1}{\omega_{\mathbf{k}+q} + \omega_{\mathbf{k}'-q}}, \]  

(2.17b)

\[ \tau_2^{\eta \nu} = \int \frac{d^3q}{V_{\text{BZ}}} e^{-i\mathbf{q} \cdot (\mathbf{r}_\eta - \mathbf{r}_\nu)} \frac{1}{\omega_{\mathbf{k}+q} + \omega_{\mathbf{k}'-q}}. \]  

(2.17c)
Denote:

\[ B_{\eta\nu} = \tau^\eta_2 V(r_{\nu}) + \delta_{\eta\nu}, \quad (2.18a) \]

\[ C_{\eta} = V(r_{\eta})\tau^\eta_1. \quad (2.18b) \]

The above equations are now organized into a matrix form:

\[
\begin{pmatrix}
B_{11} & \cdots & B_{1z} & \tau_1^1 \\
\vdots & \ddots & \vdots & \vdots \\
B_{z1} & \cdots & B_{zz} & \tau_z^z \\
C_1 & \cdots & C_z & \tau_0
\end{pmatrix}
\begin{pmatrix}
A_1 \\
\vdots \\
A_z
\end{pmatrix}
= 
\begin{pmatrix}
1 \\
\vdots \\
1
\end{pmatrix}, \quad (2.19)
\]

By solving the linear equations Eq. (2.19), we obtain all the unknown coefficients in the ansatz Eq. (2.14). Then we can substitute the values of \( \Gamma_1, \ldots, \Gamma_4 \) into the expression of effective energy, and determine which multi-\( Q \) state will be stabilized.

2.4.2 Effect of Anisotropy

In real materials, apart from the exchange interactions, there are also always exchange anisotropy present, which becomes important in certain cases (see discussion later). For simplicity, we consider short-range symmetric exchange anisotropy (cutoff at 2nd nearest neighbor):

\[
\mathcal{H}_A \propto \sum_{\langle ij \rangle} -3(S_i \cdot r_{ij})(S_j \cdot r_{ij}) \quad (2.20)
\]

such terms can arise directly from dipole-dipole interactions, or perburbatively from spin-orbit coupling\[^{15}\].

Similarly to the treatment of the Heisenberg exchange interactions, we choose the quantization axis along \([111]\) direction, and represent the spin-\( \frac{1}{2} \) operators with hard-
core bosons. In the long-wavelength limit, for both bcc and fcc lattices:

$$\mathcal{H}_A \propto \left[ (\sqrt{3}/2 + i \frac{1}{2}) b_{Q_1}^\dagger b_{-Q_1}^\dagger + (-\sqrt{3}/2 + i \frac{1}{2}) b_{Q_2}^\dagger b_{-Q_2}^\dagger - i b_{Q_3}^\dagger b_{-Q_3}^\dagger \right] + h.c \quad (2.21)$$

Then we condense the bosons by

$$\langle b_{\pm Q_n} \rangle / \sqrt{N} = \sqrt{\rho_{\pm Q_n}} \exp \left( i \phi_{\pm Q_n} \right),$$

which gives the energy correction of symmetric exchange anisotropy:

$$E_A \propto J_A \sum_n \sqrt{\rho_{Q_n} \rho_{-Q_n}} \cos(\Phi_n + 2n\pi/3 - \pi/2). \quad (2.22)$$

where $\Phi_n = \phi_{Q_n} + \phi_{-Q_n}$.

### 2.4.3 Phase Diagram

The zero-temperature phase diagram is determined by minimizing the total energy $E$ given in Eq. (2.11) \[63\]. Depending on the relative strengths of exchange interactions, one of the six possibilities in Fig. 2.3 is realized. Out of these, three condensates in particular realize vortex crystals: 3-**Q** I, 4-**Q** I, and 6-**Q** II (Fig. 2.3). Another reason for considering these states is that the latter two are further stabilized by symmetric exchange anisotropy originated from spin-orbit coupling or dipole-dipole interactions. Close to the saturation field $H_{\text{sat}}$, this exchange anisotropy yields the interaction term Eq. (2.22): Although $J_A$ is typically small, $E_A \sim |\rho|$ is linear in the boson density. Consequently, in the dilute limit ($\rho \ll |J_A/J_1| \ll 1$), it always dominates over the exchange interaction in Eq. (2.11). The calculations show that the 2-**Q** I, 4-**Q** I, and 6-**Q** II condensates are the three lowest energy states, degenerate to linear order in the density $\rho$. The degeneracy is lifted by further considering the second-order density-density interactions in Eq. (2.11), stabilizing the 6-**Q** II state over a wide range of parameters on both bcc and fcc lattices (see Fig. 2.6). On the other hand, sufficiently far away from the QCP (but still in the low density regime, $|J_A/J_1| \ll \rho \ll 1$), $E_A$ is negligible and Eq. (2.11) alone determines the ground state
configuration. The resulting phase diagrams for negligible (dominant) anisotropy are shown in the left (right) column of Fig. 2.6, respectively.

Figure 2.6: Phase diagrams of the Heisenberg model (2.1) under the nearly saturated magnetic field, where $\omega_k$ has six degenerate minima in the colored phases. (a) bcc lattice, $J_1 > 0$, no anisotropy. “NAF” denotes the case with $Q = (2\pi, 2\pi, 2\pi)$. (b) bcc lattice, $J_1 > 0$, anisotropy dominating region. (c) bcc lattice, $J_1 < 0$, without anisotropy. “Ferro” denotes $Q$ at $(0, 0, 0)$, and “PS” denotes regions where we have phase separation or bound states. (d) bcc lattice, $J_1 < 0$, anisotropy dominating region. (e) fcc lattice, $J_1 > 0$, no anisotropy. (f) fcc lattice, anisotropy dominating region.

2.5 Crystallization of Vortex Strings

We now focus on the 3-$Q$ I, 4-$Q$ I, and 6-$Q$ II states that realize vortex crystals whose spin structures on [111] layers are illustrated in Fig. 2.7. We find that the vortex
and antivortex cores form regular lattices on every layer. Vortices and antivortices correspond to different signs $\kappa = \pm 1$ of the vector spin chirality $S_i \times S_j$ when we circulate $(i \rightarrow j)$ around the vortex core. Explicitly, in the vicinity of the (anti)vortex core $\arctan(S_i^y/S_i^x) = \arg\langle b_1^\dagger \rangle \sim \kappa \varphi + \gamma + \delta(\varphi)$, where $\gamma$ is the helicity [64] and $\delta(\varphi)$ is a $2\pi$-periodic function of the polar angle $\varphi$ around the (anti)vortex core in the [111] plane such that $\int_0^{2\pi} d\varphi \delta(\varphi) = 0$ and $|\delta(\varphi)| \ll 2\pi$. For a given chirality, the (anti)vortices can have different relative helicities, as we show in Fig. 2.7; the 3-\textit{Q} I state includes three types of vortices with helicities that differ by $2\pi/3$ in each [111] plane (the same is also true for the antivortices), see Fig. 2.7a; similarly, two types of (anti)vortices appear in the 4-\textit{Q} I state with helicities that differ by $\pi$ [Fig. 2.7b]; the 6-\textit{Q} II state contains three types of vortices with helicities that differ by $2\pi/3$ and two types of antivortices with helicities that differ by $\pi$ [Fig. 2.7c].

The vortex cores are strings that extend along the third dimension. These strings form different patterns for each multi-\textit{Q} condensate. The vortex and antivortex strings form parallel straight lines along the [111] direction in the 3-\textit{Q} I and 4-\textit{Q} I states. The same is true for the antivortices of the 6-\textit{Q} II states. However, the vortex strings form a more exotic pattern in the 6-\textit{Q} II state. As is shown in Fig. 2.8, the vortex strings cross each other and the helicities of the crossing vortices and antivortices are shifted by $\pi$. This unusual behavior arises from the fact that the six \textit{Q} vectors are noncoplanar. In contrast, when the condensate \textit{Q} vectors are on the same plane in the reciprocal space, as is the case with the 3-\textit{Q} I and 4-\textit{Q} I states, and those considered in Ref. [48], the vortex strings are straight lines along the high-symmetry axis. The helicity of each (anti)vortex increases linearly in the layer index for the 3-\textit{Q} I state. In contrast, the helicity of each (anti)vortex is shifted by $\pi$ between consecutive layers of the 4-\textit{Q} I state. This alternation arises from the
Figure 2.7: Spin structures in the vortex crystal phases on [111] layers ($\mathbf{H} \parallel [111]$); only the $xy$ components are plotted. The color intensity denotes the boson density $\rho$ (the transverse spin components are $\sim \sqrt{\rho}$) and the bright spots denote fully polarized spins. The circles with (without) crosses denote vortex (antivortex) cores. Different colors of the circles denote different helicities $\gamma$ (see text). (a) The 3-$\mathbf{Q}$ I state on the bcc lattice for $|Q_n| \ll 1$. (b) The 4-$\mathbf{Q}$ I state on the bcc lattice for $|Q_n| \lesssim 2\pi$. (c) The 6-$\mathbf{Q}$ II state on the bcc lattice for $|Q_n| \ll 1$. 
fact that $|Q_n| \lesssim 2\pi$ and each [111] layer belongs to a sublattice of the bcc lattice. In the 6-$Q$ II state, the helicity of each (anti)vortex is constant except for crossing points where it is shifted by $\pi$.

### 2.6 Conclusion and Discussion

While in this chapter we only focus on the regions where $\omega_k$ has six degenerate minima, there are other regions in the phase diagrams where vortex crystals should also arise. For example, $\omega_k$ with eight minima is realized in a region next to the 6-minimum region on both the bcc and fcc lattices, and a 12-minimum case also occurs on the bcc lattice. A similar calculation can be applied to these cases in order to obtain the stable multi-$Q$ orderings.

We note that when some of the effective interactions are attractive (typically the case with ferromagnetic exchanges), the system may undergo a first order phase transition at the saturation field $[65, 61]$. This implies that it is unstable towards phase separation if one fixes the particle number (i.e., the $z$ component of magnetization) in the canonical ensemble. An alternative scenario is a continuous transition associated with the condensation of multimagnon bound states $[65]$. We have verified that none of these cases takes place for antiferromagnetic nearest neighbor interactions ($J_1 > 0$) in both the bcc and fcc lattices. For ferromagnetic nearest neighbor interactions ($J_1 < 0$), on the other hand, the phase separation occurs in a large region of the phase diagram, as indicated in Figs. 2.6(c) and 2.6(d).

Although crossings of vortex lines have been observed in superconducting vortex glasses and liquids $[66, 67]$, we are unaware of the existence of 3D vortex crystals like the one shown in Fig. 2.8. These vortex crystals can be detected with neutron diffraction in single-domain samples. Materials with more than two degenerate min-
Figure 2.8: (a)-(d) The sequence of layers along the [111] direction for the 6-$Q$ II state on the bcc lattice in the case $|Q_n| \ll 1$. Several intervening layers between (a) and (b), etc., are omitted for simplicity. The arrows indicate how the vortex cores (circles with crosses) move from one layer to the next layer above. The lines of the antivortex cores (circles without crosses) are parallel to the [111] direction. The region enclosed by a square follows one of the antivortex core. The $\pi$ helicity shift occurs below the layer (c). (e) Three-dimensional picture of the vortex string structure. The strings corresponding to antivortices in (b)-(d) are not shown.
ima in the magnon dispersion of the fully saturated state can be identified directly by measuring the inelastic neutron scattering spectrum at \(|H| > |H_{\text{sat}}|\), or indirectly, by extracting the exchange constants from the zero-field inelastic neutron scattering spectrum. Nuclear magnetic resonance (NMR) also allows us to distinguish among different multi-\(Q\) orderings because the NMR line shape is in general qualitatively different for single-, double-, and three-\(Q\) orderings.

Our results indicate that these materials are strong candidates to exhibit magnetic vortex crystals just below their saturation field. While here we have considered the particular cases of bcc and fcc lattices as examples, the general principle can be directly extended to other highly frustrated structures, such as hexagonal close-packed and pyrochlore lattices, which are also common in nature. Based on our calculations, it is expected that exchange anisotropy (due to e.g., dipolar interactions) will select a double-\(Q\) magnetic ordering or a magnetic vortex crystal. The selection mechanism between these two competing phases is provided by the effective interaction between magnons, which ultimately depends on the details of the exchange couplings. There are several candidate materials that comprise highly frustrated 3D lattices of rare-earth magnetic ions. Because the exchange anisotropy is expected to be stronger in these ions due to a large spin-orbit interaction, the vortex crystal phase could extend over a wider window of magnetic field values.
Chapter 3

Spin dynamics of a J1-J2-K model for the paramagnetic phase of iron pnictides

3.1 Introduction

The emergence of superconductivity in iron pnictides \cite{17, 68} near an antiferromagnetically ordered state \cite{69} in the phase diagram suggests strong interplay between the superconductivity and magnetism in these materials. Elucidating the magnetic excitations is therefore important for understanding not only the overall microscopic physics of these systems but also the mechanism of superconductivity. In the parent compounds, the observed \((\pi, 0)\) antiferromagnetic order arises either within a weak-coupling approach invoking a Fermi surface nesting,\cite{70, 71, 72} or from a strong-coupling approach whose starting point is a local moment \(J_1 - J_2 - K\) model.\cite{18, 73, 74, 75, 76, 77, 78, 79}

The strong-coupling approach is based on the proximity of the metallic ground state of the parent pnictides to a Mott localization transition, which gives rise to quasilocal magnetic moments.\cite{18, 77, 80, 81} This incipient Mott picture corresponds to a ratio of \(U\) (a measure of the Coulomb repulsions and Hund’s couplings among the Fe 3d electrons) to \(t\) (the characteristic bandwidth of the Fe 3d electrons) which is not too far below the Mott threshold \(U_c/t\), which is usually of order unity. This is supported by many experimental observations. For instance, the room-temperature electrical resistivity of parent iron pnictides is so large (even when the residual resistivity is rel-
atively small signaling the smallness of elastic scattering) that the extracted mean-free path of quasiparticles would be comparable to the Fermi wavelength; this is typical of bad metals near a Mott transition. Similarly, the Drude weight in optical conductivity \[82, 83\] is strongly suppressed from its non-interacting counterpart, providing a direct measure of the proximity to the Mott transition. This is further corroborated by the the temperature-induced spectral weight transfer,\[83, 84, 85\] which is also characteristic of metals near a Mott transition. In the spin sector, large frequency spin waves near the Brillouin zone boundary have been observed by inelastic neutron scattering (INS) measurements in the magnetically ordered state of several 122 iron pnictide compounds.\[86\] Both the large spectral weight and the relatively-small spin damping suggest quasi-localized moments, which are expected near the Mott transition where the spin excitations arise out of incoherent electronic excitations. Additional evidence for the incipient Mott transition picture has come from the observation of a Mott insulating phase in the iron oxychalcogenides.\[87\] This material contains an expanded Fe square lattice compared to the iron pnictides, which reduces \(t\), thereby enhancing \(U/t\) beyond \(U_c/t\) (Ref. \[87\]). Likewise, the Mott insulating behavior of the alkaline iron selenides \[88\] can also be interpreted as the result of a reduced effective \(t\) and, correspondingly, an enhanced \(U/t\) beyond \(U_c/t\).\[89, 90\]

In the vicinity of \(U_c/t\), where correlations are strong, it is natural that the spin Hamiltonian contains not only two-spin interactions, such as \(J_1\) and \(J_2\) Heisenberg exchange between nearest- and next-nearest- neighbor spins on a square lattice, but also interactions involving higher number of spins. These naturally include, for instance, the ring-exchange coupling involving four spins on a plaquette, and the biquadratic coupling of the form \(-K(S_i \cdot S_j)^2\) in systems with spin size \(S \geq 1\).\[11\] The subject of the present study is to show how such non-Heisenberg interactions, particularly the
biquadratic interaction, influence the spin dynamics in the paramagnetic phase.

Spin dynamics in the parent iron pnictides have been most extensively studied in the low-temperature state \((T < T_N)\) with both antiferromagnetic order and orthorhombic structural distortion. Here, the INS experiments up to high energies (on the order of 200 meV) show that the spin wave excitations in these compounds are highly anisotropic, with a dispersion which can be understood in terms of an anisotropic \(J_{1x} - J_{1y} - J_2\) model with \(J_{1x} \neq J_{1y}\).\(^{[86, 91, 92]}\) The presence of anisotropy in the nearest-neighbor coupling is consistent with the orthorhombic structure and orbital ordering \(^{[93, 94, 95, 96]}\). Detailed theoretical studies of the magnetic excitations in the ordered phase have been carried out in such a \(J_{1x} - J_{1y} - J_2\) model,\(^{[97]}\) and in a \(J_1 - J_2 - K\) model.\(^{[98, 99]}\) It should also be noted that terms such as the biquadratic coupling could be inferred from the sublattice angle dependence of the ground-state energy in LSDA calculations \(^{[100]}\), and were shown to appear naturally as a result of the orbital ordering between Fe \(d_{xz}\) and \(d_{yz}\) orbitals \(^{[96]}\).

The focus of the present study is instead on the spin dynamics in the paramagnetic phase of the parent iron pnictides, which has only recently been studied experimentally. The initial work by Diallo \textit{et al.}\(^{[101]}\) measured the spin dynamics of CaFe\(_2\)As\(_2\) at relatively low energies, below 70 meV. Theoretically, from the study of spin dynamics in the paramagnetic phase of the \(J_1 - J_2\) model (with or without an additional fermion damping) \(^{[102]}\), it was shown that the experimentally observed elliptical features of the spin spectral weight in momentum space are well-described by this model and we determined the change to the elliptical features at high energies.

More recently, Harriger \textit{et al.}\(^{[103]}\) reported measurements of the spin dynamics in the paramagnetic phase up to high energies (above 200 meV) in BaFe\(_2\)As\(_2\). The INS measurements confirmed the quasi-two-dimensional spin dynamics found at low
energies, and characterized the evolution of the low-energy elliptic features as they expand towards the zone boundary as the energy is raised, and determined the high-energy dispersion which appears to require a $J_{1x} \neq J_{1y}$ description even though the paramagnetic phase has a tetragonal structure. Similar data have also been reported by Ewings et al. in SrFe$_2$As$_2$. Theoretically, Park et al. analyzed the spin dynamics in the paramagnetic state within a dynamical mean-field theory (DMFT) for interactions $U/t \lesssim U_c/t$, demonstrating that the DMFT approach captures key features of the neutron scattering results, including the ellipticity of the map of the structure-factor peak in the Brillouin zone.

In this chapter, we study the spin dynamics of the $J_1 - J_2 - K$ model in the tetragonal paramagnetic phase using both modified spin wave (MSW) and Schwinger boson mean-field (SBMF) theories. The results from the two methods are in very good quantitative agreement with each other. We show that, for a moderate biquadratic coupling $K$, the dynamical structure factor $S(q, \omega)$ has not only elliptic features near $(\pi, 0)$, which expand with increasing energy and split into peaks surrounding $(\pi, \pi)$, but also an anisotropic distribution of the spectral weight that is larger along the major axis of each ellipse than along its minor axis. These properties agree well with the INS experiments.

The remainder of the chapter is organized as follows. In Sec. 3.2 we introduce the $J_1 - J_2 - K$ model and describe the MSW and SBMF theories used in this chapter. In Sec. 3.3 we show how the biquadratic coupling $K$ influences the mean-field phase diagram and magnetic excitation spectrum. In Sec. 3.4 we calculate the dynamical structure factor $S(q, \omega)$. We also show that the spectral weight exhibits anisotropic features, discuss the evolution of the anisotropic features with increasing excitation energy, and explain how these properties arise from our theory. In Sec. 3.5 we first
discuss some possible generalizations of the $J_1 - J_2 - K$ model we are studying in this
chapter. In the same section, we then consider the effect of itinerant electrons, and
compare our study with other theoretical approaches to the spin dynamics. Sec. 3.6
is devoted to a comparison with the INS experiments on the paramagnetic phases of
the parent 122 iron pnictides, and Sec 3.7 contains a few concluding remarks.

