

Numerical Studies of Models for Strongly Correlated Electrons Using Monte Carlo Mean Field and Density Matrix Renormalization Group Methods

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To my family

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Abstract

The interplay of magnetism and superconductivity in the iron-based superconductors is investigated numerically using Monte Carlo mean field and density matrix renormalization group methods. The finite temperature magnetic and transport properties of the 2D oxypnictides are calculated using Monte Carlo mean field methods, capturing many experimentally relevant phases and predicting a new phase dubbed “orbital selective directional conductor”. Finally, pairing tendencies are explored using exact diagonalization and density matrix renormalization group for low dimensional iron-based superconductor, BaFe_2S_3 , predicting hole pairs arranged along the diagonals or rungs of the ladders in real space. This dissertation further adds to the evidence that superconductivity in the iron based compounds has its origin in the magnetic fluctuations that are driven by correlations.

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Chapter 1

Introduction

Identifying the mechanism driving unconventional superconductivity remains among the most important unsolved problems of condensed matter physics. The discovery of superconductivity in copper oxide layers (cuprates) induced a tremendous effort in trying to explain the driving mechanism of those unconventional superconductors [8, 21], which added a new branch of research to condensed matter physics: high T_c superconductivity (HTc-Scs). After much effort in cuprates, an iron (Fe) based compound, fluorine doped LaFeAsO (oxypnictide), was shown to have a superconducting transition (T_c) of 26 K [51]. This finding renewed the excitement in the condensed matter community, leading to the discovery of many other iron based superconductors (Fe-Scs) [50]. There are two main classes of Fe-Scs: pnictides and chalcogenides. Pnictides refer to the bonding of iron ($3d^6$ valence) with that of arsenic or phosphorus ($3p^3/4p^3$ valence) (Fig. 1.1). Chalcogenides refer to the bonding of iron ($3d^6$ valence) with that of sulfur, selenium or tellurium ($3p^4$, $4p^4$ or $5p^4$ valence). In this thesis, our main focus will be on AFe_2As_2 (2D layered pnictides) and on BaFe_2S_3 quasi-1D ladders (chalcogenides).

1.1 Iron based and copper based superconductors

Unlike the conventional superconductors, both HTc-Scs have a magnetic ground-state at low temperatures in the parent compounds. In fact, cuprates and Fe-Scs have a qualitatively similar phase diagram (Fig. 1.2). With an exception of LiFeAs [84, 123], the common

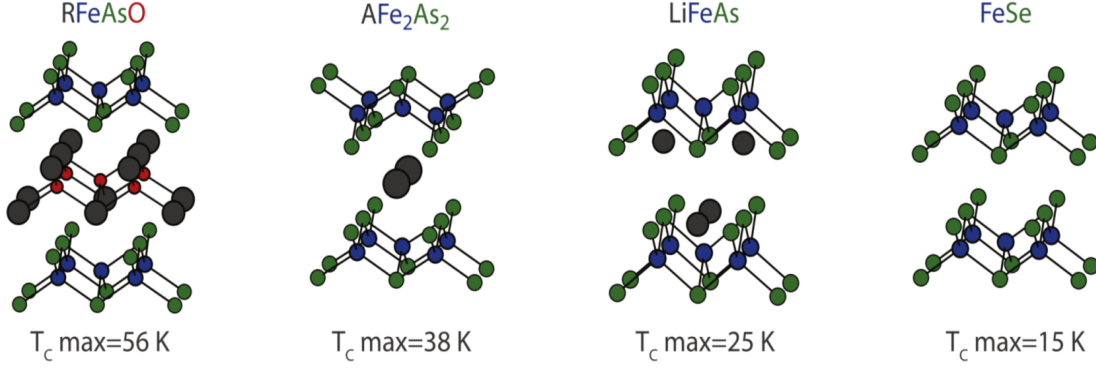


Figure 1.1: The atomic structures of three different pnictide compounds and one chalcogenide (FeSe), where the blue circles represent iron atoms, and green circles represent the ligand atoms (As, Se). The corresponding critical temperature for the superconducting transition is also listed. In the second chapter of this thesis, we focus on the 2D layers formed by the Fe₂As₂. In all Fe-Scs, iron atoms form a 2D layer structure. These structures are reproduced from Ref. [64].

qualitative features are (1) a magnetic dome near the doping corresponding to the parent compound, (2) superconductivity emerging upon doping in the 2D planes and (3) a finite temperature structural phase transition. These qualitative similarities imply a common underlying mechanism of superconductivity, and therefore it was natural to use the results of cuprates as a guide for Fe-Scs. Despite the qualitative similarities between cuprates and pnictides, there are some important differences. Cuprates are known to form the Néel type anti-ferromagnetic (AFM) order in the 2D CuO₂ planes [39], while pnictides are known to have a $(\pi, 0)$ stripe collinear type antiferromagnetic order at low temperatures in the 2D FeAs planes [36, 27] (Fig. 1.3).

Perhaps the most important difference is that the parent compound of cuprates are Mott-insulators [21] while, surprisingly, the pnictide parent compounds are bad-metals [45, 27]. This distinction in the transport properties can be understood by examining the d orbitals contribution to the Fermi surface. The LDA calculations of the cuprate's band structure show that *only* the correlated d orbital is $d_{x^2-y^2}$ but the oxygen content is significant [97, 59, 119]. One can also deduce the same conclusion from a simple atomic picture. The crystal field induced by local octahedral cage around copper atom split the five-fold degeneracy of the copper 3d orbitals. The nine valence electrons of Cu⁺² fills the local 3d orbitals according to the Hund's rules and crystal fields of each orbital (Fig. 1.4a). This electronic arrangement

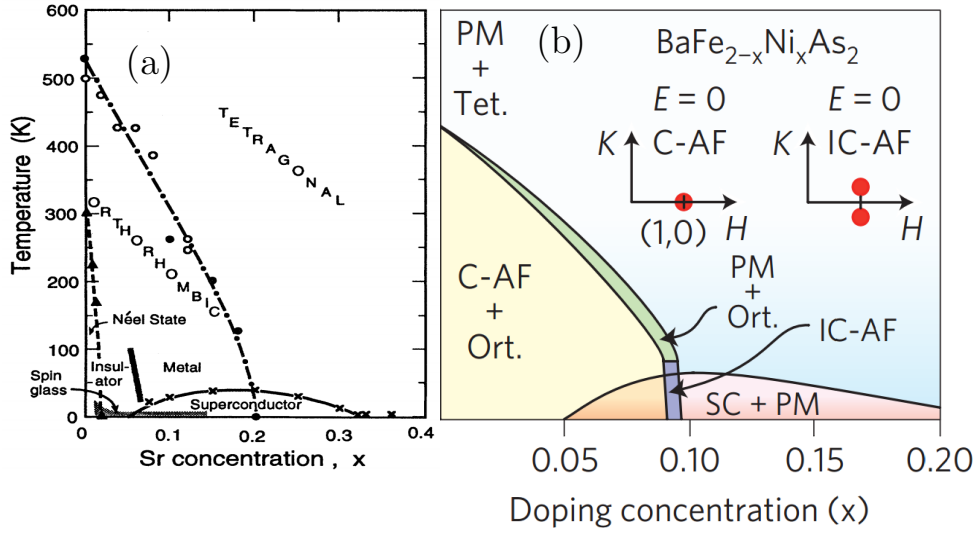


Figure 1.2: (a) phase diagram of the cuprate $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$. (b) Phase diagram of $\text{BaFe}_{2-x}\text{Ni}_x\text{As}_2$ [36]. Both materials have similar phase transitions: tetragonal to orthorhombic, superconducting, and magnetic. These plots are reproduced from Ref. [21] and [27].

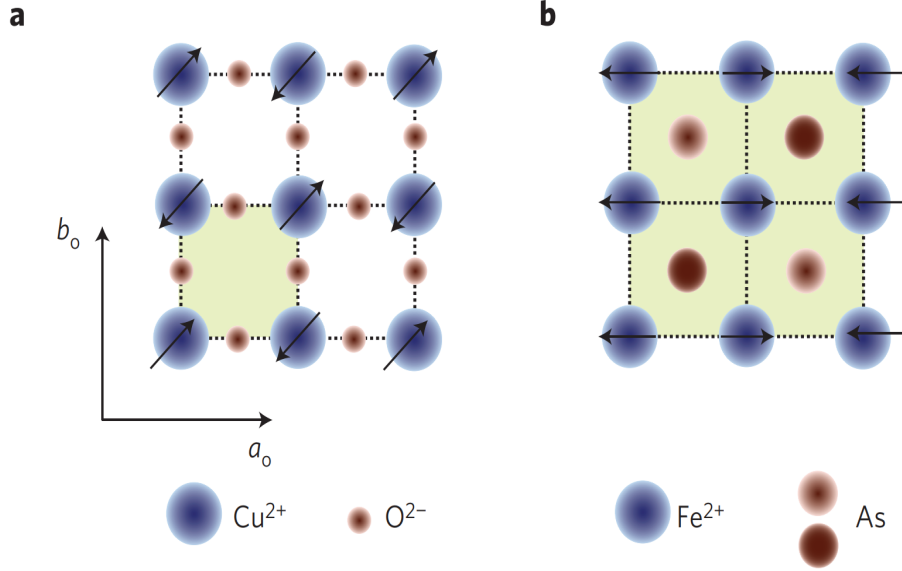


Figure 1.3: Magnetic structure of the (a) copper oxide planes in the cuprates, and (b) iron arsenide planes in the pnictides. In cuprates, the local moments are formed on the copper atom, and they are arranged in the Néel type antiferromagnetic ordering corresponding to a peak at $\mathbf{Q} = (\pi, \pi)$ in the spin structure factor. Similarly, the local moments in pnictides are formed on the iron atom with $\mathbf{Q} = (\pi, 0)$ or $(0, \pi)$ type colinear AFM ordering, i.e., AFM order along one axis and FM order along the other axis. These plots are reproduced from Ref. [27].

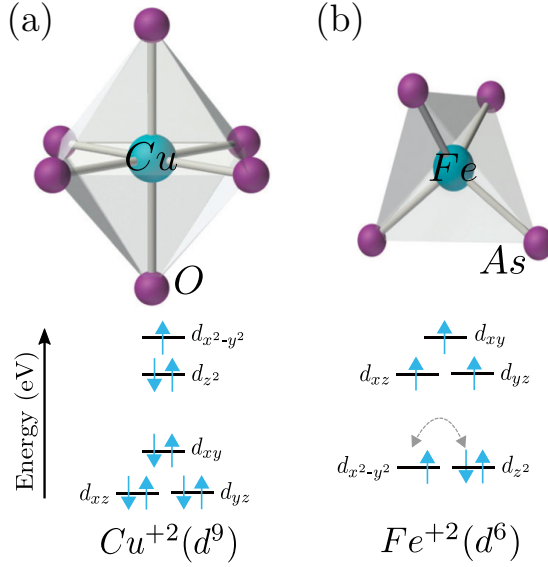


Figure 1.4: (a) Octahedral cage of the Cu atom and (b) tetrahedral cage of the Fe atom in the pnictides with the corresponding atomic energy level arrangement of the five d orbitals. The grey arrow indicates two degenerate arrangement of the down-spin in the lower orbitals. These plots are reproduced from Ref. [96].

leads to 1 valence electron in the $d_{x^2-y^2}$ orbital. Therefore, cuprate parent compounds are often modeled using a half-filled 1-orbital Hubbard Model in two dimensions, in turn explaining the AFM Mott insulator physics of cuprates.

In contrast, first principle calculations of LaFeAsO show that the Fermi surface has contributions from all five $3d$ orbitals of the iron atom [45, 11, 116, 140]. Unlike cuprates, the iron (Fe^{+2}) atoms in the Fe-Scs have 6 valence electrons leading to at least four half-filled orbitals where electrons are arranged, according to the crystal field splitting of the tetrahedral cage around the iron atom and the Hund's rules (Fig. 1.4b). This drastically different electronic structure ultimately leads to transport properties that are different from the cuprates. In fact, this difference in the Fermi surface topology also changes the pairing symmetries of the superconducting state. While it is well agreed by experts that the d -wave pairing is responsible for superconducting transition in cuprates, pairing symmetry is still a subject of debate for pnictides, where $s^\pm$ and d -wave pairing symmetries have been proposed. Multiple active orbitals accompanied by the local crystal fields make the theoretical modeling of pnictides very challenging.

