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Magnetic frustration, Kondo effect, and superconductivity in strongly correlated electron systems

by

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Abstract

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The substantial effect of quantum fluctuations in strongly correlated electron materials often results in a rich phase diagram with many interesting states of matter. This thesis aims to study some of the mechanisms that give rise to such phenomena.

In Part I, we focus on iron-based superconductors, and use bilinear-biquadratic spin models to study the different magnetic orderings present in both iron pnictides and chalcogenides, specifically FeSe and Fe ($\text{Te}_{1-x}\text{Se}_x$). After benchmarking different methods for the theoretical representation of electron spins, we find our models give good qualitative descriptions of the phases observed in the aforementioned materials. We also find that the dynamical spin structure factors are in agreement with experimental inelastic neutron scattering (INS) results. In Part II, we switch our focus to so-called heavy fermion materials, where the strong interactions between electrons endow the charge carriers with a very heavy mass. In particular, we investigate the multichannel Kondo model, which is appropriate to describe real materials with multiple conduction electron bands. When *just* enough of these bands are present to *exactly* screen the impurity spin, the heavy fermion Fermi liquid state is formed. We present a novel technique for solving this problem in both one- and two-impurity systems. In the latter case, we quantify the transition from the phase with strong magnetic correlations between the two local moments to the heavy fermion Fermi liquid regime. Extending this model further, we aim to capture the superconducting correlations between the conduction electrons on the two impurities. Using the language of auxiliary particles, called *bosons* and *holons*, we find that superconductivity does indeed arise in the region where correlations between bosons and between holons are *both* present, provided that an attractive interaction between the latter exceeds a certain minimum value.

The phenomena described in this thesis provide just a few examples that showcase the richness in the quantum world of strongly interacting particles. We hope that the theoretical methods developed in this work will help shed more light on these quantum phenomena.

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Preface

A large volume of the work compiled in Part I of this thesis has been included in the following publications:

- Patricia Bilbao Ergueta and Andriy H. Nevidomskyy, *Ising-nematic order in the bilinear-biquadratic model for the iron pnictides.* Phys. Rev. B 92, 165102 (2015).
- [2] Patricia Bilbao Ergueta, Wen-Jun Hu, and Andriy H. Nevidomskyy, Unified spin model for magnetic excitations in iron chalcogenides.
 Phys. Rev. B 96, 174403 (2017).

The work presented in Part II will be in turn included in a future publication:

 [3] Patricia Bilbao Ergueta and Andriy H. Nevidomskyy, Superconductivity in the 2-impurity multichannel Kondo model, in preparation (2019).

Part I

Magnetic frustrations on a square lattice and application to iron-based superconductors

Chapter 1

Origin and Models of Magnetic Frustration

1.1 Magnetic Couplings

The magnetic interactions that arise at the atomic level in materials originate from properties in principle, not necessarily related to magnetic phenomena. Loosely speaking, when deriving the main different types of magnetic couplings that we find in materials, one needs to consider both the Coulomb *interaction* energy and the *kinetic* energy of the electron hopping between sites, along with the Pauli Exclusion Principle. Of course, all the exchange mechanisms described below are just idealized versions of the actual phenomena that are found in real materials and, as such, must only be taken as approximations.

1.1.1 Direct (Coulomb) Exchange

The *direct exchange* (also called *Coulomb exchange* due to the vital role that this force plays in the mechanism) is due to the interplay of repulsion that electrons feel against each other and their fermionic nature. The effect of the direct exchange is best understood considering the limit case of two electrons with single-particle orbitals that are nearly degenerate. Their space wave functions would then be almost orthogonal but highly overlapping at the same time.

In this situation, the larger the probability to find the electrons close together, the stronger the contribution from the electromagnetic repulsion to the total energy will be. This is when the Pauli Exclusion Principle comes into play. Because two electrons of the same spin will avoid occupying the same position in space, the repulsive *interaction* energy will be minimized in the case where the two electrons are in a *triplet* state. Mathematically, the symmetric nature of the spin wave-function forces the spatial wave-function to be antisymmetric, which must vanish at its center, which ensures that the two electrons never meet.

One of the clearest examples of the direct exchange in action is, probably, Hund's first rule and second rules [1], which describe the aforementioned phenomenon for any number of degenerate orbitals, giving preference to electrons occupying different orbitals with the same spin before admitting any double occupations.

1.1.2 Kinetic Exchange

The kinetic exchange also shows the Pauli Exclusion Principle at work but, in this case, when it's applied to the hopping of electrons between atomic sites. In a second order process, electrons can virtually hop momentarily to a neighboring site and in doing so, spread out its position, reducing the total energy. However, hopping to nearest-neighbors with the same spin is prohibited by the Exclusion Principle, so that an antiferromagnetic order is promoted in compounds that benefit from this mechanism.

The simplest case in which one can see this mechanism in action, described by Heitler and London[2] in 1927, is the H_2 molecule. On a lattice, however, this phenomenon is realized in Hubbard's model[3, 4, 5]:

$$\mathcal{H} = -t \sum_{\langle i,j \rangle} \sum_{\sigma=\uparrow\downarrow} \left(c_{i\sigma}^{\dagger} c_{j\sigma} + c_{i\sigma} c_{j\sigma}^{\dagger} \right) + U \sum_{i} n_{i\uparrow} n_{i\downarrow}$$
(1.1)

Where U represents the Hubbard energy, which comes from electron-electron repulsion, while t is the electron hopping energy. In this case, the decrease in energy can easily be calculated to second order and takes the value: $J = \frac{4t^2}{U}$. This results in an effective Heisenberg Hamiltonian:

$$\mathcal{H} = J \sum_{i,j} \boldsymbol{S}_i \cdot \boldsymbol{S}_j, \qquad (1.2)$$

Superexchange

In materials where orbital localization suppresses the hopping between sites, the above mechanism is not feasible as is. However, a fourth order process of *virtual* hopping can occur between neighboring orbitals, with eventually also results in an effective antiferromagnetic exchange.

This phenomenon is observed in transition metal compounds with very localized d-orbitals in cations, where an indirect exchange by means of the anion orbital is responsible for this superexchange [6, 7].

1.1.3 Double (Indirect) Exchange

In contrast to the direct exchange mentioned above, the double exchange mechanism acts *indirectly* through a combination of the Direct (Coulomb) exchange on an atom and the kinetic coupling to neighbors via electron hopping. The Kondo coupling is a manifestation of this double exchange. Although we will devote Part II of this manuscript to the study of a certain version of the Kondo Hamiltonian, we briefly touch of how the RKKY interaction arises from the Kondo coupling.

The **RKKY** interaction

The Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction, taking its name from the contributors to its derivation [8, 9, 10], is a second order effect of the Kondo exchange between magnetic ions and charge carriers. The Kondo mechanism induces Friedel oscillations [11] in the spin density by polarizing the conduction electrons around said ion. This new spin distribution then will interact with another, neighboring ion in the lattice. It is worth noting that the RKKY coupling can be either ferro- or antiferromagnetic in nature since its sign depends on the distance between the impurities involved in the exchange.

1.2 Geometric frustration

In a lattice of localized spins, one or more of the exchanges described in the previous section will coexist, coupling spins to successive neighbors in the lattice. When all of the interactions are ferromagnetic, the ground state of the system is trivial since having all of the lattice spins aligned in the same direction will always minimize the ground state energy. However, if at least some of the exchanges between spins are antiferromagnetic, it is sometimes not possible to minimize the energies of all bonds at the same time. This is what we call *geometric frustration*.

1.2.1 Triangular Lattice

The quintaessential system used to illustrate frustation is probably that of a triagular lattice with an Ising Hamiltonian [12]:

$$\mathcal{H} = J \sum_{\langle i,j \rangle} \sigma_i \sigma_j, \tag{1.3}$$

where $\sigma_{i,j}$ can only take two values: $\sigma_{i,j} = \pm 1$. Looking at just one of the Ising triangles (see Fig. 1.1), one can easily understand where the problem lies.

While we can always minimize the interactions between two of the bonds, when it comes to choosing the orientation of the spin on the remaining, third site, any option will leave us with a penalty in energy due to the necessary ferromagnetic alignment of one of the bonds.



Figure 1.1: Two possible configurations of an antiferromagnetic Ising triangle.

1.2.2 The $J_1 - J_2$ Model on a Square Lattice

For the case of the Heisenberg interactions described above, the simplest model that showcases frustration is that of competing antiferromagnetic interactions to nearest and next-to-nearest neighbors (J_1 and J_2 , respectively):

$$\mathcal{H} = J_1 \sum_{\langle i,j \rangle} \boldsymbol{S}_i \cdot \boldsymbol{S}_j + J_2 \sum_{\langle \langle i,j \rangle \rangle} \boldsymbol{S}_i \cdot \boldsymbol{S}_j, \qquad (1.4)$$

In order to figure out the ground state of this system, we can study the smaller case of a square plaquette. Here, in the limit where $J_1 >> J_2$, all of the spins can form an antiferromagnetic bond with their nearest neighbors and the energy penalty due to the next-to-nearest neighbor ferromagnetic bonds is negligible since $J_2/J_1 \ll 1$.



Figure 1.2: Two possible configurations of the frustrated Heisenberg square plaquette: a) Néel order and b) *columnar* antiferromagnetic order.

For the particular case where J_2 vanishes completely, we have a perfect example of a system with antiferromagnetic (AFM) couplings but no frustration. That is, at least one AFM exchange is needed to generate frustration in a system, but AFM couplings don't *guarantee* frustration. This type of arrangement in a lattice is called Néel ordering.

In the opposite limit, when $J_2 >> J_1$, the coupling between nearest neighbors is negligible and the two sublattices effectively decouple. Now, in principle, this would leave the angle between the aforementioned sublattices as a free parameter, allowing any orientation between nearest neighbors (provided that the rest of the sublattice follows this axis). However, it turns out that through the so-called *order by disorder* mechanism [13, 14], quantum fluctuations favor the coplanar arrangements, giving the lattice what is called a *columnar* antiferromagnetic ordering. This consists of ferromagnetically aligned spin stripes arranged in an alternating order, so that the bonds in the axis perpendicular to the stripes are all antiferromagnetic.

Throughout this work, we will build upon this basic frustrated Hamiltonian into more complex models including Heisenberg couplings to further neighbors in the lattice and between planes, as well as terms with a coupling *biquadratic* on the spins, since we will be working with spin values larger than S > 1/2. Biquadratic terms are unnecessary for the case of S = 1/2 since their only effect is to renormalize the value of the preceeding Heisenberg coupling and accordingly, don't introduce any new physics.

Chapter 2

Magnetism and frustration in iron-based compounds

2.1 General properties of the iron-based compounds

The discovery of high- T_c superconductivity in iron-based compounds [15, 16] gathered plenty of attention around these materials, in part due to their shared characteristics with the cuprate superconductors.

Although the specifics of each compound vary, all iron-based superconductors are quasi-two-dimensional crystals with stacked iron-pnictogen or iron-chalcogen planes [see panel a) in Fig. 2.1]. It is the spins in the iron sites [see panel b) in Fig. 2.1] that exhibit the long-range ordering, with the upper and lower pnictide or chalcogen element atoms helping to mediate the exchanges between them.

Just like their antecessors [17], the ground state of the iron-based parent com-



Figure 2.1: a) Plane structure of a generic iron-based superconductor. b) Upper view of said plane. It is spins in the Fe sites that acquire long-range order.

pounds is antiferromagnetically ordered [18]. However, while the former exhibit a regular Neél pattern, the latter can take on a wide variety of spin structures, which we will study in Chapters 4 and 5 below.

2.2 Iron pnictides

The ground state of the parent compounds of iron pnictide superconductors is a columnar antiferromagnet (CAFM), a structure consisting of stripes of spins of opposite orientation [see panel b) in Fig. 1.2] and with an ordering wave vector of $\boldsymbol{Q} = (\pi, 0)$ or $\boldsymbol{Q} = (0, \pi)$. In their transition from a paramagnetic to an AFM regime, the pnictides also experience a structural transition from a tetragonal to an orthorhombic phase. However, the two transition temperatures, T_N and T_s , respectively, do not coincide. While $T_s \geq T_N$, the close proximity between the two temperatures obscures the specific origin of this transition. Above the structural transition, INS experiments have found that the C_4 symmetry remains broken. Indeed, Lu *et al.* [19] observed nematic fluctuations up to a temperature T^* , well above both the magnetic and structural transition temperatures.

In addition to their magnetic properties, the iron pnictide family exhibits a "bad metal" behavior, which several authors [20, 21, 22, 23] have explained through a possible proximity of the parent compounds to a Mott localization transition. Evidence towards this prediction comes from the suppression of the Drude peak in optical conductivity measurements [24, 25] and the spectral weight transfer induced by temperature [26, 27].

2.3 Iron chalcogenides

Despite their multiple similarities, the iron chalcogenides exhibit certain important behaviors that contrast with those in the iron pnictides studied above. Namely, the chalcogenides are much more correlated materials than the pnictides and their magnetic orderings greatly differ as well.

While the ground state of the iron pnictides' parent compounds is always a columnar antiferromagnet (CAFM) [18, 28, 29] with alternating stripes of opposite spins, the magnetic ordering found in the ground state of $Fe_{1+y}Te$ has a *double* stripe structure instead [30, 31, 32]. However, not all chalcogenide compounds exhibit the same (if any) magnetic ordering. Upon doping the aforementioned compound with Selenium in order to obtain $Fe(Te_{1-x}Se_x)$, the long-range order disappears [33, 34, 35, 36, 37] at a certain concentration x and all the way up to FeSe. However, INS studies have found large finite-energy spectral weight at wave vectors characteristic of a CAFM in the latter compound [38, 39, 40], which suggests the proximity to this particular magnetic ordering. Indeed, magnetism can actually be induced by applying hydrostatic pressure to the FeSe samples [41, 42, 43, 44].

Chapter 3

Theoretical methods for the study of magnetic frustrations

3.1 Hamiltonian models

The two types of Hamiltonian terms we will work with are the usual Heisenberglike term, *bilinear* in the spin operators:

$$\mathcal{H}_{\rm bl} = J \sum_{i,j} \boldsymbol{S}_i \cdot \boldsymbol{S}_j, \qquad (3.1)$$

And a *biquadratic* term with a coupling of value K:

$$\mathcal{H}_{\mathrm{bq}} = K \sum_{i,j} \left(\boldsymbol{S}_i \cdot \boldsymbol{S}_j \right)^2.$$
(3.2)

We detail the specific motivations for including terms of this type below, in Section 4.1. Unlike the Heisenberg coupling J above [of order $\mathcal{O}(S^0)$], we will take K to be of order $\mathcal{O}(\frac{1}{S^2})$. This will become relevant when discussing the methods involving a semi-classical, large-S approach, which will involve expansions up to different orders of the parameter $\frac{1}{S}$. In practice, it will be useful to use the expanded form of these terms using the S^z , spin "raising" and "lowering" operators:

$$S^+ \equiv S^x + iS^y \tag{3.3}$$

$$S^{-} \equiv S^{x} - iS^{y}, \tag{3.4}$$

In the case of ferromagnetic ordering, the expression for the dot product is obtained immediately:

$$\left(\boldsymbol{S}_{i} \cdot \boldsymbol{S}_{j}\right)^{\mathrm{FM}} = \frac{1}{2} \left(S_{i}^{+} S_{j}^{-} + S_{i}^{-} S_{j}^{+} \right) + S_{i}^{z} S_{j}^{z}.$$
(3.5)

In the antiferromagnetic case, however, we deal with a *bipartite* lattice $(i \in A, j \in B)$. Because of the opposite spin orientations between sublattices A and B we must first perform a π rotation along one of the axis perpendicular to the spin

ordering vector on one of them. Without loss of generality, we choose to rotate the axis for sublattice B around the \hat{x} direction, which sends the spin components to:

$$S_j^x \to S_j^x, S_j^y \to -S_j^y, S_j^z \to S_j^z \tag{3.6}$$

$$S_j^+ \to S_j^-, S_j^- \to S_j^+. \tag{3.7}$$

This finally leaves us with the following expression for the dot product in the case of an antiferromangetic ordering:

$$\left(\boldsymbol{S}_{i} \cdot \boldsymbol{S}_{j}\right)^{\text{AFM}} = \frac{1}{2} \left(S_{i}^{+} S_{j}^{+} + S_{i}^{-} S_{j}^{-} \right) - S_{i}^{z} S_{j}^{z}.$$
(3.8)

We now go on to describe some of the main approaches to tackle this problem using various *bosonic* spin representations.

3.2 Holstein-Primakoff representation (spin waves)

One simple approach for describing systems with a broken symmetry is to study the quantum fluctuations around the classic value of the order parameter. In the case of spin systems, we call these fluctuations *spin waves*. The simplest method to describe these spin fluctuations was introduced by Holstein and Primakoff [45]. They chose the following representation for the spin components in terms of the bosonic operator a:

$$S^{+} = \left(\sqrt{2S - a^{\dagger}a}\right)a\tag{3.9}$$

$$S^- = a^\dagger \sqrt{2S - a^\dagger a} \tag{3.10}$$

$$S^z = S - a^{\dagger}a \tag{3.11}$$

where, without loss of generality, the broken symmetry of the spin has been taken along the z-axis.

Intuitively, $n_a \equiv a^{\dagger}a$ describes the quantized "number" of deviations from the classically ordered spin, resulting in a reduced value for the spin component $S^z \leq S$. For their part, S^+ and S^- destroy and create a fluctuation "unit" respectively. This is consistent with the spin component being *raised* by S^+ and *lowered* by S^- .

Because the z-component of spin has a minimum value of $S_{\min}^Z = -S$, the Fock space must be reduced to:

$$\{|n_a\rangle\} = \{|0\rangle, |1\rangle, \dots, |2S\rangle\}$$

$$(3.12)$$

if one wishes to retain a physical solution. It's precisely the square roots $(\sqrt{2S - n_a})$ in the definitions of the raising and lowering spin operators that act as projectors to ensure the resulting states belong to the aforementioned reduced, physical Fock space.

Spin wave theory, being semi-classical in nature, approaches the problem using a large-S approximation. Thus, the usual techniques involve expanding the square

root $\sqrt{2S - a^{\dagger}a}$ in powers of $\mathcal{O}\left(\frac{1}{S}\right)$:

$$\sqrt{2S - a^{\dagger}a} \simeq \sqrt{2S} \left(1 - \frac{a^{\dagger}a}{4S} - \frac{a^{\dagger}aa^{\dagger}a}{32S^2} + \cdots \right)$$
(3.13)

up to the desired order. This, or course, destroys the constraint on the Fock space so that it is now necessary to check that the final results remain physical.