3.2 Model and Methods

The $J_1 - J_2 - K$ model for spin $S \geq 1$ is defined on a two-dimensional (2D) square
lattice with the following Hamiltonian:

$$H = J_1 \sum_{i,\delta} \mathbf{S}_i \cdot \mathbf{S}_{i+\delta} + J_2 \sum_{i,\delta'} \mathbf{S}_i \cdot \mathbf{S}_{i+\delta'} - K \sum_{i,\delta} (\mathbf{S}_i \cdot \mathbf{S}_{i+\delta})^2,$$

where $J_1$ and $J_2$ respectively denote the antiferromagnetic exchange couplings between
spins located in the nearest neighbor ($\delta = \hat{x}, \hat{y}$) and next-nearest neighbor ($\delta' = \hat{x} \pm \hat{y}$)
sites. $K$ is the coupling for the biquadratic interaction between the nearest neighbor
spin pairs.

To fully explain the experimentally observed $(\pi, 0, \pi)$ antiferromagnetic order, an
exchange coupling along the third dimension, $J_z$ should also be included. However, we
find the model defined in Eq. (3.1) already allows us to understand the experimentally
observed quasi-2D spin dynamics. Hence, we concentrate on this 2D model in the
main text.

The Hamiltonian of Eq. (3.1) is studied using both MSW[106, 107] and SBMF[5]
methods. Here, we focus on the parameter regime where the ground state has a
collinear $(\pi, 0)$ antiferromagnetic order, and decompose the biquadratic interaction
term of the Hamiltonian using two Hubbard-Stratonovich fields $\Gamma_{i,\hat{x}(\hat{y})}$. The effective
Hamiltonian reads as

\[ H = J_1 \sum_{i,\delta} S_i \cdot S_{i+\delta} + J_2 \sum_{i,\delta'} S_i \cdot S_{i+\delta'} - 2K \sum_{i,\delta} \Gamma_{i,\delta} S_i \cdot S_{i+\delta} + K \sum_{i,\delta} \Gamma_{i,\delta}^2 \]  

(3.2)

At the mean-field level, the Hubbard-Stratonovich fields are treated as static quantities, and can be expressed using equal-time spin correlators as: \( \Gamma_{i,\delta} = \langle S_i \cdot S_{i+\delta} \rangle \). The Hubbard-Stratonovich transformation itself is exact. The static approximation is made in accordance with the level of approximation inherent to the MSW and SBMF methods, which incorporate static self-energies for the respective boson fields. As shown below, our approach has two important features: i) it is capable of studying the Ising correlations at nonzero temperatures; and ii) the MSW and SBMF approaches yield consistent results.

### 3.2.1 The modified spin wave theory

The MSW theory \cite{106, 107} has been applied to the \( J_1 - J_2 \) model in Ref. \cite{102}. In this approach, a local spin quantization axis is defined at each site along the classical ordering direction \( \Omega_{cl}^i \). The Hamiltonian in Eq. (3.2) is then expressed in terms of Dyson-Maleev (DM) bosons via a local DM transformation:

\[ S_i \cdot \Omega_{cl}^i = S - a_i^\dagger a_i, \]

\[ S_i^+ = \sqrt{2S} (1 - a_i^\dagger a_i/2S)a_i, \]

and \( S_i^- = \sqrt{2S} a_i^\dagger \). Minimizing the free energy under the constraint of zero sublattice magnetization \( \langle S - a_i^\dagger a_i \rangle = 0 \) by introducing a Lagrange multiplier \( \mu \), and with respect to \( \Gamma_\delta \) (\( = \Gamma_{i,\delta} \), by assuming translational symmetry), we obtain

\[ \Gamma_x = \cos^2 \frac{\phi}{2} f_x^2 - \sin^2 \frac{\phi}{2} g_x^2, \]

\[ \Gamma_y = \sin^2 \frac{\phi}{2} f_y^2 - \cos^2 \frac{\phi}{2} g_y^2, \]

(3.3)

where \( \phi = \arccos(\Omega_{cl}^i \cdot \Omega_{cl,\delta}^i) \), \( f_\delta = \langle a_i^\dagger a_{i+\delta} \rangle \) and \( g_\delta = \langle a_i a_{i+\delta} \rangle \) are the ferromagnetic and antiferromagnetic bond operators, respectively. Minimizing the free energy with
respect to \( \phi \) gives either \( \sin \phi = 0 \) for nonzero \( f_\delta \) and \( g_\delta \), or \( \phi \) can be arbitrary if \( f_\delta = g_\delta = 0 \). This defines two phases separated by a mean-field temperature scale \( T_{\sigma_0} \): at \( T > T_{\sigma_0} \), \( \phi \) is arbitrary, and the system has \( C_{4v} \) lattice rotational symmetry; while for \( T < T_{\sigma_0} \), the \( C_{4v} \) symmetry is broken and the system is Ising ordered, corresponding to either \( \phi = 0 \) or \( \phi = \pi \). In MSW theory, the Ising order parameter can be defined as \( \sigma = 2(\cos^2 \frac{\phi}{2}(f_x^2 + g_y^2) - \sin^2 \frac{\phi}{2}(f_y^2 + g_x^2)) \). From Eq. (3.3), if we define \( \Gamma_\pm = (\Gamma_x \pm \Gamma_y)/2 \) as the symmetric and antisymmetric Hubbard-Stratonovich fields, we find that \( \Gamma_- = \sigma/4 \).

Minimizing the free energy with respect to other variational parameters, we obtain a set of self-consistent equations:

\[
f_\delta = m_0 + \frac{1}{N} \sum_{k} \frac{\varepsilon_k}{\varepsilon_k} \left( n_k + \frac{1}{2} \right) \cos(\mathbf{k} \cdot \mathbf{\delta}), \tag{3.4}
\]

\[
g_\delta = m_0 + \frac{1}{N} \sum_{k} \frac{\varepsilon_k}{\varepsilon_k} \left( n_k + \frac{1}{2} \right) \cos(\mathbf{k} \cdot \mathbf{\delta}'), \tag{3.5}
\]

\[
S + \frac{1}{2} = m_0 + \frac{1}{N} \sum_{k} \frac{\varepsilon_k}{\varepsilon_k} \left( n_k + \frac{1}{2} \right), \tag{3.6}
\]

where \( N \) is the total number of lattice sites, \( \mathbf{\delta} = \hat{x}, \hat{y}, \) and \( \mathbf{\delta}' = \hat{x}, \hat{y}, \hat{x} \pm \hat{y} \). In Eqs. (3.4)-(3.6),

\[
A_k = 2 \sin^2 \frac{\phi}{2} \tilde{J}_{1x} g_x \cos k_x + 2 \cos^2 \frac{\phi}{2} \tilde{J}_{1y} g_y \cos k_y \\
+ 4 J_2 g_{x+y} \cos k_x \cos k_y, \tag{3.7}
\]

\[
B_k = 4 J_2 g_{x+y} - \mu + 2 \sin^2 \frac{\phi}{2} (\tilde{J}_{1x} g_x - \tilde{J}_{1y} f_y(1 - \cos k_y)) \\
+ 2 \cos^2 \frac{\phi}{2} (\tilde{J}_{1y} g_y - \tilde{J}_{1x} f_x(1 - \cos k_x)), \tag{3.8}
\]

and the Bogoliubov angle \( \theta_k \) is defined via \( \tanh 2\theta_k = A_k/B_k \). The boson dispersion \( \varepsilon_k = \sqrt{B_k^2 - A_k^2} \) and the boson number \( n_k = [\exp(\varepsilon_k/k_B T) - 1]^{-1} \). At \( T = 0 \), the spectrum of the DM bosons becomes gapless at wave vector \( \mathbf{Q} \) and \( \mathbf{0} \). This corresponds to a long-range antiferromagnetic order at \( \mathbf{Q} \neq 0 \) with a nonzero spontaneous
magnetization \( m_0 \). In this case, the summation \( \sum_k' \) runs over all \( k \) values that make \( \varepsilon_k > 0 \), and the contribution from the \( \varepsilon_k = 0 \) terms is taken into account separately by \( m_0 \). For \( T > 0 \), \( m_0 = 0 \), and the system is paramagnetic. Here the summation is performed in the full momentum space. In the presence of a small third-dimension coupling \( J_z \), there will be a nonzero mean-field Néel temperature, \( T_{N0} \).

Note that these self-consistent equations are exactly the same as those for the isotropic \( J_1 - J_2 \) model. But the definitions of \( A_k \) and \( B_k \) are different. In Eqs. (3.7) and (3.8) above, we defined the effective exchange couplings \( \tilde{J}_{1x} \) (\( \tilde{J}_{1y} \)) along the \( x(y) \) direction as follows:

\[
\tilde{J}_{1x(y)} = J_1 - 2K\Gamma_{x(y)},
\]

expressed in terms of the Hubbard-Stratonovich fields \( \Gamma_{x(y)} \) of spin-spin correlators in Eq. (3.3). Although in the \( J_1 - J_2 - K \) model the bare nearest neighbor exchange coupling \( J_1 \) is still isotropic, a nonzero biquadratic coupling \( K \) leads to an anisotropic effective coupling \( \tilde{J}_{1x} \neq \tilde{J}_{1y} \) in the Ising ordered phase where \( \Gamma_x \neq \Gamma_y \), i.e. the nearest-neighbor spin correlators along \( x \) and \( y \) are unequal, similarly to the situation found originally\[19\] for the \( J_1 - J_2 \) model.

### 3.2.2 The Schwinger boson theory

In the Schwinger boson representation\[108\], the SU(2) spin operators are rewritten in terms of two Schwinger bosons via the transformation: 

\[
S_i^z = \frac{1}{2}(a_i a_i - b_i b_i), \quad S_i^+ = a_i b_i, \quad S_i^- = b_i a_i.
\]

To limit the boson Hilbert space to the physical sector, a constraint

\[
a_i a_i + b_i b_i = 2S
\]

is imposed on each site. This can be generalized to the case of either SU(\( N \))\[109\] or SP(\( N \))\[110, 111\] spins, in either case there will be \( N \) boson degrees of freedom at each site. For the experimentally observed \( \mathbf{Q} = (\pi, 0) \) or \( (0, \pi) \)
antiferromagnetic collinear phase in the 122 parent compounds, the \((ab)\)-plane spin-spin correlations are expressed as:

\[
S_i \cdot S_j = -(1 - \Theta(i, j))[2\hat{g}_{ij}\hat{g}_{ij} - S^2] + \Theta(i, j)[2\hat{f}_{ij}\hat{f}_{ij} - S(S + 1)],
\]

where \(\hat{f}_{ij} = \frac{1}{2}(a_i^\dagger a_{i+\Delta} + b_i^\dagger b_{i+\Delta})\) and \(\hat{g}_{ij} = \frac{1}{2}(a_i b_{i+\Delta} - b_i a_{i+\Delta})\) are respectively the ferromagnetic and antiferromagnetic bond operators. The function \(\Theta(i, j) = 1\) if \(i\) and \(j\) are on the same stripe sublattice, and \(\Theta(i, j) = 0\) if \(i\) and \(j\) are on different stripe sublattices. The Hubbard-Stratonovich field is then \(\Gamma_{\delta} = |\hat{f}_{\delta}|^2 - |\hat{g}_{\delta}|^2\), and in the case of \((\pi, 0)\) ordering we find

\[
\Gamma_x = -g_x^2, \\
\Gamma_y = f_y^2.
\]

Comparing this to Eq. (3.3), we see that the spin correlators coincide with those in the MSW theory if one sets \(\phi = \pi\). Similarly, the case of \((0, \pi)\) ordering corresponds to \(\phi = 0\) in Eq. (3.3). In both cases, \(\Gamma_x\) and \(\Gamma_y\) have opposite sign, leading to the anisotropy in the effective spin-spin exchange couplings \(\tilde{J}_{1x} \neq \tilde{J}_{1y}\), from Eq. (3.9).

By introducing Fourier transformation \([112]\)

\[
a_i = \frac{1}{\sqrt{N}} \sum_k a_k e^{i(k-\frac{\Delta}{2})r_i}, \quad \text{Eq. (3.12)}
\]

\[
b_i = \frac{1}{\sqrt{N}} \sum_k b_k e^{i(k+\frac{\Delta}{2})r_i}, \quad \text{Eq. (3.13)}
\]

and making a Bogoliubov transformation to a new quasiparticle creation/annihilation operators \(\alpha_k = \cosh \theta_k a_k + i \sinh \theta_k b_{-k}^\dagger\), one arrives at the mean-field free energy
density, which can be generalized to the Sp\((N)\) form \[111\]

\[
F_{MF} = \frac{NT}{N} \sum_{\mathbf{k}} \ln \left[ 2 \sinh \left( \frac{\omega_{\mathbf{k}}}{2T} \right) \right] + N\lambda \left( S + \frac{1}{2} \right) - \frac{Nz}{8} \sum_{\delta} \delta J_{\delta} (|f_{\delta}|^2 - |g_{\delta}|^2),
\]  

(3.14)

where \(z\) is the coordination number, and \(\lambda\) is the Lagrange multiplier associated with the imposed constraint that on average, the number of bosons per site \(\sum_{\sigma=1}^{N} n_{i\sigma} = NS\). Here \(\omega_{\mathbf{k}} = \sqrt{(B_{\mathbf{k}} - \lambda)^2 - A_{\mathbf{k}}^2}\) is the dispersion of the Bogoliubov quasiparticles, expressed \[112\] in terms of the variables

\[
A_{\mathbf{k}} = i \sum_{\delta} J_{\delta} g_{\delta} e^{-i k \cdot \delta}; \quad B_{\mathbf{k}} = \sum_{\delta} J_{\delta} f_{\delta} e^{-i k \cdot \delta};
\]

(3.15)

the Bogoliubov angle \(\tanh 2\theta_{\mathbf{k}} = A_{\mathbf{k}}/(\lambda - B_{\mathbf{k}})\). The dispersion relation \(\omega_{\mathbf{k}}\) explicitly depends on the ordering wave-vector \(\mathbf{Q}\) and has minima around \(\mathbf{k} = \pm \mathbf{Q}/2\). In the regime when \(J_2 > J_1/2\), the minimization of the free energy results in \(\mathbf{Q} = (0, \pi)\) or \((\pi, 0)\). For example, for \(\mathbf{Q} = (\pi, 0)\), the expressions for \(A_{\mathbf{k}}\) and \(B_{\mathbf{k}}\) become:

\[
A_{\mathbf{k}} = 2\tilde{J}_1 g_x \sin k_x + 4J_2 g_{x+y} \sin k_x \cos k_y,
\]

(3.16)

\[
B_{\mathbf{k}} = 2\tilde{J}_1 f_y \cos k_y.
\]

(3.17)

In the large-\(N\) limit of the Sp\((N)\) spin, the mean-field free energy Eq. (3.14) becomes exact.\[108, 111\] The observable magnetic excitation spectrum is obtained from \(\omega_{\mathbf{k}}\) by a \(\mathbf{Q}/2\) shift: \(\varepsilon_{\mathbf{k}} = \omega_{\mathbf{k}-\mathbf{Q}/2}\). At \(T = 0\), the magnetic order results in the gapless Goldstone modes at \(\mathbf{k} = 0\) and \(\mathbf{Q}\), as expected. The SBMF theory is known to reproduce well the spectrum of spin waves in both ferro- and antiferro-magnets.\[108, 112\]

Below, we focus on the paramagnetic phase at \(T > 0\), with short-range \(\mathbf{Q} = (\pi, 0)\) antiferromagnetic correlations (the case \(\mathbf{Q} = (0, \pi)\) is obtained by \(C_4\) lattice rotation). We obtain the following self-consistent equations from the saddle-point minimization
of the free energy Eq. (3.14):

\[ f_\delta = \frac{1}{N} \sum_k \frac{B_k - \lambda}{\omega_k} \left( n_k + \frac{1}{2} \right) \cos (k \cdot \delta), \]

(3.18)

\[ g_{\delta'} = \frac{1}{N} \sum_k \frac{A_k}{\omega_k} \left( n_k + \frac{1}{2} \right) \sin (k \cdot \delta'), \]

(3.19)

\[ S + \frac{1}{2} = \frac{1}{N} \sum_k \frac{B_k - \lambda}{\omega_k} \left( n_k + \frac{1}{2} \right), \]

(3.20)

where \( \delta = \hat{y}, \) and \( \delta' = \hat{x}, \hat{x} \pm \hat{y}. \) Under the transformation \( B_k - \lambda \rightarrow B_k \) and \( k \rightarrow k - Q/2, \) Eqs. (3.16)-(3.20) in the SBMF theory and Eqs. (3.4)-(3.8) in the MSW mean-field theory have exactly the same form in the short-range \((\pi, 0)\) correlated paramagnetic phase. Therefore, the two methods yield exactly the same mean-field phase diagram and boson dispersion, as corroborated by explicit numerical comparison. We further verified that these two theories give similar results for the spin dynamics of the \( J_1 - J_2 - K \) model.

### 3.3 Mean-field phase diagram and excitation spectrum

Since INS measurements suggest \( J_1 \sim J_2 \) for several 122 compounds, our discussion on the \( J_1 - J_2 - K \) model is focused on this parameter regime. Fig. 3.1 shows the mean-field phase diagram of the 2D \( J_1 - J_2 - K \) model using the MSW method for \( S = 1 \) and \( J_1/J_2 = 1. \) We identify three different phases. Phase I corresponds to the \((\pi, 0)/(0, \pi)\) antiferromagnetically long-range ordered phase; it exists only at \( T = 0 \) in the 2D model. Phase II and phase III are both paramagnetic. They are separated by a mean-field Ising transition temperature \( T_{\sigma 0}. \) We find that for \( J_1/J_2 = 1, \) this transition is first-order, as shown in Fig. 3.2. But it can be either first-order or second-order for \( J_1/J_2 \lesssim 0.9, \) as discussed in more detail in Appendix 3.8. In the low-temperature phase II, either \( f_x \neq f_y \) or \( g_x \neq g_y \) (see Fig. 3.2), corresponding...
Figure 3.1: Mean-field phase diagram in the MSW theory for $S = 1$ and $J_1/J_2 = 1$. Phases I, II, and III respectively denote the $(\pi,0)/(0,\pi)$ long-range antiferromagnetically ordered state (at $T = 0$), the Ising ordered paramagnetic state, and the isotropic paramagnetic state. The solid red curve refers to the mean-field temperature scale $T_{\sigma_0} = T_0$. In the shaded region the effective exchange coupling $\tilde{J}_{1y} < 0$.

Figure 3.1: Mean-field phase diagram in the MSW theory for $S = 1$ and $J_1/J_2 = 1$. Phases I, II, and III respectively denote the $(\pi,0)/(0,\pi)$ long-range antiferromagnetically ordered state (at $T = 0$), the Ising ordered paramagnetic state, and the isotropic paramagnetic state. The solid red curve refers to the mean-field temperature scale $T_{\sigma_0} = T_0$. In the shaded region the effective exchange coupling $\tilde{J}_{1y} < 0$.

to an Ising ordered phase with either $(\pi,0)$ or $(0,\pi)$ short-range antiferromagnetic correlations. This Ising ordered phase already exists in the isotropic $J_1 - J_2$ model. But here, we find that a nonzero $K$ enhances $T_{\sigma_0}$, and $K$ drives the effective nearest-neighbor exchange couplings to be anisotropic. As shown in Fig. 3.2 in the $(\pi,0)$ Ising ordered phase (corresponds to $\phi = \pi$), the effective coupling $\tilde{J}_{1y}$ can even be ferromagnetic. This is important for understanding the experimentally observed anisotropic magnetic excitations at high energies in Ca-122 and Ba-122. Phase III at $T > T_{\sigma_0}$ is the Ising disordered paramagnetic phase. In this phase the effective nearest-neighbor exchange couplings are isotropic because the nearest-neighbor bond correlators are zero. But the next-nearest-neighbor bond correlations may still be finite in this phase. One may define another temperature scale $T_{0}$, above which the next-nearest-neighbor bond correlations vanish and the system are decoupled into isolated local moments. Note that $T_{0}$ does not refer to a phase transition, and the
discontinuity of the bond correlations at $T_0$ is an artifact of the mean-field theory. \cite{106}

In general, $T_0$ and $T_{\sigma 0}$ are two different temperature scales satisfying $T_0 \geq T_{\sigma 0}$. \cite{102,111}

But for $J_1/J_2 \gtrsim 0.9$, $T_0 = T_{\sigma 0}$ for any $K/J_2$ ratio, as shown in Fig. 3.2. The phase diagram obtained in the SBMF theory is identical to the one shown in Fig. 3.1.