In addition, the tetragonal to orthorhombic transition in the pnictides is accompanied by the nematic phase leading to a $(\pi, 0)$ collinear antiferromagnet upon cooling (Fig. 1.2b), while a decoupled magnetic and structural transition is seen in cuprates with a Néel type AFM at low temperatures (Fig. 1.2a). The nematic order in pnictides is defined by the breaking of the Z_2 rotational symmetry of the lattice, leading to an alignment of spins along an axis without a long-range magnetic ordering. The breaking of the Z_2 symmetry in pnictides also gives rise to the in-plane resistivity anisotropy [18, 37]. These combined degrees of freedom together with a bad-metal parent compound suggests that (unlike cuprates) pnictides are neither weakly nor strongly interacting.

1.2 The role of interactions (correlations)

The AFM phase in the parent compound of cuprates results from the super-exchange mechanism, making it a candidate for strong interactions [21]. However, the metallic nature of the pnictide parent compounds leads to the expectation that the AFM ordering is driven by Fermi surface nesting rather than strong interactions. This claim was strongly supported by angle-resolved photoemission spectroscopy (ARPES) experiments [100], but later it was shown to produce “incomplete” results [33, 139], highlighting the presence of electron correlations. Additionally, pnictides undergo tetragonal to orthorhombic structure transition (T_s), generally before the magnetic transition (T_N). For example, the $T_s = 160$ K while $T_N = 145$ K for *LaFeAsO* [77]. The mechanism for structural transition cannot be explained by simple itinerant electron models (weak interactions/correlations), further emphasizing the importance of electron correlations. The x-ray emission spectroscopy showed the presence of robust local moment per iron atom in all the Fe-Scs at the room temperature [44, 12] (Fig. 1.5). Hence it is important that models with local moments and itinerant electrons should be considered for a model study of pnictides. Comparing these results to a generalized proposed phase diagram of the multi-orbital orbital Hubbard model [27] imply that the model study of Fe-Scs *must use intermediate to strong electron interactions*, required for the formation of local moments at room temperature.

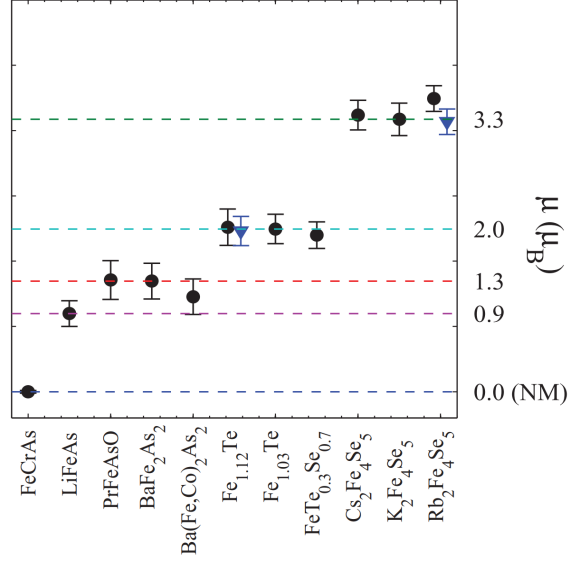


Figure 1.5: The magnitude of local moments at room temperature (circles) and at 15K (triangles) measured for various iron based compounds using the x-ray emission spectroscopy. Pnictide compounds are measured to have approximately $1\mu_B$ per iron while chalcogenides compounds have approximately $3\mu_B$. This plot is reproduced from Ref. [44].

1.3 Quasi-one-dimensional systems

In the context of the cuprates, finding crystal structures simpler than layers but still with intriguing quantum mechanical many-body properties proved to be a fruitful path for progress in that field. One of the reasons is that theorists can perform model Hamiltonian calculations with more accuracy in quasi-one-dimensional systems. In fact, spin-1/2 Cu-oxide two-leg ladders have been much studied in cuprates because of their unusual spin gap, induced by the ladder geometry [26, 25, 22]. Rung spin-singlets dominate the Cu-oxide-ladder spin state, and it was theoretically predicted that such a system should have a tendency to superconductivity upon doping (Fig. 1.6a). This was verified in high pressure experiments at ~ 3 GPa for the case of $\text{Sr}_{0.4}\text{Ca}_{13.6}\text{Cu}_{24}\text{O}_{41.84}$, reporting a critical temperature of 12 K [130]. Due to its quasi-one-dimensional character, it was possible to employ a variety of accurate many-body techniques for ladders, showing agreement between theoretical predictions and experimental results, a result that provided considerable support to the notion that superconductivity in cuprates originates in AFM fluctuations.

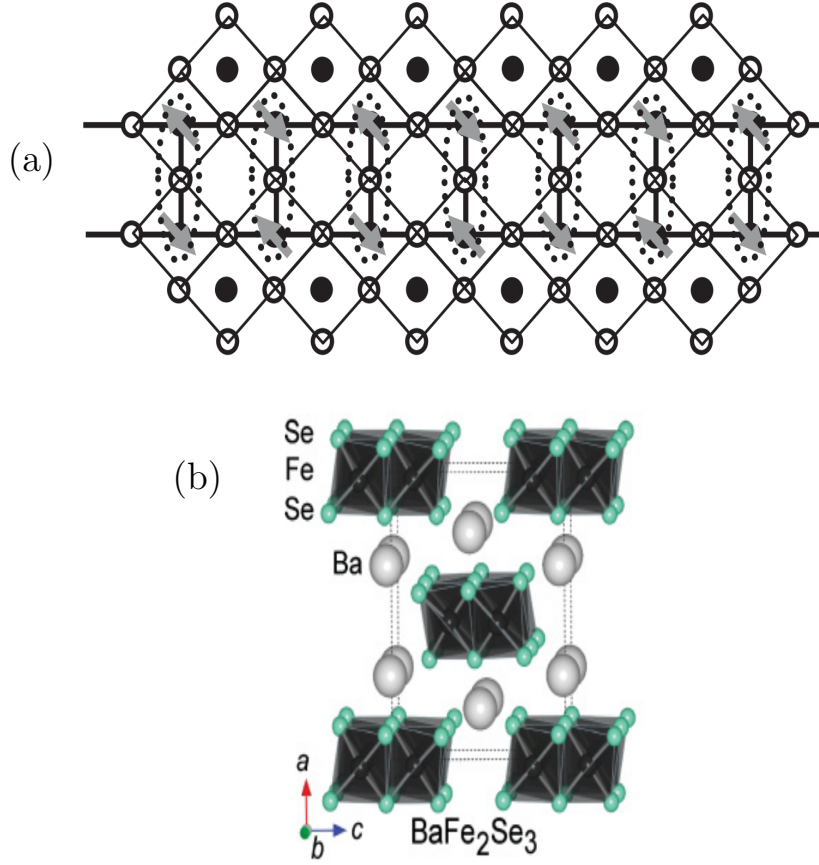


Figure 1.6: (a) Crystal structure of the ladder layer of $\text{Sr}_{0.4}\text{Ca}_{13.6}\text{Cu}_{24}\text{O}_{41.84}$ where the oxygen atom (white filled circles) between the two legs of the ladder provides a bridge for electrons to tunnel. The antiferromagnetic ordering on the ladder is shown by the preformed rung singlets in the undoped compound. (b) Crystal structure of the BaFe_2Se_3 where the legs of the iron ladder are along the 'b' axis, and the ladders are separated by the Barium atoms. These plots are reproduced from Ref. [132] and [16].

These important earlier results in the context of copper-oxide ladders suggest that progress in the understanding of iron-based superconductors would be possible if similar quasi-one-dimensional structures could be prepared and theoretically studied. For this reason, considerable interest was generated by recent studies of BaFe_2Se_3 (Fig. 1.6b) because this material contains double chains made of $[\text{Fe}_2\text{Se}_3]^{2-}$ blocks separated by Ba [16, 58, 106, 85, 15, 30, 67, 69, 78]. The resulting structure contains two extended Fe-Fe directions (the “legs”) connected by Fe-Fe bonds of similar strength (the “rungs”) thus defining two-leg ladders very similar to those in the cuprates. A difference is that an oxygen makes the Cu-Cu bridge in the cuprates’ ladders (Fig. 1.6a), while in chalcogenides the bridges between irons are provided by selenium that is located up and down the middle of the iron plaquettes (see Figures 1.6b and 1.1). Thus, as in their two-dimensional counterparts, electronic hoppings of similar strength are to be expected for the chalcogenides not only along legs and rungs but also along the plaquette diagonals between the iron atoms.

BaFe_2Se_3 is an insulator, with an activation energy between 0.13 eV [85] and 0.178 eV [58], bulk long-range AFM order at ~ 250 K induced by weak residual interladder interactions, and low-temperature magnetic moments $\sim 2.8\mu_B$ [16, 58, 106]. Remarkably, neutron diffraction studies reported a dominant magnetic order at low temperature involving blocks of four iron atoms with their moments aligned, coupled antiferromagnetically along the ladder direction [16, 85]. When K replaces Ba, thus leading to KFe_2Se_3 , the magnetic state changes to an arrangement where the spins in the same rung are coupled ferromagnetically but they are antiferromagnetically ordered in the long ladder direction [15] (similar to the BaFe_2S_3 ladders, Fig. 1.7a). Theoretical studies primarily employing the Hartree Fock approximation [67] unveiled a rich phase diagram for two-leg ladder multiorbital Hubbard models, with a plethora of phases including both states already found in the Ba- and K-based ladders as well as several other competitors. These exotic spin arrangements arise from frustrating tendencies between the staggered AFM state that dominates at small Hund coupling and the ferromagnetic (FM) state stable at large Hund coupling [67]. Hartree-Fock results for layers [65] and chains [66] also suggest a complex landscape of competing magnetic states in those geometries.

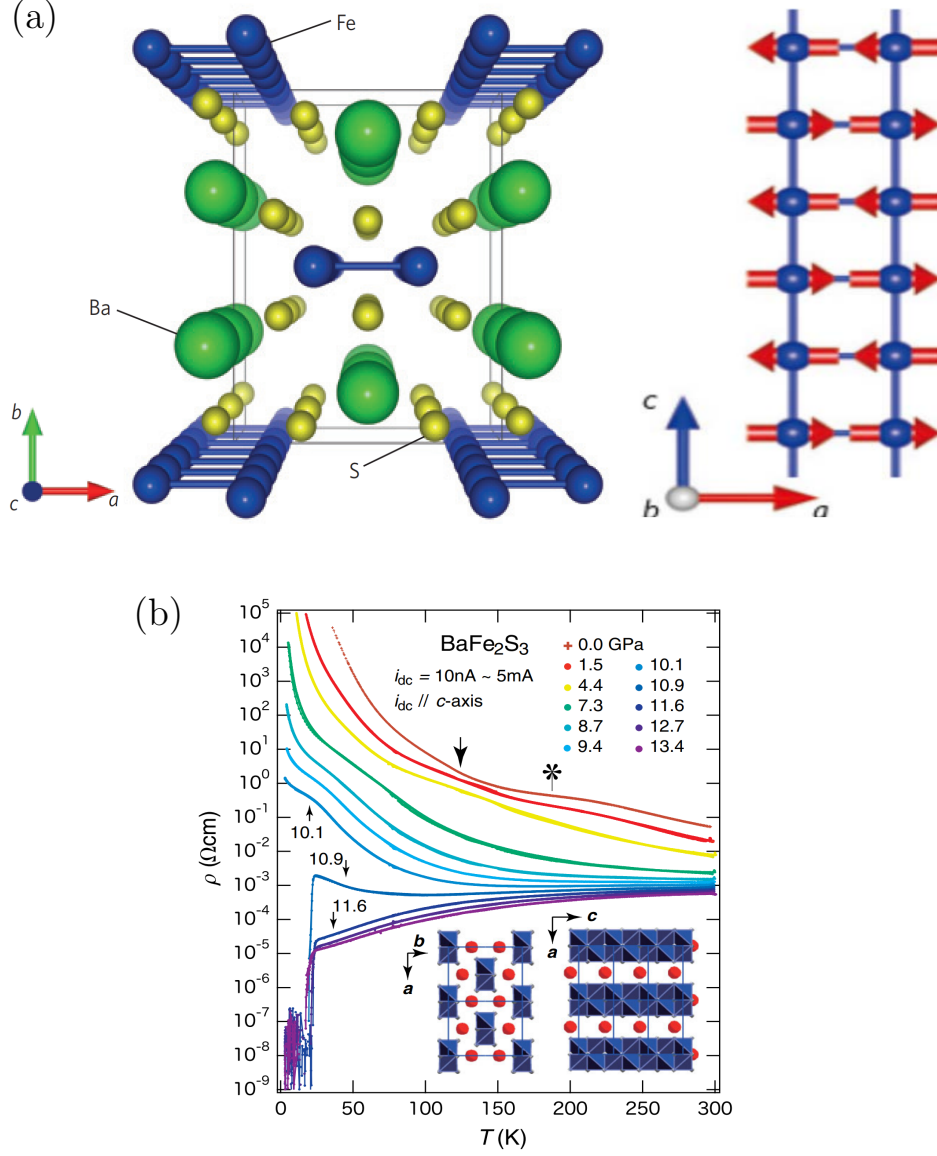


Figure 1.7: (a) Crystal structure of the BaFe_2S_3 forming iron ladders along the 'c' axis, and the $(\pi, 0)$ magnetic ordering where spins are antiferromagnetically aligned in the leg direction of the ladder and ferromagnetically aligned along the rung of the ladder. (b) Resistivity measurements along the leg of the ladder as a function of temperature, for various applied pressures. The sharp down turn in the resistivity at 10.9 GPa show the phase transition to a superconductor. These plots are reproduced from Ref. [122] and [141].