3.2.1 Linear Spin Waves (LSW)

The lowest truncation, up to $\mathcal{O}(S)$ gives the following bilinear and biquadratic Hamiltonians:

$$\mathcal{H}_{\rm bl}^{\rm FM} = NJS^2 - JS \sum_{\langle i,j \rangle} \left(a_i^{\dagger} a_i + a_j^{\dagger} a_j \right) - \left(a_i^{\dagger} a_j + a_j^{\dagger} a_i \right)$$
(3.14)

$$\mathcal{H}_{\rm bl}^{\rm AFM} = -NJS^2 + JS \sum_{\langle i,j \rangle} a_i^{\dagger} a_i + a_j^{\dagger} a_j + a_i^{\dagger} a_j + a_j^{\dagger} a_i \qquad (3.15)$$

$$\mathcal{H}_{bq}^{FM} = NKS^4 - 2KS^3 \sum_{\langle i,j \rangle} \left(a_i^{\dagger} a_i + a_j^{\dagger} a_j \right) - \left(a_i^{\dagger} a_j + a_j^{\dagger} a_i \right)$$
(3.16)

$$\mathcal{H}_{bq}^{AFM} = NKS^4 - 2KS^3 \sum_{\langle i,j \rangle} a_i^{\dagger} a_i + a_j^{\dagger} a_j + a_i^{\dagger} a_j + a_j^{\dagger} a_i, \qquad (3.17)$$

where we have used the fact that the biquadratic coupling is or the order $K \sim 1/S^2$ and N is the total number of sites. All these can be solved exactly in both cases. In particular, due to the appearance of off-diagonal terms, we must use a

Bogoliubov transformation in the antiferromagnetic case. This will be necessary on any system whose Hamiltonian has at least one antiferromagnetic term.

3.2.2 Non Linear Spin Waves (NLSW)

Going one order further $[\mathcal{O}(S^0)]$ leaves us with the following terms instead:

$$(\boldsymbol{S}_{\boldsymbol{r}} \cdot \boldsymbol{S}_{\boldsymbol{r}'})^{FM} = S^{2} - S \left[\left(a_{\boldsymbol{r}}^{\dagger} a_{\boldsymbol{r}} + a_{\boldsymbol{r}'}^{\dagger} a_{\boldsymbol{r}'} \right) - \left(a_{\boldsymbol{r}}^{\dagger} a_{\boldsymbol{r}'} + a_{\boldsymbol{r}'}^{\dagger} a_{\boldsymbol{r}} \right) \right] + a_{\boldsymbol{r}}^{\dagger} a_{\boldsymbol{r}'}^{\dagger} a_{\boldsymbol{r}} a_{\boldsymbol{r}'} - \frac{1}{4} \left(a_{\boldsymbol{r}}^{\dagger} a_{\boldsymbol{r}}^{\dagger} a_{\boldsymbol{r}} a_{\boldsymbol{r}} a_{\boldsymbol{r}'} + a_{\boldsymbol{r}}^{\dagger} a_{\boldsymbol{r}'}^{\dagger} a_{\boldsymbol{r}} a_{\boldsymbol{r}} + a_{\boldsymbol{r}'}^{\dagger} a_{\boldsymbol{r}'}^{\dagger} a_{\boldsymbol{r}'} a_{\boldsymbol{r}'} a_{\boldsymbol{r}'} a_{\boldsymbol{r}'} a_{\boldsymbol{r}'} \right)$$

$$(3.18)$$

$$(\boldsymbol{S}_{\boldsymbol{r}} \cdot \boldsymbol{S}_{\boldsymbol{r}'})^{AFM} = -S^2 + S \left(a_{\boldsymbol{r}}^{\dagger} a_{\boldsymbol{r}} + a_{\boldsymbol{r}'}^{\dagger} a_{\boldsymbol{r}'} + a_{\boldsymbol{r}} a_{\boldsymbol{r}'} + a_{\boldsymbol{r}}^{\dagger} a_{\boldsymbol{r}'}^{\dagger} \right) - a_{\boldsymbol{r}}^{\dagger} a_{\boldsymbol{r}'}^{\dagger} a_{\boldsymbol{r}} a_{\boldsymbol{r}'} - \frac{1}{4} \left(a_{\boldsymbol{r}}^{\dagger} a_{\boldsymbol{r}} a_{\boldsymbol{r}} a_{\boldsymbol{r}'} + a_{\boldsymbol{r}}^{\dagger} a_{\boldsymbol{r}}^{\dagger} a_{\boldsymbol{r}'} a_{\boldsymbol{r}} + a_{\boldsymbol{r}'}^{\dagger} a_{\boldsymbol{r}'} a_{\boldsymbol{r}} a_{\boldsymbol{r}'} + a_{\boldsymbol{r}'}^{\dagger} a_{\boldsymbol{r}'}^{\dagger} a_{\boldsymbol{r}'} a_{\boldsymbol{r}'} + a_{\boldsymbol{r}'}^{\dagger} a_{\boldsymbol{r}'}^{\dagger} a_{\boldsymbol{r}'} a_{\boldsymbol{r}'} \right)$$

$$(3.19)$$

$$\begin{bmatrix} \left[\left(\mathbf{S}_{r} \cdot \mathbf{S}_{r'} \right)^{2} \right]^{FM} = S^{4} - 2S^{2} \left(S - 1 \right) \left[\left(a_{r}^{\dagger} a_{r} + a_{r'}^{\dagger} a_{r'} \right) - \left(a_{r}^{\dagger} a_{r'} + a_{r'}^{\dagger} a_{r} \right) \right] + S^{2} \left(1 + 6a_{r}^{\dagger} a_{r'}^{\dagger} a_{r} a_{r'} \right) - S^{2} \left[\frac{5}{2} \left(a_{r}^{\dagger} a_{r}^{\dagger} a_{r} a_{r'} + a_{r'}^{\dagger} a_{r'}^{\dagger} a_{r} a_{r} + a_{r'}^{\dagger} a_{r'}^{\dagger} a_{r'} a_{r'} + a_{r'}^{\dagger} a_{r'}^{\dagger} a_{r'} + a_$$

$$\left[\left(\boldsymbol{S}_{\boldsymbol{r}} \cdot \boldsymbol{S}_{\boldsymbol{r}'} \right)^2 \right]^{AFM} = S^4 - 2S^2 \left(S - 1 \right) \left(a_{\boldsymbol{r}}^{\dagger} a_{\boldsymbol{r}} + a_{\boldsymbol{r}'}^{\dagger} a_{\boldsymbol{r}'} + a_{\boldsymbol{r}} a_{\boldsymbol{r}'} + a_{\boldsymbol{r}}^{\dagger} a_{\boldsymbol{r}'}^{\dagger} \right) + S^2 \left[S^2 \left(1 + 6a_{\boldsymbol{r}}^{\dagger} a_{\boldsymbol{r}'}^{\dagger} a_{\boldsymbol{r}} a_{\boldsymbol{r}} - a_{\boldsymbol{r}'} \right) + S^2 \left[S^2 \left(a_{\boldsymbol{r}}^{\dagger} a_{\boldsymbol{r}} a_{\boldsymbol{r}} a_{\boldsymbol{r}'} + a_{\boldsymbol{r}}^{\dagger} a_{\boldsymbol{r}'}^{\dagger} a_{\boldsymbol{r}'} + a_{\boldsymbol{r}}^{\dagger} a_{\boldsymbol{r}'}^{\dagger} a_{\boldsymbol{r}'} + a_{\boldsymbol{r}'}^{\dagger} a_{\boldsymbol{r}'}^{\dagger} a_{\boldsymbol{r}'} \right) \right]$$

$$\left(a_{\boldsymbol{r}}^{\dagger} a_{\boldsymbol{r}}^{\dagger} a_{\boldsymbol{r}} a_{\boldsymbol{r}} + a_{\boldsymbol{r}'}^{\dagger} a_{\boldsymbol{r}'} a_{\boldsymbol{r}'} + a_{\boldsymbol{r}'} a_{\boldsymbol{r}'} a_{\boldsymbol{r}'} + a_{\boldsymbol{r}'}^{\dagger} a_{\boldsymbol{r}'}^{\dagger} a_{\boldsymbol{r}'}^{\dagger} a_{\boldsymbol{r}'} \right) \right]$$

$$(3.21)$$

One then can use Wick's theorem to decouple all terms and recover a Hamiltonian bilinear in the creation and anihilation operators a_r^{\dagger} and a_r . In this decoupling, we introduce the following averages:

$$n = \left\langle a_{\mathbf{r}}^{\dagger} a_{\mathbf{r}} \right\rangle$$

$$f_{\hat{\mathbf{r}}} = \left\langle a_{\mathbf{r}}^{\dagger} a_{\mathbf{r}+\hat{\mathbf{r}}} \right\rangle = \left\langle a_{\mathbf{r}} a_{\mathbf{r}+\hat{\mathbf{r}}}^{\dagger} \right\rangle$$

$$g_{\hat{\mathbf{r}}'} = \left\langle a_{\mathbf{r}} a_{\mathbf{r}+\hat{\mathbf{r}}'} \right\rangle = \left\langle a_{\mathbf{r}}^{\dagger} a_{\mathbf{r}+\hat{\mathbf{r}}'}^{\dagger} \right\rangle.$$
(3.22)

Here, $f_{\hat{r}}$ measures the correlations between neighbors along an axis given by the director \hat{r} where the bonds are *ferromagnetic*. The parameter $g_{\hat{r}'}$ does the same

job for an axis of *antiferromagnetic* bonds. The anomalous form of this average is only due to the rotation of π performed on one of the sublattices in the case of antiferromagnetic orderings. Once decoupled, the system can then be solved with a series of self-consistent equations whose form depends on the particular Hamiltonian.

However, if one wishes to save themselves some work, there is a way to simplify these calculations. By using the so-called Hubbard-Stratonovich transformation to decouple the biquadratic terms as follows:

$$(\boldsymbol{S}_{\boldsymbol{r}} \cdot \boldsymbol{S}_{\boldsymbol{r}'})^2 \simeq 2 \langle \boldsymbol{S}_{\boldsymbol{r}} \cdot \boldsymbol{S}_{\boldsymbol{r}'} \rangle \, \boldsymbol{S}_{\boldsymbol{r}} \cdot \boldsymbol{S}_{\boldsymbol{r}'} - \langle \boldsymbol{S}_{\boldsymbol{r}} \cdot \boldsymbol{S}_{\boldsymbol{r}'} \rangle^2 \,, \qquad (3.23)$$

Where the Hubbard-Stratonovich averages themselves can also be expressed in terms of the order parameters we described above.

$$\Gamma_{\hat{r}} = \langle \boldsymbol{S}_{\boldsymbol{r}} \cdot \boldsymbol{S}_{\boldsymbol{r}+\hat{\boldsymbol{r}}} \rangle = (S - n + f_{\hat{\boldsymbol{r}}})^2$$

$$\Gamma_{\hat{\boldsymbol{r}}'} = \langle \boldsymbol{S}_{\boldsymbol{r}} \cdot \boldsymbol{S}_{\boldsymbol{r}+\hat{\boldsymbol{r}}'} \rangle = -(S - n - g_{\hat{\boldsymbol{r}}'})^2.$$
(3.24)

Again, \hat{r} denotes a direction along which the bonds between spins are ferromagnetic and \hat{r}' indicates an antiferromagnetically aligned axis instead. In what follows, we will call this last method, the Hubbard-Stratonovich (HS) or simply decoupled (SD) approach, in order to distinguish it from the full decoupling (FD) used originally. As we will see in Section 3.2.3, using the HS simplification comes at the expense of sacrificing some of the accuracy in the results. The rest of the details for this method, which depend on the particular Hamiltonian, are described on Appendix A.

3.2.3 Comparing the different order results

Before we proceed further, it is useful to compare the effects of higher order expansions with those of the regular linear spin-wave theory (LSWT). To do so, we use one of the simplest models to include both frustration and a biquadratic coupling:

$$\mathcal{H} = J_1 \sum_{\langle i,j \rangle} \boldsymbol{S}_i \cdot \boldsymbol{S}_j + J_2 \sum_{\langle \langle i,j \rangle \rangle} \boldsymbol{S}_i \cdot \boldsymbol{S}_j - K \sum_{\langle i,j \rangle} \boldsymbol{S}_i \cdot \boldsymbol{S}_j$$
(3.25)

We study this Hamiltonian on a square lattice, in the $J_2/J_1 > 1/2$ regime, where the system acquires an antiferromagnetic *columnar* order at T = 0 and plot the results of the two approaches to the NLSW as well as the LSW dispersion for the cases of spin S = 1 (Fig. 3.1) and S = 2 (Fig. 3.2).



Figure 3.1: Dispersions using results from the linear spin waves and both approaches to the non-linear spin waves for the case of S = 1.



Figure 3.2: Dispersions using results from the linear spin waves and both approaches to the non-linear spin waves for the case of S = 2.
Immediately, we can appreciate the big improvement that the FD method offers over the HS approach. Indeed, save for the lifting of the goldstone mode at (π, π) , the results obtained using a HS transformation almost overlap with the LSW dispersions for all K values. In particular, they replicate[46] the appearance of maximum at the zone boundary $[M = (\pi, \pi)]$ with an increasing value of K.

On the other hand, the dispersions that we obtain with a FD correctly account for quantum fluctuations. This results on the suppression of the aforementioned maximum, with a local minimum being observed at the (π, π) point for all values of K.

Finally, while the discrepancies between the HS (along with the LSW) results and those obtained with a FD increase with the value of K in both cases, the overall differences are smaller in the case of S = 2 than that of S = 1, indicating a smaller effect of quantum fluctuations in the former system. This accounts for the fact that increasing values of spin get progressively closer to the large-S limit, thus approaching a more classical behavior where quantum fluctuations acquire a more secondary role.

3.3 Dyson-Maleev (DM) bosons

Developed by Dyson and Maleev [47, 48, 49] in 1956-57, the Dyson-Maleev bosons offer a representation of spin where no expansion around any small parameter is needed. Because the expressions are obtained by slightly modifying those given by Holstein and Primakoff, this representation is sometimes given the name of "modified spin wave". Ironically, the motivation behind the Dyson-Maleev bosons is precisely to abandon a semi-classical approach in favor of a mean field technique that allows us to study a spin system even in regimes with no broken symmetry, precisely where the spin wave small fluctuation approach does not work. Because of this, we will use the Dyson-Maleev bosons both in magnetically ordered phases as well as in paramagnetic regimes. The spin operators are now defined as follows:

$$S^{+} = \sqrt{2S} \left(1 - \frac{a^{\dagger}a}{2S} \right) a$$
$$S^{-} = \sqrt{2S}a^{\dagger}$$
$$S^{z} = S - a^{\dagger}a$$
(3.26)

The square roots present in the Holstein-Primakoff approach have now disappeared but the price to pay is that the spin "raising" and "lowering" operators are not conjugates of each other anymore. However, when applying this representation to both bilinear and biquadratic terms, the Hamiltonian remains hermitian, so that we can proceed with the method at hand. Just like in the case of the non-linear spin waves, this technique leads to terms with more than two bosonic operators, which we must decoupled in order to obtain a solvable, bilinear effective Hamiltonian. Both the HS and FD approaches are possible here but we restrict ourselves to the latter due to its proven superior accuracy. Specific details can be found in Appendix B.

3.4 Schwinger bosons

Unlike all previously described approaches, the Schwinger boson representation introduces two different bosonic operators, a and b to describe the spin components:

$$S^+ = a^{\dagger}b \tag{3.27}$$

$$S^- = b^\dagger a \tag{3.28}$$

$$S^{z} = \frac{1}{2}(a^{\dagger}a - b^{\dagger}b)$$
(3.29)

Intuitively, a^{\dagger} creates on "unit" of spin up and b^{\dagger} does the same for a spin down. As such, the total number of bosons in both modes must satisfy: $n_a + n_b = 2S$ [see panel a) in Fig. 3.3]. Just like in the case of the Dyson-Maleev representation, this approach can be used for paramagnetic regimes as well as magnetically ordered ones due to the lack of an expansion around a small parameters. However, if one decides to do so, it is possible to condense one of the bosons by expressing its value in terms of fluctuations in the remaining mode: $a = \sqrt{2S - n_b}$. By doing so, we immediately recover the Holstein-Primakoff representation above.

3.5 Generalized spin waves (flavor waves)

In the case of the Schwinger bosons above, the spin operators are always given by the same expressions, using an SU(2) representation To derive the generalized spin waves however, we start by a generalized SU(N) representation of the spin operators:

$$S_{mm'}^{x} = \delta_{m(m'-1)} \frac{\sqrt{(m+1)(2S-m)}}{2} + \\ + \delta_{(m+1)m'} \frac{\sqrt{(m'+1)(2S-m')}}{2} \\ S_{mm'}^{y} = \delta_{m(m'-1)} \frac{\sqrt{(m+1)(2S-m)}}{2i} - \\ - \delta_{(m+1)m'} \frac{\sqrt{(m'+1)(2S-m')}}{2i} \\ S_{mm'}^{z} = \delta_{mm'}(S-m)$$

$$(3.30)$$

The constraint on the physical space is now given by:

$$\sum_{m=0}^{N-1} b_{rm}^{\dagger} b_{rm} = N\mathcal{S} \tag{3.31}$$

By condensing one of the bosons, one can then expand its corresponding operators using an approach similar to that of the Holstein-Primakoff representation.

$$b_{r0}^{\dagger} = b_{r0} = \sqrt{1 - \sum_{m=1}^{N-1} b_{rm}^{\dagger} b_{rm}} \simeq 1 - \frac{1}{2} \sum_{m=1}^{N-1} b_{rm}^{\dagger} b_{rm}$$
(3.32)

In order to work with both magnetic and non-magnetic phases alike, it is useful to recast the $J_{ij} - K_{ij}$ model in terms of the traceless symmetric quadrupolar tensor:

$$Q^{\alpha\beta} = S^{\alpha}S^{\beta} + S^{\beta}S^{\alpha} - \frac{2}{3}S(S+1)\delta_{\alpha\beta}, \qquad (3.33)$$

whose 5 independent components can be cast into the following 5-component vector $Q \equiv [(Q^{xx} - Q^{yy})/2, (2Q^{zz} - Q^{xx} - Q^{yy})/2\sqrt{3}, Q^{xy}, Q^{yz}, Q^{xz}]$. Using the identity: $2(\mathbf{S}_i \cdot \mathbf{S}_j)^2 = \mathbf{Q}_i \cdot \mathbf{Q}_j - \mathbf{S}_i \cdot \mathbf{S}_j + \frac{8}{3}$ for the case of S = 1, a general $J_{ij} - K_{ij}$ Hamiltonian can be written as follows:

$$\mathcal{H} = \frac{1}{2} \sum_{i,j} \left(J_{ij} - \frac{K_{ij}}{2} \right) \boldsymbol{S}_i \cdot \boldsymbol{S}_j + \frac{1}{4} \sum_{i,j} K_{ij} \left(\boldsymbol{Q}_i \cdot \boldsymbol{Q}_j + \frac{8}{3} \right).$$
(3.34)

We note that a biquadratic term results in an *effective* renormalized coupling $J_{ij} - \frac{K_{ij}}{2}$, similar to those in Section 3.2.3. The specific spin wave dispersions for each particular model described in the chapters below can be found in Appendix C.