The finite coupling $K$ not only changes the phase boundary of the mean-field phase diagram, but can also dramatically influence the boson excitation spectrum. In Fig. 3.3(a), we show the dispersions of the DM and Schwinger bosons along two high-symmetry directions in momentum space for various $K$ values in phase II with $\phi = \pi$ using the same parameters as in Fig. 3.1. We see that the dispersion in Schwinger boson theory matches the one in the MSW theory exactly.
Figure 3.3: (a): MSW dispersion along high symmetry directions in the paramagnetic Brillouin zone for the 2D $J_1 - J_2 - K$ model at $S = 1$, $J_1/J_2 = 1.0$, $T/J_2 = 1.0$, and various $K$ values. For comparison, the red dashed curve shows the dispersion in the SBMF theory for the same parameters and $K/J_2 = 0.8$. The gaps at $(0,0)$ and $(\pi,0)$ are too small to be seen in the figure. (b): The symbols show the dispersion from the INS data at $T = 150$ K in BaFe$_2$As$_2$, taken from Ref. [103]. The data can be fit by any of the theoretical dispersion curves [as in (a)] that lie within the shaded region.

The dispersion shows a gap at $(\pi,0)$ (and also at $(0,0)$), with the size

$$\Delta_1 = \sqrt{-\mu(8J_2g_{x+y} + 4\tilde{J}_{1x}g_x - \mu)}.$$  \hspace{1cm} (3.21)

At low temperatures the gap is small since $\mu \to 0$ as $T \ll T_{\sigma 0}$. In this limit, the excitation near $(\pi,0)$ can be approximated by $\varepsilon_k = \sqrt{v_{1x}^2(\pi - k_x)^2 + v_{1y}^2k_y^2 + \Delta_1^2}$, where the velocities are respectively:

$$v_{1x} = 4J_2g_{x+y} + 2\tilde{J}_{1x}g_x,$$

$$v_{1y} = \sqrt{(4J_2g_{x+y} + 2\tilde{J}_{1x}g_x)(4J_2g_{x+y} - 2\tilde{J}_{1y}f_y) + 2\tilde{J}_{1y}f_y\mu}.$$ \hspace{1cm} (3.22)

(3.23)

The excitation develops to the gapless Goldstone mode at $T = 0$. At $(\pi,\pi)$ (and also at $(0,\pi)$) the dispersion has a different gap

$$\Delta_2 = \sqrt{(8J_2g_{x+y} - 4\tilde{J}_{1y}f_y - \mu)(4\tilde{J}_{1x}g_x - 4\tilde{J}_{1y}f_y - \mu)}.$$ \hspace{1cm} (3.24)
The features that $v_{1x} \neq v_{1y}$ and $\Delta_1 \neq \Delta_2$ already exists in the isotropic $J_1 - J_2$ model. In the $J_1 - J_2 - K$ model, $\Delta_1$ at $(\pi,0)$ is only weakly affected by $K$ because it is dominated by $\mu$. But $\Delta_2$ at $(\pi,\pi)$ is strongly influenced. It increases with $K$. For sufficiently large $K$, approximately where $\tilde{J}_{1y}$ changes sign to be ferromagnetic (the shaded region in Fig. 3.1), the dispersion at $(\pi,\pi)$ turns from a local minimum to a maximum, as shown in Fig. 3.3(a). Similar behavior in the spin-wave dispersion of the $J_1 - J_2 - K$ model has also been discussed in Ref. [98] in the antiferromagnetically ordered phase, but our results apply to the paramagnetic phase.

### 3.4 Dynamical spin structure factor

In order to investigate the magnetic excitations, which are directly accessible by INS measurements, we have calculated the magnetic structure factor $S(q,\omega)$. Our main interest is to understand the experimentally observed anisotropic feature of the magnetic excitations in the paramagnetic phase above the Néel temperature. As already discussed in Sec. 3.3 in this temperature regime, the most relevant factor for the in-plane anisotropy is the Ising order. Therefore, we will concentrate our discussion on the magnetic structure factor in phase II of the 2D $J_1 - J_2 - K$ model. In this phase $S(q,\omega)$ has the same form in both MSW and Schwinger boson theories,

$$S(q,\omega) = 2\pi C \frac{C}{N} \sum'_{\mathbf{k}} \sum_{s,s'=\pm 1} \left[ \cosh(2\theta_{\mathbf{k}\mathbf{q}} - 2\theta_{\mathbf{k}}) - ss' \right]$$

$$\times \delta(\omega - s\varepsilon_{\mathbf{k}+\mathbf{q}} - s'\varepsilon_{\mathbf{k}}) n_{\mathbf{k}+\mathbf{q}}^s n_{\mathbf{k}}^{s'},$$

(3.25)

where $\sum'$ refers to the summation over the magnetic Brillouin zone corresponding to the $(\pi,0)$ order, which is enclosed by $-\pi/2 \leq k_x \leq \pi/2$, and $-\pi \leq k_y \leq \pi$. $n_{\mathbf{k}}^+ = n_{\mathbf{k}} + 1$ and $n_{\mathbf{k}}^- = n_{\mathbf{k}}$. $C = 1$ in the MSW theory, and $C = 3/2$ in the Schwinger
Figure 3.4: Constant energy cuts of the rotational symmetrized spin dynamical structure factor in the momentum space in the MSW theory for $S = 1$, $J_1/J_2 = 1.0$, $K/J_2 = 0.8$, and $T/J_2 = 1.0$. The corresponding energies are respectively $\omega = 4J_2$ in (a), $\omega = 10J_2$ in (b), $\omega = 11.5J_2$ in (c), $\omega = 12J_2$ in (d). In all the panels, a broadening factor $0.5J_2$ has been used for the convenience of calculation.
boson theory with SU(2) symmetry. We see from Eq. (3.25) that the contribution to $S(q, \omega)$ comes from two-boson processes. Hence, in general cases the peak of $S(q, \omega)$ does not follow the boson dispersion. But at low temperatures, the largest contribution to $S(q, \omega)$ in the summation over $k$ comes from the term at $k = (0, 0)$ since the small gap $\Delta_1$ at this point results in a large boson number $n_k$. To satisfy the energy conservation in the $\delta$ function, $S(q, \omega)$ must be peaked at $\omega \approx \varepsilon_q$. Actually this leads to a two-peak structure corresponding to $s' = \pm 1$ near $\omega = \varepsilon_q$ for a fixed $q$. But the separation of these two peaks is proportional to $\varepsilon_{(0,0)}$ and is very small at low temperatures. In the numerical calculations performed, the gap between the two peaks is healed by substituting the delta function by a Lorentzian with a small broadening width. As a result of this small broadening, $S(q, \omega)$ only shows a single peak structure. Therefore, in this limit the peak positions of $S(q, \omega)$ follow the boson dispersion.

To better discuss the anisotropic distribution of the spectral weight in momentum space, we plot the constant energy cuts of the calculated $S(q, \omega)$ at a fixed temperature $T < T_{\sigma_0}$ in Fig. 3.4. At low energies, the peaks of $S(q, \omega)$ form an elliptic ring centered at $(\pi, 0)$ (and also its symmetry related point $(0, \pi)$ after rotation symmetrization), as displayed in Figs. 3.4(a),(b). The elliptic feature is a consequence of the anisotropic correlation lengths in the Ising ordered phase, and the ellipticity near $(\pi, 0)$ is proportional to $\xi_x/\xi_y = v_{1x}/v_{1y}$, which is not sensitive to temperature since the mean-field parameters are only weakly temperature dependent for $T < T_{\sigma_0}$ (Fig. 3.2). The ellipticity also only weakly depends on $K$: for $J_1/J_2 = 1$, we find

*The factor $C = (N + 1)/N$ in a SU(N) or Sp(N) SBMF theory. Hence $S(q, \omega)$ satisfies the sum rule only when $N \to \infty$. In the SU(2) SBMF theory, $C = 3/2$ violating the sum rule is a known issue. See Ref. [108] for more detail.
\[\frac{\xi_x}{\xi_y} \simeq 1.7 \text{ at } K = 0, \text{ and } \frac{\xi_x}{\xi_y} \simeq 1.4 \text{ at } K/J = 0.8.\]

With increasing energy, the ellipse centered around \((\pi, 0)\) expands towards the Brillouin zone boundary, as seen in Figs. 3.4(a)-(d). For sufficiently large energy, the spectral weight reduces greatly along the \(q_x\) direction, and the \(S(q, \omega)\) is peaked near \(q_y = \pm \pi/2\) along the \(q_y\) direction (Fig. 3.4(c)). The elliptical peak feature appears to have been split into two parts in the direction of its major axis. As the energy gets close to \(\varepsilon(\pi, \pi)\), the two peaks move towards \((\pi, \pm \pi)\), forming patterns that are centered around \((\pi, \pm \pi)\); cf. Figs. 3.4(c)-(d). In our theory, there are two factors that contribute to this anisotropic distribution of the spectral weight along the ellipses. Firstly, for \(\omega > \varepsilon(\pi/2, 0)\), along the \(q_x\) axis the energy conservation in the \(\delta\) function of Eq. (3.25) can only be satisfied when \(k \neq (0, 0)\). A nonzero \(k\) corresponds to a smaller \(n_k\), which greatly reduces \(S(q_x, \omega)\). Along the \(q_y\) axis, however, \(S(q_y, \omega)\) is not reduced because the \(k = (0, 0)\) mode can still satisfy the energy conservation. Secondly, for a given \(k\), the coherence factor \(\cosh(2\theta_k - 2\theta_{k+q})\) along the ellipse is also anisotropic. To see this, recall that the largest contribution to \(S(q, \omega)\) is from the \(k = (0, 0)\) term in Eq. (3.25). For simplicity, we take a single mode approximation, namely, \(S(q, \omega)\) can be approximated by this \(k = (0, 0)\) term. Then the ellipse showing spectral weight peaks is determined by \(\varepsilon_q = \omega\), and the coherence factor \(\cosh(2\theta_k - 2\theta_{k+q}) \propto (B_q - A_q)/\varepsilon_q\). For \(\sqrt{q_x^2 \xi_x^2 + q_y^2 \xi_y^2} \ll T/\Delta_1\), \(B_q - A_q \approx \Delta - \varepsilon_q/\Delta - 2\tilde{J}_1 f_y q_y^2\), where \(\Delta = 8J_2 g_{x+y} + 4\tilde{J}_1 x g_x\). Since \(\tilde{J}_1 y < 0\) for the choice of model parameters, it is easy to see that along the ellipse \(\varepsilon_q = \omega\), the maximum of the coherence factor is located along the \(q_y\) axis but not the \(q_x\) axis. Since within the single mode approximation, \(S(q, \omega)\) is proportional to the coherence factor, \(S(q, \omega)\) is also anisotropic along the ellipse. Note that at low energies \(\sqrt{q_x^2 \xi_x^2 + q_y^2 \xi_y^2} \ll T/\Delta_1\), \(\tilde{J}_1 y f_y q_y^2 \ll \Delta\), so the anisotropy is very small. This coherence-factor-induced anisotropy becomes sizable
when the ellipse is large (for $\sqrt{q_x^2 s_x^2 + q_y^2 s_y^2} \gtrsim T/\Delta_1$).

### 3.5 Discussions

#### 3.5.1 The effects of spin size

![Figure 3.5](image.png)

Figure 3.5: (a): MSW dispersion along high symmetry directions in the paramagnetic Brillouin zone for the 2D $J_1 - J_2 - K$ model at $S = 2$, $J_1/J_2 = 1.0$, $T/J_2 = 1.0$, and various $K$ values. (b): The ratio of $\tilde{J}_{1y}/\tilde{J}_{1x}$, showing the anisotropy in effective exchange couplings, see Eq. (3.9), as a function of the biquadratic coupling for $S = 1$ and $S = 2$.

Besides the $S = 1$ results shown in Sec. 3.3 and Sec. 3.4, we have also studied the $J_1 - J_2 - K$ model with larger spin sizes, and found the mean-field phase diagram is similar to the one in Fig. 3.1. Approximately, $T_0$ and $T_0$ are increased by a factor of $S(S + 1)/2$. The boson dispersion shown in Fig. 3.5(a) also exhibits the similar features as in the $S = 1$ case. In Fig. 3.5(b) we compare the ratio of the effective nearest neighbor couplings $\tilde{J}_{1y}/\tilde{J}_{1x}$, defined in Eq. (3.9), for $S = 1$ and $S = 2$ (at
zero temperature). We find that with increasing $S$, the minimal $K/J_2$ value where $\tilde{J}_{1y}$ becomes ferromagnetic is dropped from $K/J_2 \approx 0.53$ to $K/J_2 \approx 0.13$. Hence, we conclude that the anisotropy of the effective exchange couplings induced by non-Heisenberg coupling $K$ is more significant for larger spin size $S$.

We can further compare our MSW result at $T = 0$ with the one in a recent MSW study, which used a mean-field treatment that is different from ours.\textsuperscript{99} The two theories yield exactly the same results when the spin size $S \to \infty$. For finite spin sizes, by comparing the behavior of $\tilde{J}_{1y}/\tilde{J}_{1x}$ ratio in Fig. 3.5(b) and the corresponding results in Ref. \textsuperscript{99}, we observe that the two theories give qualitatively similar results for the anisotropy in the exchange couplings: The biquadratic coupling $K$ reduces the ratio of the effective ratio $\tilde{J}_{1y}/\tilde{J}_{1x}$. Quantitatively, there are some differences between the two approaches. In particular, while for $S > 1$, the ratio $\tilde{J}_{1y}/\tilde{J}_{1x}$ changes sign at a finite $K$ value in both theories, this sign change does not appear for $S = 1$ in Ref. \textsuperscript{99}.

3.5.2 Generalizations of the $J_1 - J_2 - K$ model

Several remarks on the $J_1 - J_2 - K$ model studied in this chapter. From the incipient Mott picture, when the system is in the vicinity of $U_c/t$, the spin Hamiltonian contains interactions involving more than just two spins. To see this, we start from a multi-orbital Hubbard model on a square lattice, and assume that Hund’s rule coupling locks the spins in different orbitals to a high spin state. Then we may obtain a spin-only Hamiltonian by integrating out the fermion degrees of freedom based on perturbation in $t/U$. To the $t^2/U$ order, we obtain the usual $J_1 - J_2$ Heisenberg interaction between nearest- and next-nearest-neighbor spins. The next-order terms appear in the order $t^4/U^3$, and include the biquadratic $K$ term as well as the ring exchange interactions.
Here, we have focused on the effects of the biquadratic interaction. The influence of the ring exchange interactions in the regime we are considering is briefly discussed in Appendix 3.9.

To fully understand the antiferromagnetic \((\pi, 0, \pi)\) order revealed in the experiments, the 2D \(J_1 - J_2 - K\) model needs to be extended to the 3D case by including an interlayer coupling \(J_z\). A nonzero \(J_z\) will support the antiferromagnetic order up to the Néel temperature \(T_N\). In the mean-field treatment, the antiferromagnetic order emerges at a mean-field Néel temperature \(T_{N0}\). The details of the effects of the interlayer coupling \(J_z\) to the magnetic phase diagram of the \(J_1 - J_2 - K\) model and the magnetic excitation spectrum is further discussed in Appendix 3.10.

When fluctuations beyond the mean-field level are taken into account, the actual Néel and Ising transition temperatures, \(T_N\) and \(T_\sigma\) can be well below their mean-field values. The mean-field temperatures \(T_{N0}\) and \(T_{\sigma0}\) then correspond to some crossover temperature scales, below which the fluctuating order have significant effects. The fluctuating anisotropic effects we have presented will be dominant in the temperature regime \(T_N < T < T_{\sigma0}\)\(^{102}\).

### 3.5.3 Effect of itinerant electrons

Within the bad-metal description of the iron pnictides, the quasi-localized moments are coupled to itinerant electrons with a spectral weight that depends on the proximity to the Mott transition. A convenient way to describe the effect of itinerant electrons on the spin dynamics is to reformulate the results of the local-moment-based calculations in terms of a non-linear sigma model, and introduce into the latter a damping caused by the itinerant electrons; for details, we refer to Ref. \(^{102}\). Well below \(T_{\sigma0}\) and in the vicinity of \((\pi, 0)\), the effects of the itinerant electrons are described in terms of
the effective action for the staggered magnetization $M$:

$$S[M] = T \int dq \sum_l [r + \Delta r + v^2_{lx}q_x^2 + v^2_{ly}q_y^2 + \omega_l^2 + \gamma |\omega_l|] M^2 + u M^4 + O(M^6).$$

Here, $M = m + m'$ is the sum of $m$ and $m'$, the $O(3)$ vectors respectively for the magnetizations of the two decoupled sublattices on the square lattice, and $\omega_l$ the Matsubara frequency. This action arises in a “$w$-expansion”, which is based on a proximity to the Mott transition and is described in Refs. \[77, 78\]; it has the form of the usual $\sigma$-model \[113\]. In the first term, $\Delta r > 0$ is a mass shift and $\gamma$ describes the strength of spin damping from coupling to fermions. (See Fig. 3.6) At relatively low energies, this introduces a procedure that can be used to describe the broadening of the spin spectral peaks in momentum space due to coupling to itinerant electrons. \[102\]

![Diagramm of the second-order contribution to the effective action in Eq. (3.26) due to coupling to fermions.](image)

We should emphasize that this procedure is a qualitative treatment of the spin damping. Incorporating the full details of the electronic bandstructure will introduce momentum-dependence of the damping rate, making it possible to generate the type of anisotropic damping that was proposed phenomenologically by Harriger et al. \[103\].

Comparing our results for the $J_1 - J_2 - K$ model in Fig. 3.4(a)-(d) with those of the $J_1 - J_2$ model (Fig. 4 of Ref. \[102\]) shows that, the biquadratic term itself brings out an anisotropy in the spectral weight of the elliptic peaks. The spectral weight is larger along the major axis of the ellipse than along its minor axis. This anisotropy
goes in the same direction as that of the experimental data on BaFe$_2$As$_2$, illustrated in Fig. 3.4(f). We therefore conclude that both the ellipticity and intensity anisotropy of the spectral peaks in momentum space are controlled by the exchange interactions.

We note that the Ising order parameter is also coupled to the itinerant electrons. Since the Ising order parameter breaks the $C_{4v}$ symmetry, it couples to those spin singlet fermion bilinears that correspond to the $B_{1g}$ representation. Consequently a nonzero Ising order parameter will induce a nonzero $d_{x^2-y^2}$ nematic charge density $(\cos k_x - \cos k_y) c_{k,\alpha}^\dagger c_{k,\alpha}$ for all the d-orbital electrons, where $\alpha$ is the orbital index. In addition the Ising order parameter will induce a nonzero charge density imbalance $c_{kxz}^\dagger c_{kxz} - c_{kyz}^\dagger c_{kyz}$ between the $d_{xz}$ and the $d_{yz}$ orbitals, which is also referred as the ferro-orbital order. As a result the spin fluctuations from the incoherent degrees of freedom can give rise to an orbitally ordered, charge nematic metal, with anisotropic transport properties. In a model with 3D coupling (see Appendix 3.10), the coupling to the itinerant electrons will reduce the Néel transition temperature from its mean-field value $T_{N0}$ to $T_N < T_{N0}$, through the positive $\Delta r$ noted above. It will likewise decrease the Ising transition temperature from its mean-field value $T_{\sigma0}$ to $T_{\sigma} < T_{\sigma0}$. However, the correlation lengths are still sizeable and should be anisotropic up to $T_{\sigma0}$.[102] This implies that, in the 3D model with three-dimensional coupling, we expect anisotropic magnetic excitations to exist from $T_N$ all the way up to the crossover temperature scale $T_{\sigma0}$, in the absence of a static Ising order.