Recently, an unexpected experimental result has been reported using BaFe_2S_3 [122, 141], that has a two-leg ladder structure (Fig. 1.7a). This material was found to become superconducting at a pressure above 10 GPa with an optimal critical temperature $T_c = 24$ K (Fig. 1.7b). The *same material*, but at ambient pressure, is a Mott insulator with the same magnetic order as KFe_2Se_3 namely involving FM rung and AFM leg spin correlations with a critical temperature ~ 120 K, according to power neutron diffraction studies [122] (Fig. 1.7a). *These discoveries unveiled the first iron-based superconductor that does not rely on a square lattice structure of irons*, opening an intriguing avenue of research similar to the one opened with the discovery of superconductivity in Cu oxide ladders in the context of the cuprates.

Theoretical studies in this area of research mainly use model Hamiltonians since there are no *ab initio* techniques that can handle with sufficient accuracy the correlation effects caused by the Coulombic charge repulsion among the electrons. The case of the Hubbard model with only one active orbital ($d_{x^2-y^2}$) has been widely studied in the context of copper-based high-temperature superconductors, and a variety of exciting results and predictions have been unveiled [21, 56, 107]. A significant fraction of those studies, however, arise from approximate analytic many-body techniques that are difficult to control since there is no obvious small parameter to guide expansions when one is dealing with correlated electrons. For this reason, considerable efforts have been devoted to the use of computational techniques to study Hubbard-like models [21]. Alas, the exact computational methods are not without severe limitations as well. For example, the Lanczos method is restricted to small clusters [21] while the density matrix renormalization group (DMRG) is restricted to quasi-one-dimensional systems [133]. An alternative is the determinant quantum Monte Carlo (DQMC) technique [10, 135, 91] that can handle the one-orbital Hubbard model in dimensions larger than one and employ clusters of a reasonable size. However, DQMC presents the infamous “sign problem” which severely restricts its range of applicability. For instance, deviations from the particle-hole symmetric model, such as when electronic hopping beyond the nearest neighbors is introduced, or when doping away from half filling is attempted, severely restricts the temperature range where DQMC can be applied [135, 63].

Considerable theoretical progress has been achieved in this context via the use of mean field approximations of several varieties [120, 50, 23, 47, 115]. This is mainly because of the

need to incorporate several $3d$ iron orbitals in the model Hamiltonian for iron based HTc-Sc. It is well known that the Hubbard model for pnictides must have a minimum of two iron orbitals: d_{xz} and d_{yz} , while most experts agree that at least a third orbital d_{xy} should also be incorporated [20]. Moreover, the crystal structure indicates that hopping amplitudes must involve both Fe-Fe nearest and next-nearest neighbors processes (Fig. 1.1). All these factors are detrimental to the performance of Lanczos, DMRG, and DQMC techniques, and the applications of these methods have been limited in the context of the iron-based superconductors. In fact, in a recent review [27], a crude drawing of the phase diagram of a multi-orbital Hubbard model was sketched by hand based on physical expectations, but this prediction has yet to be confirmed due to the lack of reliable techniques for the calculations.

This thesis addresses the numerical study of the iron-based superconductors using the multi-orbital Hubbard model employing the Monte-Carlo mean-field method and the density matrix renormalization group. We shall first introduce benchmarking results of the newly developed method that mixes classical Monte Carlo and mean field approximations (MCMF) on a 1-orbital Hubbard Model in chapter 2. Then, we apply MCMF to a more realistic 2-orbital model for iron-based oxypnictides superconductors to study its magnetic and transport properties in the parent compound, shown in chapter 3. Additionally, we study the magnetic and superconducting properties of BaFe_2S_3 ladder using a combination of *ab initio* and density matrix renormalization group (DMRG) using a realistic 2-orbital Hubbard model, shown in chapter 4. Finally, in an attempt to understand the pairing mechanism of the BaFe_2S_3 ladder, we study the pairing mechanism in a two-orbital Hubbard model on a chain (chapter 5).

Chapter 2

One Orbital Monte Carlo-Mean Field (MCMF)

This chapter is a modified version of Physical Review B **90**, 205133 (2014) [82].

2.1 Introduction

The essence of the computational method described here is to set up the mean field equations for the problem at hand, and then raise the mean field parameters, such as the effective staggered magnetic field that appears for an antiferromagnetic (AFM) state, which is then treated via Monte Carlo simulations at finite temperatures. These classical variables play the role of the spin in the resulting spin-fermion-like model. For the fermions, the resulting Hamiltonian is quadratic and can be solved numerically via library subroutines or other procedures. This methodology was proposed in a study of the competition between AFM and superconducting (SC) tendencies in the one-orbital Hubbard model [76]. In this earlier work, both the staggered AFM field and a complex field representing the SC order parameter deduced from the Bogoliubov de Gennes (BdG) equations were introduced and handled via Monte Carlo simulations.

As we will show, an interesting result is that this computational procedure captures the highly nontrivial nonmonotonic behavior of the Néel temperature (T_N) with increasing Hubbard U at half filling, in excellent agreement with DQMC. This is a dramatic

improvement over standard Hartree-Fock mean field techniques that incorrectly predict a smooth increase of T_N with U . This up and down behavior of T_N with U was also observed recently in a similar study of the negative U Hubbard model [125] (note that there is a mapping between positive and negative U), and in early studies of models for d-wave superconductivity with increasing pairing attraction [76, 2, 75]. In addition, many other observables calculated within this approach, such as the specific heat, are in qualitative, and often quantitative, agreement with DQMC, as shown below. Moreover, the spin-fermion model also allows for the calculation of dynamical observables directly in real time and frequency. We demonstrate this here by calculating the single particle density of states. Finally, we further examine the utility of this approach by examining the Hubbard model with longer range hopping. In this case, DQMC cannot be applied due to a severe fermion sign problem [21] but our method still works. In summary, the simple combination of Monte Carlo and mean field methods allows for a proper treatment of real space fluctuations as well as the temperature effects in Hubbard models, including the study of regimes where the relevant correlations, such as the spin correlations, are of short range in space. Although it will be computationally demanding, after the success of the test presented here, the method will be ready to be implemented for multi orbital Hubbard models of relevance in, *e.g.*, iron superconductors, where not much is known about their thermodynamic behavior.

This chapter is organized as follows. The Hubbard model and the proposed technique are discussed in Appendix A. The technique is formally introduced by using the Hubbard Stratonovich variables employed in Ref. [126]. The main results are presented in Sec. 2.2, starting with the case of three dimensions and its comparison with DQMC. This is followed by a presentation of results for the two-dimensional case in Sec. 2.4, as well as results for a Hubbard model with hopping beyond nearest neighbors where DQMC suffers from a severe sign problem in Sec. 2.5. Finally, at the end of this chapter we conclude with a brief summary.

2.2 Three-dimensional lattice

Lets start by introducing the $T/t - U/t$ phase diagram at half filling in three dimensions. In figure 2.1, we show T_N as a function of U/t where the plotted results are from MCMF (filled

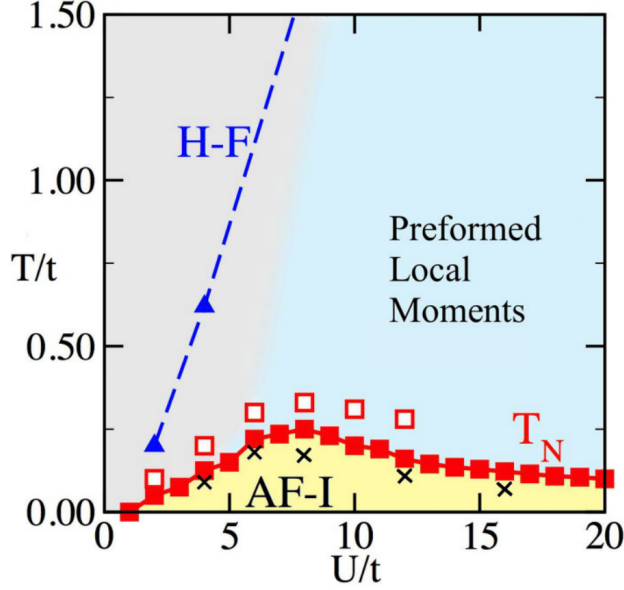


Figure 2.1: The T/t - U/t phase diagram for the one-band Hubbard model. The solid red squares show the dependence of T_N on U/t obtained using the MCMF technique on 4^3 clusters. The crosses are estimations of T_N obtained from finite-size scaling. The AF-I region denotes the Néel type AFM phase with long-range order and insulating characteristics. The open squares are the T_N obtained from the DQMC method, from Ref. [118]. The light blue region depicts the regime of preformed local moments above the AF-I phase. The dashed line shows the T_N obtained from the simplistic Hartree-Fock calculation at finite temperature where the critical temperature incorrectly grows linearly with U/t at large U/t . The determination of the crossover between the gray and blue regions, and the fact that the MCMF local moment region coincides with HF T_N at temperatures much larger than typical T_N scales, are discussed in the text.

squares), DQMC(open squares) [118], Hartree-Fock (blue line) and finite-size scaling (black X). The most important result of our technique is that it can capture the non-monotonic behavior of the T_N with increasing U . This result can not be obtained by a simple Hartree-Fock mean field theory. Hartree-Fock predicts T_N grows linearly as a function of U . Unlike Hartree-Fock mean field theory, MCMF includes the thermal fluctuations and therefore it correctly predicts the presence of a low energy scale that regulates the Néel temperature at large U/t , where the Hubbard model effectively becomes the Heisenberg model governed by super-exchange $J \sim \frac{t^2}{U}$. The comparison with DQMC also provides additional evidence that our technique is not only qualitatively correct, but it provides reasonably quantitative values of T_N . MCMF finite size scaling results for T_N are consistently underestimated. Nevertheless, for the purpose of testing the method and its potential application to the multiorbital fermionic models, this degree of accuracy is quite acceptable.

Another important feature missing in the standard finite-temperature HF approach is the presence of the local moments above T_N . In Fig. 2.1, the blue shaded region shows the formation of the local moments at high temperatures. In the HF approach, the moment ordering and the formation occurs at same temperature upon cooling, which is in disagreement with the results of DQMC [92].