3.6 Side-by-side

The earlier differences between the two approaches to NLSW are reminiscent of the work by Stanek *et al.* [46], who compared the dispersions obtained with a full decoupling of the Dyson-Maleev bosons with those of the usual Schwinger boson approach. In a similar analysis, we compare those same Dyson-Maleev results with those of the remaining spin representations described above [see panel a) in Fig. 3.4]. Note that in the case of the GSW we only plot the dispersion corresponding to the lowest energy mode.



Figure 3.3: The Schwinger boson approach (left) allows only for fluctuations around the classical vector field. For S = 1, the GSWT (right) includes an extra mode of fluctuations that don't modify the value of S_z . The constraint on the physical space still remains, so that the maximum number of fluctuation units (the steps, in this simplified representation) is still 2S.

Just as the full decoupling (FD) of the Holstein-Primakoff (HP) bosons, both of the newly added methods (namely, the FD of the DM bosons and the GSW) give an accurate estimate of the effect of quantum fluctuations, evidenced again by the suppression of the maximum at the zone boundary. In the case of the DM approach, this is due to the use of a *full* decoupling of the biquadratic term, which allows us to account for *all* spin correlations. In the case of the GSW, the correct dispersions are a result of including the most general order parameter, in contrast with the expansion around only the *classical* vector field used in all other methods. As an aside, the dispersions resulting from a FD of the HP bosons seem to actually *overestimate* the



Figure 3.4: a) Dispersion for all the described spin representations. In b), we plot the value of the normalized dispersion maxima for increasing values of the biquadratic coupling K.

effect of quantum fluctuations compared to other methods. This could be attributed to the absence of all the necessary dispersion modes, which the GSWT correctly accounts for.

To put an end to our analysis and in the interest of completeness, we briefly touch on the result for a simpler J - K model with a Néel ground state:

$$\mathcal{H} = J \sum_{\langle i,j \rangle} \boldsymbol{S}_i \cdot \boldsymbol{S}_j - K \sum_{\langle i,j \rangle} \boldsymbol{S}_i \cdot \boldsymbol{S}_j$$
(3.35)

Stanek *et al.* [46] used this Hamiltonian as well to initially benchmark the DM and SB approaches. This allows us to compare their results to those obtained with out methods of choice. In this case, we plot the value of the dispersion maxima at the

Method	Slope
DM (Stanek <i>et al.</i>)	1.2771
SB (Stanek <i>et al.</i>)	2.6567
NLSW (Simple Decoupling)	2.3284
NLSW (Full Decoupling)	0.7319
DM	1.2771
GSW	1.0000

Table 3.1: Comparison of the spin-wave dispersion slopes $d\omega_{max}/dK$ calculated with different methods in this work and in Ref. [46] (Stanek *et al.*). Here ω_{max} is the maximum of the spin-wave dispersion at (π, π) and the slope is obtained from a linear fit of the data in panel b) of Figure 3.4.

edge of the Brillouin zone for several values of K [see panel b) in Fig. 3.4], normalized with respect to the K = 0 case. As expected, the results mimic those obtained for the previous, frustrated model. The FD of the NLSW and DM bosons, as well as the GSW, all produce similar slopes, while the SD of the NLSW overestimates the value of the maxima by an approximate factor of 2 (see Table 3.1 for the exact numerical values). Yet again, we attribute this discrepancy to the inability of the Hubbard-Stratonovich method to properly account for the quantum fluctuations. The SB approach also suffers from this pitfall, with a slope close to that produced by the simply decoupled NLSW.

We also compute the value of the slope $d\omega_{\max}(K)/dK$ (see Table 3.1) to obtain a quantitative measure of the results described above. This also allows us to numerically prove the equivalency of our technique for the DM bosons (self-consistent system of Euler-Lagrange equations) to that of Stanek *et al.* (free energy minimization).

Chapter 4

Ising-nematic order in the iron pnictides

4.1 Model and solutions

In order to study the magnetism in the parent compounds of the iron pnictides, we propose the frustrated Heisenberg $J_1 - J_2$ model with a biquadratic coupling Kon a lattice of quasi-local moments:

$$\mathcal{H} = J_1 \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j - K \sum_{\langle i,j \rangle} (\mathbf{S}_i \cdot \mathbf{S}_j)^2 + J_2 \sum_{\ll i,k \gg} \mathbf{S}_i \cdot \mathbf{S}_k + J_c \sum_i \mathbf{S}_i \cdot \mathbf{S}_{i+\hat{z}}$$
(4.1)

The reasons for this particular choice are many. To begin with, and just as various other authors have done[50, 51, 52, 20, 53, 54, 21, 55], we use a system of localized

spins. This is due to the prediction of the proximity of the parent compounds to a Mott localization transition (see Section 2.2 for more details). The two terms encompassing the frustrated nature of the problem, namely J_1 and J_2 give us the ground state we base all of our calculations on. We choose $J_1 = J_2$ for the rest of this work, obtaining a J_2/J_1 ratio well into the $J_2/J_1 > 1/2$ regime where the spins arrange into a *columnar* antiferromagnetic (CAFM) structure with alternating spin stripes of opposite orientation.

We also set the spin value to S = 1 for all the results that follow. Physically, this is because the Fe ion in the pnictides is in a Fe²⁺ oxidation state, leaving two electrons in xz and yz orbitalas that are strongly coupled via Hund's interaction. This means that the value S = 1 we're choosing is justified as the *effective* spin of the local moment on each Fe site. This value for the effective spin S also agrees with results from INS experiments[56], which found an integrated spin spectral weight of $3\nu_B$ per Fe on the pnictide parent compounds. Indeed, it follows from the formula for the magnetic moment of a spin:

$$\left\langle m^2 \right\rangle = \left(g\nu_B\right)^2 S(S+1),\tag{4.2}$$

with g = 2[57] that S = 1 gives, in fact, the best approximation.

Beyond the basic frustrated Heisenberg Hamiltonian, we include the biquadratic term to nearest neighbors since an *effective* anisotropic coupling is necessary for agreement with INS data [58, 59], in particular to correctly describe the dispersion of the spin excitations near the Brillouin zone boundary. Without it, we would need to set $J_1^x \neq J_1^y$, with widely different values for each [58, 60, 61], even beyond the structural transition, which is unphysical. In fact, even below T_s , the small $(\delta \leq 1\%)$ orthorrombic distortion makes any significant difference between the values of J_1 in each direction highly unlikely. The addition of a biquadratic coupling Kdynamically produces the desired effective anisotropic coupling to nearest neighbors without directly manipulating the values of J_1 . Finally, we include an exchange term of strength J_c between the pnictogen planes to capture the three-dimensional nature of the real compounds.

Because we will study the system for a wide range of temperatures, ranging from below the magnetic transition temperature (T_N) to above that of the structural transition (T_s) , we need a method that works beyond the ordered phase. This means that we must discard the semi-classical spin wave approaches that depend on the fluctuations around the ordered moment to be small. Because the Dyson-Maleev bosons proved superior to the Schwinger bosons in the treatment of quantum fluctuations (see Section 3.6 above), we choose the former approach for the remainder of this work. The details of the derivations and the spin excitation dispersions are given in Appendix B.

4.2 Magnetic and nematic transitions

As explained on Section 2.2, part of the interest in the iron pnictides lies on the difference between the magnetic and structural transition temperatures (T_N and T_s , respectively). Moreover, the Ising-nematic phase has been observed even beyond the

structural phase transition. Because of this, it is natural to study the evolution of order parameters that measure both the magnetic order and the nematicity of the system. Choosing the *staggered* magnetization (m_s) for the former is straightforward. In the case of nematicity, we choose to use the difference between the correlations along the two crystallographic directions:

$$\Gamma_{y} - \Gamma_{x} \equiv \langle \boldsymbol{S}_{r} \cdot \boldsymbol{S}_{r \pm \hat{\boldsymbol{y}}} \rangle - \langle \boldsymbol{S}_{r} \cdot \boldsymbol{S}_{r \pm \hat{\boldsymbol{x}}} \rangle$$

$$(4.3)$$

By plotting the evolution of these two parameters with temperature, we can easily obtain the values of the two transition temperatures. We use the usual nomenclature T_N for the temperature where the staggered magnetization m_s vanishes. It is less clear whether we can identify the temperature our nematic parameter vanishes with the structural transition temperature T_s or another, higher temperature T^* up to which nematicity survives [19]. Because of this, we choose the alternate notation T_{σ} . For a fixed value of $K = 0.01J_2$ (see Fig. 4.1), the difference between the two transition temperatures greatly reduces every time J_c increases by an order of magnitude. Note that we normalized the temperature axis so that $T_N = 1$. This allows use to study the *absolute* differences among the nematic ranges without those being obscured by variations on m_s . Note that the discontinuous first order transition that can be observed for the nematic order parameter is only due to the mean-field nature of our method.

More generally, plotting the *actual* transition temperature values as we vary the interlayer coupling (see Fig. 4.2) allows us to obtain a clearer view of the existence



Figure 4.1: Evolution of the nematic order parameter $(\Gamma_y - \Gamma_x)$ and staggered magnetization (m_s) at $K = 0.01J_2$ with temperature normalized to $T_N = 1$, for increasing values of J_c . The interplanar coupling rapidly reduces the nematic range with each order of magnitude, having it completely vanish at $J_c \sim 0.1J_2$.

range of each regime. It turns out that the variation in the temperature range for the Ising-nematic phase is mainly mediated by the temperature of the magnetic transition instead. As expected, the magnetically ordered range rapidly increases with J_c . The mechanism by which the interplanar coupling stabilizes magnetic order is related to the dimensionality of the problem. In practice, the addition of a finite J_c takes the system from two to three dimensions. Even though the discrete Z_2 symmetry can still be broken in a two-dimensional system at finite temperatures [62], this is not the case for a continuous symmetry by virtue of the Mermin-Wagner theorem [63]. Thus, unlike a purely Ising-nematic phase, true long-range magnetic order can't strictly exist in two dimensions. Because the value of J_c is, in a way, a



Figure 4.2: Evolution of both the magnetic and nematic transition temperatures $(T_N$ and T_{σ} , respectively) for biquadratic coupling values of K = 0 (a) and $K = 0.01J_2$ (b). A finite interplanar coupling effectively changes the dimensionality of the system, thus stabilizing the magnetic order. This results in a rapid narrowing of the range where pure nematicity exists. For finite values of K, this range virtually disappears for appreciable values of the interlayer coupling.

measure of the system dimensionality, the sharp increase in the range where magnetic order exists when $J_c \rightarrow 0$ is a direct consequence of this fact.

When a biquadratic coupling is not present [Fig. 4.2 a)], there still exists a small range of pure nematicity at the reasonably physical value of $J_c \sim 0.1 J_2$. When K is turned on, however, the Ising-nematic phase quickly disappears for these values, even for the very small value of $K = 0.01 J_2$ [Fig. 4.2 b)]. In principle, this is an unexpected find since previous fits on the INS data [64] have given estimates of about $K \sim 0.6 J_1$ for the biquadratic coupling. However, the samples where these nematic fluctuations have been detected at a temperature T^* well above T_N [19] (and T_s , for that matter) were all detwinned by applying an uniaxial pressure on them, which explicitly breaks



Figure 4.3: Phase diagrams for interlayer coupling values of $J_c = 0.01J_2$ (a) and $J_c = 0.1J_2$ (b). The range of temperatures in which a purely nematic regime exists quickly decreases with the value of the biquadratic coupling K. When the interplanar coupling reaches values of $J_c = 0.1J_1$, the nematic phase has almost disappeared and the magnetic and nematic transition temperatures $(T_N \text{ and } T_{\sigma})$ coincide for almost the entire range of K.

the C_4 symmetry. Because of this, T^* is a more appropriate measure of a crossover rather than a true phase transition, and in consequence, could be greatly affected by the value of the applied pressure. Since our model doesn't include the effect of this external variable, this suggests that our definition of T_{σ} might be better suited to describe the structural transition temperature T_s . If this is the case, the very narrow ranges observed for the purely nematic phase are in good agreement with the close proximity observed between T_N and T_s .

If we instead fix the value of J_c and study the evolution of both the magnetic and nematic transition temperatures as a function of K instead (see Fig 4.3), we find, yet again, that the range of temperatures where a pure Ising-nematic order exists shrinks rapidly as the value of the biquadratic coupling increases. This is

especially noticeable when we choose the more physical value of $J_c = 0.1J_2$ for the interplanar coupling, in which case the nematic regime vanishes almost completely. These results are in stark contrast with those of Yu et al. [65], who found an increase in both transition temperatures with K. However, this work used a simpler version of the the Dyson-Maleev approach, decoupling the biquadratic term by means of a Hubbard-Stratonovich transformation, similar to our simply decoupled NLSW. This method leads to a more straightforward Hamiltonian, with effective couplings of values $[J_1^{\text{eff}}]_{x(y)} = J_1 - 2K\Gamma_{x(y)}$. Because $\Gamma_x < 0$, it is clear by these expressions that the solutions lead to enhanced couplings on the x-direction while the y-direction coupling is diminished, leading to a more stable breaking of the C_2 symmetry the larger the value of K gets. Our technique, although proven to be more accurate in describing quantum fluctuations (see Section 3.6), leads to more cumbersome expressions so that one cannot a priori predict the effects of K on the anisotropy. We conclude that, within our approach, considerable fine-tuning of both the interlayer exchange J_c and the biquadratic coupling K are necessary for the existence of a pure Ising-nematic phase.

4.3 Dynamical structure factors

We now go on to compare the experimentally measured dynamical structure factors (these are proportional to the differential cross sections in inelastic neutron scattering experiments) with those predicted by our model (see Appendix B for expressions). In all cases, we choose a Lorentzian broadening with a width of value $\gamma = 0.5J_2$ to account for both Landau damping and the finite experimental resolution. We also rotationally symmetrize the results *a posteriori* so that we can properly compare them with twinned samples.

We begin by tackling the results at low temperatures, comparing the predictions of our method at T = 0 with the findings by Harriger *et al.* [59] (see Fig. 4.4). To do so, we choose four different cuts of increasing energy ω , where we should observe maxima at positions \boldsymbol{q} where $\omega \approx \omega_{\boldsymbol{q}}$ as a consequence of energy conservation. Indeed, the rings the appear at the lower energy cuts [Figs. 4.4 a) and b)] are centered at the two degenerate ordering vectors $\boldsymbol{q}_1 = (\pi, 0)$ and $\boldsymbol{q}_2 = (0, \pi)$, as expected, and in good agreement with the data [Figs. 4.4 e) and f)]. Both our theoretical predictions and the experimental results showcase elliptical rings as a result of the anisotropy of the system. As we increase the energy ω of the cuts, the positions of the maxima begin to shift towards the magnetic zone boundary and the rings lose its characteristic shape as they expand. For high enough energies [Figs. 4.4 c) and d)], the structure factor peaks now reside at the $\boldsymbol{q} = (\frac{\pi}{2}, \frac{\pi}{2})$ position. This is in semi-qualitative agreement with the data [Figs. 4.4 g) and h)], where the discrepancies are likely due to a larger effective broadening γ resulting from Landau damping.

In order to study the evolution of the dynamical structure factor at finite temperatures, we now choose to compare our theoretical predictions with the INS measurements performed on BaFe_{2-x}Ni_xAs₂ by Lu *et al.* [19]. In the temperature range where our model allows for a pure Ising-nematic regime $(T_N < T < T_{\sigma})$, we do indeed find a C_2 anisotropy in the structure factor [see Fig. 4.5 a)], as expected and in agreement with the data [Fig. 4.5 c)]. This feature disappears at higher temperatures both in our model [Fig. 4.5 b)] and as measured experimentally [Fig. 4.5 d)].



Figure 4.4: Side-by-side comparison of theoretical calculations (left side) and experimental results (right side) [59] for several energy cuts of the low temperature dynamical structure factors. We fix the value of $K = 0.8J_2$ and choose the energy values of a) $\omega = 2J_2$, b) $\omega = 3J_2$, c) $\omega = 5J_2$, and d) $\omega = 6J_2$ for the aforementioned cuts.



Figure 4.5: Calculated dynamical structure factor [a) and b)] with set values of S = 1, K = 0, and $J_c = 0.1J_2$ for the parameters of our model. The cuts are taken at energy $\omega = 0.5J_2$. Temperatures are a) $T = 1.043T_N$ [indicated in Fig. 4.2 a)] and b) $T = T_{\sigma} = 1.085T_N$. Panels c) and d) are experimental INS data on detwinned BaFe_{2-x}Ni_xAs₂ samples above the Néel temperature $T_N = T_s \sim 138K$ [19].

Chapter 5

Chalcogenides

5.1 Model and solutions

In order to study the ground states of the iron chalcogenides, we propose the following spin Hamiltonian:

$$\mathcal{H} = J_1 \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + K_1 \sum_{\langle i,j \rangle} (\mathbf{S}_i \cdot \mathbf{S}_j)^2 + J_2 \sum_{\langle \langle i,j \rangle \rangle} \mathbf{S}_i \cdot \mathbf{S}_k + K_2 \sum_{\langle \langle i,j \rangle \rangle} (\mathbf{S}_i \cdot \mathbf{S}_j)^2 + J_3 \sum_{\langle \langle \langle i,j \rangle \rangle \rangle} \mathbf{S}_i \cdot \mathbf{S}_j$$
(5.1)

Just as the model used for the pnictides, here we include both bilinear and biquadratic terms in the spins as well. However, we have extended the range of the J_m and K_n constants up to one neighbor further (m = 3 and n = 2, respectively) further. The need for additional parameters in this model with respect to that in the previous chapter shouldn't come as a surprise, considering the additional magnetically ordered phases observed in the chalcogenide family (see Section 2.3).