### 3.5.4 Comparison with other approaches

Our studies in the $J_1 - J_2 - K$ model, with or without the coupling to the itinerant electrons, are very different from purely itinerant studies with $U/t$ much smaller than $U_c/t$. Because the Fermi surface comprises small electron and hole pockets, such
calculations are expected to yield very small spin spectral weight. Experimentally, the total spectral weight is known to be large, with an effective moment that is larger than 1 $\mu_B$/Fe in CaFe$_2$As$_2$ (Ref. [86]). Such a large spectral weight arises naturally in our approach using as the starting point the $J_1 - J_2$ model (with or without the $K$ term).

Our approach can be compared more closely with that of the DMFT studies of Ref. [105], in which the ratio of the effective interaction (combined Coulomb and Hund’s interactions) to the characteristic bandwidth is close to the Mott-transition value, $U/t \lesssim U_c/t$. The proximity to the Mott transition ensures that a large part of the electronic spectral weight lies in the incoherent regime, which will naturally give rise to a large spin spectral weight. The consistency of the momentum-dependence determined by the DMFT calculations and that of our $J_1 - J_2 - K$ calculations further suggests the compatibility of the two approaches. There are some important differences, however. In the DMFT calculation, the anisotropy of the structure factor has been attributed to the geometry of the Fermi surface(s). The $J_1 - J_2 - K$ results however tie the anisotropy of the spin spectral weight in momentum space with the Ising correlations.

Experimentally, the Ising correlations can be very naturally connected with the $x - y$ anisotropy observed in ARPES [114] and transport [115] measurements in the detwinned 122 iron pnictides at temperatures above $T_N$. A recent theoretical calculation [116] shows how resistivity anisotropy in the tetragonal phase above $T_N$ follows from the existence of the Ising correlations discussed here.
Figure 3.7: Evolution of $S(q, \omega)$ in the paramagnetic phase of the $J_1 - J_2 - K$ model, showing that the elliptical features near $(\pi, 0)$ at low energies (top panels) are split into features that are centered around $(\pi, \pi)$, as the energy is increased towards the zone-boundary spin-excitation energy (bottom panels). This trend is consistent with the inelastic neutron scattering experiments, shown in the box on the right for two energies measured in the paramagnetic phase of BaFe$_2$As$_2$ (data taken from Ref. [103]). (a)-(d): Same as Fig. 3.4(a)-(d), but with damping $\gamma = 3J_2$. (e)-(f): The INS data at $T = 150$ K in BaFe$_2$As$_2$, taken from Ref. [103]. The energy transfer is $\omega = 50 \pm 10$ meV in (e), and $\omega = 150 \pm 10$ meV in (f). Here we find that the best agreement between theory and experimental data achieves when taking $J_2 \approx 13$ meV in the model.
3.6 Comparison with experiments on the paramagnetic phases of parent 122 iron pnictides

Spin dynamics in the paramagnetic phase of the parent 122 iron pnictides has been recently studied via INS measurements. For CaFe$_2$As$_2$, spin dynamics at low energies (below 70 meV) has been studied by Diallo et al. It is found that the peaks of the dynamical structure factor form anisotropic elliptic features at low energies, similar to the results in the antiferromagnetic phase. More recently, Harriger et al. measured the spin dynamics of BaFe$_2$As$_2$ up to 200 meV. At low energies, they found the distribution of spectral weights in the momentum space forms similar elliptic feature as in the CaFe$_2$As$_2$ case. With increasing energy, the elliptic feature expands towards the Brillouin zone boundary. Moreover, they determined the magnetic dispersion to be peaked (or flat-topped) near ($\pi, \pi$). Similar results have also been reported for SrFe$_2$As$_2$.

Our study on the $J_1 - J_2 - K$ model have already provided valuable information for understanding these experimental observations. In real materials, the various fluctuation mechanisms and the coupling to fermions/phonons will reduce the Néel and Ising transition temperatures. However, below the mean-field Ising temperature $T_{\sigma_0}$, the effective couplings between the nearest neighbors are always anisotropic. Hence we expect the magnetic fluctuations to be anisotropic for $T_N(\leq T_\sigma) < T < T_{\sigma_0}$, which corresponds to the upper portion of region II in Fig. 3.1. This anisotropy is reflected in the spin dynamics in the paramagnetic phase.

To be specific, the anisotropic elliptic feature at low energies observed in CaFe$_2$As$_2$ and other parent 122 compounds can already been understood within the $J_1 - J_2$ model. We have shown in Fig. 3.4 that the $J_1 - J_2 - K$ model gives the similar
low-energy elliptic feature. It will be important to measure the spin dynamics at high energies in this material.

Our calculated evolution of this elliptic feature as the energy is raised in the $J_{1}-J_{2}-K$ model can be systematically compared with the experimental observations in BaFe$_2$As$_2$ and SrFe$_2$As$_2$. To see this, we fit the peak positions of calculated $S(q, \omega)$ to the experimental magnetic excitation dispersion data in BaFe$_2$As$_2$, from which we can extract the best fitted values of the exchange couplings. Assuming $S = 1$, we find the fitted exchange couplings are $J_2 = 17 \pm 4$ meV, $J_1/J_2 = 1.0 \pm 0.5$, and $K/J_2 = 0.8 \pm 0.3$. We find that a very broad range of the $J_1/J_2$ ratio can all fit the experimental data quite well. As illustrated in Fig. 3.3(b), any dispersion curve within the shaded region fits the experimental data within error bars. But to fit the dispersion data near the local maximum at $(\pi, \pi)$, a moderate $K/J_2$ ratio is necessary. For $S = 1$, we find $K/J_2 \approx 0.8$ fits the data the best. On the other hand, for $S = 2$, the best fitted $K/J_2$ ratio is substantially reduced to about 0.2.

For BaFe$_2$As$_2$, detailed measurements in the momentum space have been reported by Harriger et al. [103]. This allows us to see that the agreement between our theory and the experiment is not only for the dispersion, but also for the anisotropic distribution of the spectral weight of $S(q, \omega)$ in momentum space.

In order to make a comparison with experimental data, we use Eq. (3.25) in the calculation of $S(q, \omega)$ and approximate the delta function by the following Lorentzian broadening

$$\delta(\omega - \Delta \varepsilon) \rightarrow \frac{1}{\pi} \frac{\gamma}{(\omega - \Delta \varepsilon)^2 + \gamma^2}. \quad (3.27)$$

Here we have assumed that the broadening mainly comes from the damping effect due to coupling to itinerant electrons. It is then reasonable to take the phenomenological broadening factor to be the damping $\gamma$ introduced in Eq. (3.26) since in either the
MSW or Schwinger boson theory, the damping is still due to the same bubble in Fig. 3.6. Calculating the magnitude of $\gamma$ requires a detailed microscopic theory and is beyond the scope of this work, however we can use Ref. [102] for reference, where it has been determined that $\gamma/J_2 \approx 3$ for CaFe$_2$As$_2$. Here we assume that this ratio still holds for BaFe$_2$As$_2$ and the damping is isotropic. In Figs. 3.7(a)-(d) we replott the theoretical dynamical spin structure factor in Fig. 3.4 with this damping factor, and compare them with the experimental data in Ref. [103]. At low energies, our theory correctly captures the elliptical feature centered at $(\pi, 0)$ as displayed in Figs. 3.7(a),(b). Experimentally, this is seen as a filled elliptical spot due to damping effect, which is also shown in our theoretical plot in Fig. 3.7(a). The evolution of the elliptical feature with increasing energy is also consistent with the experimental observation: as the ellipse expands towards zone boundary, it gradually splits into two parts, and forms a pattern around $(\pi, \pi)$ (see Figs. 3.7(c), (d), and (f)). We reiterate that such anisotropic features are the properties of our $J_1 - J_2 - K$ model either with an isotropic or without additional damping due to itinerant electrons. While anisotropic damping proposed in Ref. [103] could reinforce the effect, it is not necessary to understand the INS experiments. In CaFe$_2$As$_2$ the elliptical feature around $(\pi, 0)$ persists up to high energies, while in BaFe$_2$As$_2$, this elliptical feature splits into two parts at intermediate energy. [103] These two different behaviors can both be understood within our $J_1 - J_2 - K$ model with similar, nearly isotropic damping but different $K$ values.

3.7 Conclusions

In this chapter we have investigated the finite temperature spin dynamics of a $J_1 - J_2 - K$ antiferromagnetic Heisenberg model using both MSW and SBMF theories.
The spin dynamics obtained from these two methods are similar to each other.

We have found that by including a moderate biquadratic coupling \( K \), the magnetic excitation spectrum of the \( J_1 - J_2 - K \) model is anisotropic below a mean-field Ising transition temperature \( T_{\sigma 0} \). As in the case of the \( J_1 - J_2 \) model \[102\], the peak of the low-temperature dynamical structure factor \( S(q, \omega) \) contains elliptical features near \((\pi, 0)\) in the paramagnetic Brillouin zone at low excitation energies. However, unlike the pure \( J_1 - J_2 \) model, the spectral intensity also displays anisotropy along the ellipse, with the intensity being higher along the major axis than that along the minor axis. This spectral anisotropy accounts for the observed particular way in which the low-energy elliptical features, centered around \((\pi, 0)\), expand towards the zone boundary as the energy is increased towards the zone-boundary spin-excitation energy. It also gives rise to a particular form of high-energy spectral features that are centered around \((\pi, \pi)\).

We have also compared our calculated dynamical spin structure factor of the \( J_1 - J_2 - K \) model with the recent inelastic neutron-scattering measurements in the paramagnetic phases of the 122 iron pnictides \[103, 101, 104\]. The theoretical results provide a very natural understanding of the salient features of the experiments.

### 3.8 Ising transition at small \( J_1/J_2 \) ratios

We find that the nature of the mean-field Ising transition at \( T_{\sigma 0} \) depends on both \( J_1/J_2 \) and \( K/J_2 \) ratios. At \( K = 0 \) and \( J_1/J_2 \lesssim 0.9 \), we find \( T_{\sigma 0} < T_0 \), and the Ising transition at \( T_{\sigma 0} \) is always second-order (Fig. 3.8). When \( J_1/J_2 \gtrsim 0.9 \), \( T_{\sigma 0} \) meets \( T_0 \) and the Ising transition becomes first-order. This is an artifact of the mean-field approximation since the transition at \( T_0 \) is always first-order.\[106, 107\] Still for \( J_1/J_2 \lesssim 0.9 \), increasing \( K \) from zero, the transition at \( T_{\sigma 0} \) changes from second-order
to first-order when $K$ is bigger than a bicritical point $K_c$. As shown in Fig. 3.8 for $J_1/J_2 = 0.6$, $K_c/J_2 \approx 0.04$. At $K \gtrsim K_c$, $T_{\sigma 0} < T_0$. This suggests that the Ising transition near $K_c$ is not influenced by $T_0$, but the order of this transition is tuned by $K$. Hence the first-order transition at $T_{\sigma 0}$ is not an artifact of the mean-field treatment.

Figure 3.8: Mean-field magnetic phase diagram in the MSW theory for $S = 1$, $J_1/J_2 = 0.6$. The dashed blue and dashed dotted brown curves refer to the mean-field temperature scales $T_{N0}$ and $T_0$, respectively. The thicker solid red curve refers to a second-order Ising transition at $T_{\sigma 0}$, while the thinner solid red curve refers to a first-order transition. In the shaded region, the effective exchange coupling $\tilde{J}_{1y} < 0$.

3.9 Effects of ring exchange couplings

Besides the quadratic and biquadratic interactions, other interactions involving more than two spins can also appear in the spin Hamiltonian in the vicinity of Mott transition. For instance, the four-spin ring exchange interaction can appear as a consequence of the fourth-order perturbation associated with the electron hopping process. We can consider the effects of a four-spin ring exchange process on the spin dynamics by adding a term $K \square \sum_{i,j,k,l}[(S_i \cdot S_j)(S_k \cdot S_l) - (S_i \cdot S_l)(S_j \cdot S_k) + (S_i \cdot S_j)(S_k \cdot S_l)]$ to
the Hamiltonian, where $K_{\square} > 0$, and the sites $(i, j, k, l)$ are the vertices of a square plaquette, labeled clockwise. The four spin ring exchange competes against $J_1$ and $J_2$ and tends to weaken the antiferromagnetic order coming from $J_1$ or $J_2$. In the linear spin wave description of the $(\pi, 0)$ ordered state, we obtain the effective exchange constants \( \tilde{J}_{1x} = J_1 + 2(K - K_{\square})S^2 \), \( \tilde{J}_{1y} = J_1 - 2(K - K_{\square})S^2 \), and \( \tilde{J}_2 = J_2 + K_{\square}S^2 \), and a reduced spin gap at $(\pi, \pi)$. This trend also persists in the paramagnetic state, and reduces the size of the Ising order parameter. For consistency with the experimental results we require $K > K_{\square}$.

### 3.10 Effects of interlayer exchange coupling

![Figure 3.9: Mean-field magnetic phase diagram in the MSW theory for $S = 1$, $J_1/J_2 = 1$, and an interlayer exchange coupling $J_z/J_2 = 0.1$. The dashed blue and solid red curves refer to the mean-field temperature scales $T_{N0}$ and $T_{c0}$, respectively. In the shaded region, the effective exchange coupling $\tilde{J}_{1y} < 0$.](image)

The real materials have a 3D tetragonal structure. In the $J_1 - J_2 - K$ model, the 3D effects can be studied by extending the model to include a finite interlayer exchange interaction $J_z \sum_i \mathbf{S}_i \cdot \mathbf{S}_{i+\hat{z}}$. In 3D the long-range antiferromagnetic phase survives at
finite temperature up to the Néel temperature $T_N$. In MSW and SBMF theories, the mean-field Néel temperature $T_{N0}$ is determined by the onset of spontaneous sublattice magnetization $m_0$. In general, $T_{N0} \leq T_\sigma \leq T_0$. The modification to our discussion in Sec. 3.2 comes through an additional interlayer antiferromanetic bond correlation parameter $g_z$. In the presence of $J_z$, the self-consistent equations of Eqs. (3.4)-(3.6) and Eqs. (3.18)-(3.20) are unchanged, but the expressions for $A_k$ and $B_k$ are modified according to

$$A_k^{3D} = A_k + 2J_z g_z \cos k_z$$  \hspace{1cm} (3.28)  \\
$$B_k^{3D} = B_k + 2J_z g_z,$$  \hspace{1cm} (3.29)
in the MSW mean-field theory, and

\[ A^{3D}_k = A_k + 2J_z g_z \sin k_z \]  
\[ B^{3D}_k = B_k. \]  

in the SBMF theory.

In Fig. 3.11 we show the phase diagram at the experimentally suggested ratio \( J_z/J_2 = 0.1 \). Similar to the 2D case, the mean-field phase diagram consists of an Ising and Néel ordered antiferromagnetic phase (I), an Ising ordered but Néel disordered paramagnetic phase (II), and an Ising and Néel disordered paramagnetic phase (III), separated by mean-field temperatures \( T_{N0} \) and \( T_{\sigma 0} \) (see also Fig. 3.10). For the parameters in Fig. 3.9 the transitions are both first-order, and both \( T_{N0} \) and \( T_{\sigma 0} \) increase with \( K \). For \( K/J_2 \gtrsim 0.2 \), \( T_{N0} \) meets \( T_{\sigma 0} \), and there is only a single transition between phases I and III. The absence of phase II in this regime is an artifact of the mean-field theory, since \( T_{\sigma 0} \) is always bounded above by the mean-field scale \( T_0 \).

In connection to the real materials, we note that the structural and magnetic
transitions in the 1111 pnictides are well separated. But in 122 compounds, they are either very close to each other, or become a single first-order transition. This can be understood in terms of the present theory, provided $J_z$ is stronger in the 122 materials. By comparing Fig. 3.9 and Fig. 3.2 we see that the magnetic transition is closer to the Ising transition for a larger $J_z$. Recent experiments also show that the electron doping may cause the separation of the structural and magnetic transition temperatures in Ba(Fe,Co)$_2$As$_2$ system[117]. The similarity between this behavior and the $K$ dependence of $T_{\sigma 0}$ and $T_{N0}$ in the phase diagram of Fig. 3.9 suggests the possibility that electron doping is positively correlated with a reduction of the biquadratic interaction. It would then be interesting to reveal the link between them in future experimental and theoretical studies.

In Fig. 3.11 we show the low-temperature boson dispersions of the 3D model for various $K$ values along two high-symmetry directions in the $k_z = \pi$ plane. Aside from a larger gap at $(0,0,\pi)$, the dispersion is very similar to the one in 2D: the dispersion is highly anisotropic, and with increasing $K$, the local minimum at $(\pi,\pi,\pi)$ turns to a maximum. This is not too surprising because the in-plane anisotropy is a consequence of the 2D Ising-type fluctuations, and is not sensitive to the interlayer exchange coupling.
Chapter 4

Spin Ferroquadrupolar Order in the Nematic Phase of FeSe

4.1 Introduction

Superconductivity in the iron-based superconductors \cite{17, 118} is widely recognized to have spin fluctuations at its origin \cite{119, 120}, as it develops after the suppression of columnar antiferromagnetism (CAFМ) by doping or applied pressure on the parent compounds \cite{69, 121, 122, 123}. The CAFМ phase is characterized by the magnetic Bragg peaks at wave-vectors $Q_{1,2} = (\pi, 0)/(0, \pi)$ in the one-iron Brillouin zone, seen ubiquitously in different families of the iron pnictides and chalcogenides \cite{69, 124, 125}. The discovery of superconductivity in stoichiometric FeSe thus came as a surprise, because the long-range magnetic order is conspicuously absent in this material \cite{126, 127, 128, 129, 130, 131}. Another important feature, universally observed across different families of iron-based superconductors, is the appearance of an electronic nematic phase \cite{115, 132, 114, 133} which spontaneously breaks the lattice $C_4$ rotational symmetry. Usually, nematicity appears in close proximity to magnetism above the Néel temperature, which we considered in Chapter 3. However in FeSe, the nematic phase appears without any accompanying magnetism and coexists with superconductivity \cite{127, 128, 129, 130}. It is thus important to understand the origin of this non-magnetic nematic phase, in particular to gain insight into its effect on superconductivity.
It turns out that magnetic order can be induced by applying hydrostatic pressure to FeSe \cite{127, 128, 129}. It has also been suggested based on \textit{ab initio} calculations that the non-magnetic phase in FeSe lies in close proximity to the CAFM phase \cite{134, 135, 136}. Further evidence of proximity to long-range magnetic order comes from inelastic neutron scattering (INS) experiments, which found large spectral weight at wavevectors $Q_{1, 2}$ \cite{137, 138, 139, 140}. Two natural questions arise: In the theoretical phase diagram, is there a non-magnetic phase that neighbors on the CAFM? And furthermore, how does such a non-magnetic phase give rise to nematicity?

In an attempt to answer these questions, several theoretical scenarios have been proposed for non-magnetic ground states that may appear as a result of frustration: a nematic quantum paramagnet \cite{141}, a spin quadrupolar state with wave-vectors $Q_{1, 2}$ \cite{142}, or a staggered dimer state \cite{143}. In all three cases, the ground state wavefunction was designed to explicitly break the $C_4$ symmetry, thus resulting in nematicity. Alternatively, instead of being the ground state property, nematicity can also be induced as a result of anisotropic thermal \cite{144, 146} or possibly quantum fluctuations.

In this chapter, we investigate the frustrated bilinear-biquadratic Heisenberg model used by many authors to model iron pnictides and chalcogenides \cite{75, 98, 145, 141, 142}, and show that the most likely non-magnetic state that agrees qualitatively with the INS data on FeSe is the spin ferroquadrupolar (FQ) phase. By using variational mean-field, flavor-wave expansion, and the density matrix renormalization group (DMRG) calculations, we firmly establish that the FQ phase is situated in close proximity to the CAFM state in the phase diagram and is readily accessible in the realistic parameter regime of the model. The experimentally observed onset of magnetism in FeSe under applied pressure \cite{127, 128, 129} is thus interpreted as the transition between
the proposed FQ phase and CAFM. The calculated dynamical spin structure factors
agree qualitatively with the INS data \cite{137,138,139,140}, exhibiting pronounced
maxima of the scattering intensity at the gapped $Q_{1,2}$ points. We note that this
is in contrast with the antiferroquadrupolar (AFQ) scenario, which has negligible
spectral weight at these wavevectors \cite{142}. Furthermore, we demonstrate that FQ
order is robust with respect to $C_4$ symmetry breaking environment, and can thus
support nematicity, regardless of its microscopic origin. Additionally, we find that
the density-density interactions between $Q_{1,2}$ modes are highly repulsive within the
FQ phase and diverge upon approaching the FQ/CAFM phase boundary, providing
a scenario in which quantum fluctuations in FQ are the origin of nematicity.