(i) *Local moments and magnetic order*: Typical structure factors $S(\mathbf{q})$ for $\mathbf{q} = (\pi, \pi, \pi)$ are shown in Fig. 2.2. Here, we observe that T_N increases as a function of U for $U/t < 8$, but decreases for $U/t > 8$. This data was used to produce filled squares in figure 2.1, which shows the non-monotonic behavior of T_N as a function of U/t . We also plot the moment formation vs. temperature (Fig. 2.2). Here, the system-averaged local moment is defined as $M = \langle (n_\uparrow - n_\downarrow)^2 \rangle = \langle n \rangle - 2\langle n_\uparrow n_\downarrow \rangle$ with $\langle n \rangle = \langle n_\uparrow + n_\downarrow \rangle$. Since $2S_z = n_\uparrow - n_\downarrow$ and we have a rotationally invariant system, then $M = 4\langle S_z^2 \rangle = 4\langle (\mathbf{S} \cdot \hat{\Omega})^2 \rangle$, where $\hat{\Omega}$ is an arbitrary unit vector.

Note that for a half-filled uncorrelated case ($\langle n \rangle = 1, U/t = 0.0$) the double occupancy $\langle n_\uparrow n_\downarrow \rangle$ can be decomposed into $\langle n_\uparrow \rangle \langle n_\downarrow \rangle = 1/4$. Since the large temperature limit is equivalent to an uncorrelated system limit, we can observe that the double occupancy is approximately 1/4 (Fig. 2.2c), making the magnetic moment reach its uncorrelated value of 1/2 (Fig. 2.2b). On the other hand, for large U/t and very low T/t , the double occupancy

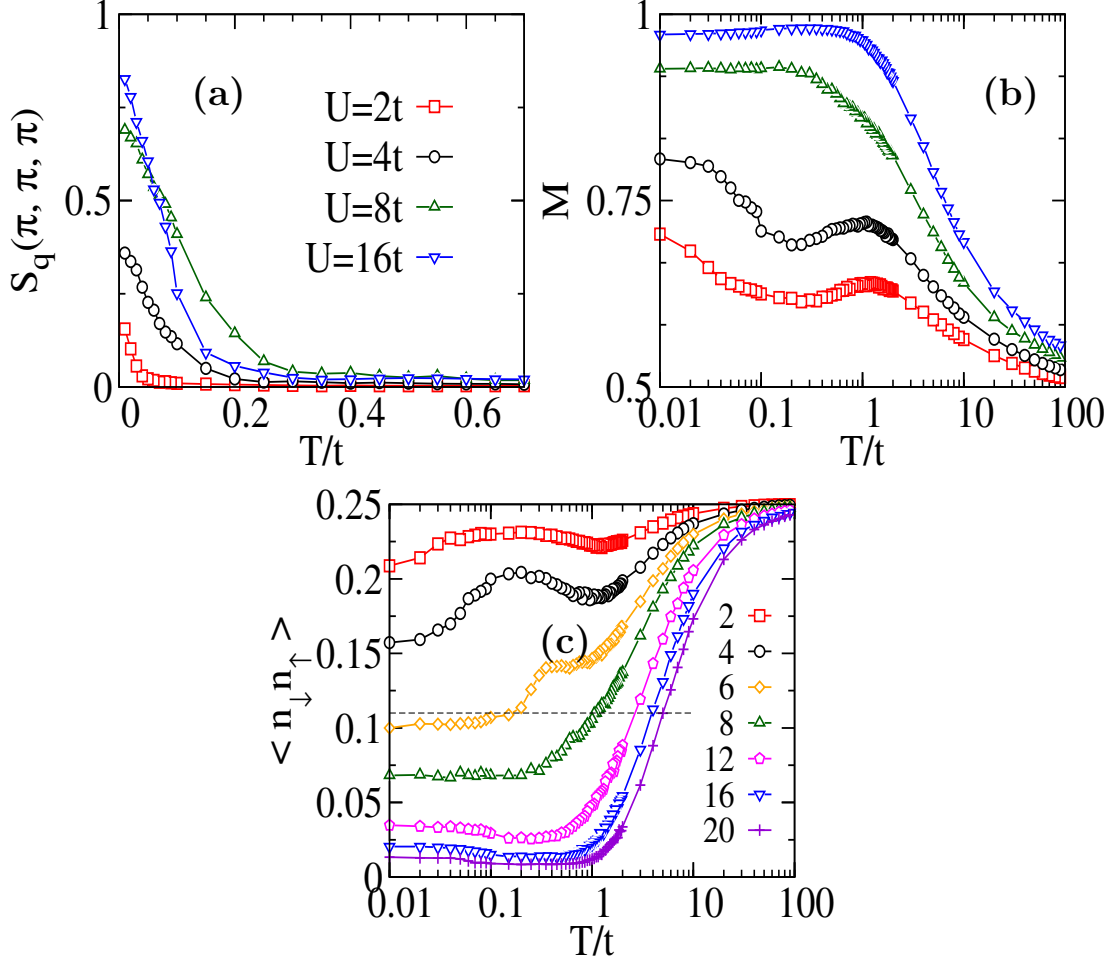


Figure 2.2: (a) The magnetic structure factor $S(\mathbf{q})$ for $\mathbf{q} = (\pi, \pi, \pi)$, obtained at various U/t s as indicated. The data are from 4^3 clusters with 4000 MC sweeps at every temperature, while cooling the system down from high to low temperatures under Monte Carlo. (b) The corresponding local moments M vs. temperature. We capture the feature that at large U/t the peak in the moment size shifts to nonzero temperature. This effect, due to the setting of long-range order, was reported before in the DQMC studies of Ref. [108]. At small U/t the overall shape is also in good agreement with the DQMC data, indicating that the MCMF method indeed captures the essence of the problem. Panel (c) shows the expectation value of double occupation for the various U/t 's indicated. The thin dashed line indicates a cutoff discussed in the text.

is much suppressed and $M \sim 1$, *i.e.*, the $U/t = \infty$ result. It is also important to notice that the increase in the auxiliary field magnitude at high temperature, due to large thermal fluctuations, does not imply an increasing magnetic moment. As seen in Fig. 2.2b, the magnetic moment M saturates at its uncorrelated value of $1/2$ at high temperature, while the magnitude of the auxiliary moment increases as a function of temperature for large temperatures, as discussed later.

We notice that $M(T)$ has some features at intermediate temperatures that evolve with U/t . In the intermediate temperature range, we see two kinds of behavior: while M has a minima at $U/t = 4.0$, M has maxima at finite $T/t \sim 0.1$ and ~ 0.5 for $U/t = 8, 16$, respectively. It is known that at large value of U/t the system can be approximated by a spin-1/2 Heisenberg antiferromagnet. At small but finite temperature, the AFM order is perturbed by excitations, this perturbed state suppress the virtual exchange due to the Pauli exclusion principle, which can be seen in the minima of double occupancy at large values of U/t (fig. 2.2c). This increases the degree of localization and promotes larger on-site moment size thereby pushing the maxima of M to finite temperature. These two features were also reported before in two-dimensional DQMC studies [92], increasing the evidence that MCMF captures the essence of the problem.

(ii) *Specific heat*: The temperature evolution of the local moment in Fig. 2.2b shows a continuous increase with decreasing temperature up to $T/t = 1$. But this does not provide clear information on the crossover location between regimes with and without local moments. To address this issue, and to further test the MCMF method, we calculated the specific heat C_v vs. temperature for different values of U/t . Here, it is expected that C_v vs. temperature should have a two peak structure [108, 92], the peak at high temperature corresponding to moment formation and the peak at low temperature corresponding to moment ordering at large U/t .

In Fig. 2.3a, the specific heat vs. temperature is shown for a 4^3 system, where we find the expected two peak structure. The locus of the low-temperature peak corresponds well with the T_N shown in Fig. 2.1. The high-temperature peak positions vs. U/t are in Fig. 2.3b. Here we also show the HF data with open squares. Clearly, beyond $U/t = 10$ the MCMF result coincides with the HF result. At lower values of U (below $4t$), the high-temperature peak

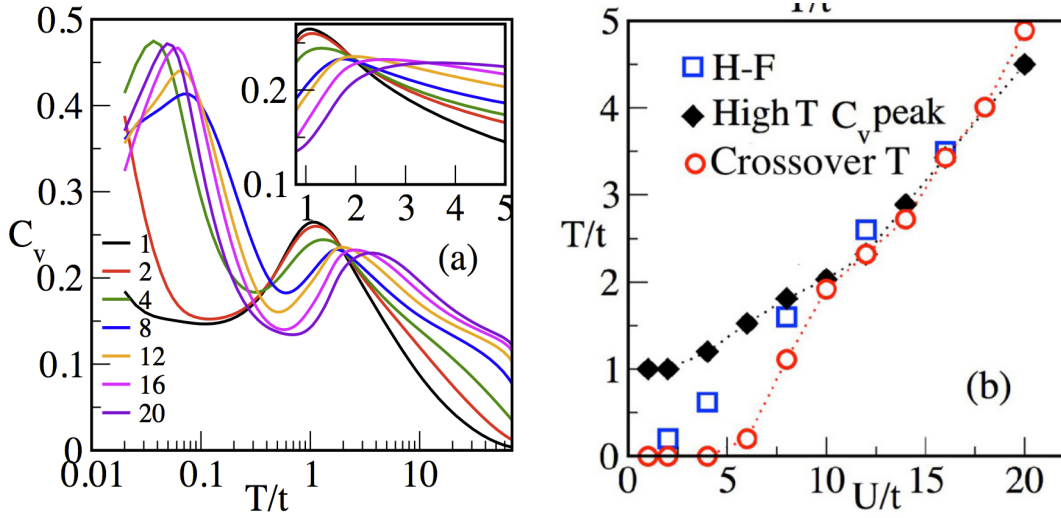


Figure 2.3: (a) Specific heat vs. temperatures for different U values. Two peak structures are observed: at large U/t the high-temperature peak corresponds to the moment formation while the lower one corresponds to moment ordering. The inset shows the universal crossing of the different C_v curves at $T/t \sim 2.0$. Panel (b) shows the position of the high-temperature peak varying U . The Hartree-Fock results are shown with open squares. At large U , beyond $6t$, these peak positions are close to the mean field results. At low U , the high-temperature peak position saturates to $1t$, while the low- T peak approaches zero. The nonmerging of the two peaks in three dimensions is in agreement with DQMC data in two dimensions. The low-temperature peak corresponds to T_N in Fig. 2.1. The open circles in (b) show the crossover temperature from the no local moments regime to a region of preformed moments as obtained from the data on double occupation shown in Fig. 2.2c. In (a) the data shown are a smoothed version of the actual data to reduce statistical fluctuations.

appears to saturate to $T/t = 1$. On the other hand, the low-temperature peak is suppressed to zero with low T/t . We note that we were unable to reliably carry out the numerical derivative below $T/t = 0.02$, but the trend of the low-temperature peak shifting towards zero is apparent here and is also in Fig. 2.1 (solid, red squares). Thus, in the present study we report that the high- and low-temperature peaks do not merge with reducing temperature at small U/t in three dimensions. (This is also the case in two dimensions, which is discussed later.) Previous studies have not agreed on this issue: dynamical mean field theory (DMFT) [42, 17, 131] and Lanczos on one-dimensional chains [113] find the two peaks merging together with reducing U while a DQMC study in two dimensions [92] agrees with our conclusions. Here we have extended the results to three dimensions.

Another feature arising from the independence of the high-temperature entropy is a universal [31, 92] crossing in C_v . In two-dimensional DQMC, this occurs at $T/t \sim 1.6$, with a spread in temperature of $\sim 0.2t$. This has been observed in DMFT [42, 17, 131] as well. We find a similar crossing both in three and two dimensions. In three dimensions the crossing is at approximately $T/t = 2.0$ and has a small spread for low U values, while at larger U there seems to be a systematic increase to higher temperature with increasing U . This last conclusion was reported earlier as well [31]. For our main purpose of testing the MCMF method, in two dimensions once again our results agree well with DQMC data, as discussed later.