To avoid any confusion, we must also comment on a small change in notation. Because in Chapter 4, we only used negative values for the constant K, we explicitly added the a minus sign in front of the biquadratic term on Eq. 4.1 for convenience. We then went on to exclusively use positive values for the newly defined K througout the aforementioned chapter. However, in order to accommodete the full range of possible values for the biquadratic constants, and following the prevalent notation in more recent literature[66, 67, 68, 69], we now treat the signs of the bilinear and biquadratic constants on equal footing.

Building up on a variational search for possible magnetically ordered phases on a 4x4 Fe plaquette (performed by Zhentao Wang, see Acknowledgements), we use the generalized spin wave or flavor wave method described in Section 3.5 for the rest of our calculations.

5.2 Phase diagrams

Starting from the mean field results [Fig. 5.1, panels a) to d)], if we look at smaller values of $|K_1|$ (this corresponds to the top part of the phase diagrams), we can easily understand how the coupling between third nearest neighbors (J_3) drives the transition between successive magnetically ordered phases. At small J_3 and $|K_1|$ (top left of the diagrams), the system we are looking at is not dissimilar to the one we studied in Chapter 2.2 to describe the iron pnictides. Because we have chosen



Figure 5.1: K_1 vs. J_3 phase diagrams resulting from both the mean field [panels a) through d)] and the flavor wave [panels e) to h)] approaches.

values for J_1 and J_2 so that: $J_2/J_1 = 0.8 > 0.5$, we find the expected *columnar* antiferromagnetic phase (CAFM), as depicted in Fig. (5.2). In this regime, the bonds between third nearest neighbors are always ferromagnetic (FM). Because J_3 favors antiferromagnetic (AFM) bonds instead, the CAFM phase can remain energetically favorable only up to a certain value of J_3 . Unsurprisingly, its region of existence expands with increasing K_2 , due to the enhancement of the AFM bonds (in this case, between second nearest neighbors) discussed in Section 3.5.

Once J_3 becomes large enough, the system transitions into a staggered dimer (SD) phase. Here, the spins in the antiferromagnetically ordered axis remain unchanged while half of the bonds in the axis perpendicular to this lose their FM ordering (see Fig. 5.2 for a schematic depiction of this transition). Along this axis then, all third nearest neighbor bonds have now become AFM. Increasing J_3 even further will eventually result in AFM bonds between third nearest neighbors in *both* axes,



Figure 5.2: Simplified depiction of the transitions between the magnetically ordered phases, where the arrows point in the direction of increasing J_3 , the coupling between third nearest neighbors. Initially, on the FM bonds in one direction change, giving rise to the *staggered dimer* phase (DS). Eventually, the bonds become AFM in *both* directions and the system transitions into a *double stripe* or *plaquette* state.

resulting in the so-called *double stripe* (DS) or *plaquette* (PL) phases, which are energetically degenerate within the mean field approach. A simplified depiction of this transition is also included in Fig. 5.2.

Following up on the discussion about the effect of $|K_2|$ started above, we note than an increasing value of this coupling partially destabilizes the SD phase, by effectively weakening its FM bonds to second neighbors. This, combined with the enhanced stability of the CAFM results in a direct transition from the latter phase to either a DS or a PL regime, without the SD phase existing in between. This same effect is responsible for the shrinking of the DS and PL regions as $|K_2|$ increases.

The influence of $|K_1|$ is most noteworthy on the leftmost side of the diagram, where J_3 is small, and for the case of $K_2 = 0$. In this case, provided J_3 is small enough to be neglected, we are effectively left with the $J_1 - J_2 - K$ model that we used before (see Chapter 2.2). In the case of the pnictides though, we never studied the system for very large values of the biquadratic coupling to nearest neighbors (first neighbors). It turns out that, even for a ratio of $J_2/J_1 > 1/2$, the system eventually still transitions into a Néel state, provided the aforementioned biquadratic coupling assumes large enough values. The reason for this is simple. In both the CAFM and the Néel cases, a finite biquadratic coupling K_1 results in a stronger bond between nearest neighbor AFM bonds. This effect actually stabilizes both phases, by enhancing the anisotropy in the CAFM case and by doing the opposite in the Néel case. However, the latter phase has twice as many AFM bonds than the former. The FM bonds get weakened but the effect is not as stark, so that, for sufficiently large values of K_1 , the system favors the isotropic configuration.

As mentioned in Section. 3.5, the flavor wave approach needs the assumption of a particular state, since all consequently derived expressions depend on it. In this sense, the mean field approach is more powerful since it can *predict* new orders and produce a simplified phase diagram by just finding the energetically favorable state in each region. However, as far as this mean field method is concerned, all regimes can in principle exist throughout the entire phase diagram and it's only their ground state energy differences that matter. On the other hand, it is the quantum fluctuations that we can only compute using the flavor waves that are necessary to find the *true* regions where each phase can exist. This is because the flavor waves offer us a tool, i.e. the spin excitation dispersions, that indicates where each state becomes unstable.

With this, we go on to study the effects that fluctuations have on the regions of stability of each phase [see Fig. 5.1, panels e) to h)]. Qualitatively, the behavior of all phases remains the same, with changes to their phase boundaries. This is expected since quantum fluctuations, when pronounced enough, will destroy the long range order of the system. If we begin by our most recent discussion, we can see that both the CAFM and Néel stability regions have expanded. This suggests that the strengthening effect that biquadratic terms have on AFM bonds is actually *enhanced* by quantum fluctuations. In this case and in others throughout the phase diagram, the addition of quantum fluctuations caused some of the existence regimes to overlap between phases. As usual, the phase with lowest *total* energy is plotted in the overlapping area. That means that we take both the mean field ground state as well as the *zero point* energies into account. The latter one can only be obtained by means of the flavor waves.

This is particularly important in the case of the DS and PL phases, which are degenerate at the mean field level. However, the two energies split once quantum fluctuations are added. For small $|K_2|$, it is the PL regime that is energetically favorable, while this effect reverses for large values of $|K_2|$. However, the stability range of the PL state remains larger than that of the DS phase in all cases. In the case where both stability ranges almost completely overlap $[K_2 = -0.3, \text{ depicted in Figure 5.1 f}]$, the difference in energies is also the smallest one of all cases. Some of these energy differences between the DS and PL phases can be seen in Figure 5.3.



Figure 5.3: Zero point energy evolution with K_1 for the plaquette (solid line) and double stripe (dashed line) phases and across a cut through constant $J_3 = 1.0$. Panel a) shows the results for $K_2 = 0$, and panel b) those for $K_2 = K_1$.

This plot also showcases the first order nature of the transition between the phases.

Finally, although relatively trivial, it is worth mentioning that in both the mean field and flavor wave approaches, the area where ferroquadrupolar order (FQ) exists increases with K_1 and K_2 . This is evident from the Hamiltonian model itself and fluctuations don't seems to have a significant effect on altering this.

5.2.1 Incommensurate phases

The unshaded regions of the diagram indicate our inability to find stability of any of the phases in those areas. As mentioned earlier, we can only use the flavor wave method over ground states already predicted. From this, we can conclude that the phases existing in those regions couldn't be predicted by our earlier, variational method.



Figure 5.4: a) y-component of the instability vectors at the FQ phase boundary as a function of J_3 . This is done for the case of $K_2 = K_1$. b) Phase diagram for $K_2 = K_1$ [shown also in Fig. 5.1 h)], indicating the regions of the phase boundary for which each instability vector in obtained.

One possibility is that of *incommensurate* phases. These cannot exist in the finite 4x4 Fe-site cluster that was used in the variational method. In addition to this, there is also experimental evidence for the possibility of their existence. In particular, according to INS experiments [70, 71, 72], the high temperature spin structure factor in Fe(Te_{1-x}Se_x) may be incommensurate. We can study the the Q vectors at which the instability occurs in the flavor wave dispersion (the value of the dispersion at vector Q becomes imaginary beyond the boundary). The results are shown in Figure 5.4.

Throughout the leftmost side of the diagram, the instability vector can be written as: $\mathbf{Q} = (\pi, \delta)$, where δ has two contrasting behaviors. For very small J_3 , where the FQ phase border touches the CAFM region, $\delta = 0$, as expected due to the lack of an intermediate between the two. However, past a threshold value, δ increases smoothly towards the value of $\delta = \pi/2$, although never quite reaches it. This is also in agreement with the proximity of the FQ region boundary to the SD phase [with ordering vectors $\boldsymbol{Q} = (\pi, \frac{\pi}{2})$ and $\boldsymbol{Q} = (\frac{\pi}{2}, \pi)$] for those values of J_3 .

On the rightmost ride of the diagram, the instability vector takes a completely different for, which we can write as $\mathbf{Q} = (\frac{\pi}{2} + \delta, \frac{\pi}{2} + \delta)$. Yet again, this points towards an intermediate incommensurate phase with ordering vectors close to those of the DS, which is precisely the phase boundary that follows closely that of the FQ state for these J_3 values. These incommensurable wave vectors will also be observable in the study of the dynamical spin structure factors discussed in Section 5.3 below.

5.3 Dynamical structure factors

If the ground state of FeSe is indeed that of ferroquadrupolar order [67], then the increase in value of the third neighbor coupling J_3 would mimic the doping of the aforementioned compound with Te. We now proceed to study the evolution of the dynamical spin structure factor along a cut of value $K_1 = K_2 = -1$, and the transition from FQ to double stripe order.

The FQ ground state, while maintaining the time-reversal symmetry, breaks the spin-rotational symmetry of the Hamiltonian. Because of this, the expected Goldstone modes at Q = 0 have vanishing intensity [73, 74] in the static limit, which is consistent with the absence of Bragg peaks in FeSe under ambient pressure [75, 76]. This feature is replicated by our model, as can be seen in panels a), b), and c) of Fig. 5.3, which lie within the FQ region.



Figure 5.5: Dynamical spin structure factors along a cut through $K_1 = K_2 = -1$ in the phase diagram. Panels a) to c) correspond to points within the FQ region for increasing J_3 and panel d) showcases the results within the DS phase.

For small values of J_3 , the structure factor showcases two very close minima. One of them, at vector $\mathbf{Q} = (\pi, 0)$, clearly hints at the proximity to the CAFM zone boundary. The appearance of the other minima is reminiscent of our findings in Section 5.2.1. In fact, if we write its position in an analogous form $\mathbf{Q} = (\pi, \delta)$, we find that δ indeed increases as we move to the right along the cut never to fully reach $\delta = \frac{\pi}{2}$, as predicted. By the second panel b), it has already become more pronounced than the minumum at $\mathbf{Q} = (\pi, 0)$. It is here that we also observe the appearance of yet another minimum at position $\mathbf{Q} = \left(\frac{\pi}{2} + \delta, \frac{\pi}{2} + \delta\right)$, with small but finite δ . This is again in agreement with our previous study of possible incommensurate phases. This minimum goes on to become the absolute minimum as we reach proximity to the boundary of the DS phase. Once we cross the phase boundary into the DS region, the minimum becomes the true Goldstone mode at $\mathbf{Q} = \left(\frac{\pi}{2}, \frac{\pi}{2}\right)$.

Chapter 6

Conclusions

In this first part of the thesis, we have studied two different families of iron-based superconductors, namely the iron pnictides and the iron chalcogenides, using different variations of a frustrated spin Hamiltonian and different bosonic spin representation techniques.

We began by putting the various spin representations to the test by comparing their spin excitation spectrum. Varying the value of spin allowed us to pinpoint the effect of fluctuations, and thus to identify the approaches that offer a more accurate description. These methods, namely, the full decoupling of the non-linear spin waves, the Dyson-Maleev bosons (also fully decoupled), and the generalized spin waves also proved superior at capturing the effect of biquadratic terms. With this knowledge at hand, we restricted ourselves to using only two out of the three aforementioned techniques in the rest of our study. While both the of the methods that need a full decoupling also produce systems of self-consistent equations to be solved, the application of the generalized spin waves to a problem is immediate. For this reason, we keep it our main method of choice whenever possible (within magnetically ordered regions) and resort to the Dyson-Maleev approach only when working in a region with no broken symmetries.

By using the fully decoupled version of the later, we were able to improve upon earlier work [65], and in doing so, to obtain a qualitative agreement with both spin wave dispersions and dynamical structure factors measured by INS experiments on iron pnictide compounds. Even though we found a small region of pure Ising-nematic order, we concluded that its existence requires a careful fine-tuning of both the biquadratic coupling K and the interplanar exchange J_c . This leads us to believe a single orbital orbital model might be inadequate to describe the complete system, and further work that involves the multi-orbital physics of the problem is needed.

When it comes to the model proposed for the iron chalcogenides, we found a phase diagram much richer than that of the pnictides, with many different magnetic orderings (See Fig. 5.1), including a regime with ferroquadrupolar order. Working within the areas with a broken symmetry, we used the generalized spin wave approach to study the shifts in the phase boundaries beyond those predicted by a simply variational approach. This resulted in new phase diagrams with the possibility of incommensurate orderings. We found that the Te doping responsible for the transition from the non-magnetic FeSe to a $Fe(Te_{1-x}Se_x)$ showcasing long range order in the form of a spin double stripe space can be roughly modeled by increasing the value of the coupling between third nearest neighbors.

Part II

Superconductivity in Multi-Impurity Kondo Model

Chapter 7

Introduction to the Kondo Effect

7.1 The resistance minimum and the Kondo solution

The main source of resistivity in metals is the scattering off of phonons in the lattice. As temperature decreases, this scattering dies off and, in principle, would produce a diverging conductivity. In real materials, however, conduction electrons scatter off of defects and impurities leaving a saturated, residual resistivity even at low temperatures.

But observations starting on 1934 [77] and forward disagreed with this model. Instead of observing that constant resistance, their experiments found that, at low enough temperatures, the resistivity started increasing again, creating a characteristic minimum. These experiments also noticed the strong dependence of this minimum on the concentration of impurities, pointing to those as the origin of this phenomenon. To explain this, Kondo [78] studied the effect that a single magnetic impurity would have within a non-magnetic metal. This interaction can be described by means of the following Hamiltonian [79]:

$$H = \sum_{\boldsymbol{k},\alpha} \epsilon_{\boldsymbol{k}} c^{\dagger}_{\boldsymbol{k}\alpha} c_{\boldsymbol{k}\alpha} + J_K \sum_{\boldsymbol{k}\boldsymbol{k}'\alpha\beta} \boldsymbol{S} \cdot \left(c^{\dagger}_{\boldsymbol{k}\alpha} \frac{\boldsymbol{\sigma}_{\alpha\beta}}{2} c_{\boldsymbol{k}'\beta} \right).$$
(7.1)

The first term is the usual kinetic term for the conduction electrons and it's the second term that describes the Kondo interaction between the impurity spin and the conduction electrons, at a strength given by the coupling J_K , . Using perturbation theory, Kondo found an expression for the resistivity that indeed depended on the magnetic impurity density, entering in the form of constants a and b:

$$\rho(T) = \rho_{\text{host}}(T) + a\rho_0 J_K^2 + b\rho_0^2 J_K^3 \log\left(\frac{D}{T}\right).$$
(7.2)

Here, ρ_{host} refers to the resistivity of the ideal metal, and ρ_0 and D correspond to the density of states at the Fermi level and the bandwidth, respectively. At low temperatures, the logarithmic term dominates and eventually produces the observed increase in resistivity. Physically, this can be modeled as an exchange between the spin of the impurity and those of the conduction electrons. The resistivity in the material is then raised by these correlations when they become strong enough.
7.2 The Kondo problem and the Anderson solution

As a consequence of the pertubative nature of its solution, Kondo's approach results in a diverging resistivity at low temperatures. The approximate temperature below which perturbation theory fails is known as the *Kondo temperature* (T_K) and is approximately given by:

$$T_K \sim D e^{-\frac{1}{\rho_0 J_K}} \tag{7.3}$$

Using the so-called "poor man's scaling", Anderson [80] fixed this divergence by predicting the existence of a *Kondo singlet*, composed of the impurity and one of the conduction electron's spins, at low enough temperatures. Because the spin of the impurity is now compensated, the impurity itself behaves in a non-magnetic way in this regime. Later on, Wilson [81] confirmed this result by using the numerical renormalization group.

7.3 Heavy fermion materials

Some of the best candidates to exhibit the behavior predicted by the Kondo model are the so-called heavy fermion materials. Heavy fermion compounds are materials composed of rare-earth or actinide elements, thus containing f-electrons. They behave like a metal at low temperatures, but the values for the physical quantities are strongly renormalized. These properties were first observed by Andres *et al.* [82] in 1975 on a $CeAl_3$ compound. The 4-f and 5-f electrons in these materials are strongly localized, so that a periodic Kondo Hamiltonian offers a possible way to describe their low temperature properties.

In addition to the Kondo effect characteristic of these type of materials, some heavy fermion compounds also exhibit superconductivity. In systems such as those in the CeMIn₅ series (M=Co,Rh,Ir), this superconductivity has been observed in the vicinity of a magnetic quantum critical point[83].

7.4 The multichannel Kondo model

Initially introduced by Nozières and Blandin [84], the single impurity multichannel Kondo model involves K identical bands (or *channels*) of conduction electrons of spin size S interacting antiferromagnetically with a local moment of the same spin value through the Kondo mechanism:

$$H = \sum_{\boldsymbol{k},\nu,\alpha} \epsilon_{\boldsymbol{k}} c^{\dagger}_{\boldsymbol{k}\nu\alpha} c_{\boldsymbol{k}\nu\alpha} + J_K \sum_{\nu} \boldsymbol{s}_{\nu} \cdot \boldsymbol{S}$$
(7.4)

The initial study using a large-N approach was carried out by Cox and Ruckenstein [85], who confirmed the non-Fermi liquid behavior of the system. It was Parcollet *et al.* [86] who introduced a bosonic representation of the local moment, with Rech *et al.* [87] using this formalism to characterize the three possible regimes we describe below. A crucial point to understand this classification is that we need N = 2S + 1 particles of spin S to form a *singlet*. That is, our local moment needs an extra P = 2S electrons for a singlet to exist.

7.4.1 Underscreened

When the number of channels K is smaller than P, the number of bands can't provide enough electrons to fully screen the impurity by forming a Kondo singlet. Thus, a portion of the impurity remains unscreened and the system takes on the behavior of a single local moment.