4.2 Model

We use a bilinear-biquadratic Heisenberg model \cite{75,98,145,141,142} to investigate
the ground state properties and spin dynamics:

$$H = \frac{1}{2} \sum_{i,j} J_{ij} S_i \cdot S_j + \frac{1}{2} \sum_{i,j} K_{ij} (S_i \cdot S_j)^2,$$

(4.1)

where $S_i$ is the quantum spin-1 operator on site $i$. In the present study, the interactions
are limited to the 1st and 2nd nearest neighbors: $J_{ij} = \{J_1, J_2\}$, $K_{ij} = \{K_1, K_2\}$.

The quadrupolar operators are traceless symmetric tensors $Q^{\alpha\beta} \equiv S^\alpha S^\beta + S^\beta S^\alpha - \frac{4}{3} \delta^{\alpha\beta} (\alpha, \beta = x, y, z)$. Only five of these tensors are linearly independent, which are
convenient to cast in a 5-vector form:

\[
Q = \begin{pmatrix}
Q^{x^2-y^2} \\
Q^{3z^2-r^2} \\
Q^{xy} \\
Q^{yz} \\
Q^{zx}
\end{pmatrix} = \begin{pmatrix}
\frac{Q^{x^2}-Q^{y^2}}{2} \\
\frac{2Q^{x^2}-Q^{z^2}-Q^{y^2}}{2\sqrt{3}} \\
Q^{xy} \\
Q^{yz} \\
Q^{zx}
\end{pmatrix} = \begin{pmatrix}
(S^x)^2 - (S^y)^2 \\
\frac{3(S^x)^2-2}{\sqrt{3}} \\
S^x S^y + S^y S^x \\
S^y S^z + S^z S^y \\
S^z S^x + S^x S^z
\end{pmatrix}
\] (4.2)

For spin-1, we use the identity:

\[
(S_i \cdot S_j)^2 = \frac{1}{2} Q_i \cdot Q_j - \frac{1}{2} S_i \cdot S_j + \frac{4}{3},
\] (4.3)

we can rewrite model Eq. (4.1) as

\[
\mathcal{H} = \frac{1}{2} \sum_{i,j} \left( J_{ij} - \frac{K_{ij}}{2} \right) S_i \cdot S_j + \frac{1}{4} \sum_{i,j} K_{ij} \left( Q_i \cdot Q_j + \frac{8}{3} \right).
\] (4.4)

A time reversal invariant basis for spin-1 is used in this work, \(|\alpha\rangle = \{ |x\rangle, |y\rangle, |z\rangle \}\), defined as a unitary transformation from the regular \(|S_z\rangle\) basis:

\[
|x\rangle = i \frac{|1\rangle - |\bar{1}\rangle}{\sqrt{2}}, \quad |y\rangle = \frac{|1\rangle + |\bar{1}\rangle}{\sqrt{2}}, \quad |z\rangle = -i |0\rangle.
\] (4.5)

Arbitrary single site state can be represented by a unit-length director \(\vec{d}_i\) in this basis \(|\vec{d}_i\rangle = \sum_\alpha d_\alpha |\alpha\rangle\).

### 4.3 Variational Mean-field Calculation

Given a spin state parametrized by director \(\vec{d}_i\), the energy of the model Eq. (4.4) can be readily calculated at the mean-field level by decoupling \(\langle S_i \cdot S_j \rangle \approx \langle S_i \rangle \cdot \langle S_j \rangle\) and similarly for \(\langle Q_i \cdot Q_j \rangle\). Such mean-field decoupling is justified in a minimally
entangled long-range order state, for which the wavefunction can be written in a separable form \(|\Psi\rangle = \prod_i |\vec{d}_i\rangle\) \cite{146}. The mean-field ground state energy density is given by:

\[
E_0 = \frac{1}{2N} \sum_{i,j} \left[ J_{ij} |\langle \vec{d}_i | \vec{d}_j \rangle|^2 - (J_{ij} - K_{ij}) |\langle \vec{d}_i | \vec{d}^*_j \rangle|^2 + K_{ij} \right],
\]

(4.6)

where \(N\) stands for the total number of lattice sites.

We then perform a variational search by minimizing Eq. (4.6) with respect to \(\vec{d}_i\), where the directors \(\vec{d}_i\) are restricted on \(2 \times 2\) and \(4 \times 4\) unit cells with periodic boundary condition. The purely quadrupolar states are identified with vanishing magnetic moment:

\[
\langle S_i \rangle \equiv 2 \text{Re}[\vec{d}_i] \times \text{Im}[\vec{d}_i] = 0, \forall i.
\]

Among the quadrupolar states, one distinguishes a FQ phase, with all directors parallel, and more general AFQ phases with non-collinear directors. The familiar magnetic phases corresponds to dipolar moment \(|\langle S_i \rangle| = 1, \forall i\) with a spin structure factor characterized by the Bragg peaks. In general, one also encounters states that contain a mixture of magnetic and quadrupolar moments with \(0 < |\langle S_i \rangle| < 1\) on all sites, or states which have purely magnetic/quadrupolar moments only on partial sites, or even so-called semi-ordered states with undetermined \(|\langle S_i \rangle|\) \cite{146}.

The variational mean-field phase diagram is given in Fig. 4.1 obtained for antiferromagnetic \(J_1 > 0\) and \(J_2/J_1 = 0.8\), which were deduced by fitting the INS spectra for BaFe\(_2\)As\(_2\) \cite{103} to the \(J_1 - J_2 - K_1\) spin model \cite{98, 145}. Due to the fact that FeSe lies in proximity to CAFM, we do not expect its parameters to deviate dramatically from those deduced in Refs. \cite{98, 145}, and we have also verified that the magnetic and quadrupolar phases in Fig. 4.1 are robust to small variations of \(J_2/J_1\). Remarkably, Fig. 4.1 shows that the only non-magnetic phase in close proximity to CAFM is the FQ phase, with both phases realized at negative biquadratic interaction \(K_1\). We note that \(K_1 < 0\) is generically expected from the fitting of the INS spectra in the iron
pnictides/chalcogenides [98, 145], with the ratio $|K_1|/J_1$ of order 1. This is indeed consistent with the location of CAFM region in Fig. 4.1. No other purely quadrupolar phases were found; in particular the AFQ($\pi$, 0)/0, $\pi$) phase, expected to be realized for positive $K_2$ [142] turns out to be unstable to the admixture of the magnetic moment, resulting in a mixed magnetic/quadrupolar state with $0 < |\langle S_i \rangle| < 1$ (grey region in Fig. 4.1) [147].

Figure 4.1: Variational mean-field phase diagram of the Hamiltonian Eq. (4.1) with $J_1 = 1$, $J_2 = 0.8$ and periodic boundary condition (2 × 2 and 4 × 4 unit cells yield exactly the same results). The dashed lines denote shifted phase boundaries when breaking $C_4$ symmetry in Eq. (4.1) by hand, using $J_{x,y}^z = (1 \pm 0.2)J_1$.

For completeness, we also write down the energies of several typical phases of
interest:

\[ FM : E_0 = 2J_1 + 2J_2 + 2J_3 + 2K_1 + 2K_2 + 2K_3; \]
\[ \text{AFM}(\pi, 0)/(0, \pi) : E_0 = -2J_2 + 2J_3 + 3K_1 + 4K_2 + 2K_3; \]
\[ \text{AFM}(\pi, \pi) : E_0 = -2J_1 + 2J_2 + 2J_3 + 4K_1 + 2K_2 + 2K_3; \]
\[ FQ : E_0 = 4K_1 + 4K_2 + 4K_3; \]
\[ \text{AFQ}(\pi, 0)/(0, \pi) : E_0 = 3K_1 + 2K_2 + 4K_3; \]
\[ \text{AFQ}(\pi, \pi) : E_0 = 2K_1 + 4K_2 + 4K_3. \] (4.7)

By restricting ourselves to consider only these phases listed in Eq. (4.7), we can obtain a biased mean-field phase diagram Fig. 4.2. We note that the AFQ \((\pi, 0)/(0, \pi)\) phase in Fig. 4.2 is energetically unfavorable in the full variational treatment, and will be replaced by the region \(0 < |S_i| < 1\) in Fig. 4.1.

![Figure 4.2](image-url)

**Figure 4.2**: Biased mean-field phase diagram obtained from comparing energies listed in Eq. (4.7), with \(J_1 = 1\), \(J_2 = 0.8\), and \(J_3 = K_3 = 0\).
4.4 Density Matrix Renormalization Group (DMRG)

Since the variational mean-field calculation only takes into account minimally entangled mean-field states, the results in Fig. 4.1 may be energetically unfavorable upon quantum fluctuations. To verify the stability of the FQ phase, we have performed the SU(2) DMRG calculations [148, 149, 150, 151] on \( L \times 2L \) rectangular cylinders with \( L = (4, 6, 8) \) near the mean-field FQ/CAFM phase boundary. We keep up to 4000 SU(2) states, leading to truncation errors less than \( 2 \times 10^{-5} \) in all data points presented in this work. In Fig. 4.3, we show both the static spin and quadrupolar structure factors, defined as

\[
m^2_S(q) = \frac{1}{L^4} \sum_{ij} \langle S_i \cdot S_j \rangle e^{iq(r_i-r_j)},
\]

\[m^2_Q(q) = \frac{1}{L^4} \sum_{ij} \langle Q_i \cdot Q_j \rangle e^{iq(r_i-r_j)},\]

where \( i, j \) are only partially summed on \( L \times L \) sites in the middle of the cylinder, in order to reduce boundary effects [152, 26, 150, 153]. Fig. 4.3(a)(b) show the results for \( m^2_S(q) \) in the FQ and CAFM phases, respectively; Fig. 4.3(c)(d) depict \( m^2_Q(q) \) in these two phases. Since \( m^2_S(q) \) and \( m^2_Q(q) \) are maximized near \( (0, \pi) \) and \( (0, 0) \) respectively, we fix \( q \) at these two momenta, and perform finite size scaling analysis of \( m^2_S(q) \) and \( m^2_Q(q) \) in Fig. 4.3(e)(f). For large negative \( K_1 \), it is clearly shown that the \( m^2_S(0, \pi) \) is suppressed from \( L = 4 \) to 8, and vanishes in the thermodynamic limit by extrapolation; while \( m^2_Q(0, 0) \) remains finite, confirming FQ as the underlying phase. For small negative \( K_1 \), \( m^2_S(0, \pi) \) remains finite in the thermodynamic limit, confirming the corresponding phase to be CAFM. We note that the DMRG yields a larger FQ region with the FQ/CAFM boundary found at \( K_1 > -1.4 \), compared to

\*\( L \) represents the size of y-direction which has periodic boundary condition
the mean-field prediction of $K^c_1 = -1.6$ in Fig. 4.1.

Figure 4.3: Static spin and quadrupolar structure factors obtained from DMRG on RCL–2L cylinders with $J_1 = 1, J_2 = 0.8, K_2 = -1$. (a)(b) $m^2_S(q)$ for $L = 8$. (c)(d) $m^2_Q(q)$ for $L = 8$. (e)(f) Finite-size scaling of $m^2_S(q = (0, \pi))$ and $m^2_Q(q = (0, 0))$ as a function of the inverse cylinder width, where the lines are guide to the eye.

4.5 Spin Excitations and Comparisons to Inelastic Neutron Scattering Experiments

Having established FQ as a stable non-magnetic phase in close proximity to CAFM, we turn to the analysis of its magnetic excitations.

To be general, we formulate the calculation with an applied magnetic field $-h \sum_i S_i^z$; then the directors in the FQ phase become:

$$\vec{d}_i = (\cos(\mu/2), i \sin(\mu/2), 0),$$  (4.9)
where \( \mu \) is determined by minimizing Eq. [4.6] which in an FQ state gives:

\[
\sin \mu = \frac{h}{4(J_1 + J_2 - K_1 - K_2)}.
\] (4.10)

Then we use the flavor-wave technique, which represents the local spin and quadrupolar operators \( O_i \) in terms of three flavors of Schwinger bosons in the fundamental representation of SU(3) [146, 154, 155, 156]:

\[
O_i = \sum_{\alpha \beta} b_{i,\alpha}^\dagger O_{\alpha \beta}^{\alpha \beta} b_{i,\beta},
\] (4.11)

subject to the constraint \( \sum_{\alpha} b_{i,\alpha}^\dagger b_{i,\alpha} = 1 \).

Further, we perform a unitary transformation according to the directors in the magnetic field:

\[
\tilde{b}_i = \mathcal{V}_i^\dagger b_i,
\] (4.12a)

\[
\tilde{S}_i^\nu = \mathcal{V}_i^\dagger S_i^\nu \mathcal{V}_i,
\] (4.12b)

\[
\tilde{Q}_i^\nu = \mathcal{V}_i^\dagger Q_i^\nu \mathcal{V}_i,
\] (4.12c)

where the transformation matrix \( \mathcal{V}_i \) is identical on all sites \( i \) in an FQ state:

\[
\mathcal{V}_i = \begin{pmatrix}
  i \sin(\mu/2) & 0 & \cos(\mu/2) \\
  \cos(\mu/2) & 0 & i \sin(\mu/2) \\
  0 & 1 & 0
\end{pmatrix}.
\] (4.13)

The third component of \( b_i \) is condensed, by expanding

\[
\tilde{b}_{i,3} = \tilde{b}_{i,3} = \sqrt{1 - \tilde{b}_{i,1}^\dagger \tilde{b}_{i,1} - \tilde{b}_{i,2}^\dagger \tilde{b}_{i,2}}.
\] (4.14)

The Hamiltonian expanded up to quadratic level can be diagonalized by the Bogoliubov transformation

\[
\alpha_{q,a} = \cosh \theta_{q,a} b_{q,a} - \sinh \theta_{q,a} b_{q,a}^\dagger,
\] (4.15)
up to a constant it gives:

\[ H_{tw} = \sum_{a=1,2} \sum_q \omega_{q,a}(\alpha_{q,a}^\dagger \alpha_{q,a} + 1/2), \]  
(4.16)

where the dispersion \( \omega_{q,a} \):

\[ \omega_{q,a} = \sqrt{(2t_{aa}(q) + \lambda_{aa})^2 - 4\Delta_{aa}(q)^2}, \]  
(4.17)

and the Bogoliubov coefficients:

\[ \tanh 2\theta_{q,a} = -\frac{2\Delta_{aa}(q)}{2t_{aa}(k) + \lambda_{aa}}, \]  
(4.18)

where \( t_{ab}(q), \Delta_{ab}(q) \) and \( \lambda_{ab} \) are \( 2 \times 2 \) diagonal matrices in FQ:

\[ t_{11}(q) = (J_1 \cos^2 \mu + K_1 \sin^2 \mu)(\cos q_x + \cos q_y) \]
\[ + 2(J_2 \cos^2 \mu + K_2 \sin^2 \mu) \cos q_x \cos q_y, \]  
(4.19a)

\[ t_{22}(q) = J_1(\cos q_x + \cos q_y) + 2J_2 \cos q_x \cos q_y, \]  
(4.19b)

\[ \Delta_{11}(q) = (K_1 - J_1) \cos^2 \mu (\cos q_x + \cos q_y) \]
\[ + 2(K_2 - J_2) \cos^2 \mu \cos q_x \cos q_y, \]  
(4.19c)

\[ \Delta_{22}(q) = (K_1 - J_1) \cos \mu (\cos q_x + \cos q_y) \]
\[ + 2(K_2 - J_2) \cos \mu \cos q_x \cos q_y, \]  
(4.19d)

\[ \lambda_{11} = -8(J_1 + J_2 - K_1 - K_2) \sin^2 \mu \]
\[ - 4(K_1 + K_2) + 2h \sin \mu, \]  
(4.19e)

\[ \lambda_{22} = -4(J_1 + J_2 - K_1 - K_2) \sin^2 \mu \]
\[ - 4(K_1 + K_2) + h \sin \mu. \]  
(4.19f)

The dynamical spin structure factor at \( T = 0 \) is defined as:

\[ S^{\alpha\beta}(q, \omega) = \sum_f \langle \text{g.s.}|S^{\alpha}(q)|f\rangle \langle f|S^{\beta}(-q)|\text{g.s.}\rangle \delta(\omega - E_f + E_g). \]  
(4.20)
The spin operators in Eq. (4.20) are represented by the SU(3) bosons, keeping only the linear order terms (neglecting the two-magnon continuum and the constant background):

\begin{align}
S_x(q) &= -\sin \frac{\mu}{2} \left( \tilde{b}_{-q,2}^\dagger + \tilde{b}_{q,2} \right), \\
S_y(q) &= -i \cos \frac{\mu}{2} \left( \tilde{b}_{-q,2}^\dagger - \tilde{b}_{q,2} \right), \\
S_z(q) &= i \cos \mu \left( \tilde{b}_{-q,1}^\dagger - \tilde{b}_{q,1} \right). 
\end{align}

(4.21a, 4.21b, 4.21c)

Then Eq. (4.20) can be written down explicitly:

\begin{align}
S^{xx}(q, \omega) &= \sin^2 \frac{\mu}{2} \frac{2t_{22}(q) + \lambda_{22} - 2\Delta_{22}(q)}{\omega_{q,2}} \delta(\omega - \omega_{q,2}), \\
S^{yy}(q, \omega) &= \cos^2 \frac{\mu}{2} \frac{2t_{22}(q) + \lambda_{22} + 2\Delta_{22}(q)}{\omega_{q,2}} \delta(\omega - \omega_{q,2}), \\
S^{zz}(q, \omega) &= \cos^2 \frac{\mu}{2} \frac{2t_{11}(q) + \lambda_{11} + 2\Delta_{11}(q)}{\omega_{q,1}} \delta(\omega - \omega_{q,1}). 
\end{align}

(4.22a, 4.22b, 4.22c)

With zero magnetic filed, the dispersion $\omega_{q,a}$ are degenerate in flavor index $a = \{1, 2\}$, shown in Fig. 4.4(a). Since FQ phase spontaneously breaks the spin-rotational symmetry, there are two gapless Goldstone modes at $q = 0$. However there is no Bragg peak as the dynamical spin structure factor $S(q, \omega)$ shown in Fig. 4.4(b) has a vanishing spectral weight ($\propto |q|$) at $q=0, \omega=0$ because of the conservation of time reversal symmetry in quadrupolar states \[155, 154, 157, 158\]. In Fig. 4.4(b), we see large spectral weight at $Q_{1,2}$ at low energy due to the proximity to the CAFM phase. The spectral weight further shifts towards $(\pi, \pi)$ when increasing $\omega$ (see Fig. 4.4(c-f)), closely tracking the INS results on FeSe \[137, 138, 139, 140\]. We note that in the AFQ $(\pi, 0)/(0, \pi)$ phase proposed in Ref. \[142\], one would expect Goldstone modes with zero spectral weight at $Q_{1,2}$, which would contradict the large-intensity dispersing feature near $Q_{1,2}$ found in the INS data on FeSe.
Figure 4.4: Dispersion and dynamical spin structure factor in the FQ phase obtained from flavor-wave calculation with $J_1 = 1, J_2 = 0.8, K_1 = -1.65, K_2 = -0.8$. (a) Dispersion plotted in the 1st BZ. (b) Energy-momentum dependence of $S(q, \omega)$. (c)-(f) Constant-energy cuts of $S(q, \omega)$ in $q$-space. (c) $\omega/J_1 = 2$. (d) $\omega/J_1 = 4$. (e) $\omega/J_1 = 6$. (f) $\omega/J_1 = 8$. A Lorentzian broadening factor $\lambda = 0.8J_1$ is used for approximating the delta-functions.
With finite magnetic field, since we break the time-reversal symmetry by hand, then the degeneracy of the two Goldstone modes is lifted (see Fig. 4.5). This effect can be verified with future experiments to distinguish if the non-magnetic state is indeed FQ.

\[ J_1 = 1, J_2 = 0.8, K_1 = -1.65, K_2 = -0.8, h = 2(J_1 + J_2 - K_1 - K_2), \]

and a Lorentzian broadening factor \( \lambda = 0.8J_1 \).