(iii) *Crossover temperatures:* At large U , the high temperature peak of C_v corresponds to the moment formation. Below $U/t = 10$, however, this high-temperature peak deviates from the linear behavior seen in Fig. 2.3b and eventually saturates to $T/t = 1$. The approach of the peak location to $T \sim t$ at small U indicates that considerable contribution to this peak comes from electron delocalization. For this reason, for $U/t < 10$, the high-temperature peak cannot be used as a reliable indicator of local moments. Thus, we use the double occupation as plotted in Fig. 2.2c, as an alternative indicator. To do so, we need to choose a cutoff because the local moment formation is not abrupt but occurs with continuity. This cutoff is shown in Fig. 2.2c, with a horizontal dashed line. For a given U , the temperature where double occupation goes below the cutoff is taken to be the crossover temperature to a region with preformed local moments. In principle, the choice of such a

cutoff is arbitrary, however, the C_v calculations serve as a guide. To address this issue, we chose a cutoff value such that the location of the high-temperature peak in temperature and the crossover temperature from the cutoff coincide at large U . The crossover temperature for all other U values are obtained from this fixed cutoff, which is plotted in Fig. 2.3b with open circles. In the same figure, we also show that the crossover temperature is in good agreement with the high temperature C_v peak. The crossover temperature for $U/t = 6$ is very close to the corresponding T_N in Fig. 2.1. For lower U values, large double occupation considerably suppresses the local moment formation.

(iv) *Density of states:* In the half-filled Hubbard model, the charge gap is directly related to the existence of the local moments regardless of magnetic order. This charge gap manifests as a gap (zero spectral weight in a finite energy range) in the DOS at $T = 0$. With increasing temperature, this hard gap softens and is replaced by a pseudogap, with the spectral weight in the gap gradually increasing with increasing temperature. At large U/t this monotonic behavior is seen in Fig 2.4a from our MCMF results. The DOS is displayed up to $T = 0.55t$, but the monotonicity persists to higher temperatures. In contrast, at $U/t = 4$, shown in Fig. 2.4b, the pseudogap spectral weight has a nonmonotonic behavior: for $T > 0.17t$ the spectral weight at $\omega - \mu = 0$ decreases with increasing temperature, while for $T < 0.17t$ the spectral weight decreases with decreasing temperature. Since the spectral weight at $N(\omega = 0)$ results from the scattering of the electrons from the classical field, in Fig. 2.4c we show the evolution of the corresponding system-averaged auxiliary field values, $\langle |\mathbf{m}| \rangle$. For $U/t = 2$ and 4, $\langle |\mathbf{m}| \rangle$ has minima at $T/t = 0.05$ and 0.1, respectively. For $U/t = 8$ and 14, $\langle |\mathbf{m}| \rangle$ for $T/t \lesssim 0.5$.

The behavior shown in Fig. 2.4 can be explained as follows. At high enough temperatures with negligible local moments, the value of $\langle |\mathbf{m}| \rangle$ is governed by thermal fluctuations. At these temperatures the auxiliary fields behave as harmonic oscillators with a mean amplitude proportional to $\sqrt{T/U}$. Thus, $\langle |\mathbf{m}| \rangle$ grows with increasing temperature. On the other hand, the $T = 0$ equilibrium value of $\langle |\mathbf{m}| \rangle$ is directly proportional to U , as seen in Fig. 2.4c. For smaller values of U , thermal fluctuations dominate to low enough temperatures, causing $\langle |\mathbf{m}| \rangle$ to reduce to values smaller than their $T = 0$ value. On further reduction in temperature, these thermal fluctuations are suppressed and $\langle |\mathbf{m}| \rangle$ starts to increase towards its mean

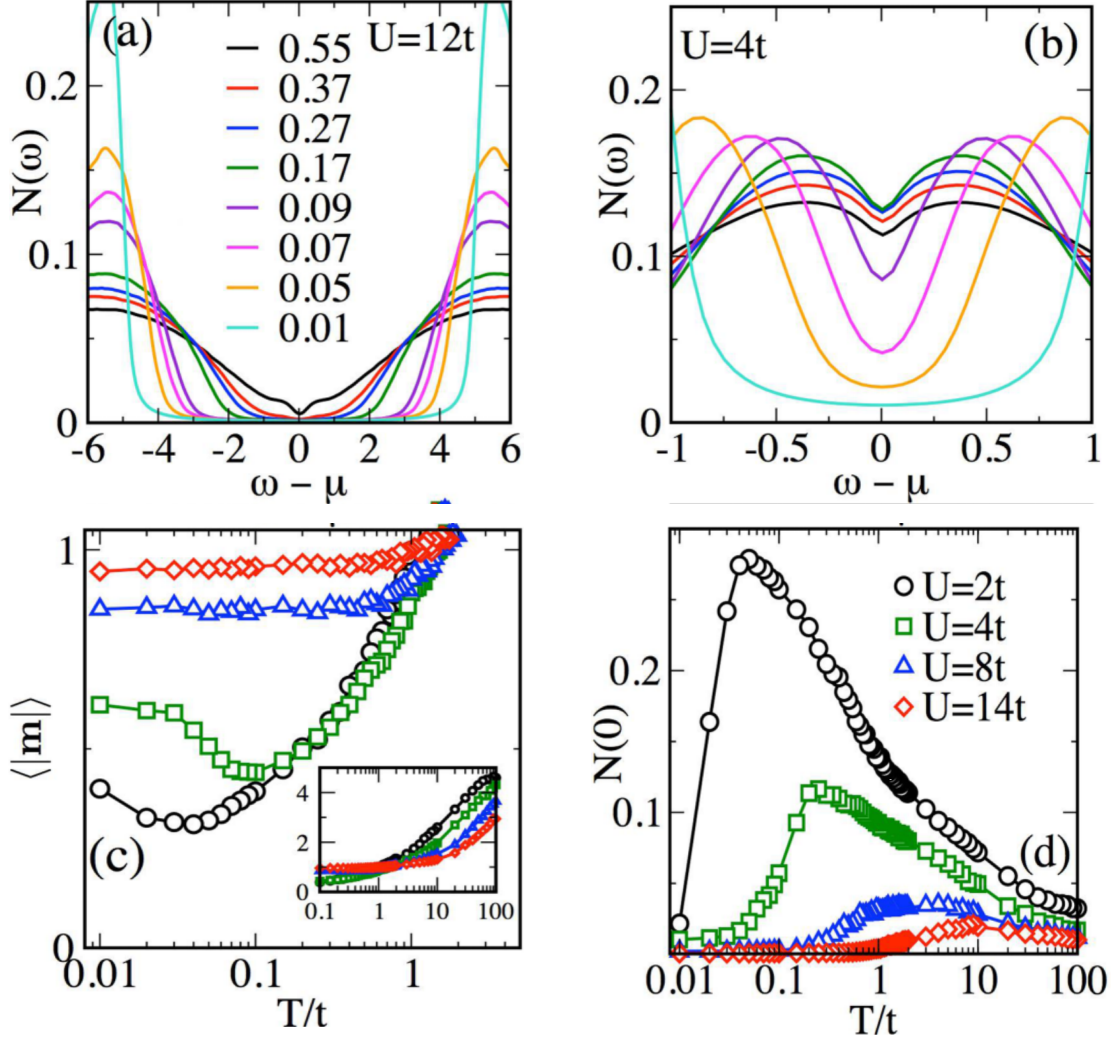


Figure 2.4: Density of states $N(\omega)$ for (a) $U/t = 12$ and (b) $U/t = 4$ at the temperatures indicated in panel (a). At large $U/t = 12$, the Hubbard gap is gradually filled up due to thermal fluctuations. The weight at $\omega - \mu = 0$ monotonically increases with increasing temperature. A DOS pseudogap is seen above $T_N \sim 0.2t$. At a smaller coupling $U/t (= 4)$, while the gap is filled similarly to the large U case with the increase of temperature, above T_N we observe a nonmonotonicity in the dependence of the zero-energy weight with temperature. (c) Magnitude of the auxiliary classical fields averaged over the lattice $\langle |\mathbf{m}| \rangle$ vs temperature for the U values in (d). At large temperature the thermal fluctuations cause ($\langle |\mathbf{m}| \rangle$) to grow linearly with temperature for all the U 's shown (inset). The reason for this temperature dependence of $\langle |\mathbf{m}| \rangle$ and its correlation with $N(\omega = 0)$ is discussed in the text. (d) Shows the $N(\omega = 0)$ feature remarked in panel (b) for different U values. This nonmonotonicity was reported before in a DQMC study; see Ref. [108].

value at $T = 0$. The minima in $\langle |\mathbf{m}| \rangle$ vs. temperature corresponds to the location of the maxima in $N(0)$ in Fig. 2.4d for $U/t = 2$ and 4. This indicates that the $N(0)$ suppression at high temperature results from the scattering of electrons from thermally fluctuating large $\{\mathbf{m}_i\}$ fields, while at small temperatures, the reduction in $N(0)$ results from the depletion of spectral weight due to the opening of the Mott gap. The peak in the $N(0)$ occurs between the two regimes.

With increasing U , the dominance of thermal fluctuations in governing $\langle |\mathbf{m}| \rangle$ is pushed to progressively higher temperatures as is also seen from the peaks of $N(0)$ for $U/t = 8$ and 16 in Fig. 2.4d. At these temperatures $\langle |\mathbf{m}| \rangle$ is higher than their $T = 0$ values, thus no minima are found for these cases in Fig. 2.4c. We stress that the high-temperature increase in the auxiliary field magnitude does not imply an increasing magnetic moment. As seen in Fig. 2.2b the magnetic moment M saturates at its uncorrelated value of $1/2$ at high temperature. The nontrivial effect of the fluctuations in the auxiliary fields is in the DOS, in the low-temperature feature in M at small U , and possibly in the conductivity. Another feature observed in the inset of Fig. 2.4c is that the magnitude of the auxiliary fields vs. temperature for different U 's cross between $T/t = 1$ and $T/t = 2$. Since at large T , $\langle |\mathbf{m}| \rangle$ grows as $\sqrt{T/U}$, the auxiliary fields magnitude for smaller U grows more rapidly than those for larger U . At small temperatures, however, the $\langle |\mathbf{m}| \rangle$ values are directly proportional to U as discussed before, naturally explaining the observed crossing. Note that this crossing coincides with the universal crossing of the specific heat in Fig. 2.3b.

(v) *Real-space spin correlation.* Figure 2.5a shows the spin-spin correlation $C(|r|)$ at $T/t = 0.4 > T_N/t$ for different values of $|r|$. The special case $|r| = 0$ corresponds to M and with increasing U/t , $C(|r| = 0)$ saturates. The most prominent real-space AFM correlation at this temperature is for $C(|r| = 1)$. While it is almost zero for $U/t \leq 4$, it increases as a function of U/t , reaches a maximum at $U \sim 8t$, and then reduces with further increases in U/t . The large U/t suppression is due to the t^2/U suppression of the spin ordering stiffness. While a similar trend is seen for larger $|r|$, the magnitude of the correlation is greatly suppressed. In Fig. 2.5b we show the evolution of $C(|r|)$ with temperature at a typical large value of U/t . While the magnitude of the moment, given by $C(|r| = 0)$, increases slightly with temperature, the short-range correlations are suppressed rapidly beyond T_N .