7.4.2 Overscreened

On the other hand, when there are even *more* channels than the strictly necessary to form a singlet (K > P), the impurity is *overscreened* and the system exhibits non-Fermi liquid behavior.

7.4.3 Exactly Screened

When K = P, there are *just enough* conduction electron channel to exactly compensate the spin of the impurity. It is in this case that the Kondo singlet it formed, consisting of the impurity spin and those of the electrons that each channel supplies. The rest of the conduction electrons then scatter off of the singlet and the system behaves like a Fermi liquid.

Chapter 8

The single impurity system

8.1 Model and solutions

We begin by considering the single-impurity multichannel Kondo Hamitonian[86, 87] with K channels of conduction electrons and a local moment of spin S located at the origin, and expressed in terms of bosonic operators. The number of spin flavors is then fixed to: N = 2S + 1.

$$\mathcal{H} = \sum_{\boldsymbol{k},\nu,\alpha} \epsilon_{\boldsymbol{k}} c^{\dagger}_{\boldsymbol{k}\nu\alpha} c_{\boldsymbol{k}\nu\alpha} + \frac{J_K}{N} \sum_{\nu,\alpha,\beta} \psi^{\dagger}_{\nu\alpha} \psi_{\nu\beta} b^{\dagger}_{\beta} b_{\alpha} - \lambda (n_b - 2S)$$
(8.1)

The first term corresponds to the kinetic energy (ϵ_k) of the conduction electrons, which carry both channel (ν) and spin (α) indices. From now on, K will denote the number of channels and N the number of spin flavors, which must be fixed to: N = 2S + 1. Because K is of order N and we are working on a large-N framework, the finite quantity $\kappa \equiv \frac{K}{N}$ will be of better use in what follows. Since we will work in the *exactly* screened regime only, we also have that: $\kappa = n_b \equiv \frac{2S}{N}$. So in a sense, κ is a measure of both the channel number *and* the spin size.

The second term in the Hamiltonian encompasses the effects of the Kondo interaction. We use the Schwinger boson representation to write local moment spin $S_{\alpha\beta} = b^{\dagger}_{\alpha} b_{\beta} - \delta_{\alpha\beta}/N$

The third term is added in order to enforce the physical constraint into the Hilbert space.

In order to decouple the interaction, we use the standard Hubbard-Stratonovich technique by introducing the new *holon* field χ_{ν} . The holons are of fermionic nature but only carry a channel index. This transforms the Kondo term above into the following expression:

$$H_K = \sum_{\nu,\alpha} \frac{1}{\sqrt{N}} [(\psi_{\nu\alpha}^{\dagger} b_{\alpha}) \chi_{\nu}^{\dagger} + \text{H.c.}] + \sum_{\nu} \frac{\chi_{\nu}^{\dagger} \chi_{\nu}}{J_K}$$
(8.2)

Provided that the spinless fermionic fields are given by: $\chi^{\dagger}_{\nu} = \frac{J_K}{\sqrt{N}} \sum_{\beta} \psi_{\nu\beta} b^{\dagger}_{\beta}$. The propagators for each particle are given by the following expressions:

$$G_b(i\nu_n) = \frac{1}{i\nu_n + \lambda - \Sigma_b(i\nu_n)}$$
(8.3)

$$G_{\chi}(i\omega_n) = -\frac{J_K}{1 + J_K \Sigma_{\chi}(i\omega_n)}$$
(8.4)

$$G_c(i\omega_n) = \frac{G_c^0(i\omega_n)}{1 + G_c^0 \Sigma_c(i\omega_n)}$$
(8.5)

Where the self-energies satisfy the following relations [87]:

$$\Sigma_b(\tau) = -kG_c(\tau)G_\chi(\tau) \tag{8.6}$$

$$\Sigma_{\chi}(\tau) = G_c(-\tau)G_b(\tau) \tag{8.7}$$

In practice, we obtain the expression in the real τ -space by performing Fourier transforms on the above expressions written in terms of imaginary Matsubara frequencies. As far as the conduction electron self-energy goes, because there is no sum over K or S involved on its calculation, it remains of order $\mathcal{O}(\frac{1}{N})$. In the large-N limit then, we can neglect it and we will work with the *bare* electron propagator instead:

$$G_c(i\omega_n) \approx G_c^0(i\omega_n) = \sum_{\boldsymbol{k}} \frac{1}{i\omega_n - \epsilon_{\boldsymbol{k}}}$$
(8.8)



Figure 8.1: a) Evolution of the magnetic susceptibility in time for several values of κ . At low temperatures, it acquires the constant value, characteristic of a Fermi liquid regime. For high temperatures, however, a typical local moment susceptibility appears. In panel b), all $T\chi$ ultimately reach a constant value, thus proving the 1/T dependence.

The final step to solve the problem is to obtain the value of the chemical potential λ . We do so by imposing the following constraint [86]:

$$\kappa = n_b = \sum_n G_b(i\omega_n) \tag{8.9}$$

8.2 Susceptibilities

After obtaining the value of the chemical potential λ , we can now plot the magnetic susceptibility (see Fig. 8.1). At low temperatures we observe a constant behavior in the susceptibility, consistent with the Pauli susceptibility characteristic of



Figure 8.2: Evolution of the a) magnetic susceptibility as $T \to 0$ and b) the temperature at which the susceptibility reaches its maximum value as κ increases.

a Fermi liquid, hinting at the expected Kondo screening.

Now, when it comes to the large temperature region, we easily identify Curie-Weiss Law ruling the behavior of the tails of the curves. To check that the tail does indeed have a $\sim 1/T$ dependence, we plot $T\chi$ as well on the right panel of Figure 8.1.

It is also illuminating to study the evolution of the values of the susceptibility at certain key points, namely the $T \to 0$ and large-T limits as well as its maximum value χ_{max} . As we can see in Figure 8.2, all of these quantities increase with $\kappa = n_b$, which a measure of the spin size. χ_0 , the susceptibility in the low temperature limit $[\chi(T \to 0)]$ is nothing but the Pauli susceptibility of a Fermi liquid whereas $C = T\chi_{\infty}$ is the so-called Curie constant. By tracking the temperature at which each maxima happens [panel b) in Fig. 8.2] we can trace a semblance of a phase diagram since the maxima approximately signals the change in behavior of the susceptibility itself.



Figure 8.3: a) Evolution of the magnetic susceptibility in time for several values of D. b) J_K dependence on the parameter D, which measures half of the bandwidth.

The range of temperatures for which the Kondo regime exists then, slowly decreases as the the value of κ increases.

We can also take a quick look at the effect that D, half of the bandwidth has on the susceptibilities. Plotting the temperature evolution of χ for various values of D[see panel a) in Fig. 8.3] reveals that its influence is minimal. At high temperatures, this effect is to be expected since the susceptibility follows the usual Curie behavior which depends only on the local moments. Thus, no parameter characterizing the conduction electrons (such as, in this case, the bandwidth D) should affect its value. At low temperatures, on the other hand, the susceptibility only depends on the Kondo temperature. In our work, this is set to T_K and serves as the unit in which all other quantities are measured (that is, we set $T_K = 1$ throughout the remainder of this work). Because of this, the T_K dependence is absorbed in the definition of temperature. Finally, by using the approximate formula for the value of the Kondo temperature:

$$T_K = D e^{-\frac{1}{\rho_0 J_K}},\tag{8.10}$$

we can obtain the value of the Kondo coupling J_K as a function of D:

$$J_{K} = \frac{1}{\rho_{0} \log(D)} = \frac{2D}{\log(D)}$$
(8.11)

As seen in panel b) of Figure 8.3, J_K increases roughly linearly with D, at least for reasonable values of the bandwidth. Because this is just an approximate formula, we may vary the prefactor for J_K without affecting results qualitatively.

Chapter 9

The two-impurity system

9.1 The Heisenberg term and the new solutions

For two impurities, the Hamiltonian has independent Kondo terms for each local moment, along with a Heisenberg interaction between the two, of coupling strength J_H . It takes the following form:

$$\mathcal{H} = \sum_{\boldsymbol{k},\nu,\alpha} \epsilon_{\boldsymbol{k}} c^{\dagger}_{\boldsymbol{k}\nu\alpha} c_{\boldsymbol{k}\nu\alpha} + \frac{J_K}{N} \sum_{\nu,\alpha,\beta} (\psi^{\dagger}_{1\nu\alpha} \psi_{1\nu\beta} b^{\dagger}_{1\beta} b_{1\alpha} + \psi^{\dagger}_{2\nu\alpha} \psi_{2\nu\beta} b^{\dagger}_{2\beta} b_{2\alpha}) - \lambda (n_{b1} + n_{b2} - 4S) - \frac{J_H}{N} B^{\dagger}_{12} B_{12}$$

$$(9.1)$$

Where we chose the same Lagrange multiplier for each impurity: $\lambda_1 = \lambda_2 \equiv \lambda$ due to symmetry under the exchange $1 \leftrightarrow 2$ and the boson pair operators are defined by: $B_{12} = \sum_{\alpha} \operatorname{sgn}(\alpha) b_{1\alpha} b_{2\bar{\alpha}}$. We decouple the last interaction term using the mean-field parameter $\Delta_b = -J_H \frac{\langle B_{12} \rangle}{N}$:



Figure 9.1: Feynman diagrams used to derive the Gorkov equations [88, 89, 90]. Here, we have chosen Δ to be a real quantity.

$$-\frac{J_H}{N}B_{12}^{\dagger}B_{12} \to \Delta_b^* B_{12} + B_{12}^{\dagger}\Delta_b + \frac{N|\Delta_b|^2}{J_H}$$
(9.2)

Using the Gorkov [88, 89, 90] method, we can obtain the new bosonic Green's functions as follows. The Feynman diagrams in Figure 9.1 help us write the following system of new Dyson's equations, which now include the order parameter Δ_b , for the propagators:

$$G_b(i\nu_n) = G_b^0(i\nu_n) + G_b^0(i\nu_n)\Sigma_n(i\nu_n)G_b(i\nu_n) + G_b^0(i\nu_n)\Delta G_b(i\nu_n)$$
(9.3)

$$F_b(i\nu_n) = G_b^0(i\nu_n)\Sigma_n(i\nu_n)F_b(i\nu_n) + G_b^0(i\nu_n)\Delta G_b(-i\nu_n)$$
(9.4)

Solving the system results in the new expressions for the normal and anomalous propagators:

$$G_b(i\nu_n) = \frac{-i\nu_n + \lambda - \Sigma_b(-i\nu_n)}{[i\nu_n + \lambda - \Sigma_b(i\nu_n)][-i\nu_n + \lambda - \Sigma_b(-i\nu_n)] - |\Delta_b|^2}$$
(9.5)

$$F_b(i\nu_n) = \frac{\Delta_b}{[i\nu_n + \lambda - \Sigma_b(i\nu_n)][-i\nu_n + \lambda - \Sigma_b(-i\nu_n)] - |\Delta|^2}$$
(9.6)

Finally, we impose self-consistency on the new order parameter Δ_b by requiring:

$$\Delta_b = -\frac{J_H}{N} \langle B_{12} \rangle = J_H \sum_n F_b(i\nu_n) =$$

$$= 2\Delta_b \sum_n \frac{1}{[i\nu_n + \lambda - \Sigma_b(i\nu_n)][-i\nu_n + \lambda - \Sigma_b(-i\nu_n)] - |\Delta_b|^2}$$
(9.7)

9.2 Results

As far as the two-impurity susceptibility goes, yet again, for large enough temperatures, the system showcases the same ~ 1/T behavior [see panel b) in Fig. 9.2] as in the previous one-impurity case. Unlike in the case of one impurity though, the destruction of the Kondo effect for large enough temperatures is not enough for this behavior to set, unless for very small J_H values. However, for large enough J_H , the $\Delta_b \neq 0$ state dominates over the Kondo regime at low temperatures. In this case, it is necessary for the magnetic correlations between the local moment spins to disappear ($\Delta_b = 0$) in order for the Curie behavior to be observable.

This is made clear when comparing the two panels in Figure 9.2. Indeed, the temperature at which Δ_b vanishes [panel a) in Fig. 9.2] coincides exactly with the



Figure 9.2: Evolution of Δ_b [panel a)] and χ [panel b)] with the renormalized temperature T/J_H for various values of the J_H coupling between local moments spins and for the case of $\kappa = 0.3$. For small values of $1/J_H$ (strong coupling between the two impurities), we note that the temperature at which the order parameter vanishes coincides with the point where the behavior of χ abruptly changes. This abrupt behavior, however, smooths out as the value of $1/J_H$ increases. Indeed, as the coupling between the two local moments weakens, the impurities start to decouple and the susceptibility starts to showcase a smooth maximum, just like the ones we observed in Fig.8.1, for the case of a single impurity.

point where we observe a sudden change in the behavior of χ [panel b)] for the smaller values of $1/J_H$. However, as the strength of the J_H coupling decreases (that is, for larger values of $1/J_H$), the coupling between the two impurities weakens and a smooth maximum, not unlike that for the one impurity susceptibilities, begins to appear.

Computing the evolution of Δ_b for sufficient values of J_H , we can obtain a diagram (see Fig. 9.3) that gives a more general description of the ranges of existence of Δ_b . Again, we observe the agreement between the results of our method and those by Rech *et al.* [87]. As expected, spin size stabilizes the regime with magnetic corre-



Figure 9.3: Diagram showcasing the range in temperature and inverse J_H coupling over which Δ_b is finite for the indicated values of κ . As mentioned before, here and throughout the rest of the thesis, the value of the Kondo temperature is set to $T_K = 1$. The colored areas indicate the regions where $\Delta_b \neq 0$.

lations between the localized spins, so that Δ_b remains finite for higher temperatures as $\kappa = n_b$ increases. Surprisingly, when it comes to the boundary dependence on J_H , larger spin sizes actually *shrink* the range of J_H values within which the correlations survive.

The diagram above is, however, not the full story. While the situation is trivial in regions where $\Delta_b = 0$ — the two impurities decouple completely so we effectively have two single impurity systems such as the one described in the previous chapter. Thus, in the $\Delta_b = 0$ regions, the system will exhibit a Kondo behavior, providede that the value of the temperature is low enough for a Kondo singlet to form.



Figure 9.4: Color coded phase shift evolution with both the value of J_H and the renormalized temperature T/J_H for the case of $\kappa = 0.3$.

Once Δ_b acquires a finite value, however, things are not so simple, since the Kondo regime can, in principle, coexist with correlated impurities. One way to elucidate to what extent both regimes coexist is to compute the holon phase shifts δ_{χ} (see Fig. 9.4). A phase shift of value $\delta_{\chi} = \pi$ will indicate the presence of the Kondo effect. We can calculate this phase shift δ_{χ} by using the following formula:

$$\delta_{\chi} = \operatorname{Im} \ln \left[1 + J_K \Sigma_{\chi} (0 - i\delta) \right] = \operatorname{Im} \ln \left[-J_K^2 G_{\chi}^{-1} (0 - i\delta) \right]$$
(9.8)

However, the argument in the retarded Green's function used in the expression above is given as a *real* frequency in contrast to the imaginary Matsubara frequencies we have been previously using. Thus, in order to compute this quantity in the real



Figure 9.5: Holon phase shift evolution with $1/J_H$ for the following temperatures (in units of T_K): a) 0.15, b) 0.25, c) 0.4, and d) 0.6. A least-mean square fit to a function of the form 0.5[tanh (a + bx) + 1] is plotted as a red line along the data points (blue dots). As temperature increases, the transition to the Kondo regime becomes less abrupt.

space, we use the method of the Padé approximants (see Appendix D) applying it to either the self-energy or the full Green's function. As expected, as $1/J_H$ increases (that is J_H decreases, moving from left to right of the plot), that phase shift raises from $\delta_{\chi} = 0$ to $\delta_{\chi} = \pi$, confirming the Kondo nature of the state at small J_H s. By choosing a constant temperature cuts (see Figure 9.5), one can observe that the change between the phase shift values becomes more sudden as temperature decreases. This is consistent with the existence of a quantum phase transition [91, 92, 93].

We can now draw our attention to the four cuts included at different values of the inverse coupling $(1/J_H = 0.25, 1/J_H = 0.55, 1/J_H = 0.85, \text{ and } 1/J_H = 1.15)$. We choose these specific values in order to compare these results with those on Fig. 9.2 above. At a temperature of $T/J_H = 0.1$ (also indicated in the diagram with a green dashed line), $\Delta_b \neq 0$ for all cases. If the two regimes (Kondo and coupled impurities) were completely incompatible, one would imagine the phase shift to strictly vanish for all these cases. However, the actual results are very different. Even at the same temperature, the value of the phase shift varies widely. For example, while $\delta_{\chi} = 0$ for the smallest $1/J_H$ and $\delta_{\chi} = \pi$ for the largest, the value of the phase shift steadily increases between those two limit value with no abrupt transition. That is, the stronger the Heisenberg coupling between impurities there is, the smaller the phase-shift we obtain. This indicated that indeed, both regimes can coexist and, as expected, the weaker the coupling between impurities is, the easier it is for the Kondo regime to penetrate beyond the $\Delta_b = 0$ boundary and into the region with a finite Δ_b .

However, this last calculation also highlights one of the drawbacks of choosing to work in the imaginary space for our method, in contrast to the real frequency approach of Rech *et al.* [87]. Unlike all the previous quantities we have calculated, the results of the Padé approximants are *extremely* sensitive to the Green's Function's exact values. Because of this, a number of frequencies of the order of *at least* $N \sim$ 5×10^4 is necessary to begin obtaining somewhat reliable results. As such, our method is better suited to calculate quantities that can be written in terms of Matsubara frequencies only.

Chapter 10

Adding superconductivity to the system

As we pointed out in Section 7.3, unconventional superconductivity arises in the 115 family of the Cerium-based heavy fermion materials when in the proximity of a magnetic quantum critical point. Throughout the two previous chapters, we have already introduced the two main competing mechanisms in these systems. On the one hand, the Kondo coupling is the essence of the heavy fermion behavior, while the RKKY spin-spin coupling governs the magnetism. Using the framework we have already set up, we now proceed to study the possibility of a superconducting regime.