4.6 Interplay of FQ and Nematicity

Having demonstrated that FQ phase is indeed consistent with the INS results on FeSe \cite{137, 138, 139, 140}, we now ask further whether FQ phase can coexist with nematicity observed in FeSe. We apply \( C_4 \) breaking exchange anisotropy in Eq. (4.1), using \( J_1^{x,y} = (1 \pm 0.2)J_1 \) in the variational mean-field calculation. This results in the shift of the phase boundaries (shown with dashed lines in Fig. 4.1) and although the FQ phase shrinks slightly, it clearly remains stable in a large portion of the mean-field phase diagram.
We now turn to the microscopic origin of nematicity in FeSe – can FQ order be the reason for the discrete $C_4$ symmetry breaking? Unlike other proposals starting with nematic spin wavefunctions in the ground state [141, 142, 143], in the flavor wave theory up to bilinear terms in Eq. (4.16), the spin correlations in FQ phase are $C_4$ symmetric. This does not mean that the FQ ground state cannot spontaneously break this symmetry and in fact, it turns out that higher order interactions (mode-mode coupling) become increasingly important when approaching the FQ/CAFM phase boundary. Collecting up to the 4th order terms in the flavor wave theory [147], we obtain

$$H_{\text{4th}} = H_{\text{fw}} + H_{\text{int}}$$

where

$$H_{\text{int}} = \frac{1}{N} \sum_{abcd} \sum_{k_1, k_2, q} V_{ab}^{cd}(k_1, k_2, q) \alpha^{\dagger}_{k_1 + q, a} \alpha^{\dagger}_{k_2 - q, b} \alpha_{k_2, c} \alpha_{k_1, d},$$

(4.23)

where only five combinations of $\{abcd\}$ are nonzero: $\{1111\}$, $\{2222\}$, $\{1122\}$, $\{2211\}$ and $\{1221\}$. Above, only particle number conserving terms have been kept for simplicity.

In terms of Schwinger bosons, we can define a nematic order parameter as $\langle \Delta \rangle = \sum_a \langle n_{Q_1,a} - n_{Q_2,a} \rangle$, where $\langle \ldots \rangle$ denotes the expectation value in the full interacting Hamiltonian $H_{\text{4th}}$, and $n_{q,a} = \alpha^{\dagger}_{q,a} \alpha_{q,a}$ is the boson density operator of flavor $a$ at momentum $q$. If we stop at the quadratic level of flavor wave theory, then $\langle \Delta \rangle_{\text{fw}} \equiv 0$ due to the Bose-Einstein condensation at $q = (0,0)$. Once interactions are taken into account in $H_{\text{4th}}$, the condensate will become depleted, resulting in a finite boson density at the local minima $Q_{1,2}$ of the spectrum in Fig. 4.4(a) and thus making it possible, in principle, that $\langle \Delta \rangle \neq 0$. To see how this may occur, we consider the density-density interactions between the $Q_{1,2}$ modes, which can be extracted from Eq. (4.23) as:

$$H_{\text{int}} = \bar{V}(n_{Q_1,1}n_{Q_2,1} + n_{Q_1,2}n_{Q_2,2}) + \bar{V}' n_{Q_1,1}n_{Q_2,2} + \ldots,$$

(4.24)
where the intra-flavor and inter-flavor interactions $\tilde{V}$ and $\tilde{V}'$ are expressed through $V_{cd}^{ab}(k_1, k_2, q)$ in Eq. (4.23).

The values of $\tilde{V}$ and $\tilde{V}'$ are plotted in Fig. 4.6. Intriguingly, they are repulsive in the region $K_1 > -3$, and diverge when approaching the FQ/CAFM phase boundary at $K_1^c = -1.6$, resulting in a $C_4$ symmetry-breaking imbalance in boson occupation $n_{Q_1} \neq n_{Q_2}$. Since sufficiently strong (not necessarily diverging) interactions can commonly trigger diverging susceptibilities, we expect the renormalized nematic susceptibility to diverge before reaching the FQ/CAFM phase boundary, resulting in a finite nematic window $K_1^N < K_1 < K_1^c$ inside the FQ phase. The existence of such a window should be carefully verified by further analytical and numerical efforts, which will be a subject of future work. We note that while the present study is limited to second-neighbor interactions, our mean-field analysis shows that inclusion of third neighbor $K_3^3(S_i \cdot S_j)^2$ term with $K_3 < 0$ will further favor FQ over magnetic phases, possibly leading to a wider nematic region.
4.7 Conclusion and Discussion

Direct experimental measurements of quadrupolar orders are typically difficult, due to the negligible spectral weight of the spin structure factor near the ordering wave-vector. A possible way to visualize such “ghost” modes is by applying a magnetic field: the degeneracy of the two flavors will be lifted, and one of the Goldstone modes acquires a gap and a visible spectral weight \[157, 158\], as we demonstrate in \[147\]. More direct evidence can be gained from the quadrupolar structure factor, which should exhibit Bragg peaks at the ordering wave-vector, and in principle can be measured by resonant inelastic X-ray scattering experiments \[159\].

In summary, we showed that FQ phase lies in close proximity to CAFM in the phase diagram of a bilinear biquadratic spin-1 model and that it is stable in a realistic range of the model parameters, as verified by both the mean-field and DMRG methods. The dynamical spin structure factor \(S(q, \omega)\) inside the FQ phase is shown to be qualitatively consistent with the recent INS results on FeSe. While at the quadratic level the FQ ground state does not explicitly break the \(C_4\) lattice symmetry, we demonstrate that the quantum fluctuations result in repulsive density-density interactions between \(Q_{1,2}\) magnon modes, whose strength diverges on approaching the FQ/CAFM phase boundary. This suggests the existence of a finite window inside the non-magnetic FQ phase where the \(C_4\) symmetry is spontaneously broken. Further studies are necessary to establish such nematic window unequivocally, however, even if the nematicity is driven by other sources (for example, local strains due to lattice imperfections; or orbital ordering, as proposed in the light of recent nuclear magnetic resonance \[160, 161\] and ARPES \[162\] experiments), the incipient nematic order will couple to the symmetry-breaking quantum fluctuations that we found in the FQ phase. Our calculations show that the FQ order is robust with respect to
such $C_4$ breaking environments and can coexist with nematicity.
Chapter 5

Orbital nematic order and interplay with magnetism in the two-orbital Hubbard model

5.1 Introduction

Nematicity, defined as spontaneous breaking of the four-fold rotational $C_4$ symmetry down to $C_2$, has been recently observed in the electronic properties of Fe-based superconductors\[1, 68\]. Experimental efforts, including transport measurements\[115, 132, 163, 164, 165, 133, 166, 167\], optical conductivity\[132, 168, 169, 170\], scanning tunneling microscopy\[171, 172, 173\], neutron scattering\[103, 174, 175\], quantum oscillations\[176\], magnetic torque measurements\[177\], and angle-resolved photoemission spectroscopy (ARPES) measurements\[178, 114, 179\], have reported the electronic in-plane anisotropy, mostly in the compounds of the BaFe$_2$As$_2$ (122) family, even at temperatures higher than the lattice structural transition\[115, 133, 177, 175, 114, 179\]. Upon electron doping, the resistivity anisotropy is first enhanced in the underdoped region, but then suppressed upon further doping into the superconducting region\[115\]; while hole doping appears to suppress the resistivity anisotropy even while below the structural phase transition\[165\].

Origins of the observed anisotropy have been discussed in the context of lattice, magnetic and orbital fluctuations\[75, 76, 78, 180, 181, 182, 94, 183, 95, 184, 185\], and there are also debates whether the nematicity is intrinsic or if it comes from the anisotropic impurity scattering\[167, 173, 186\]. Recent resistivity $\rho$ measurements
under fixed strain\[133\] $\delta = (a - b)/(a + b)$ ($a$ and $b$ are the lattice constants) have detected divergent nematic susceptibility $d\rho/d\delta$, proving that the nematicity is of electronic origin rather than due to an elastic lattice instability.

One possible mechanism behind this electronic nematicity is the so-called Ising-nematic (also referred to as “spin nematic”) scenario, based on the discrete Ising symmetry breaking between two columnar antiferromagnetic (CAF) ordering wavevectors, $Q_1 = (\pi, 0)$ and $Q_2 = (0, \pi)$. In the ordered CAF phase, the magnetization $M(r) = M_1 e^{iQ_1 \cdot r} + M_2 e^{iQ_2 \cdot r}$ breaks the $C_4$ symmetry whenever $M_1 \neq M_2$, and neutron scattering finds either $M_1 = 0$ or $M_2 = 0$ in the iron pnictides\[30, 103\]. The nematic order parameter is defined as the difference in spin correlations along the $\hat{x}$ and $\hat{y}$ axes:

$$\psi = \sum_i |M(i) \cdot M(i + \hat{x}) - M(i) \cdot M(i + \hat{y})| \propto |M_1|^2 - |M_2|^2.$$  \hspace{1cm} (5.1)

However, the $C_4$ symmetry can be spontaneously broken even above the Néel temperature ($T_N$) due to anisotropic spin fluctuations, as was first pointed out by Chandra, Coleman and Larkin\[19\], and later applied to the iron pnictides\[75, 76, 78, 116, 187\]. Recently, these anisotropic spin fluctuations have been imaged directly\[175\] using the inelastic neutron scattering in uniaxial-pressure detwinned samples of BaFe$_2$$_{2-x}$Ni$_x$As$_2$.

On the other hand, it has been proposed that the electronic nematicity may stem from unequal population of the $d_{xz}$ and $d_{yz}$ orbitals, resulting in the ferro-orbital ordering\[94, 180, 181, 182, 183, 95\]. The order parameter is the orbital polarization $p = \langle n_{xz} - n_{yz} \rangle$, which explicitly breaks the $C_4$ symmetry. Experimentally, polarization-dependent ARPES found orbitally-polarized Fermi surfaces inside the CAF phase of the parent compound BaFe$_2$As$_2$\[178\]. The unambiguous splitting of Fe $d_{xz}$ and $d_{yz}$ orbitals has also been observed by ARPES above the structural transition temperature $T_s$ in the detwinned samples of lightly Co-doped\[114\] and P-doped\[179\]
BaFe$_2$As$_2$.

The driving force for the electronic nematic transition is very difficult to determine because of the strong coupling between internal spin and orbital degrees of freedom. Indeed, on the symmetry grounds, Landau free energy will contain a linear coupling between the Ising-nematic order parameter $\psi$ and the orbital ordering $p$:

$$\Delta F = \eta p \cdot \psi \propto \eta p \cdot \langle M_1^2 - M_2^2 \rangle .$$

Independent of the sign of the coupling constant $\eta$, such a term in the free energy will result in a non-vanishing value of the orbital polarization whenever $\psi$ takes on a non-zero value, and the other way round. It has been proposed that this “chicken and egg” problem may in principle be resolved by comparing the rates of divergence in the orbital and Ising-spin susceptibilities on approaching the transition, however this approach would only work provided the coupling constant $|\eta|$ is not too large.

In order to disentangle the effects of the magnetic and orbital ordering, it is useful to consider the compounds where the two phases are clearly separated. One such example is stoichiometric FeSe, which we considered in Chapter 4. FeSe undergoes a structural transition at around $T_s \approx 90$ K without any sign of the antiferromagnetic ordering. Recent ARPES measurements on FeSe show a clear splitting of $\sim 50$ meV between the energies of the Fe $d_{xz}$ and $d_{yz}$ orbital bands, which sets in at about $T_s$. Importantly, this measured energy splitting is more than five times larger than expected from density-functional theory (DFT) calculations considering the orthorhombic lattice distortion alone. The likely orbital nature of the structural transition in FeSe is also corroborated by recent nuclear magnetic resonance (NMR) and shear modulus measurements. In Chapter 4 we have discussed about the spin aspect in the nematic phase of FeSe, which is possibly a spin ferroquadrupolar phase. But we haven’t fully addressed the origin of nematicity,
which could be rooted in the multi-orbital physics in FeSe.

Another example is NaFeAs of the 111 family with \( T_s = 53 \) K significantly higher than the Néel temperature \( T_N = 40 \) K. Recent ARPES measurements in NaFeAs indicate\cite{190, 191} that the orbital order develops exactly at or slightly above \( T_s \) and appears to trigger the antiferromagnetic order at the lower temperature \( T_N \), due to the nesting of the two-fold anisotropic Fermi surface \cite{190}. One is tempted therefore to interpret this result based on the orbital-driven scenario\cite{190}. Intriguingly, scanning tunneling spectroscopy finds evidence of local electronic nematicity up to temperatures twice \( T_s \), in the nominally tetragonal phase\cite{192}.

The above experimental observations raise a possibility that it may indeed be possible to stabilize ferro-orbital order in the absence of long-range magnetic ordering. It is the purpose of this chapter to address this question theoretically within the minimal two-orbital model.\cite{193, 194} While the two-orbital model is known to have a number of limitations in describing the iron-based superconductors (for instance, resulting in a wrong number of Fermi pockets and missing the \( d_{xy} \) orbital contents on the Fermi surface), the primary reason for using this model here is its conceptual simplicity. We do not presume that such a simple model can describe the realistic electronic properties of, e.g. FeSe or LiFeAs. Rather, the question we aim to address can be phrased as follows – what is the minimal theoretical model (whether or not applicable to the iron pnictides) that can support orbital nematic ordering in the absence of magnetism? In this chapter, we show that the two-orbital model is sufficient to describe this physics under realistic values of intra-orbital and inter-orbital Coulomb repulsion. Generalization of these results to a realistic five-orbital model of the iron pnictides or iron chalcogenided will be the subject of future work.

We used the combination of RPA and non-perturbative quantum cluster calcula-
lations (VCA) to study the effect of electron interactions and doping on both the antiferromagnetism and ferro-orbital ordering at zero temperature. Within the two-orbital model, we find that the orbital nematic order strongly depends on inter-orbital Hubbard repulsion and Hund’s coupling term, and its dependence on electron and hole doping is not symmetric. In the undoped and hole-doped system, we find that orbital order coexists with magnetic order, as observed ubiquitously in the 122 family of iron pnictides. Our key finding is that sufficient electron doping stabilizes the orbital nematic phase while suppressing the antiferromagnetic ordering, suggestive of the experimental observations of orbital order in Co-doped LiFeAs\cite{195} and FeSe\cite{162,161,160}.

This chapter is organized as follows: In Section 5.2 we introduce the two-orbital Hubbard model as a starting point for our calculations; in Section 5.3 and 5.4 we study the orbital nematic order and its interplay with the magnetic order by using RPA and VCA methods, respectively; in Section 5.5 we discuss our results in the context of the related theoretical and experimental work, and we finally draw the conclusions in Section 5.6.

5.2 Model

To study the orbital nematic order in Fe-based superconductors, we start from the minimal two-orbital Hubbard model capturing the itinerant electrons with onsite electron-electron interactions:

\[
H = H_0 + U \sum_{i,\alpha} n_{i\alpha \uparrow} n_{i\alpha \downarrow} + \left( U' - \frac{J}{2} \right) \sum_{i,\alpha < \beta} n_{i\alpha} n_{i\beta} \\
- 2J \sum_{i,\alpha < \beta} \mathbf{S}_{i\alpha} \cdot \mathbf{S}_{i\beta} + J' \sum_{i,\alpha < \beta} (c_{i\alpha \uparrow}^\dagger c_{i\alpha \downarrow}^\dagger c_{i\beta \downarrow} c_{i\beta \uparrow} + h.c.)
\] (5.3)
Here \( i \) is the site label, \( \alpha, \beta \in \{xz, yz\} \) stand for the orbital indices. \( U \) and \( U' \) are respectively the intra- and inter-orbital Hubbard interaction, \( J \) stands for the Hund’s coupling and \( J' = J \) stands for the pair hopping term. \( H_0 \) is the non-interacting two-orbital Hamiltonian from Ref. [193]:

\[
H_0 = \sum_{k\sigma} \psi_{k\sigma}^\dagger \left[ \epsilon_+(k) \mathbb{1} + \epsilon_-(k) \tau_3 + \epsilon_{xy}(k) \tau_1 \right] \psi_{k\sigma} \tag{5.4}
\]

with \( \psi_{k\sigma}^\dagger = [c_{xz,\sigma}^\dagger(k), c_{yz,\sigma}^\dagger(k)] \), where \( \sigma \) is the spin label, and

\[
\epsilon_\pm(k) = \frac{\epsilon_x(k) \pm \epsilon_y(k)}{2}
\]

\[
\epsilon_x(k) = -2t_1 \cos k_x - 2t_2 \cos k_y - 4t_3 \cos k_x \cos k_y \tag{5.5}
\]

\[
\epsilon_y(k) = -2t_2 \cos k_x - 2t_1 \cos k_y - 4t_3 \cos k_x \cos k_y
\]

\[
\epsilon_{xy}(k) = -4t_4 \sin k_x \sin k_y
\]

The values of the hopping parameters have been kept fixed throughout the context: \( t_1 = -0.25eV, t_2 = 0.325eV, t_3 = t_4 = -0.2125eV \).

The reason behind studying the two-orbital (as opposed to the full five-orbital) model is the universally accepted fact that the major contribution to the Fermi surface comes from the iron \( t_{2g} \) orbitals \( (d_{xz}, d_{yz}, d_{xy}) \), whereas the \( e_g \) orbital weight is very small[70, 196]. Additionally, the \( d_{xz} \) and \( d_{yz} \) orbitals carry most of the spectral weight[70] and the \( d_{xy} \) orbital can thus be neglected in the first approximation. While it is true that in order to obtain all the Fermi pockets observed in ARPES, one needs to consider all 5 Fe orbitals[197], here we chose to focus on the two-orbital model in the hope that it captures the salient features of nematicity in the iron pnictides, especially because the ferro-orbital nematic order only affects the two \( d_{xz} \) and \( d_{yz} \) orbitals in question. We expect that our central results will remain unaltered upon inclusion of the other orbitals, however demonstrating this explicitly will be the subject of future work. We note that the present approach is similar in spirit to the weak-coupling
approaches [144, 187, 198, 120] which start from the band picture of a hole pocket around the Γ-point and electron pockets in the corners of the Brillouin zone, in a sense that these approaches also deal with a reduced Hilbert space of typically two bands (irrespective of their orbital contents). Nevertheless, even such a simplified two-band approach is known to produce reliable results which are largely unaffected by inclusion of other bands [120].

We point out that another, more pragmatic reason for limiting the present consideration to two orbitals is because the five-orbital model is well beyond the computational demands of the state-of-the-art variational cluster approximation used in this work (see section 5.4 for more detail). Studying antiferromagnetism necessitates the use of a 4-site cluster, which when combined with 5 orbitals per Fe site, would result in an effective 20-“site” Hubbard model that lies well beyond the present limits of either the exact diagonalization or continuous-time quantum Monte Carlo solver [199] used in VCA or in cluster-DMFT.

The inter- and intra-orbital interaction strengths in Eq. (5.3) are not independent of each other. In the atomic limit, a well known relation $U' = U - 2J$ holds, which ensures orbital rotational invariance [200]. In a solid, the electron-electron interactions are screened, meaning that the above relation between $U'$ and $U$ may not be obeyed exactly. Below, we shall investigate the phase diagram of the model Eq. (5.3) treating $U'$ and $U$ as independent parameters. However later on, when studying the effect of interactions on nematicity and antiferromagnetism, we shall use the relation $U' = U - 2J$ which we expect to hold approximately in the iron pnictides. The values of interactions for the 122 Fe-pnictides in the two-orbital model are approximately $U = 2$ eV, $U' = 0.6$ eV, $J = 0.7$ eV. Note that we chose the interaction $U$ to be somewhat lower than $U = 2.7$ eV calculated within the ab initio constrained-
RPA scheme [201], which is consistent with the smaller effective bandwidth when considering only \( d_{xz} \) and \( d_{yz} \) orbitals in Eq. (5.3), compared to the width of all 5 Fe bands. When we attempted to use larger values of \( U \gtrsim 2.5 \text{ eV} \), the variational cluster calculations (see Sec. 5.4 below) indicate the parent compound to be a Mott insulator. Therefore, to keep the system metallic in agreement with experiments, we were forced to choose a lower value of \( U = 2 \text{ eV} \).