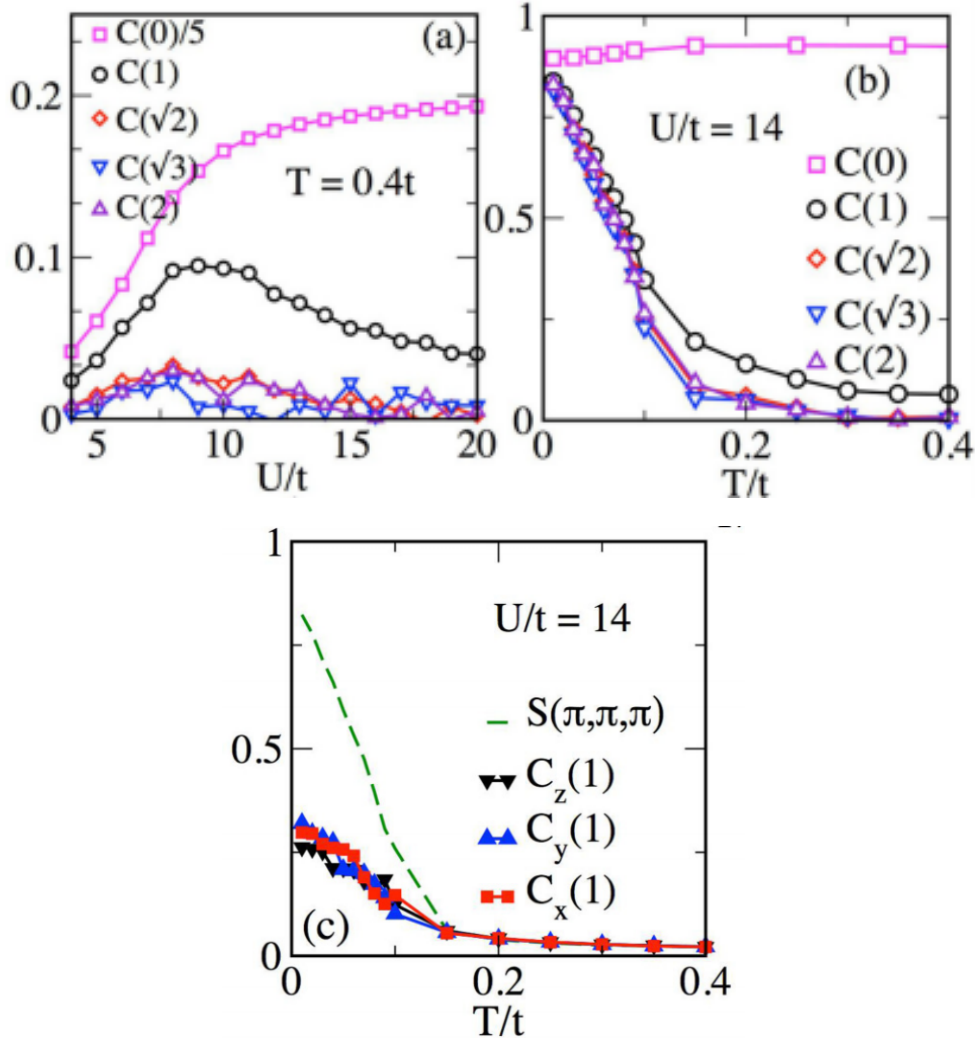


Figure 2.5: (a) Real-space spin-spin correlations $C(|r|)$, for $|r| = 0, 1, 2, 3, 2$, at $T/t = 0.4$, *i.e.*, a temperature above T_N . See the text for the definition of $C(|r|)$. The $|r| = 0$ curve corresponds to the square of the local moment and shows that the size of the preformed magnetic moment increases with U/t and saturates beyond $U/t \sim 8$, *i.e.*, where T_N is maximized. The rest of the curves show the real-space AFM correlations, $q = (\pi, \pi, \pi)$, among the moments. Again the correlations are the largest for $U/t \sim 8$. On the large U side the decrease in the correlation results from thermal fluctuations competing with the AFM spin order stiffness which scales as t^2/U . (b) Shows the dependence of $C(|r|)$ on temperature for large $U/t (= 14)$. The magnitude of the moment is almost independent of the temperature, while there is a clear short-range AFM correlation between the moments at all temperatures shown. Longer range correlations for $|r| > 1$ are suppressed rapidly above $T_N \sim 0.12t$. (c) The real-space correlations $C(|r| = 1)$ using only the x , y , or z components of the spin. The data confirm explicitly the rotational invariance expected to exist in the mean field Hamiltonian. The AFM structure factor is also displayed for comparison. Results are similar for $|r| > 1$ as well.

The increase in $C(|r = 0|)$ or the size of the local moment were discussed earlier. Only $C(|r = 1|)$ is robust above T_N . Finally in Fig. 2.5c we also show individually the x , y , and z components of $C(|r|)$ for $|r| = 1$ for $U = 14t$. This confirms explicitly the rotational invariance of the calculation.

Summarizing this subsection, we have established that the nonmonotonic dependence of T_N on U , the physics of preformed local moments, and the pseudogap features in the DOS can all be captured within the MCMF method

2.3 Traveling Cluster Approximation

ED+MC is numerically expensive since an exact diagonalization must be performed at every step in the process. The numerical cost of a sequential sweep scales as $\mathcal{O}(N^3)N$, with N the total number of lattice sites. To overcome this $\mathcal{O}(N^4)$ scaling we employ a recently developed variation of real-space ED+MC that scales linearly with the system size [55]. This technique, called traveling cluster approximation (TCA), defines a region (the traveling cluster) around the site where a MC update is attempted. A change is proposed and the update is accepted or rejected based on the energy change computed within the traveling cluster, thus bypassing the costly diagonalization of the full system. Only when observables are calculated, after equilibrium has been reached, is a full system diagonalization performed. This adds only a few hundred full system diagonalizations to the computational cost. For TCA, the computation cost of ED for a system with N sites is $\mathcal{O}(N_c^3)$, where N_c is the traveling cluster size where the traveling cluster is defined using periodic boundary conditions. The cost of a full sweep of the lattice is $N_c^3 \times N$ or linear in N as opposed to N^4 . This allows us to solve much larger systems. We now discuss our benchmarks for the TCA and the results on large two- and three-dimensional lattices.

(1) *Benchmarking.* Let us begin by comparing the results from TCA with ED+MC. In Fig. 2.6 we compare various observables on two-dimensional 8^2 clusters with periodic boundary conditions. Results are shown for two different traveling cluster sizes, namely $N_c = 4^2$ (squares) and $N_c = 8^2$ (triangles), while results for the full ED+MC are given as the solid lines. Figure 2.6a shows $S(q)$ for $\mathbf{q} = (\pi, \pi)$, Figs. 2.6b and 2.6c show the DOS,

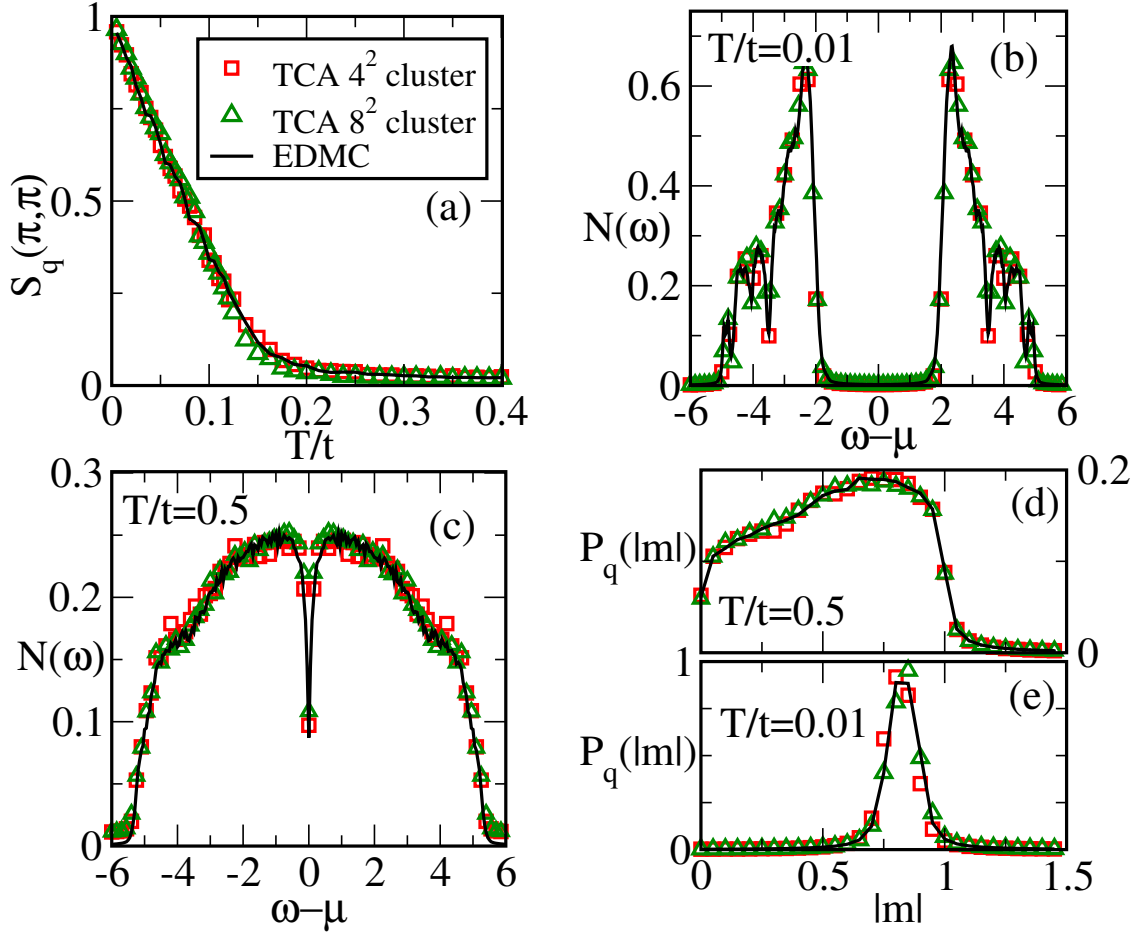


Figure 2.6: Comparison of results for an 8^2 system at $U/t = 6$, obtained using ED+MC (solid line) and TCA with two different sizes of traveling cluster sizes, 4^2 (squares) and 8^2 (triangles). Panel (a) shows $S(\pi, \pi)$, (b) and (c) show the DOS, while (d) and (e) display the $P_q(|\mathbf{m}|)$, for the three cases. The DOS and $P_q(|\mathbf{m}|)$ are shown at low $T(= 0.01t)$ and high $T(= 0.5t)$ temperatures, as indicated.

and Figs. 2.6d and 2.6e show $P_q(|\mathbf{m}|)$ at low ($T = 0.01t$) and high ($T = 0.5t$) temperatures. All of the results are for $U = 6t$. The T_N obtained is about $0.15t$ from both methods. The Mott gap is found to be about $4t$ at low temperature [see 2.6b].

The pseudogap feature at $T/t = 0.5$ in Fig. 2.6c is also captured very accurately within TCA. Finally, all sites at low temperature show $|\mathbf{m}_i| \sim 1$, which evolves into a broad distribution at high temperatures. We find a satisfactory agreement between ED+MC and TCA data for all the observables and at all temperatures. Furthermore, these results show that employing a 4^2 traveling cluster is adequate as the results are virtually indistinguishable from those obtained using a 8^2 traveling cluster. In the following we will employ 4^2 and 4^3 traveling clusters in two and three dimensions, respectively.

(2) *Finite-size scaling.* With TCA-based MCMF now we can study up to 16^3 lattices. As a result the $S(\pi, \pi, \pi)$ data are available for $N = 4^3$ to 16^3 system sizes. Moreover, the magnetic structure factor obtained with the TCA agrees with the ED+MC data at all temperatures. This indicates that finite-size effects associated with the TCA do not affect the finite temperature evolution of the magnetic state. Hence we can employ a finite-size scaling analysis to the TCA data in order to obtain the Néel temperature in the thermodynamic limit.

On a finite system, estimates of T_N can be obtained either from an inspection of the $S(\pi, \pi, \pi)$ data or from the maxima of thermodynamic quantities such as the specific heat or the magnetic susceptibility. Then, assuming that the correlation length $\xi(T_N(L) - T_N^{Thermo}) = aL$ on a L^3 system, and given that $\xi(x) \propto |x|^\nu$, one arrives at the scaling form, $T_N(L) = T_N^{Thermo} + bL^{1/\nu}$. Here, L denotes data from a L^3 cluster size. We plot the finite cluster Néel temperatures against $1/L$ and use T_N^{Thermo} , b , and ν as fit parameters. A typical data fit is presented in Fig. 2.7b. For reference, we provide the $S(\pi, \pi, \pi)$ data for different system sizes in Fig. 2.7a. The crosses in Fig. 2.1 are T_N^{Thermo} obtained from this finite-size scaling analysis.