10.1 The holon coupling term

In an analogous way in which we added magnetic correlations into the system, we now introduce a term which couples the holons in the system with an attractive strength of value g. We propose that this term may emerge from terms arising in higher-order expansion of the Schrieffer-Wolf projection from the Anderson model. Particularly, just as the second order expansion accounts for the RKKY coupling, we expect a further, *fourth* order expansion to account for the holon-holon coupling. We decouple it using the same method as before:

$$-\frac{g}{N}H_{12}^{\dagger}H_{12} \to \Delta_{\chi}^{*}H_{12} + H_{12}^{\dagger}\Delta_{\chi} + \frac{N|\Delta_{\chi}|^{2}}{g}$$
(10.1)

Where now $H_{12} = \chi_1 \chi_2$ and the order parameter is given by $\Delta_{\chi} = g \frac{\langle H_{12} \rangle}{N}$. Using an analogous derivation to that in the previous chapter, we arrive to the new expressions for the holon propagators:

$$G_{\chi}(i\omega_n) = -\frac{J_K \left[1 + J_K \Sigma_{\chi}(-i\omega_n)\right]}{\left[1 + J_K \Sigma_{\chi}(i\omega_n)\right] \left[1 + \Sigma_{\chi}(-i\omega_n)\right] + \left|\frac{\Delta_{\chi}}{J_K}\right|^2}$$
(10.2)

$$F_{\chi}(i\nu_n) = \frac{\Delta_{\chi}}{[1 + J_K \Sigma_{\chi}(i\omega_n)][1 + \Sigma_{\chi}(-i\omega_n)] + |\frac{\Delta_{\chi}}{J_K}|^2}$$
(10.3)

Note the different units for the new order parameter Δ_{χ} , resulting from the rather unusual choice of units for the holon propagators. The new self-consistent equation for Δ_{χ} reads as follows:



Figure 10.1: Feynman diagrams used to derive the conduction electron anomalous propagator.

$$\Delta_{\chi}(T) = g\Delta_{\chi}(T)\sum_{n} \frac{1}{[1 + J_K \Sigma_{\chi}(i\omega_n)][1 + \Sigma_{\chi}(-i\omega_n)] + |\frac{\Delta_{\chi}(i\omega_n)}{J_K}|^2}$$
(10.4)

Unlike in the case of the previously presented order parameter Δ_b , one must be careful when performing the sums in this case. In order for those not to diverge, we must introduce a cutoff frequency ω_D (somewhat analogous to the Debye frequency, hence our notation). In particular, we renormalize the order parameter used in the summations $[\Delta_{\chi}(i\omega_n) = \Delta_{\chi}(T)\sigma(i\omega_n)]$ using the following regularizer function:

$$\sigma(i\omega_n) = \frac{\omega_D}{\sqrt{\omega_D^2 + \omega_n^2}} \tag{10.5}$$

Finally, the superconducting order parameter Δ_{SC} will be calculated using the following formula [see Figure 10.1, where we take only the leading order on $G_c(i\omega_n)$]:

$$\Delta_{SC} = \sum_{n} F_c(i\omega_n) = \frac{G_c^0(i\omega_n)\Omega(i\omega_n)G_c^0(-i\omega_n)}{1 - G_c^0(i\omega_n)\Sigma_c(i\omega_n)}$$
(10.6)

Here, we have omitted the 1/N prefactor in order to properly compare Δ_{SC}



Figure 10.2: Evolution of Δ_b , Δ_{χ} , Δ_{SC} with the renormalized temperature T/g for a) $1/J_H = 0.05$ and b) $1/J_H = 0.15$. All quantities are normalized with respect to their maximum values so they can fit in the same plot.

to the two previously presented order parameters. This is because, in the large-N formalism, Δ_{SC} is of an order higher in the 1/N expansion parameter. $\Omega_c(i\omega)$ is the anomalous self-energy, which results of the convolution between the boson and holon anomalous propagators in τ -space.

10.2 Results

We now begin by plotting the evolution of all order parameters with the renormalized temperature T/g. These quantities are Δ_b , Δ_{χ} , and Δ_{SC} , which measure the correlations between bosons, holons, and conduction electrons, respectively. We normalize all results to the maximum value of each parameter so they can be shown in the same plot (see Fig. 10.2). For smaller values of $1/J_H = 0.05$ (larger values of



Figure 10.3: Phase diagrams for a) $1/J_H = 0.05$ and b) $1/J_H = 0.15$.

 J_H , left panel in Fig. 10.2), the correlations between bosons survive beyond those between holons, so that it is the latter that play a decisive part in the existence range of superconductivity. On the other hand, for larger values of $J_H = 0.15$ (smaller J_H , right panel in Fig. 10.2), the boson-boson correlations vanish at smaller temperatures and so does superconductivity with them.

Just like before, gathering data for sufficient values of g results in the phase diagrams shown in Figure 10.3. In both cases, as temperature increases, the region of stability in g of the magnetically correlated phase shrinks. As for the correlations between holons, the range is smaller all over for the case of a smaller J_H [1/ J_H = 0.15 on panel b) of Fig. 10.3], as expected. Intriguingly, a minimum value of g is needed for the holon correlations to exist at all. This is remarkably different from the case of BCS superconductivity, where an infinitesimal value of an attractive interaction is enough to yield superconductivity for electrons in a Fermi surface. This effect happens due to the presence of the so-called Cooper logarithm. However, with our introduction of a cutoff frequency ω_D , the famous logarithm doesn't arise, leaving us with a threshold value in the coupling in order for superconductivity to emerge.

As far as Δ_{χ} goes, we focus on two main features. First, there is a minimum value of g below which the phase with correlations between holons doesn't exist. Second, its behavior varies little for different values of J_H . Qualitatively, once a phase boundary appears for a finite temperature, the increase of the boundary line in temperature increases linearly with q until it reaches a *plateau*. This is indicative of the phase boundary temperature being of order g ($T_{\chi}/g = \text{constant}$) at large g, when the effect of the bosonic correlations becomes negligible and the holons are effectively free. Depending on the value of the J_H coupling, this transition between a varying and a constant phase boundary can happen before (larger J_H) or after (smaller J_H) Δ_b vanishes. In the former case, the boundary of the superconducting regime is allowed to reach its maximum possible value set by the existence of a finite Δ_{χ} before decreasing again. In the latter case, however, the disappearance of the correlations between local moment spins suppresses the superconductivity before it has the chance to reach its potential maximum temperature. Because of this, we conclude that a strong correlation between the localized spins plays a crucial role in maximizing the temperature up to which superconductivity can survive.

Chapter 11

Conclusions

In this part, we have studied the multichannel Kondo model in the exactly screened regime and for the cases of one and two impurities. First and foremost, this allowed us to assess the validity of our approach to the problem to that of previous authors [87]. In particular, Rech *et al.* solved the system using self-consistent integrals of propagators in their real frequency representation while we used an iterative method consisting of performing sums over the Matsubara frequencies in the imaginary axis. In the one impurity case, studying the local moment susceptibility allowed us to estimate the region where the Kondo regime exists, as well as its variation with spin size.

The two impurity system introduces the order parameter that measures the correlations between the two impurity spins. In this case, the existence of a Kondo phase depends not only on temperature but on the value of the J_H coupling between the local moment spins. When this J_H reaches a threshold value, the two impurity



Figure 11.1: Evolution of the phase diagram for $\text{CeRh}_{1x}\text{Co}_x\text{In}_5$ with Co doping [94]. As x increases, the magnetism get suppressed and the system becomes superconducting.

spins align with opposing direction and the Kondo regime disappears. A change in the holon phase shift also confirms the suppression of the Kondo effect.

Finally, adding an attractive coupling g between the holons gives rise to superconductivity in the system. However, there a few requirements for this phenomenon. On the one hand, both the correlations between local moment spins (measured by Δ_b) as well as those between holons (measured by Δ_{χ}) must *both* be present. On the other hand, superconductivity also disappears when the holon-holon coupling moves below a minimum value. This rather unusual feature is due to the lack of a Cooper logarithm in the case of the holons. Because of this, we conclude that relatively careful fine tuning of both the J_H and g couplings is needed for superconductivity to exist within a reasonable range in this system.

It is a possibility then, to use the J_H/g ratio as a measure of doping in real materials. Indeed, in the Ce-115 family of compounds, the interplay between magnetism and superconductivity is dependent on the composition. As an example, Co doping in the CeRh_{1x}Co_xIn₅ system suppresses magnetic order and drives the system into a superconducting state (see Fig. 11.1). In this case, due to its magnetism, we would expect the J_H/g to be larger in CeRhIn₅. This suggests that the Co doping is actually responsible for fine-tuning its value so that the compound eventually superconducts.

Part III

Summary and outlook

In this work we have studied a few examples of phenomena observed in strongly correlated electron materials. I devoted the first part to iron-based superconductors and studied their phase diagrams by means of a localized spin model of frustrated interactions. Particularly, we focused on describing magnetism in both the iron pnictides and the iron chalcogenides. Among the similarities between these two types of materials in the valence state of the Fe ions (Fe²⁺ in this case). This means that both families of compounds can be effectively described by a lattice of S = 1spins.

As such, models used to describe them admit spin-biquadratic terms in their Hamiltonian. Just as with the usual Heisenberg type spin-bilinear terms, one can, in principle, include couplings up to an arbitrary number of neighboring sites. However, the $J_1 - J_2$ Heisenberg Hamiltonian with a single biquadratic term being a simpler version of the general model, proved appropriate to compare several different common approaches used to represent spins. Doing so allowed us to select the methods most appropriate to describe quantum fluctuations in the system. After choosing the most convenient representation, we were able to reproduce experimental results such as the dynamical structure factors both below and above the magnetic transition temperature in iron pnictide materials, particularly, in the compound BaFe₂As₂ [59, 19].

The full model with additional couplings is necessary to give an accurate description of the full phase diagram in the case of the iron chalcogenides. In particular, we were able to model the transition from the non-magnetic order in FeSe to a double stripe magnetic ordering upon doping with Telurium. The observation of a columnar antiferromagnetic ordering under applied pressure was also predicted by this model. In addition to this, we studied the possibility of intermediate incommensurate phases appearing in the system.

In the second part, we turned our attention towards the Kondo effect, characteristic of heavy fermion materials. We used a novel approach to tackling the multichannel Kondo problem for a few impurities, involving sums over Matsubara frequencies in the imaginary axis. We found agreement with previous results for the susceptibility, phase diagrams and phase shifts.

Once we had put our method to test, we proceeded to add the final key element needed to describe the unconventional superconductivity showcase in certain heavy fermion compounds, namely, the Cerium-based 115 family of materials. Since we had already accounted for the Kondo and RKKY couplings, we hypothesized a pairing interaction arising from higher-than-second order expansion on the Anderson interaction to be responsible for the appearance of superconductivity. In our framework, this is represented by a coupling g between the holons, that is, the collective excitations arising from the Kondo interaction of the spin with the conduction electrons. The inclusion of this pairing indeed translated into the appearance of superconductivity under certain conditions, namely, the existence of correlations between both the bosons and the holons of the system as well as a minimum value of the coupling between the latter. Introducing more than two impurities into the system as well as different couplings between said local moments could shed light into the interplay between magnetic frustration and the Kondo effect.

Appendix A

Non linear spin waves

Starting from the Hamiltonian we used for the iron pnictides, we take all averages to be real for convenience, without loss of generality.

$$n = \langle a_{\boldsymbol{r}}^{\dagger} a_{\boldsymbol{r}} \rangle$$

$$g_{x} = \langle a_{\boldsymbol{r}} a_{\boldsymbol{r}+\hat{\boldsymbol{x}}} \rangle = \langle a_{\boldsymbol{r}}^{\dagger} a_{\boldsymbol{r}+\hat{\boldsymbol{x}}}^{\dagger} \rangle$$

$$f_{y} = \langle a_{\boldsymbol{r}}^{\dagger} a_{\boldsymbol{r}+\hat{\boldsymbol{y}}} \rangle = \langle a_{\boldsymbol{r}} a_{\boldsymbol{r}+\hat{\boldsymbol{y}}}^{\dagger} \rangle$$

$$g_{xy} = \langle a_{\boldsymbol{r}} a_{\boldsymbol{r}+\hat{\boldsymbol{x}}\pm\hat{\boldsymbol{y}}} \rangle = \langle a_{\boldsymbol{r}}^{\dagger} a_{\boldsymbol{r}+\hat{\boldsymbol{x}}\pm\hat{\boldsymbol{y}}}^{\dagger} \rangle$$
(A.1)

We assume the rest of the averages to be zero by virtue of the conservation of the total z-component of the spin $(S^z = \sum_i S_i^z)$ in each direction. In principle, both bilinear and biquadratic terms can be treated in this manner. However, for the purpose of studying the differences between the different approaches to the decoupling procedure, we also use a Hubbard-Stratonovich (HS) transformation for the biquadratic spin term:

$$\left(\boldsymbol{S}_{\boldsymbol{r}}\cdot\boldsymbol{S}_{\boldsymbol{r}'}\right)^{2}\simeq 2\left<\boldsymbol{S}_{\boldsymbol{r}}\cdot\boldsymbol{S}_{\boldsymbol{r}'}\right>\boldsymbol{S}_{\boldsymbol{r}}\cdot\boldsymbol{S}_{\boldsymbol{r}'}-\left<\boldsymbol{S}_{\boldsymbol{r}}\cdot\boldsymbol{S}_{\boldsymbol{r}'}\right>^{2}.$$
(A.2)

The remaining spin bilinears are then decoupled as usual per Wick's theorem, whereas the HS averages themselves can be expressed in terms of the mean-field parameters in Eq. (A.1) as follows:

$$\Gamma_{x} = \langle \boldsymbol{S}_{\boldsymbol{r}} \cdot \boldsymbol{S}_{\boldsymbol{r}+\hat{\boldsymbol{x}}} \rangle = -(S - n - g_{x})^{2}$$

$$\Gamma_{y} = \langle \boldsymbol{S}_{\boldsymbol{r}} \cdot \boldsymbol{S}_{\boldsymbol{r}+\hat{\boldsymbol{y}}} \rangle = (S - n + f_{y})^{2}.$$
(A.3)

The advantage of using the above HS transformation is that it results in simpler expressions for the spin-wave dispersions (see Eq. A.7 below). However, as we show in the main text, this comes at a price that the HS decoupling is much worse at capturing the spin fluctuations compared to the full decoupling (FD) method. With this proviso, we show the details of both methods below, but the reader is advised to use the FD method for accurate results.

After full use of Wick's theorem, the non-linear spin-wave theory results in the following quadratic Hamiltonian (up to inessential constant terms):

$$\mathcal{H}_{NLSW} = \sum_{\boldsymbol{k}} \left[A_{\boldsymbol{k}} \left(a_{\boldsymbol{k}}^{\dagger} a_{\boldsymbol{k}} + a_{-\boldsymbol{k}} a_{-\boldsymbol{k}}^{\dagger} \right) + B_{\boldsymbol{k}} \left(a_{\boldsymbol{k}} a_{-\boldsymbol{k}} + a_{\boldsymbol{k}}^{\dagger} a_{-\boldsymbol{k}}^{\dagger} \right) \right], \qquad (A.4)$$

which, after the Bogoliubov transformation, is expressed in terms of new boson

operators:

$$\mathcal{H}_{NLSW} = \sum_{\boldsymbol{k}} \omega_{\boldsymbol{k}} \left(\alpha_{\boldsymbol{k}}^{\dagger} \alpha_{\boldsymbol{k}} + \frac{1}{2} \right), \qquad (A.5)$$

with the spin wave dispersion:

$$\omega_{\boldsymbol{k}} = 2\sqrt{A_{\boldsymbol{k}}^2 - B_{\boldsymbol{k}}^2},\tag{A.6}$$

where the expressions for the coefficients A_k and B_k are given in Eq. (A.7) for the HS decoupling and in Eq. (A.8) for the full decoupling (FD):

$$A_{\mathbf{k}(HS)} = (J_1 - 2K\Gamma_x)(S - n - g_x) + (J_1 - 2K\Gamma_y)(S - n + f_y)(\cos k_y - 1) + 2J_2(S - n - g_{xy})$$
$$B_{\mathbf{k}(HS)} = (J_1 - 2K\Gamma_x)(S - n - g_x)\cos k_x + 2J_2(S - n - g_{xy})\cos k_x\cos k_y.$$
(A.7)

$$\begin{aligned} A_{k(FD)} &= J_1(S - n - g_x) + J_1(S - n + f_y)(\cos k_y - 1) + \\ &+ 2J_2(S - n - g_{xy}) - KS^2 \{-S + 2[1 + 5(n + g_x)]\} - \\ &- KS^2 \{-S + 2[1 + 5(n - f_y)]\}(1 - \cos k_y); \end{aligned} \tag{A.8} \\ B_{k(FD)} &= J_1(S - n - g_x) \cos k_x + 2J_2(S - n - g_{xy}) \cos k_x \cos k_y - \\ &- KS^2 \{-S + 2[1 + 5(n + g_x)]\} \cos k_x. \end{aligned}$$

Minimizing the free energy with respect to the mean-field parameters defined in Eq. (A.1), we finally arrive at a system of Euler–Lagrange equations:

$$\alpha_{x} = \frac{1}{N_{s}} \sum_{\mathbf{k}} \left(\left\langle a_{\mathbf{k}}^{\dagger} a_{\mathbf{k}} \right\rangle + \left\langle a_{\mathbf{k}} a_{-\mathbf{k}} \right\rangle \cos k_{x} \right) =$$

$$= -\frac{1}{2} + \frac{1}{N_{s}} \sum_{\mathbf{k}} \left(1 + 2n_{\mathbf{k}} \right) \frac{A_{\mathbf{k}} - B_{\mathbf{k}} \cos k_{x}}{\omega_{\mathbf{k}}};$$

$$\beta_{y} = \frac{1}{N_{s}} \sum_{\mathbf{k}} \left\langle a_{\mathbf{k}}^{\dagger} a_{\mathbf{k}} \right\rangle (1 - \cos k_{y}) = -\frac{1}{2} + \frac{1}{N_{s}} \sum_{\mathbf{k}} \left(1 + 2n_{\mathbf{k}} \right) \frac{A_{\mathbf{k}} (1 - \cos k_{y})}{\omega_{\mathbf{k}}};$$

$$\alpha_{xy} = \frac{1}{N_{s}} \sum_{\mathbf{k}} \left(\left\langle a_{\mathbf{k}}^{\dagger} a_{\mathbf{k}} \right\rangle + \left\langle a_{\mathbf{k}} a_{-\mathbf{k}} \right\rangle \cos k_{x} \cos k_{y} \right) =$$

$$= -\frac{1}{2} + \frac{1}{N_{s}} \sum_{\mathbf{k}} \left(1 + 2n_{\mathbf{k}} \right) \frac{A_{\mathbf{k}} - B_{\mathbf{k}} \cos k_{x} \cos k_{y}}{\omega_{\mathbf{k}}},$$
(A.9)

where for convenience, we have denoted: $\alpha_x = n + g_x$, $\alpha_{xy} = n + g_{xy}$, and $\beta_y = n - f_y$. The above equations are to be solved self-consistently because their right-hand side depends on the mean-field parameters themselves via Eqs. (A.7) and (A.8).