### 5.3 Random Phase Approximation

For the case when interaction is not too strong, we can treat the Hubbard terms and Hund’s term as perturbation, and perform an RPA calculation for both the spin-spin correlation function and orbital nematic density-density correlation function:

\[
\chi_{\text{spin}}(k, i\omega_n) \equiv 2 \int_0^\beta d\tau \sum_{\alpha_1\alpha_2} e^{i\omega_n \tau - i kr} \langle T\hat{S}_{\alpha_1}^z(r, \tau)\hat{S}_{\alpha_2}^z(0, 0) \rangle \tag{5.6}
\]

\[
\chi_{\text{nematic}}(k, i\omega_n) \equiv \frac{1}{2} \int_0^\beta d\tau \sum_r e^{i\omega_n \tau - i kr} \langle T(\hat{n}_{xz}(r, \tau) - \hat{n}_{yz}(r, \tau)) \cdot (\hat{n}_{xz}(0, 0) - \hat{n}_{yz}(0, 0)) \rangle \tag{5.7}
\]

Where \( \alpha_1, \alpha_2 \) are orbital indices. Summation over spin indices has been made implicit by writing \( \hat{n}_n = \hat{n}_{n\uparrow} + \hat{n}_{n\downarrow} \).

The bare spin susceptibility is of the form:

\[
\chi_{\text{spin}}^{(0)}(k, i\omega_n) \equiv \sum_{\alpha_1\alpha_2} \left( \chi^{(0)}_{\text{spin}} \right)_{\alpha_1\alpha_2} = \frac{-1}{2\beta N} \sum_{\omega_m, \alpha_{s_1s_2}} \sum_{\alpha_1\alpha_2} \left[ \sigma_{s_1s_2} \tilde{G}_0^{\alpha_1\alpha_2}(k + q, i\omega_n + i\omega_m) \cdot \sigma_{s_1s_2} \tilde{G}_0^{\alpha_2\alpha_1}(q, i\omega_m) \right] = -(D_0^{xx}(k, i\omega_n) + D_0^{yy}(k, i\omega_n) + 2D_0^{xy}(k, i\omega_n)) \tag{5.8}
\]

where \( \tilde{G}_0^{\alpha\beta} \) is the non-interacting Green’s function and the susceptibility matrix \( \chi^{(0)}_{\text{spin}} \) depends on two orbital indices, which has been summed over to obtain the final
(scalar) quantity. The explicit form of this matrix is as follows (\(\alpha_1, \alpha_2 = \{x, y\}\) is a shorthand notation for \(xz, yz\) orbitals):

\[
\chi^{(0)}_{\text{spin}} = \begin{pmatrix}
\chi^{(0)}_{xx} & \chi^{(0)}_{xy} \\
\chi^{(0)}_{yx} & \chi^{(0)}_{yy}
\end{pmatrix}
\]

\[-\begin{pmatrix}
D_{xx} & D_{xy} \\
D_{yx} & D_{yy}
\end{pmatrix}
\]  \hspace{1cm} (5.9)

Above, \(D_{\alpha\beta}^{\alpha\beta}(k, i\omega_n)\) is the 2 \(\times\) 2 matrix in the orbital basis denoting the bare susceptibility (bubble diagram):

\[
\chi^{(0)}_{\text{spin}} = - \begin{pmatrix}
\chi^{(0)}_{xx} & \chi^{(0)}_{xy} \\
\chi^{(0)}_{yx} & \chi^{(0)}_{yy}
\end{pmatrix}
\]

\[-\begin{pmatrix}
D_{xx} & D_{xy} \\
D_{yx} & D_{yy}
\end{pmatrix}
\]  \hspace{1cm} (5.9)

\[
D_{\alpha\beta}^{\alpha\beta}(k, i\omega_n) = \frac{1}{N} \sum_{q} \left( \langle \alpha | \nu_1, k + q \rangle \langle \nu_1, k + q | \beta \rangle \langle \beta | \nu_2, q \rangle \langle \nu_2, q | \alpha \rangle \right)
\times \frac{n_F(E_{\nu_1, q}) - n_F(E_{k+q, \nu_1})}{i\omega_n + E_{q, \nu_2} - E_{k+q, \nu_1}} \Gamma_{s_1 s_2}^{s_3 s_4} \Gamma_{\alpha \beta}^{s_3 s_4} \left( k, i\omega_n \right) \left( q, i\omega_m \right) (5.10)
\]

where \(E_{\nu, q}\) is the dispersion of the two non-interacting bands in Ref. [193] (\(\nu_1, \nu_2 = \{+, -\}\) are band indices):

\[
E_{\pm}(k) = \epsilon_{\pm}(k) \pm \sqrt{\epsilon_{\pm}^2(k) + \epsilon_{xy}^2(k)}. \hspace{1cm} (5.11)
\]

We note in passing that the most general bare susceptibility is a 4-point correlation function that can be written as a 4-rank tensor in both orbital indices \(\alpha_1, \ldots, \alpha_4\) and spin indices \(s_1, \ldots, s_4\) (see also Ref. [70]):

\[
(\chi_0)^{\alpha_1 \alpha_2 \alpha_3 \alpha_4}_{s_1 s_2 s_3 s_4} (k, i\omega_n)
\]

\[
= -\frac{1}{\beta N} \sum_{\omega_m, q} \Gamma_{\alpha \alpha}^{\alpha_1 \alpha_2} (k + q, i\omega_n + i\omega_m) \Gamma_{\alpha \alpha}^{\alpha_3 \alpha_4} (q, i\omega_m) (5.12)
\]

where the choice of the matrix \(\Gamma_{\alpha \alpha}^{s s'}\) depends on the type of susceptibility one calculates. For instance, in the case of spin susceptibility, \(\Gamma_{\alpha \alpha}^{s s'} = \delta_{\alpha \beta} \delta_2^{s s'}\), which is how we obtained Eq. (5.8) above.
One can similarly calculate the (bare) orbital nematic susceptibility, in which case the vertex matrix $\Gamma_{\alpha\bar{\beta}}^{ss'} = \tau_{\alpha\bar{\beta}}^{z} \delta_{ss'}$ is identity in spin indices:

$$\chi^{(0)}_{\text{nematic}}(k, i\omega_n) \equiv \sum_{\alpha_1\alpha_3} \left( \chi^{(0)}_{\text{nematic}} \right)_{\alpha_1\alpha_3}$$

$$= -\frac{1}{2\beta N} \sum_{\omega_m q} \sum_{\alpha_1\alpha_2\alpha_3\alpha_4} \left[ \tau_{\alpha_1\alpha_2}^{z} \tilde{G}_0^{\alpha_1\alpha_3}(k + q, i\omega_n + i\omega_m) \cdot \tau_{\alpha_3\alpha_4}^{z} \tilde{G}_0^{\alpha_4\alpha_2}(q, i\omega_m) \right]$$

$$= - (D_{xx}^{0}(k, i\omega_n) + D_{yy}^{0}(k, i\omega_n) - 2D_{xy}^{0}(k, i\omega_n)) \quad (5.13)$$

where

$$\chi^{(0)}_{\text{nematic}} = \begin{pmatrix} \chi^{(0)}_{xx} & \chi^{(0)}_{xy} \\ \chi^{(0)}_{yx} & \chi^{(0)}_{yy} \end{pmatrix}_{\text{nematic}}$$

$$= - \begin{pmatrix} D_{xx}^{0} & -D_{xy}^{0} \\ -D_{yx}^{0} & D_{yy}^{0} \end{pmatrix} \quad (5.14)$$

The 2x2 matrix $\chi^{(0)}_{\text{nematic}}$ is defined above such that the final results turn out to be in a simplified 2x2 matrix form (Eqs. 5.16, 5.17) although the intermediate steps in the RPA calculation actually involve more complicated tensor operations.

In order to now calculate the susceptibilities in the presence of the (weak) interactions, we sum over the RPA series of diagrams for the two susceptibilities, see Figs. 5.1 and 5.2. In the general form, the RPA renormalized susceptibility (before tracing out the external Pauli matrices) is calculated in the following way:

$$\left( \chi_{\text{RPA}} \right)_{s_1s_2s_3s_4}^{\alpha_1\alpha_2\alpha_3\alpha_4} = \left( \chi_{0} \right)_{s_1s_2s_3s_4}^{\alpha_1\alpha_2\alpha_3\alpha_4} - \left( \chi_{0} \right)_{s_1s_2t_1t_2}^{\alpha_1\alpha_2\beta_1\beta_2} V_{t_1t_2t_3t_4}^{\beta_1\beta_2\beta_3\beta_4} \left( \chi_{\text{RPA}} \right)_{t_3t_4s_3s_4}^{\beta_3\beta_4\alpha_3\alpha_4} \quad (5.15)$$

where $V$ is a tensor with 4-orbital indices $(\beta_1, \ldots, \beta_4)$ and 4-spin indices $(t_1, \ldots, t_4)$ of the interactions in the Hubbard model [70].

Following the orbital rotational invariant senario, we use $J' = J$, and all the results below are done by calculating explicitly the tensor form in Eq. (5.15), except for some digression for the case $J' = 0$ made in Eqs. (5.16)-(5.19). These simplified equations with $J' = 0$ are beneficial for an intuitive understanding of the dominant
interactions \{U, U', J\}, and we have checked that the following results (calculated with \(J' = J\)) do not change qualitatively even if we set \(J' = 0\).

For the case \(J' = 0\), after tracing out the corresponding external Pauli matrices (see Figs. 5.1, 5.2), the final results can be simplified in the 2x2 matrix form using the definitions in Eqs. 5.9 and 5.14:

\[
\chi_{\text{spin}} = \chi^{(0)}_{\text{spin}} \left( 1 - \begin{pmatrix} U & J \\ J & U \end{pmatrix} \chi^{(0)}_{\text{spin}} \right)^{-1}
\]  

\[
\chi_{\text{nematic}} = \chi^{(0)}_{\text{nematic}} \left( 1 + \begin{pmatrix} U & -2U' + J \\ -2U' + J & U \end{pmatrix} \chi^{(0)}_{\text{nematic}} \right)^{-1}
\]  

Figure 5.1: RPA diagrams for spin susceptibility.

When we sum over all components of the matrices, this yields the total RPA-renormalized (scalar) susceptibilities:

\[
\chi_{\text{spin}} = \chi_{xx} + \chi_{xy} + \chi_{yx} + \chi_{yy} = \frac{\chi^{(0)}_{\text{spin}} - 2(U - J) \det D_0}{1 - U \chi^{(0)}_{\text{spin}} - 2(U - J) D_0^{xy} + (U^2 - J^2) \det D_0}
\]  

(5.18)
\[ \chi_{\text{nematic}}(0) = \chi_{\text{nematic}}^{(0)} + 2(U + 2U' - J) \det D_0 \left/ \left\{ 1 + U \chi_{\text{nematic}}^{(0)} \right\} \right. \\
+ 2(U - 2U' + J)D_0^{xy} + [U^2 - (2U' - J)^2] \det D_0 \] (5.19)

where \( \chi_{\text{spin}}^{(0)} \) and \( \chi_{\text{nematic}}^{(0)} \) are defined in Eq. (5.8) and Eq. (5.13), and \( \det D_0 = D_0^{xx} D_0^{yy} - D_0^{xy} D_0^{yx} \). The explicit \( (k, i\omega_n) \) dependence of susceptibilities has been suppressed in Eqs. (5.18)-(5.19) for brevity.

From the expression of \( D_0^{\alpha\beta} \) (see Eq. (5.10)), Re\( D_0^{\alpha\beta} < 0 \) in general. We also notice that at \( q = 0 \), \( \det D_0(0, \omega) > 0 \), thus the ferro orbital nematic instability comes from the \(- (2U' - J)^2 \det D_0 \) term, i.e. the orbital nematic susceptibility at \( q = 0 \) only diverges when we have large enough inter-orbital Hubbard repulsion, and Hund’s coupling cannot be too large.

At small interaction strength, the spin and orbital susceptibilities are finite. Upon increasing interaction, spin susceptibility at \( Q = (\pi, 0) \) or \( (0, \pi) \) and orbital nematic susceptibility at \( q = (0, 0) \) start to diverge in a certain parameter region, implying the tendency towards columnar antiferromagnetic order and ferro orbital nematic
Figure 5.3: RPA phase diagram when fixing $J = 0$ and $\mu = 0.3625$ eV. Phases PM, CAF and Orb respectively denote the isotropic paramagnetic phase, the columnar antiferromagnetic phase, and the orbital nematic phase.

Figure 5.4: RPA phase diagram when fixing $J = 0.7$ eV and $\mu = 0.3625$ eV. Phases PM, CAF and Orb using same abbreviation as in Fig. 5.3.
Figure 5.5: The dependence of critical values of interactions on $\mu$ in the absence of Hund’s coupling, $J = 0$.

Figure 5.6: The dependence of critical values of interactions on $\mu$ for finite Hund’s coupling $J = 0.7$ eV.

order, respectively. The phase boundaries are thus given when the RPA renormalized susceptibilities diverge.

In Fig. 5.3 and Fig. 5.4 we set Hund’s coupling $J = 0$ and $J = 0.7$ eV, respectively. In both phase diagrams, we observe four separate regions: the isotropic paramagnetic
Figure 5.7: RPA phase diagram when fixing $U = 3.5\text{eV}$, $J = 0$. Notice that for sufficiently high electron doping, the orbital nematic phase without magnetic instability (yellow region) appears even when intra-orbital interaction dominates $U > U'$. Dashed vertical line marks the half-filled case (parent compound).

phase (PM), the columnar antiferromagnetic phase (CAF), the orbital nematic phase (Orb), and a region where both susceptibilities have diverged (CAF+Orb). In the CAF region, since magnetic order has already developed ($\langle M_1 \rangle \neq \langle M_2 \rangle$), the orbital nematic order should also be nonzero since there is a linear coupling between orbital order and magnetism, see Eq. (5.2). We therefore used dashed line in the phase diagrams to indicate that the CAF and CAF+Orb phases are actually not distinguishable. This is not true for the boundary between Orb and CAF+Orb phases however, since finite orbital order ($\langle p \rangle \neq 0$) does not necessarily lead to long-range magnetic order: magnetic fluctuations can break the $C_4$ symmetry, $\langle \psi \rangle \neq 0$ in Eq. (5.1), without a true magnetic order[19].

We have also studied the effect of doping on the phase diagram. Let’s denote the critical value of $U$ when the system enters the magnetic CAF phase as $U_c$ (while keeping $U' = 0$), and denote the critical value of $U'$ when the system becomes orbitally
ordered as $U'_c$ (while keeping $U = 0$). The dependence of $U_c$ and $U'_c$ on doping is plotted in Fig. 5.5 for zero Hund’s coupling and in Fig. 5.6 for a realistic value of $J = 0.7$ eV.

From Figs. 5.5 and 5.6 we see that both electron and hole doping enhance the critical value of interactions necessary to stabilize antiferromagnetic or orbital order, making it more difficult to enter the ordered phases. However, the doping effect is not particle-hole symmetric: electron doping greatly enlarges $U_c$, which means that magnetic order is suppressed much faster than orbital nematic order upon electron doping. To make this more transparent, we plotted the phase diagrams describing both the effects of interactions and doping in Fig. 5.7 and Fig. 5.8. Besides the effect that larger $U'$ makes the orbital nematic phase more stable, which is consistent with Fig. 5.3 and Fig. 5.4, we also notice that the phases are very sensitive to doping. In particular, when the system is sufficiently doped with electrons, we found that the
magnetic susceptibility is always finite, so that as a function of increasing $U' - U$, the only phase transition is from paramagnetic phase to the orbital nematic phase, without any magnetism. Interestingly, the orbital ordered phase can be stabilized even when the intra-orbital repulsion $U$ dominates ($U > U'$), see Fig. 5.7. Normally, the regime $U > U'$ is expected to be dominated by antiferromagnetism (c.f. the half-filled case, marked by a dashed vertical line), however in the case of large electron doping the propensity to magnetic ordering is strongly suppressed, resulting in a non-magnetic orbital nematic phase (yellow region in Figs. 5.7 and Fig. 5.8).

5.4 Variational Cluster Approximation

The RPA is a weak-coupling approach that only detects the tendency to certain orderings based on the divergence of the respective susceptibilities. To study the ordered phases themselves, we use the variational cluster approximation (VCA)\cite{202}, which is a non-perturbative quantum cluster method similar in spirit to the cluster dynamical mean-field theory (CDMFT)\cite{203}. Unlike in the CDMFT however, the bath degrees of freedom are not included explicitly in the VCA calculation (this can be done in VCA, see Ref. \cite{204}, however is not necessary for the present work). Rather, the effect of the bath is captured indirectly by varying the inter-cluster one-body parameters \{h\} in such a way as to minimize the free energy (Potthoff functional) $\Omega[\Sigma_{ij}(\omega)]$ calculated in the conserving approximation \cite{202}. The Potthoff functional depends on the cluster self-energy $\Sigma_{ij}(\omega, \{h\})$, which in turn depends on the variational parameters of the cluster \{h\}. These parameters are fixed from the variational principle on the Potthoff functional: $\delta \Omega[\Sigma]/\delta \Sigma = 0$ at the solution. In practical calculations, the variational principle is enforced by requiring that $\partial \Omega[\Sigma(\{h\})]/\partial \{h\} = 0$.

Formulated in this fashion, the VCA is a variational extension of the cluster pertur-
bation theory\cite{205, 206} and provides a powerful way of treating the strongly correlated lattice models with local (on-site) interactions. The VCA method has been shown to capture both the weak- and strong-coupling limit of the (one-band) Hubbard model and compares very favourably to the quantum Monte Carlo simulations. It has been used successfully to study the metal-insulator transition\cite{207, 204}, frustrated magnetism\cite{208, 209} and d-wave superconductivity in quasi-2D organic superconductors\cite{210} and in the high-Tc cuprates\cite{211, 212, 208}. It was shown in particular to capture the d-wave superconductivity of purely electronic origin and to yield the correct doping dependence\cite{212}, as well as the pseudogap feature in the quasiparticle spectral weight\cite{211, 213}. We use the exact diagonalization (ED) method based on Lanczos method to solve the quantum cluster impurity model, which offers two principal advantages over the Monte-Carlo based solvers routinely used in CDMFT: (i) there is no need for analytical continuation as the self-energy is expressed in real, not imaginary, frequency and (ii) the zero-temperature properties can be readily accessed, avoiding the infamous fermionic sign problem inherent to the quantum Monte Carlo impurity solvers.

The VCA method is particularly well suited to our task because it allows explicit treatment of a spontaneous symmetry-breaking long-range order, and has been successfully used to study magnetism\cite{214, 208, 209} and unconventional superconductivity\cite{211, 212, 208} in the Hubbard model. To study the orbital nematic order, we allow the Hamiltonian on a cluster to have a variational degree of freedom associated with a cluster Weiss field, $\Delta \hat{H}_{cl} = p_{cl} \cdot (\hat{n}_{xz} - \hat{n}_{yz})$. Note that the actual lattice Hamiltonian is unaltered, so that the $C_4$ symmetry is not explicitly broken by construction. Rather, the spontaneously broken symmetry inside the orbital nematic phase is signified by a non-zero value of the cluster Weiss field $p_{cl}$ at the variational
solution of the Potthoff functional [202]:

\[
\frac{\delta \Omega[\Sigma(p_{cl})]}{\delta p_{cl}} \bigg|_{sol} = 0. \tag{5.20}
\]

This is in difference to earlier VCA calculations of nematicity by Daghofer and collaborators [215, 216], in which the \(C_4\) tetragonal symmetry was broken by construction at the level of the original lattice Hamiltonian, by introducing anisotropy either in the onsite energy of the \(xz/yz\) orbitals, in the hopping amplitudes between the \(x\) and \(y\) directions, or in the Heisenberg exchange terms in the \(x\) and \(y\) directions. These studies do not probe the spontaneous symmetry breaking but rather investigate the response of the system to the \(C_2\) distortion, similar in spirit to the experimental studies under uniaxial stress [115] or strain [133]. The present approach, on the other hand, is faithful to the original variational idea by Potthoff [202], allowing the \(C_4\) symmetry to be broken spontaneously, by introducing the in-cluster nematic Weiss field \(p_{cl}\).