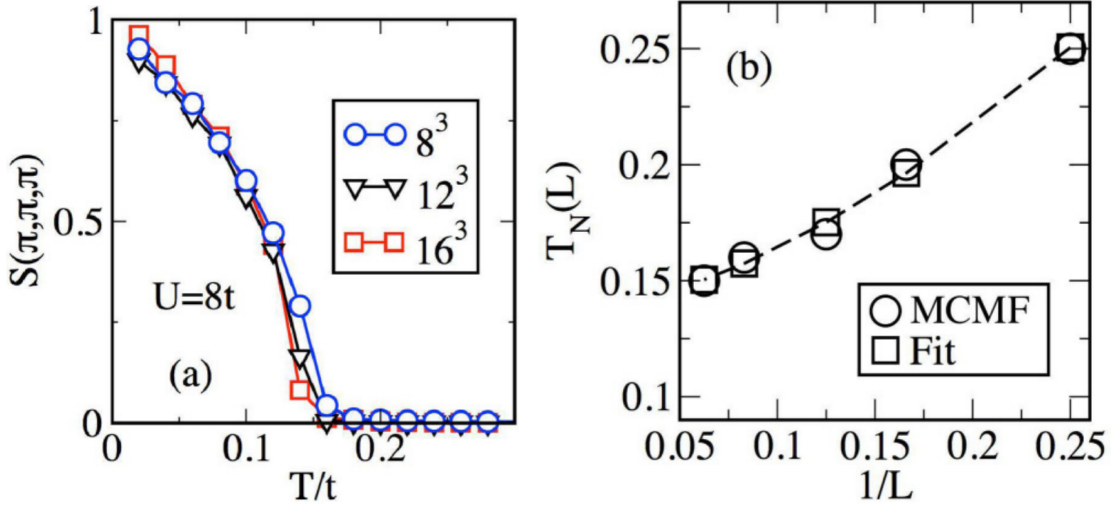


Figure 2.7: Finite-size scaling analysis for the 3D case. Panel (a) shows $S(\pi, \pi, \pi)$ for $U/t = 8$ for three system sizes obtained using TCA. To find T_N in the thermodynamic limit we fit $T_N(L)$, the Néel temperature for a cluster of size L^3 , against $1/L$. Fitting to a scaling form (see text) provides $T_{ThermoN}$, the Néel temperature in the thermodynamic limit. As a typical example, in (b) we show the MCMF $T_N(L)$ data and the fit using the scaling form for $U = 8t$. The dashed line is a guide to the eye. See text for discussion.

2.4 Two-dimensional lattice

We now turn to results for large two-dimensional system sizes using a 4^2 traveling cluster. The results shown in Fig. 2.8a are for a 32^2 system. The method can typically be pushed up to 40^2 sizes. Figure 2.8a shows the AFM Néel temperature. Note that in principle the Mermin-Wagner theorem establishes that there is no true T_N in two dimensions for an $O(3)$ magnet. This theorem is valid only for short-range spin-spin interactions, however. In our case, the integration of the fermions leads to effective spin-spin interactions at all distances, although the rate of the decay of the couplings with distance is unknown. To be cautious we should refer to this scale as T_{corr} instead of T_N , but below we will continue using the T_N notation for this temperature scale since this is the convention widely used in the literature.

Here, we observe that T_N has the correct scaling of t^2/U at large U . The corresponding spin structure factors are shown in Fig. 2.8b. To make a comparison with two-dimensional DQMC data[92], $C_v(T)$ as well as the locus of the high- and low- temperature peaks of the specific heat are shown in Fig. 2.9. As in the three-dimensional case, we observe a two-peak structure in the specific heat and also capture the universal crossing of $C_v(T)$ for different U

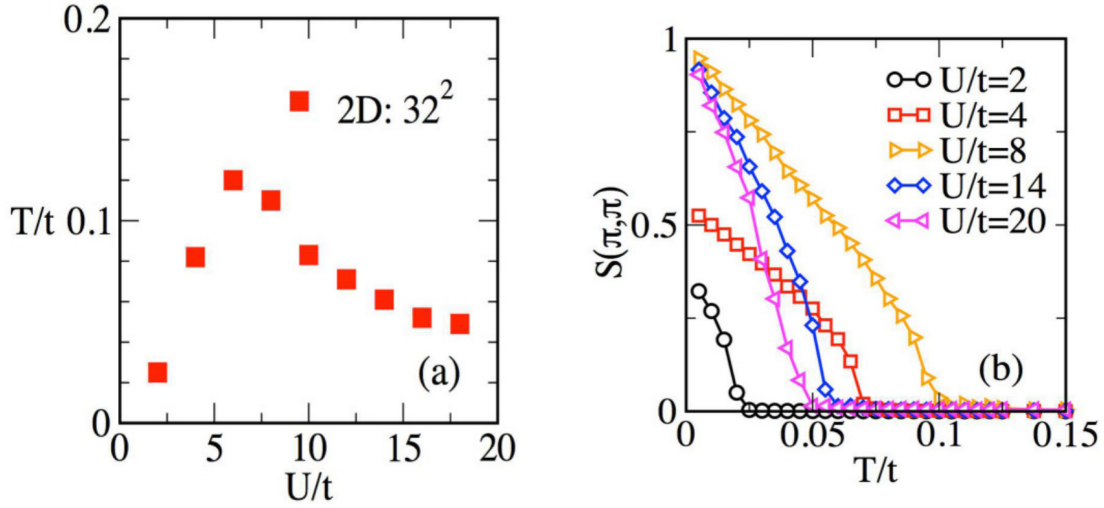


Figure 2.8: (a) T_N vs. U/t and (b) representative $S(\pi, \pi)$'s, both for a 32^2 system. The results are obtained using a 4^2 traveling cluster.

values in Fig. 2.9a. The crossing occurs at $T/t = 1.6$ and has a small spread in temperature values. These are in agreement with the DQMC data for the same system size. In Fig. 2.9b, we show the comparison between our data and the peak locations in DQMC. We find that the saturation of the high-temperature peak (T_{high}) for $U/t < 4$, persists in two dimensions. The small U saturation of T_{high} can be understood by studying the $U = 0$ limit, where the specific heat peaks at $T \sim t (= 1)$. For the behavior at large U we can analyze the limiting case of $t = 0$ (single site problem), where it is easy to see that T_{high} grows linearly proportional to U . This linear growth of T_{high} at large U is seen in Fig. 2.9b, and is in good agreement with DQMC results[92].

The large system sizes accessible to MCMF allows a detailed analysis of the spatial evolution with temperature of the $\{\mathbf{m}_i\}$ field configurations, as shown in Fig. 2.10. Here, the top and bottom panels contain the spatial maps of $|\mathbf{m}_i|$ for $U/t = 4$ and 14, respectively. The maps are shown for four different temperatures, decreasing from left to right. The temperature range here was chosen to show that in the small U case the $\{\mathbf{m}_i\}$ grows with decreasing temperature, similar to the case in three dimensions. The strong thermal fluctuations that make the MCMF approach accurate at high temperatures are clearly visible.

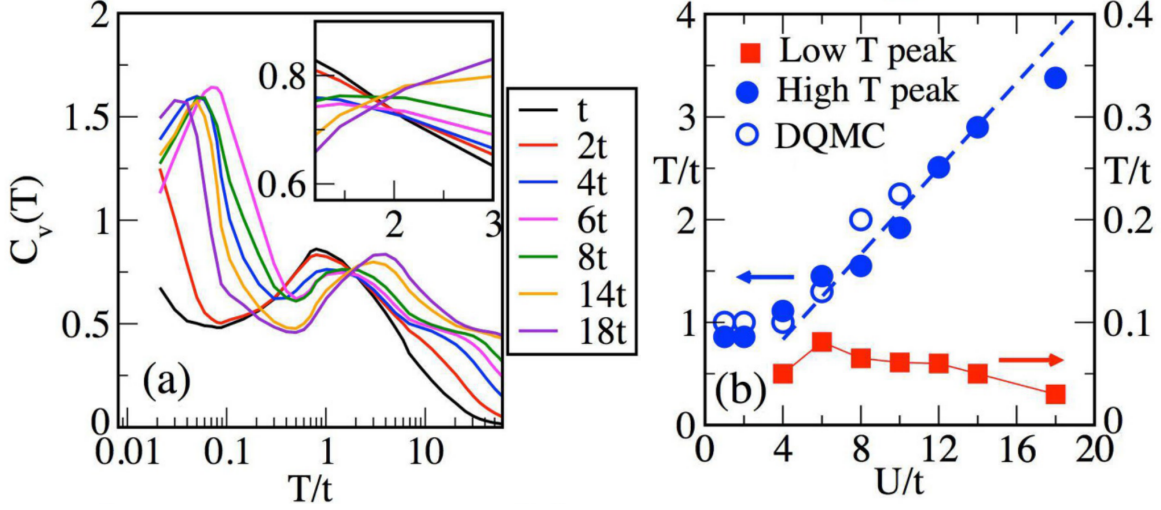


Figure 2.9: The specific-heat vs. temperature data in two dimensions for various U values. The twin peak structure and universal crossing are clearly seen. The corresponding loci of the high- and low-temperature peaks are shown in (b). Here we also show DQMC data for the high-temperature peaks. For comparison with DQMC, we show these data for a 6^2 system size. The dashed line is a guide to the eye.

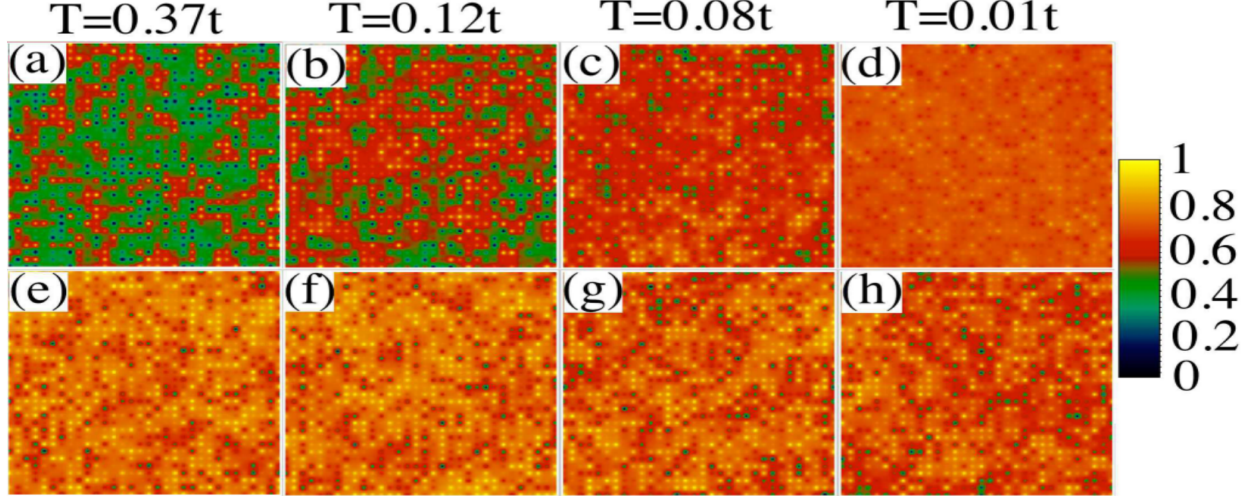


Figure 2.10: Spatial snapshots of $|\mathbf{m}_i|$ for $U/t = 4$ (top) and $U/t = 14$ (bottom). Panels (a)(d) and (e)(h) show the snapshots for $T/t = 0.37, 0.12, 0.08$, and 0.01 for the two cases, respectively. At $U/t = 14$ (bottom), values as high as $|\mathbf{m}| \sim 1.0$ exist at all temperatures shown, much above $T_N(0.06t)$. $|\mathbf{m}|$, however, grows with reducing temperature and shows thermal fluctuations at the higher temperatures for $U/t = 4$ (top). In the figure, yellow implies $|\mathbf{m}| = 1$ and black $|\mathbf{m}| = 0$. The snapshots are for a 32^2 system size.

At $U/t = 4$, the magnitude of $|\mathbf{m}|$ has a broad distribution, with regions of small and large values [see Fig. 2.10a].

At temperatures above that close to $T_N \sim 0.08t$, regions with $|\mathbf{m}| \sim 0.7$ start spanning the entire system, as exemplified in Figs. 2.10 (b) and (c). Figure 2.10(d) shows the system below T_N . These thermal fluctuations imply fluctuating spin moments in a MC snapshot; however, averaging over spin moments from many such MCMF configurations, at a fixed temperature, results in moments that are uniform in space. We stress that there is no spatial phase separation implied in these snapshots.