Appendix B

Dyson-Maleev bosons (modified spin waves)

Once again, we obtain dispersions of the form: $\omega_{\mathbf{k}} = 2\sqrt{A_{\mathbf{k}}^2 - B_{\mathbf{k}}^2}$ for the twoand three-dimensional cases, respectively:

$$A_{k(2D)} = \lambda + 2J_2(S - \alpha_{xy}) + J_1 [r_x(S - \alpha_x) + r_y(S - \beta_y)(\cos k_y - 1)]$$

$$B_{k(2D)} = J_1 r_x(S - \alpha_x) \cos k_x + 2J_2(S - \alpha_{xy}) \cos k_x \cos k_y$$
(B.1)

$$A_{\boldsymbol{k}(3D)} = A_{\boldsymbol{k}(2D)} + J_c(S - \alpha_z)$$

$$B_{\boldsymbol{k}(3D)} = B_{\boldsymbol{k}(2D)} + J_c(S - \alpha_z) \cos k_z$$
(B.2)

Where r_x , r_y stand for the following expressions:
$$r_{x} = 1 + \frac{K}{S - \alpha_{x}} \left[2S^{3} - 2S^{2}(1 + 5\alpha_{x}) + S(18\alpha_{x}^{2} + 8\alpha_{x} + 1) - 12\alpha_{x}^{3} - 9\alpha_{x}^{2} - 2\alpha_{x} \right]$$

$$r_{y} = 1 - \frac{K}{S - \beta_{y}} \left[2S^{3} - 2S^{2}(1 + 5\beta_{y}) + S(18\beta_{y}^{2} + 8\beta_{y}) - 12\beta_{y}^{3} - 9\beta_{y}^{2} - \beta_{y} \right]$$
(B.3)

Finally, we introduced the chemical potential λ to enforce the constraint $\langle S_z \rangle = 0$ in the paramagnetic regime, which results in the following equation:

$$S = \frac{1}{N_s} \sum_{\boldsymbol{k}} \left\langle a_{\boldsymbol{k}}^{\dagger} a_{\boldsymbol{k}} \right\rangle \to S + \frac{1}{2} = \frac{1}{N_s} \sum_{\boldsymbol{k}} \left(1 + 2n_{\boldsymbol{k}} \right) \frac{A_{\boldsymbol{k}}}{\omega_{\boldsymbol{k}}} \tag{B.4}$$

Thus, $\lambda = 0$ in the magnetically ordered phase. The resulting set of self-consistent equations has the same form of (A.9), with the addition of the following equation in the three-dimensional case:

$$\alpha_{z} = \frac{1}{N_{s}} \sum_{\boldsymbol{k}} \left(\left\langle a_{\boldsymbol{k}}^{\dagger} a_{\boldsymbol{k}} \right\rangle + \left\langle a_{\boldsymbol{k}} a_{-\boldsymbol{k}} \right\rangle \cos k_{z} \right) = -\frac{1}{2} + \frac{1}{N_{s}} \sum_{\boldsymbol{k}} \left(1 + 2n_{\boldsymbol{k}} \right) \frac{A_{\boldsymbol{k}} - B_{\boldsymbol{k}} \cos k_{z}}{\omega_{\boldsymbol{k}}}$$
(B.5)

We differentiate the two domains, $T < T_N$ (where n < S and $\lambda = 0$) and $T > T_N$ (where n = S and $\lambda \neq 0$).

Finally, the dynamical spin structure factors:

$$S(\boldsymbol{q},\omega) = \int dt \ e^{i\omega t} \left\langle \boldsymbol{S}_{\boldsymbol{q}}(t) \cdot \boldsymbol{S}_{-\boldsymbol{q}}(0) \right\rangle \tag{B.6}$$

are given by [65]:

$$S(\boldsymbol{q},\omega) = \frac{2\pi}{N_s} \sum_{\boldsymbol{k}} \sum_{s,s'=\pm 1} \left[\cosh\left(2\theta_{\boldsymbol{k}+\boldsymbol{q}} - 2\theta_{\boldsymbol{k}}\right) - ss' \right] \times \delta(\omega - s\epsilon_{\boldsymbol{k}+\boldsymbol{q}} - s'\epsilon_{\boldsymbol{k}}) n_{\boldsymbol{k}+\boldsymbol{q}}^s n_{\boldsymbol{k}}^{s'},$$
(B.7)

where: $n_{k}^{-} = n_{k}$ and $n_{k}^{+} = n_{k} + 1$. The angle θ_{q} is determined by the coefficients of the Bogoliubov transformation:

$$\tanh\left(2\theta_{\boldsymbol{q}}\right) = \frac{B_{\boldsymbol{q}}}{A_{\boldsymbol{q}}} \tag{B.8}$$

As mentioned in the main text, in practice, we substitute the δ function by a Lorentzian broadening of width γ in order to obtain finite results. Said function takes the following form:

$$\delta(\omega - \Delta\epsilon) \to \frac{1}{\pi} \frac{\gamma}{(\omega - \Delta\epsilon)^2 + \gamma^2}$$
 (B.9)

Appendix C

Generalized spin waves (flavor waves)

In what follows we outline the expressions describing the spin wave dispersions for all phases identified in the iron chalcogenides. To do so, we use the procedure outlined in Section 3.5 in order to obtain a quadratic Hamiltonian. In all cases, we will start by performing a unitary transformation of the bosonic and spin operators into a new basis:

$$\tilde{\boldsymbol{b}}_i = \boldsymbol{\mathcal{V}}_i^{\dagger} \boldsymbol{b}_i \tag{C.1}$$

$$\tilde{\boldsymbol{S}}_i = \boldsymbol{\mathcal{V}}_i^{\dagger} \boldsymbol{S}_i \boldsymbol{\mathcal{V}}_i,$$
 (C.2)

where the transformation matrices are determined by the directors $|\vec{d_i}\rangle$ that de-

scribe each ground state[95].

At the end of each section, when necessary, we also include the formulas for the dynamical spin structure factors, which we computed following the definition below[95]:

$$S^{\mu\nu}(\boldsymbol{q},\omega) = \chi_{\mu\nu}''(\boldsymbol{q},\omega) = \frac{N_S}{N} \sum_{\alpha\beta} \sum_{f} \left\langle \text{g.s.} | S^{\mu}_{\alpha,\boldsymbol{q}} | f \right\rangle \left\langle f | S^{\nu}_{\beta,-\boldsymbol{q}} | \text{g.s.} \right\rangle \times \delta(\omega - E_f + E_g)$$
(C.3)

Here, $|f\rangle \langle f| = 1$ is the complete set of states and the indices α, β are used to denote the different sublattices, with the ratio N/N_S keeping track of their total number.

C.1 Flavor waves for the ferroquadrupolar (FQ) phase

In the FQ state the directors $\vec{d_i}$ are identical on all sites (in total one sublattice $N/N_S = 1$). Due to SU(2) symmetry of the Hamiltonian, we can safely choose

$$\vec{d_i} = \{1, 0, 0\}.$$
 (C.4)

Correspondingly, the transformation matrix is same on every site *i*:

$$\mathcal{V}_{i} = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix}.$$
 (C.5)

The flavor wave Hamiltonian expanded up to quadratic level is given by

$$\mathcal{H}_{\text{fw}} = \sum_{\boldsymbol{q},a} \left[t(\boldsymbol{q}) + \lambda \right] \left(\tilde{b}_{\boldsymbol{q},a} \tilde{b}_{\boldsymbol{q},a}^{\dagger} + \tilde{b}_{-\boldsymbol{q},a}^{\dagger} \tilde{b}_{-\boldsymbol{q},a} \right) + \sum_{\boldsymbol{q},a} \left[\Delta(\boldsymbol{q}) \tilde{b}_{\boldsymbol{q},a}^{\dagger} \tilde{b}_{-\boldsymbol{q},a}^{\dagger} + h.c. \right] + NE_{0},$$
(C.6)

where

$$t(\boldsymbol{q}) = J_1(\cos q_x + \cos q_y) + 2J_2 \cos q_x \cos q_y + J_3(\cos 2q_x + \cos 2q_y), \quad (C.7)$$
$$\Delta(\boldsymbol{q}) = (K_1 - J_1)(\cos q_x + \cos q_y) + 2(K_2 - J_2) \cos q_x \cos q_y - J_3(\cos 2q_x + \cos 2q_y), \quad (C.8)$$
$$\lambda = -2(K_1 + K_2), \quad (C.9)$$
$$E_0 = 4(K_1 + K_2). \quad (C.10)$$

Bogoliubov transformation:

$$\alpha_{\boldsymbol{q},a} = \cosh \theta_{\boldsymbol{q}} \tilde{b}_{\boldsymbol{q},a} - \sinh \theta_{\boldsymbol{q}} \tilde{b}_{-\boldsymbol{q},a}^{\dagger}, \qquad (C.11)$$

with

$$\tanh 2\theta_{\boldsymbol{q}} = -\frac{\Delta(\boldsymbol{q})}{t(\boldsymbol{q}) + \lambda}.$$
 (C.12)

The diagonalized Hamiltonian:

$$\mathcal{H}_{\text{fw}} = \sum_{a=1,2} \sum_{\boldsymbol{q}} \omega_{\boldsymbol{q}} (\alpha_{\boldsymbol{q},a}^{\dagger} \alpha_{\boldsymbol{q},a} + 1/2) + N(E_0 - \lambda), \qquad (C.13)$$

where the dispersion ω_q is given by:

$$\omega_{\boldsymbol{q}} = 2\sqrt{[t(\boldsymbol{q}) + \lambda]^2 - \Delta^2(\boldsymbol{q})}.$$
 (C.14)

Finally, the dynamical structure factors in this case are given by:

$$\chi_{xx}''(\boldsymbol{q},\omega) = 0 \qquad (C.15)$$

$$\chi_{yy}^{\prime\prime}(\boldsymbol{q},\omega) = \chi_{zz}^{\prime\prime}(\boldsymbol{q},\omega) = \frac{t(\boldsymbol{q}+\lambda+\Delta(\boldsymbol{q}))}{\sqrt{[t(\boldsymbol{q}+\lambda)]^2 - \Delta^2(\boldsymbol{q})}}\delta(\omega-\omega_{\boldsymbol{q}})$$
(C.16)

C.2 Flavor waves for the columnar antiferromagnetic (CAFM) phase

There are in total two sublattices $N/N_S = 2$, whose directors can be chosen as:

$$\vec{d}_{i\in A} = \frac{1}{\sqrt{2}} \{0, 1, i\},$$
 (C.17)

$$\vec{d}_{i\in B} = \frac{1}{\sqrt{2}} \{0, 1, -i\}.$$
 (C.18)

Correspondingly, the transformation matrices are written below:

$$\mathcal{V}_{i\in A} = \frac{1}{\sqrt{2}} \begin{pmatrix} 0 & 0 & \sqrt{2} \\ 1 & i & 0 \\ i & 1 & 0 \end{pmatrix}, \quad (C.19)$$

$$\mathcal{V}_{i\in B} = \frac{1}{\sqrt{2}} \begin{pmatrix} 0 & 0 & \sqrt{2} \\ 1 & -i & 0 \\ -i & 1 & 0 \end{pmatrix}. \quad (C.20)$$

The quadratic terms of the resulting Hamiltonian now include cross-terms between sublattices:

$$\mathcal{H}_{\text{fw}} = \sum_{\boldsymbol{q},a} \left(t_{aa} + \lambda_{aa} \right) \left(\tilde{b}_{A,\boldsymbol{q},a} \tilde{b}_{A,\boldsymbol{q},a}^{\dagger} + \tilde{b}_{A,\boldsymbol{q},a}^{\dagger} \tilde{b}_{A,\boldsymbol{q},a} \right. \\ \left. + \tilde{b}_{B,\boldsymbol{q},a} \tilde{b}_{B,\boldsymbol{q},a}^{\dagger} + \tilde{b}_{B,\boldsymbol{q},a}^{\dagger} \tilde{b}_{B,\boldsymbol{q},a} \right)$$

$$\left. + \sum_{\boldsymbol{q},a} \Delta_{aa} \left(\tilde{b}_{A,\boldsymbol{q},a}^{\dagger} \tilde{b}_{B,-\boldsymbol{q},a}^{\dagger} + \tilde{b}_{B,\boldsymbol{q},a}^{\dagger} \tilde{b}_{A,-\boldsymbol{q},a}^{\dagger} + h.c. \right)$$

$$\left(\text{C.21} \right)$$

With the coefficients λ_{aa} , $t_{aa}(\boldsymbol{q})$ and $\Delta_{aa}(\boldsymbol{q})$ depending on the parameters of the model as follows:

 $\lambda_{11} = 2(2J_2 - K_2) - 4J_3, \qquad (C.22)$

$$\lambda_{22} = -K_1 + 2(J_2 - K_2) - 2J_3, \qquad (C.23)$$

$$t_{11}(\boldsymbol{q}) = K_1 \cos q_y, \qquad (C.24)$$

$$t_{22}(\boldsymbol{q}) = J_1 \cos q_y + J_3[\cos(2q_x) + \cos(2q_y)]$$
(C.25)

$$\Delta_{11}(\boldsymbol{q}) = K_1 \cos q_x + 2K_2 \cos q_x \cos q_y, \qquad (C.26)$$

$$\Delta_{22}(\boldsymbol{q}) = -(J_1 - K_1)\cos q_x - 2(J_2 - K_2)\cos q_x \cos q_y.$$
(C.27)

And the diagonalized Bogolibouv dispersions finally taking the following form (with a = 1, 2):

$$\omega_{\boldsymbol{q},a} = 2\sqrt{[t_{aa}(\boldsymbol{q}) + \lambda_{aa}]^2 - \Delta_{aa}^2(\boldsymbol{q})}.$$
 (C.28)

C.3 Flavor waves for the Néel phase

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In this case, both the Hamiltonian as well as the diagonalized dispersions have the same symbolic expression as in the CAFM case. However, the coefficients are now given by:

$$\lambda_{11} = 2(2J_1 - K_1) - 2(2J_2 - K_2) - 4J_3, \qquad (C.29)$$

$$\lambda_{22} = 2(J_1 - K_1) - 2J_2 - 2J_3, \tag{C.30}$$

$$t_{11}(\boldsymbol{q}) = 2K_2 \cos q_x \cos q_y, \qquad (C.31)$$

$$t_{22}(\boldsymbol{q}) = 2J_2 \cos q_x \cos q_y + J_3[\cos(2q_x) + \cos(2q_y)], \qquad (C.32)$$

$$\Delta_{11}(\boldsymbol{q}) = K_1(\cos q_x + \cos q_y), \qquad (C.33)$$

$$\Delta_{22}(\boldsymbol{q}) = -(J_1 - K_1)(\cos q_x + \cos q_y). \tag{C.34}$$

C.3.1 Flavor waves for the double stripe (DS) phase

Unlike in the previous two cases where the introduction of two sublattices was enough, four are necessary in this case. However, since there are still only two distinct directors, the previously shown transformation matrices are enough to derive the Hamiltonian. It is now convenient to write the actual Hamiltonian down so that it becomes block diagonal. This is due to the lack of cross terms between the bosonic operators of the different modes. The quadratic terms can be written in the following matricial form:

$$\mathcal{H}_{\rm fw} = 2 \sum_{\boldsymbol{q}} (\psi_{11}^{\dagger} \psi_{22}) H_{\rm fw} \begin{pmatrix} \psi_{11} \\ \psi_{22}^{\dagger} \end{pmatrix}, \qquad (C.35)$$

with the block-diagonal form of the Hamiltonian matrix explicitly written as:

$$\tilde{S}_i^{\nu} = \mathcal{V}_i^{\dagger} S_i^{\nu} \mathcal{V}_i \tag{C.36}$$

$$H_{\rm fw} = 2 \begin{pmatrix} \mathcal{J} & 0\\ 0 & \mathcal{K} \end{pmatrix}, \qquad (C.37)$$

and where: $\psi_{aa}^{\dagger} = (b_{A,q,a}^{\dagger}, b_{B,q,a}^{\dagger}, b_{C,-q,a}, b_{D,-q,a})$. The matrix elements of each 4×4 block-diagonal matrix are given by:

$$\mathcal{J}_{11} = \mathcal{J}_{22} = \mathcal{J}_{33} = \mathcal{J}_{44} = 4J_3 + K_2 \cos\left(q_x - q_y\right) \equiv \mathcal{A}, \quad (C.38)$$

$$\mathcal{J}_{12} = \mathcal{J}_{14}^* = \mathcal{J}_{21}^* = \mathcal{J}_{23} = \mathcal{J}_{32}^* = \mathcal{J}_{34} = \mathcal{J}_{41} = \mathcal{J}_{43}^* = \frac{K_1}{2} (e^{iq_x} + e^{iq_y}), \quad (C.39)$$

$$\mathcal{J}_{13} = \mathcal{J}_{24} = \mathcal{J}_{31} = \mathcal{J}_{42} = K_2 \cos\left(q_x + q_y\right) \equiv \mathcal{B}. \quad (C.40)$$

and

$$\mathcal{K}_{11} = \mathcal{K}_{22} = \mathcal{K}_{33} = \mathcal{K}_{44} =$$
$$= -(K_1 + K_2) + 2J_3 + J_2 \cos(q_x - q_y) \equiv \mathcal{C}, \qquad (C.41)$$
$$\mathcal{K}_{44} = \mathcal{K}_{44} = \mathcal{K}$$

$$\mathcal{K}_{12} = \mathcal{K}_{21}^* = \mathcal{K}_{34} = \mathcal{K}_{43}^* = \frac{1}{2} (e^{iq_x} + e^{iq_y}), \qquad (C.42)$$

$$\mathcal{K}_{14}^* = \mathcal{K}_{23} = \mathcal{K}_{32}^* = \mathcal{K}_{41} = -\frac{(J_1 - K_1)}{2} (e^{iq_x} + e^{iq_y}), \quad (C.43)$$

$$\mathcal{K}_{13} = \mathcal{K}_{24} = \mathcal{K}_{31} = \mathcal{K}_{42} =$$

$$= -(J_2 - K_2)\cos(q_x + q_y) - J_3[\cos(2q_x) + \cos(2q_y)] \equiv \mathcal{D}.$$
 (C.44)