From the solution of Eq. (5.20), we then obtain the cluster propagator and self-energy using the ED solver. The full lattice propagator \(G(K, \omega)\) is then calculated from the cluster solution by treating the one-body hopping terms between neighboring clusters as a perturbation. When the Potthoff functional is minimized at a nonzero value of \(p_{cl}\), the full lattice Hamiltonian will develop a long range order of the orbital nematicity, ie. a nonzero value of \(\langle p \rangle \equiv \langle \hat{n}_{xy} - \hat{n}_{yz} \rangle\).

In Fig. 5.9, the expectation value of the orbital nematic order parameter is calculated on the lattice, as a function of \(U' = U\). After developing a non-zero value of \(\langle p \rangle\), upon further increasing \(U' = U\), the orbital nematic order parameter attains a peak and then becomes suppressed at higher values of the interaction strength. In the calculation for a realistic value of Hund’s coupling \(J = 0.7 \text{ eV}\) (Fig. 5.10), the magnitude of order parameter \(p\) starts from a non-zero value when \(U'\) is small. This
Figure 5.9: Orbital nematic order parameter versus interaction strength from VCA calculation at half-filling. The inter-orbital Hubbard repulsion $U'$ is varied at the same time as the intra-orbital $U$: $U' = U - 2J$, with the Hund’s coupling $J$ fixed at zero. The dotted line is a guide to the eye.

Figure 5.10: Orbital nematic order parameter versus the inter-orbital Hubbard coupling $U' = U - 2J$ from VCA calculation at half-filling. Hund’s coupling is fixed at $J = 0.7eV$. The dotted line is a guide to the eye.
Figure 5.11: Change of orbital nematic order (circle) and magnetic order (square) with respect to doping from VCA calculation, when $U = 2$ eV, $U' = 0.6$ eV, $J = 0.7$ eV. The half-full symbol corresponds to not a smooth minima but a cusp in the Potthoff functional. Note that near half filling, solutions with zero staggered magnetization appear – these are unphysical and should be disregarded due to the VCA minimization algorithm becoming unstable when the chemical potential falls onto sharp maxima/minima in the density of states. This does not however affect any of our conclusions.
is somewhat unexpected from our RPA results, where Hund’s coupling $J$ slightly suppresses orbital nematic order (see Fig. 5.3, 5.4, 5.5, and 5.6). Upon further increasing $U'$, the same suppression of $p$ is observed as in the case of $J = 0$ above (see Fig. 5.9).

The above VCA calculations have probed the orbital nematic order in the absence of antiferromagnetism. Of course we know from our RPA calculations that magnetism also arises in the phase diagram of the two-orbital model. The effects of interaction on magnetic order alone (without orbital order) have been studied with VCA in Ref. [194]. The authors found that the magnetic order starts developing at intermediate interactions $U$ in the parent compounds. (Although it should be noted that in their study, the Hund’s coupling $J$ was assumed to scale with $U$ as $J = U/4$, which becomes unphysical for too small or too large values of $U$).

We will now investigate the phase diagram of the model as a function of electron doping when both the orbital order and columnar antiferromagnetism are present. We set the interaction strengths close to realistic values determined from the ab initio constrained-RPA calculations [201]: $U = 2$ eV, $J = 0.7$ eV, $U' = U - 2J = 0.6$ eV. Besides the orbital nematic Weiss field $p_{cl}$ coupled to $n_{xy} - n_{yz}$ on the cluster, we have also introduced the columnar antiferromagnetic Weiss field $M_{cl}$ coupled to $S^z_r \cos(Q \cdot r)$ on the cluster, with wave vector $Q = (\pi, 0)$ or $(0, \pi)$. A VCA variational search was then performed for both $p_{cl}$ and $M_{cl}$, to study the interplay of the two orders. The resulting doping dependence is plotted in Fig. 5.11, where the lattice chemical potential was varied to control the electron occupancy. We found coexisting orbital and antiferromagnetic orders in the parent compound at half-filling. While both types of ordering are suppressed by hole doping, the effect of electron doping is to slightly enhance the orbital nematic order while suppressing antiferromagnetism. This effect likely stems from the competition between the two orders, although the enhancement
of orbital order with electron doping could be an artifact of the simplified two-orbital model studied here.

Crucially, we find that upon electron doping, antiferromagnetism is suppressed much faster than orbital nematic phase, resulting in a region at sufficiently high electron doping ($x \gtrsim 13\%$) where orbital nematic order exists without any magnetism. This is consistent with our previous RPA finding (yellow region in Figs. 5.7 and 5.8).

5.5 Discussion

We have studied the emergence of orbital nematic order in the iron pnictide superconductors, within the framework of the two-orbital Hubbard model that captures the physics of Fe $d_{xz}$ and $d_{yz}$ orbitals. In particular, we have analyzed the dependence of the nematic order on doping and interaction strength, as well as its interplay with magnetism.

First, we studied the instabilities towards the orbital nematic order and magnetic order in the weak-coupling approach by calculating the corresponding susceptibilities using the RPA method. We recovered the results of previous studies that the ordering wave-vector is at $Q = (\pi, 0)$ or $(0, \pi)$ for columnar antiferromagnetism\cite{193} and $(0, 0)$ for ferro-orbital nematic order\cite{180}, respectively. We found that orbital nematic order strongly depends on inter-orbital Hubbard repulsion $U'$, while magnetic order depends on the intra-orbital Hubbard repulsion $U$. Both the magnetic and orbital nematic order parameters are affected by Hund’s coupling $J$: larger $J$ values tend to suppress the propensity to orbital nematic ordering and, on the other hand, enhance the magnetic susceptibility.

It has been long believed that the nematicity and antiferromagnetism coexist at low temperatures, with a wealth of experimental data supporting this in the 1111
and 122 families of iron pnictides. Within the RPA, we indeed find regions in the phase space where both the magnetic and nematic susceptibilities diverge, implying coexistence of the two order parameters. It is true that the long-range CAF phase with \( M_1 \neq M_2 \) necessarily breaks the \( C_4 \) symmetry and will generically induce a non-zero value of orbital polarization \( p \) because of the linear coupling in the Landau free energy. The converse is however not true: the ferro-orbital phase with non-vanishing \( p \), while nematic in nature, need not have long-range magnetic order. Indeed, our RPA calculations show that upon electron doping, magnetic susceptibility is suppressed much faster than orbital ordering, until for sufficiently large electron doping, only orbital order survives (See Fig. 5.5 and Fig. 5.6). Hole doping, on the other hand, does not reveal this tendency.

We note that our results do not eliminate the possibility of spin fluctuations taking part in the nematicity even if the static magnetic order is absent. Indeed, as remarked in the Introduction, ferro-orbital nematic order parameter will couple linearly to the spin-fluctuation Ising parameter, see Eq. (5.2). Indeed, as we discussed in Chapter 4, when the system is in close proximity to the antiferromagnetic phase, another possible driving force of nematicity could be quantum fluctuations in the spin ferroquadrupolar phase. Thus it is possible that both spin and orbital mechanisms are present for nematicity.

Since RPA is a weak coupling approach which works only when the long-range order is approached from the disordered phase, we have used the non-perturbative variational cluster approximation (VCA) which allowed us to study the ordered phases in the variational approach. We studied the orbital nematic and magnetic order parameters, as well as their interplay as a function of electron density (doping) and interaction strength. Our VCA calculation showed that orbital nematic order strongly
depends on inter-orbital Hubbard interaction $U'$ and Hund’s coupling $J$, similar to the RPA results. However, while Hund’s coupling suppresses orbital order within the RPA approach, this is not the case in the non-perturbative VCA calculation. In Fig. 5.10 we find a non-vanishing orbital nematic order even in the absence of inter-orbital interaction $U' = 0$ and finite Hund’s coupling $J = 0.7$ eV, implying that the effect of interactions and Hund’s term is not always captured properly by the RPA method.

The doping dependence study from VCA shows that electron and hole doping are not symmetric. Crucially, we find that upon moderate electron doping ($\gtrsim 13\%$, see Fig. 5.11), long-range magnetic order is completely suppressed, while the orbital nematic order persists, similar to our RPA results.

It is instructive to compare our VCA results with previous attempts to address nematicity with the cluster dynamical mean-field theory (CDMFT)\cite{203} and similar cluster approaches. One possible way to detect tendency to nematicity is to explicitly introduce orthorhombic distortion into the hopping terms in Eqs. (5.4, 5.5), and then study the electronic response. For the one-band Hubbard model, this has been done using CDMFT\cite{217} and dynamical cluster approximation\cite{218}. Both groups found that for a sufficiently large interaction $U$, a small orthorhombic distortion can lead to a large nematic response in the low-energy electron scattering rate. Another way to study the spontaneous development of nematicity in the one-band Hubbard model is by introducing an anisotropic hopping inside the cluster alone $\Delta \hat{H}_{cf} = \delta t \sum_r (\hat{c}^\dagger_{r+x} \hat{c}_{r+y} + \hat{c}^\dagger_{r+y} \hat{c}_{r+x}) + h.c.$, and then optimize the strength of $\delta t$ variationally in VCA\cite{219}. Using this approach, the authors found that anisotropy can develop in the overdoped region\cite{219}. However, the multi-orbital nature of the iron pnictides was not taken into account in these studies.
The nematicity in multi-band Hubbard model has been studied with VCA in Refs. [215, 216], however these authors have also explicitly broken the $C_4$ tetragonal symmetry at the level of the original lattice Hamiltonian, by introducing anisotropy either in the onsite energy of the $xz/yz$ orbitals, in the hopping amplitudes between the $x$ and $y$ directions, or in the Heisenberg exchange terms in the $x$ and $y$ directions. Because the $C_4$ symmetry is broken by construction, these studies do not probe the spontaneous symmetry breaking but rather investigate the response of the system to the $C_2$ distortion, similar in spirit to the experimental studies under uniaxial stress [115] or strain [133]. Not surprisingly, the magnitude of the induced orbital order is then proportional to the imposed strain and does not have an intrinsic value. In the present work, by contrast, the lattice expectation value of orbital order $p = \langle \hat{n}_{xz} - \hat{n}_{yz} \rangle$ is finite even at zero induced strain and is consistent with the value found by ARPES in BaFe$_2$As$_2$ [114].

We note that in a different context, nematicity in a two-orbital Hubbard model has also been studied in application to Sr$_3$Ru$_2$O$_7$ in Refs. [220, 221]. In these works, the authors studied the nematic instability using RPA and renormalization group methods. It was found that Aslamazov–Larkin-type vertex corrections result in the strong coupling between spin and orbital fluctuations, leading the authors to conclude that the spin fluctuations lie at the origin of the nematic instability. In the present work, on the other hand, we find a regime of parameters where the ferro-orbital RPA susceptibility diverges even without the vertex corrections, while the spin susceptibility remains finite (see section 5.3). This implies that the spin fluctuations may not be the primary origin of ferro-orbital nematicity in the two-orbital Hubbard Model. Furthermore, from our VCA calculations we find that the orbital nematic order persists even when magnetic ordering is fully suppressed in the electron doped region.
(see Fig. 5.11), which again suggests that magnetic fluctuations in the strongly doped regions are not the origin of orbital nematicity in our case. It should be noted that the Fermi surface topology and nesting properties in Sr$_3$Ru$_2$O$_7$ are different from the iron pnictides, so the conclusions drawn in Refs. [220], [221] do not trivially generalize to our case.

Finally, we note that other types of orbital ordering, involving $d_{xy}$ orbitals, have been proposed in the literature, [165] [185] [222] however those are beyond the scope of the two-orbital model used in this study. While it would be desirable to extend this study to include all five iron orbitals, unfortunately the VCA calculations become computationally prohibitive because of the limitations of the exact diagonalization solver when dealing with multiple orbitals. Nevertheless, as remarked earlier in Sec. 5.2 we hope that the present two-orbital model captures the salient features of nematicity in the iron pnictides, based on the dominant contribution of $d_{xz}$ and $d_{yz}$ orbitals to the Fermi surfaces of these materials. [70]

5.6 Conclusions

To summarize, we have studied the doping dependence of both the orbital nematic and antiferromagnetic orders using the RPA and non-perturbative variational cluster approximation at zero temperature, and found a region at moderately large electron doping where the orbital nematic order survives without long-range magnetism. While these results are limited to the two-orbital model, which is not sufficient to describe the realistic band structure of the iron-based superconductors, our findings are suggestive of the connection to the experimental observations of an orbital nematic phase in FeSe [162] [161] [160] without any sign of antiferromagnetism [189] [127]. This raises the question whether two-fold symmetric antiferromagnetic fluctuations
are essential for stabilizing the nematic phase, as has been argued previously within the spin-nematic scenario.\cite{78, 116, 187, 198, 188} It would appear from the present study that this may not always be the case, and while the importance of spin fluctuations is undeniable in the vicinity of the magnetic order in the 122 and 1111 families of iron pnictides, it is possible that orbital fluctuations being the key to nematicity, especially for the region away from half-filling. This situation may be more common than previously appreciated: recent ARPES measurements\cite{195} detect $d_{xz}/d_{yz}$ orbital splitting inside the superconducting phase in the parent LiFeAs as well as electron-doped LiFe$_{1-x}$Co$_x$As, with no magnetic phase nearby. Very recently, orbital ordering has also been observed\cite{223} inside the superconducting phase of the optimally doped and overdoped BaFe$_2$(As$_{1-x}$P$_x$)$_2$, far away from the magnetically ordered phase and in the same regime where the torque magnetometry detected $C_2$-symmetric spin response\cite{177}. These observations also raise the question of the interplay between orbital nematicity and superconductivity in the iron pnictides, which will be the subject of future study.
Chapter 6

Summary and Outlook

We have explored a few examples where magnetic frustration is essential for the understanding of the spin ground state properties and the corresponding excitations. In particular, we examined the effects of frustration on several types of exchange mechanisms (namely, isotropic Heisenberg and biquadratic interactions, along with anisotropic dipole-dipole interactions). As we mentioned in Chapter 1, such interactions arise naturally through kinetic, super-exchange and double-exchange mechanisms (as well as a few others), which are commonly found in the broad family of strongly correlated electron systems.

Due to the bosonic nature of magnon excitations, we have utilized the mapping from spin to boson language (for instance, the Matsubara-Matsuda transformation in Chapter 2). Through such formal manipulations, not only have we obtained a mathematically convenient description of the magnetic states, but also gained more physical insights by unifying the concepts of magnetic ordering and Bose Einstein Condensation (BEC).

In Chapter 2, we looked into the BEC transition near the saturation field. Such BEC transition is highly frustrated once there are several degenerate minima in the magnon dispersion, leading to degenerate ground state manifolds. The degeneracy can be lifted by quantum fluctuations: in the dilute limit (close to the saturation field), the sum of all the ladder-type Feynman diagrams gives the leading order energy correction to the different ground states, depending on the magnon occupations.
and relative phases of different minima. The resulting phase diagram obtained by minimizing the ground state energy contains many different phases, some of which even exhibit the exotic macroscopic crystallization of magnetic vortices. Along the magnetic field direction, these vortices form either parallel strings, if the $k$-space positions of the condensed minima are coplanar; or otherwise the vortex strings can even have modulation along the magnetic field direction.

In Chapter 3, we move one step further by studying the spin excitations in the Fe-based superconductors, whose magnetic properties are typically described by the frustrated Heisenberg Model. In particular, we are interested in the tetragonal phase of such materials, in which case the lattice structure is symmetric under $C_4$ (90 degree) lattice rotation. However, a straightforward fitting of the $C_4$-symmetric Heisenberg model to the inelastic neutron scattering (INS) experiments simply fails, and attempts have been made to fix this discrepancy by breaking the $C_4$ symmetry in the Hamiltonian by hand. Although the $C_4$ breaking version of the Heisenberg model is able to explain qualitatively the INS features, it is inconsistent with the fact that physically, there is no $C_4$ symmetry breaking source in the tetragonal phase of Fe-based superconductors. In Chapter 3, we study the effect of the nearest neighbor biquadratic term $-K(S_i \cdot S_j)^2$, which becomes non-negligible for systems close to the Mott transition (which is indeed the case for Fe-based superconductors). By utilizing the modified spin wave and the Schwinger boson mean-field techniques, we found that this higher order term induces an emergent anisotropy in the nearest neighbor Heisenberg interaction at the mean-field level. Furthermore, the calculated dynamical spin structure factor $S(q, \omega)$ from the two methods yields consistent results comparing to the INS data, showing the importance of such higher order corrections for the understanding of the spin dynamics in Fe-based superconductors.
While the spin dynamics of most Fe-based superconductors seems to be dominated by the ordered magnetic ground states with wave-vectors \((\pi, 0)/(0, \pi)\), there is one special case: the stoichiometric iron selenide FeSe shows the same structural transition as other Fe-based superconductors upon decreasing temperature without however hosting a magnetic phase. We show that such non-magnetic state can also be understood within the bilinear-biquadratic model, which contains a ferroquadrupolar (FQ) phase in its theoretical phase diagram. The FQ state has no net magnetization, but its spin fluctuations break the spin rotational symmetry and thus result in the dispersive Goldstone modes in the spectrum of excitations. We have verified the stability of the FQ phase by the state-of-the-art density matrix renormalization group calculations, and further checked that it is stable in the presence of the \(C_4\)-symmetry breaking environments from the mean-field calculation. Further evidence of the putative FQ phase in FeSe is provided from the dynamical spin structure factors calculated using the flavor wave technique. The calculated \(S(q, \omega)\) shows strong fluctuations at wave-vectors \((\pi, 0)/(0, \pi)\) at low energies, and the spectral weight further moves to \((\pi, \pi)\) at high energies. These features are qualitatively consistent with the INS data, providing evidence that FQ phase is a competitive candidate for the ground state of FeSe.

Recently, there has been increasing interest in the Fe-based superconductors regarding the appearance of the nematic phase and its origin. In the magnetically ordered phases with wave-vector \((\pi, 0)/(0, \pi)\) and their corresponding paramagnetic phases, the nematicity can be naturally induced by coupling to the anisotropic spin excitations. For the FQ phase, although the spin correlations at the mean-field level are \(C_4\)-symmetric, we showed that the quartic interactions from the flavor wave theory have strong tendency towards breaking the \(C_4\) symmetry, thus explaining the
observed nematicity in the non-magnetic phase of FeSe.

We should not be simply satisfied with the success of explaining the nematicity from spin fluctuations. In the context of Fe-based superconductors, a more careful description should include the multi-orbital nature of these materials. And as a result, the observed nematicity could also originate from the orbital physics. The simplest example is the energy splitting between \( d_{xz} \) and \( d_{yz} \) orbitals, which has been observed in the angle-resolved photoemission spectroscopy (ARPES). To address this mechanism of nematicity, we study in Chapter 5 the competition of magnetic order and orbital splitting in the two-orbital Hubbard model, by means of the random phase approximation (RPA) and the variational cluster approximation (VCA) methods. We find that magnetic order is not necessary for finite orbital splitting, for example in the heavily electron doped phase of the model.

Overall, magnetic frustration is commonly found in many families of strongly correlated electron systems, and it has far-reaching consequences for both the ground state and dynamical properties. The frustration induced magnetic states have applications in the field of spintronics, and the magnetism also has effect on the other degrees of freedom. It is known that the electron transport behavior is strongly affected near a magnetic critical point; as a specific example, we know that the unconventional superconductivity often arises near such a critical point and has roots in its interplay with frustrated magnetism. Therefore, the magnetic frustration is not only of fundamental interest to the physics community, but also finds its uses in related applications (or potential applications) in everyday life. And perhaps it is not unrealistic to say that research in the field of magnetic frustration could perhaps help reduce the human-life frustration, by providing us not only with new knowledge but also with new technologies and convenience.
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