The dispersions can be derived immediately from a standard Bogoliubov transformation of the Hamiltonian above. This is done by obtaining the eigenvalues of the new matrix resulting from the similarity transformation: $\tilde{H}_{\text{fw}} = \Theta H_{\text{fw}}$, where the matrix $\Theta = \text{diag}(1, 1, -1, -1)$. This gives the following result:

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$$\omega_{\boldsymbol{q},1,\pm} = 2\sqrt{\mathcal{A}^2 - \mathcal{B}^2 \pm 2\sqrt{\kappa_1}},\tag{C.45}$$

$$\omega_{q,2,\pm} = 2\sqrt{\mathcal{C}^2 - \mathcal{D}^2 - \frac{K_1}{2}(K_1 - 2J_1) \pm 2\sqrt{\kappa_2}},$$
 (C.46)

where κ_1 and κ_2 are given by:

$$\kappa_{1} = \frac{K_{1}^{2}}{2} (\mathcal{A}^{2} + \mathcal{B}^{2}) [1 + \cos(q_{x} - q_{y})] - \frac{K_{1}^{2} \mathcal{A} \mathcal{B}}{2} [\cos(2q_{x}) + \cos(2q_{y}) + 2\cos(q_{x} + q_{y})] - \frac{K_{1}^{4}}{16} [\sin(2q_{x}) + \sin(2q_{y}) + 2\sin(q_{x} + q_{y})]^{2}$$
(C.47)

$$\kappa_{2} = \frac{1}{2} [J_{1}^{2} \mathcal{C}^{2} + (J_{1} - K_{1})^{2} \mathcal{D}^{2}] [1 + \cos(q_{x} - q_{y})] + \frac{J_{1}(J_{1} - K_{1})\mathcal{C}\mathcal{D}}{2} [\cos(2q_{x}) + \cos(2q_{y}) + 2\cos(q_{x} + q_{y})] - (C.48) - \frac{J_{1}^{2}(J_{1} - K_{1})^{2}}{16} [\sin(2q_{x}) + \sin(2q_{y}) + 2\sin(q_{x} + q_{y})]^{2}$$

The dynamical structure factors, in this case, are:

$$\chi_{xx}''(\boldsymbol{q},\omega) = 1 \qquad (C.49)$$

$$\chi_{yy}''(\boldsymbol{q},\omega) = \chi_{zz}''(\boldsymbol{q},\omega) =$$

$$= \frac{1}{8} \sum_{i=1,4} |(V_{\boldsymbol{q}}^{1i} + V_{\boldsymbol{q}}^{2i}) - (V_{\boldsymbol{q}}^{3i} + V_{\boldsymbol{q}}^{4i})|^2 \times \delta(\omega - \omega_{\boldsymbol{q},2,+}) +$$

$$+ \frac{1}{8} \sum_{i=2,3} |(V_{\boldsymbol{q}}^{1i} + V_{\boldsymbol{q}}^{2i}) - (V_{\boldsymbol{q}}^{3i} + V_{\boldsymbol{q}}^{4i})|^2 \times \delta(\omega - \omega_{\boldsymbol{q},2,-}) \qquad (C.50)$$

Where the $V_{\pmb{q}}^{ij}$ are the coefficients of the Bogoliubov transformation that diagonalize the Hamiltonian:

$$b_{A,k} = V_{k}^{11} \alpha_{I,k} + V_{k}^{12} \beta_{I,k} + V_{k}^{13} \beta_{II,k}^{\dagger} + V_{k}^{14} \alpha_{II,k}^{\dagger} \quad (C.51)$$

$$b_{B,k} = V_{k}^{21} \alpha_{I,k} + V_{k}^{22} \beta_{I,k} + V_{k}^{23} \beta_{II,k}^{\dagger} + V_{k}^{24} \alpha_{II,k}^{\dagger} \quad (C.52)$$

$$b_{C,\mathbf{k}} = (V_{-\mathbf{k}}^{31})^* \alpha_{I,-\mathbf{k}}^{\dagger} + (V_{-\mathbf{k}}^{32})^* \beta_{I,-\mathbf{k}}^{\dagger} + (V_{-\mathbf{k}}^{33})^* \beta_{II,-\mathbf{k}} + (V_{-\mathbf{k}}^{34})^* \alpha_{II,-\mathbf{k}}$$
(C.53)

$$b_{D,\boldsymbol{k}} = (V_{-\boldsymbol{k}}^{41})^* \alpha_{I,-\boldsymbol{k}}^{\dagger} + (V_{-\boldsymbol{k}}^{42})^* \beta_{I,-\boldsymbol{k}}^{\dagger} + (V_{-\boldsymbol{k}}^{43})^* \beta_{II,-\boldsymbol{k}} + (V_{-\boldsymbol{k}}^{44})^* \alpha_{II,-\boldsymbol{k}} \quad (C.54)$$

C.3.2 Flavor waves for the $(\pi, \frac{\pi}{2})$ phase

Just like before, all the symbolic expressions are the same as those in the section above, with the coefficients of the matrix in Eq. (C.37) given by:

$$\mathcal{J}_{11} = \mathcal{J}_{22} = \mathcal{J}_{33} = \mathcal{J}_{44} = 2J_1 - K_1 \equiv \mathcal{A}, \qquad (C.55)$$

$$\mathcal{J}_{12} = \mathcal{J}_{14}^* = \mathcal{J}_{21}^* = \mathcal{J}_{23} = \mathcal{J}_{32}^* = \mathcal{J}_{34} = \mathcal{J}_{41} = \mathcal{J}_{43}^* = -\frac{K_1}{2}e^{iq_x} + K_2e^{-iq_x}\cos q \qquad (C.56)$$

$$= \frac{\pi_1}{2} e^{iq_x} + K_2 e^{-iq_x} \cos q_y, \qquad (C.56)$$

$$\mathcal{J}_{13} = \mathcal{J}_{24} = \mathcal{J}_{31} = \mathcal{J}_{42} = K_1 \cos\left(2q_y\right) \equiv \mathcal{B}.$$
 (C.57)

and the coefficients K_{ij} taking on the form:

$$\mathcal{K}_{11} = \mathcal{K}_{22} = \mathcal{K}_{33} = \mathcal{K}_{44} = J_1 - \frac{3K_1}{2} - K_2 + J_3 \cos(2q_y) \equiv \mathcal{C}, \quad (C.58)$$

$$\mathcal{K}_{12} = \mathcal{K}_{21}^* = \mathcal{K}_{34} = \mathcal{K}_{43}^* = \frac{J_1}{2}e^{iq_x} + J_2e^{-iq_x}\cos q_y, \quad (C.59)$$

$$\mathcal{K}_{14}^* = \mathcal{K}_{23} = \mathcal{K}_{32}^* = \mathcal{K}_{41} = -\frac{(J_1 - K_1)}{2}e^{iq_x} - (J_2 - K_2)e^{-iq_x}\cos q_y, \quad (C.60)$$

$$\mathcal{K}_{13} = \mathcal{K}_{24} = \mathcal{K}_{31} = \mathcal{K}_{42} = -(J_1 - K_1)\cos q_y - J_3\cos(2k_x) \equiv \mathcal{D}.$$
 (C.61)

After diagonalizing, the resulting dispersions are now:

$$\omega_{q,1,\pm} = 2\sqrt{\mathcal{A}^2 - \mathcal{B}^2 \pm \sqrt{\kappa_1}}, \quad (C.62)$$
$$\omega_{q,2,\pm} = 2\sqrt{\mathcal{C}^2 - \mathcal{D}^2 - \frac{K_1}{4}(K_1 - 2J_1) - K_2(K_2 - 2J_2)\cos^2 q_y \pm \sqrt{\kappa_2}}, \quad (C.63)$$

with:

$$\kappa_{1} = (\mathcal{A}^{2} + \mathcal{B}^{2})(K_{1}^{2} + 4K_{2}^{2}\cos^{2}q_{y}) - \frac{1}{2}(K_{1}^{2} - 4K_{2}^{2}\cos^{2}q_{y})^{2}[1 - \cos(4q_{x})] - 8\mathcal{AB}K_{1}K_{2}\cos q_{y} + (C.64) + 2(\mathcal{A}K_{1} - 2\mathcal{B}K_{2}\cos q_{y})(2\mathcal{A}K_{2}\cos q_{y} - \mathcal{B}K_{1})\cos(2q_{x})$$

$$\kappa_{2} = \mathcal{C}^{2}(J_{1}^{2} + 4J_{2}^{2}\cos^{2}q_{y}) + \mathcal{D}^{2}[(J_{1} - K_{1})^{2} + 4(J_{2} - K_{2})^{2}\cos^{2}q_{y}] - \frac{1}{2}[J_{1}(J_{1} - K_{1}) - 4J_{2}(J_{2} - K_{2})\cos^{2}q_{y}]^{2}[1 - \cos(4q_{x})] + 4\mathcal{C}\mathcal{D}[J_{1}(J_{2} - K_{2}) + J_{2}(J_{1} - K_{1})]\cos q_{y} - 2[\mathcal{C}J_{1} + 2\mathcal{D}(J_{2} - K_{2})\cos q_{y}][2\mathcal{C}J_{2}\cos q_{y} + \mathcal{D}(J_{1} - K_{1})]\cos(2q_{x})$$
(C.65)

C.3.3 Flavor Wave for the staggered dimer (SD) phase

$$\mathcal{K}_{11} = \mathcal{K}_{22} = \mathcal{K}_{33} = \mathcal{K}_{44} = \mathcal{K}_{55} = \mathcal{K}_{66} = \mathcal{K}_{77} = \mathcal{K}_{88} = 4J_3, \ (C.66)$$

$$\mathcal{K}_{15} = \mathcal{K}_{26} = \mathcal{K}_{37} = \mathcal{K}_{48} = \mathcal{K}_{51} = \mathcal{K}_{62} = \mathcal{K}_{73} = \mathcal{K}_{84} = 0, \ (C.67)$$

$$\mathcal{K}_{13} = \mathcal{K}_{28} = \mathcal{K}_{31} = \mathcal{K}_{46} = \mathcal{K}_{57} = \mathcal{K}_{64} = \mathcal{K}_{75} = \mathcal{K}_{82} = K_2 \cos(q_x + q_y), \ (C.68)$$

$$\mathcal{K}_{17} = \mathcal{K}_{24} = \mathcal{K}_{35} = \mathcal{K}_{42} = \mathcal{K}_{53} = \mathcal{K}_{68} = \mathcal{K}_{71} = \mathcal{K}_{86} = K_2 \cos(q_x - q_y), \ (C.69)$$

$$\mathcal{K}_{12} = \mathcal{K}_{25} = \mathcal{K}_{38} = \mathcal{K}_{43} = \mathcal{K}_{56} = \mathcal{K}_{61} = \mathcal{K}_{74} = \mathcal{K}_{87} = \frac{\kappa_1}{2} e^{iq_x}, \quad (C.70)$$

$$\mathcal{K}_{16} = \mathcal{K}_{21} = \mathcal{K}_{34} = \mathcal{K}_{47} = \mathcal{K}_{52} = \mathcal{K}_{65} = \mathcal{K}_{78} = \mathcal{K}_{83} = \frac{K_1}{2} e^{-iq_x}, \quad (C.71)$$

$$\mathcal{K}_{14} = \mathcal{K}_{23} = \mathcal{K}_{36} = \mathcal{K}_{45} = \mathcal{K}_{58} = \mathcal{K}_{67} = \mathcal{K}_{72} = \mathcal{K}_{81} = \frac{\mathcal{K}_1}{\frac{2}{V}} e^{iq_y}, \quad (C.72)$$

$$\mathcal{K}_{18} = \mathcal{K}_{27} = \mathcal{K}_{32} = \mathcal{K}_{41} = \mathcal{K}_{54} = \mathcal{K}_{63} = \mathcal{K}_{76} = \mathcal{K}_{85} = \frac{\mathcal{K}_1}{2} e^{-iq_y}$$
(C.73)

$$\mathcal{J}_{11} = \mathcal{J}_{22} = \mathcal{J}_{33} = \mathcal{J}_{44} = \mathcal{J}_{55} = \mathcal{J}_{66} = \mathcal{J}_{77} = \mathcal{J}_{88} =$$
$$= 2J_3 - (K_1 + K_2), \qquad (C.74)$$

$$\mathcal{J}_{15} = \mathcal{J}_{26} = \mathcal{J}_{37} = \mathcal{J}_{48} = \mathcal{J}_{51} = \mathcal{J}_{62} = \mathcal{J}_{73} = \mathcal{J}_{84} =$$

$$= -J_3[\cos(2q_x) + \cos(2q_y)], \qquad (C.75)$$

$$\mathcal{J}_{13} = \mathcal{J}_{31} = \mathcal{J}_{57} = \mathcal{J}_{75} = J_2 \cos(q_x + q_y),$$
 (C.76)

$$\mathcal{J}_{24} = \mathcal{J}_{42} = \mathcal{J}_{68} = \mathcal{J}_{86} = J_2 \cos(q_x - q_y),$$
 (C.77)

$$\mathcal{J}_{28} = \mathcal{J}_{46} = \mathcal{J}_{64} = \mathcal{J}_{82} = -(J_2 - K_2)\cos(q_x + q_y), \quad (C.78)$$

$$\mathcal{J}_{17} = \mathcal{J}_{35} = \mathcal{J}_{53} = \mathcal{J}_{71} = -(J_2 - K_2)\cos(q_x - q_y), \qquad (C.79)$$

$$\mathcal{J}_{12} = \mathcal{J}_{43} = \mathcal{J}_{56} = \mathcal{J}_{87} = \frac{\mathcal{J}_1}{2} e^{iq_x},$$
 (C.80)

$$\mathcal{J}_{21} = \mathcal{J}_{34} = \mathcal{J}_{65} = \mathcal{J}_{78} = \frac{J_1}{2} e^{-iq_x},$$
 (C.81)

$$\mathcal{J}_{14} = \mathcal{J}_{23} = \mathcal{J}_{58} = \mathcal{J}_{67} = \frac{J_1}{2} e^{iq_y},$$
 (C.82)

$$\mathcal{J}_{32} = \mathcal{J}_{41} = \mathcal{J}_{76} = \mathcal{J}_{85} = \frac{J_1}{2}e^{-iq_y},$$
 (C.83)

$$\mathcal{J}_{25} = \mathcal{J}_{38} = \mathcal{J}_{61} = \mathcal{J}_{74} = -\frac{(J_1 - K_1)}{2}e^{iq_x}, \qquad (C.84)$$

$$\mathcal{J}_{16} = \mathcal{J}_{47} = \mathcal{J}_{52} = \mathcal{J}_{83} = -\frac{(J_1 - K_1)}{2}e^{-iq_x}, \qquad (C.85)$$

$$\mathcal{J}_{36} = \mathcal{J}_{45} = \mathcal{J}_{72} = \mathcal{J}_{81} = -\frac{(J_1 - K_1)}{2}e^{iq_y}, \qquad (C.86)$$

$$\mathcal{J}_{18} = \mathcal{J}_{27} = \mathcal{J}_{54} = \mathcal{J}_{63} = -\frac{(J_1 - K_1)}{2}e^{-iq_y}$$
(C.87)

Appendix D

Padé Approximants

Padé Approximants, initially developed by Henri Padé are rational functions computed to be the best possible approximation of another given function. In general the numerator and the denominator of the approximant can be polynomials of different order but we will focus on the N-point Padé approximant:

$$C_N(z) = \frac{A_N(z)}{B_N(z)} \tag{D.1}$$

Here, $A_N(z)$ and $B_N(z)$ are both polynomials of order (N-1)/2 if N is odd and of orders (N-2)/2 and N/2, respectively is N is even. One can use this method in order to obtain an analytic continuation of a function with values at points in the complex plane. In our case, we wish to obtain the real frequency values of the propagators from the data at the imaginary Matsubara frequencies.

We now proceed to summarize a quick algorithm for computing the Padé approx-

imant of said function. This was described in [96] by Vidberg and Serene. First, we write the rational function in the form of continued fraction given by the following expression, where u_i are the values of our function (in this case, the propagators) and z_i the complex points (the Matsubara frequencies, in our case) where the function takes said values, for indices i = 1, ..., N.

$$C_N(z) = \frac{a_1}{1 + \frac{a_2(z-z_1)}{1 + \dots + a_N(z-z_{N-1})}},$$
(D.2)

We know that the function $C_N(z)$ should take the already known values at the z_i points:

$$C_N(z_i) = u_i,\tag{D.3}$$

with i = 1, ..., N. We can now obtain the a_i coefficients by using the recursion formula: $a_i = g_i(z_i)$. The values of the g_i are then given by:

$$g_1(z_i) = u_i,\tag{D.4}$$

for i = 1, ..., N, and:

$$g_p(z) = \frac{g_{p-1}(z_{p-1}) - g_{p-1}(z)}{(z - z_{p-1})g_{p-1}(z)},$$
(D.5)

for $p \ge 2$. These can be written as the following triangular matrix:

$$\begin{bmatrix} a_1 = u_1 & u_2 & u_3 & u_4 & \dots \\ 0 & a_2 & g_2(z_3) & g_2(z_4) & \dots \\ 0 & 0 & a_3 & g_3(z_4) & \dots \\ 0 & 0 & 0 & a_4 & \dots \\ 0 & 0 & 0 & 0 & \dots \\ \vdots & \vdots & \vdots & \vdots & \ddots \end{bmatrix}$$
(D.6)

whose $p_{i,j}$ coefficients can be calculated as follows:

$$p_{1,j} = u_j, \tag{D.7}$$

for j = 1, ..., N, and:

$$p_{i,j} = \frac{p_{i-1,i-1} - p_{i-1,j}}{(z_j - z_{i-i})p_{i-1,j}}$$
(D.8)

for j = 2, ..., N and i = 2, ..., j. Once we obtain these coefficients, we can use the diagonal elements a_i to finally calculate the recursive expressions for the initial fraction $C_N(z) = \frac{A_N(z)}{B_N(z)}$ and thus the value of our function in the real axis. These are given by:

 $A_0 = 0 \tag{D.9}$

$$A_1 = a_1 \tag{D.10}$$

$$A_{n+1}(z) = A_n(z) + (z - z_n)a_{n+1}A_{n-1}(z)$$
(D.11)

and:

 $B_0 = B_1 = 1$ (D.12)

$$B_{n+1}(z) = B_n(z) + (z - z_n)a_{n+1}B_{n-1}(z)$$
(D.13)

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