The corresponding distribution of the $\{\mathbf{m}_i\}$ configurations for Fig. 2.10 is shown in Fig. 2.11. In Fig. 2.11a, we observe a gradual increase in the sharpness and peak height of $P_q(|\mathbf{m}|)$ with reducing temperature. At large U , $P_q(|\mathbf{m}|)$ shows little thermal fluctuations in the temperature range shown. This corresponds to almost saturated $\langle |\mathbf{m}| \rangle$ as in three dimensions at temperatures below $T/t \sim 1$. The uniformity in the bottom panel of Fig. 2.10 translates into a sharp $P_q(|\mathbf{m}|)$ in Fig. 2.11 for typical large values of U/t , as shown in Fig. 2.11b for $U/t = 14$.

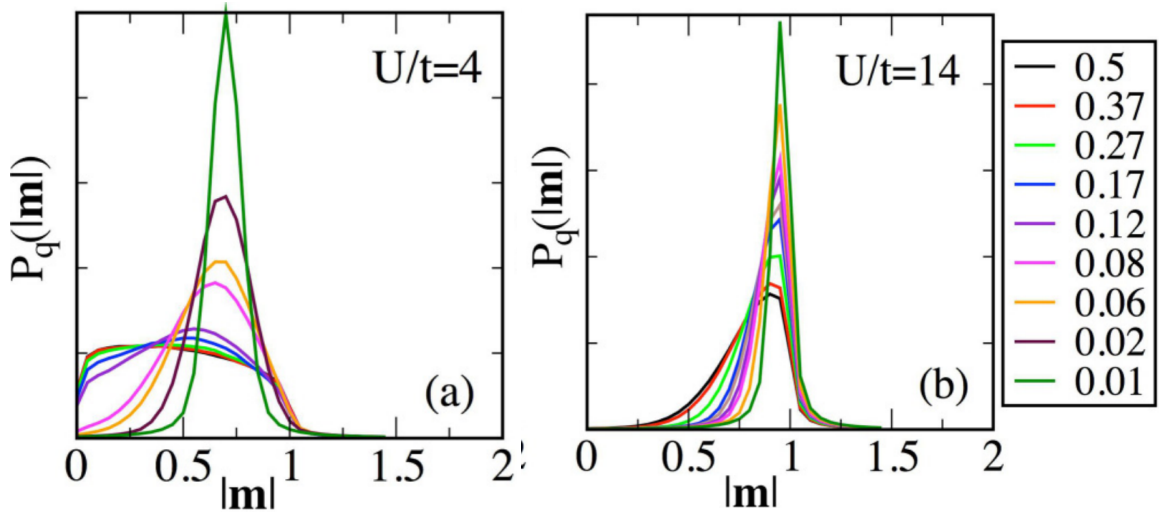


Figure 2.11: $P(q)(|\mathbf{m}|)$ for $U/t = 4$ and 14 at various temperatures indicated on the right. (a) At $U/t = 4$ and high temperature, lattice sites acquire values of $|\mathbf{m}|$ between 0 and 1 in a uniform manner. This distribution starts peaking at $T/t \sim 0.08$ which is close to T_N . At lower T , the auxiliary field distribution peaks at about $|\mathbf{m}| = 0.7$. (b) At large U/t , the $|\mathbf{m}|$ values are well defined moments (about 1) at all sites at the temperatures shown, much above T_N . There is only a small thermal broadening even at $T \sim 10T_N$.

2.5 Longer Range Hopping

In this section we extend our analysis and apply the MCMF method to study the Hubbard model on a two-dimensional square lattice with nearest-neighbor and next-nearest-neighbor hopping t and t' , respectively. The t' hopping processes have been widely considered important in the context of the cuprate superconductors, both directly in the Hubbard model[32], as well as in the $t - t' - J$ model[86]. In addition, understanding the role of t' is in general relevant to the study of frustrated systems. For this model, DQMC studies suffer a severe fermion sign problem due to the broken particle-hole symmetry introduced by t' . Thus, the ground-state properties remain inaccessible. DMFT studies have had more success but with limited or no spatial correlations[112]. There are other approaches to access the ground-state properties[127, 7, 71], but they are difficult to generalize to finite temperature. MCMF fills this void.

The MCMF approach used here reduces to unrestricted Hartree-Fock at $T = 0$, but, as shown by comparison with DQMC results earlier, it rapidly improves its accuracy with increasing temperature. Moreover, MCMF does not have a sign problem. Thus, it allows controlled calculations of both finite temperature and ground-state properties on very large two- and three-dimensional clusters, under a broad variety of circumstances. With this in mind, here we address the $U - t - t'$ model using MCMF. We also use DQMC to solve the same problem for the lowest temperature allowed by the sign problem.

(1) *Comparison with DQMC.* In Figs. 2.12a and 2.12c, we show $S(\pi, \pi)$ calculated using DQMC and MCMF, respectively, for $t' = 0$. For this case, DQMC does not have sign problems and in principle we could obtain results for lower temperatures. However, given the $[O(L)^2]$ scaling in CPU time (where L is the number of imaginary time slices[135]) and the existence of results in the literature, we stopped the DQMC calculation at $T/t \sim 0.1$. It is clear that even at these temperatures we do observe AFM correlations beginning to grow with reducing T . We also observe that magnetic correlations start to grow at a higher temperature for $U/t = 8$ compared to $U/t = 4$ and 16. This is indicative of the nonmonotonicity of T_N with U , as extensively discussed earlier. By comparison, MCMF

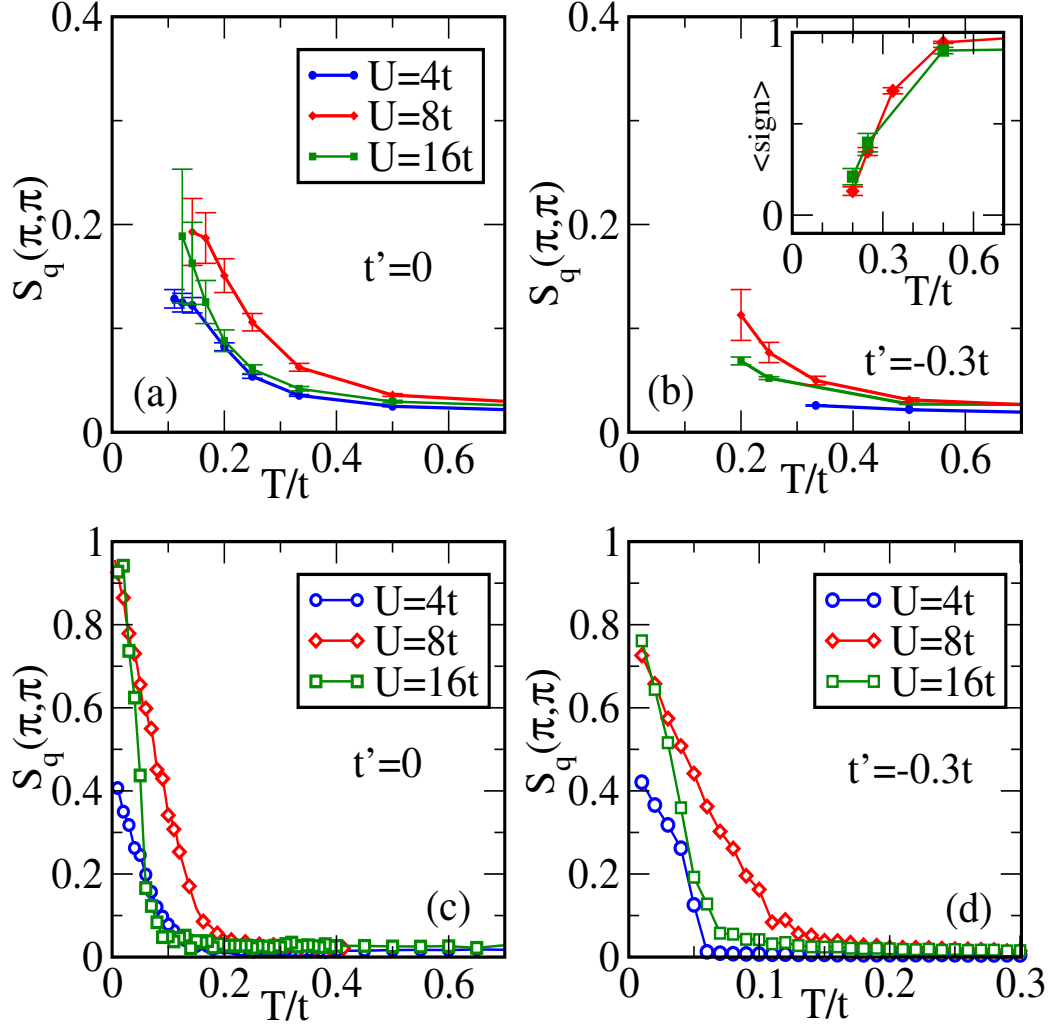


Figure 2.12: DQMC results showing $S(\pi, \pi)$ for several U 's at (a) $t' = 0$ and (b) $t' = 0.3t$. The inset in (b) contains the average sign vs. temperature. For $t' = 0.3t$, sign error increases rapidly preventing the access to low temperatures. Panels (c) and (d) show the $S(\pi, \pi)$ obtained from MCMF. The data shown here are for a 6^2 system.

ordering happens at a lower temperature. An additional difference with DQMC is the high-temperature tail seen in Fig. 2.12a, which is absent in Fig. 2.12c. As shown for three dimensions in Fig. 2.5b, however, short-range spatial correlation, in particular $C(|r| = 1|)$, survives up to high temperatures. Similar correlations survive in two dimensions as well, but the presence of quantum effects makes the AFM correlations survive to longer length scales in DQMC contributing to the high-temperature tail. In contrast, since only $C(|r| = 1|)$ is significant in MCMF, the magnetic structure factor in Fig. 2.12c has a suppressed tail. This comparison highlights the effect of the mean field approximation in MCMF at low T on long-range correlations and may explain the reduced values of T_N as compared with DQMC.

In Fig. 2.12b we present $S(\pi, \pi)$ for the physically relevant case $t'/t = 0.3$ [21]. In the inset, we show the average value of the fermion sign as a function of temperature. The loss of particle-hole symmetry causes the average sign to rapidly fall to zero. As a result, it becomes impossible to obtain reliable results below $T/t = 0.2$ using DQMC. In contrast, the MCMF approach easily captures the long-range AFM order as shown in Fig. 2.12d.

(2) *Ground-state properties.* Earlier $T = 0$ studies [71, 7, 127] have established that at small U , a small finite t' destroys magnetic order in favor of paramagnetism (PM). For t'/t below 0.7, the paramagnetic phase evolves into a $\mathbf{q} = (\pi, \pi)$ antiferromagnet with increasing U . At larger t' and larger U , there is a transition to a state that is a linear superposition of $\mathbf{q} = (0, \pi)$ and $\mathbf{q} = (\pi, 0)$ states from the PM state. Finally for U greater than $10t$, there is a possible spin-liquid phase in between the $\mathbf{q} = (\pi, \pi)$ and $\mathbf{q} = (0, \pi)/\mathbf{q} = (\pi, 0)$ phases. The Gutzwiller approximation combined with the random phase approximation (GA+RPA) also find a number of incommensurate magnetic phases sandwiched between the low U (PM) and large U [$\mathbf{q} = (\pi, \pi)$ or $\mathbf{q} = (0, \pi)/\mathbf{q} = (\pi, 0)$] orders [71].

Here, we present some of the ground-state and finite temperature properties with the goal to show the ability of MCMF to capture essential physics both at low and high temperatures. Detailed quantitative comparison with existing literature will be presented elsewhere. Figure 2.13 shows our results. In Fig. 2.13a, the locus of the $\mathbf{q} = (\pi, \pi)$ and $\mathbf{q} = (0, \pi)$ Néel temperatures is shown as a function of t'/t . The U/t values used represent small, intermediate, and large U/t regimes. At $U/t = 4$, the $\mathbf{q} = (\pi, \pi)$ phase is progressively weakened and ultimately destroyed in favor of a paramagnetic state. In Fig. 2.13c we show

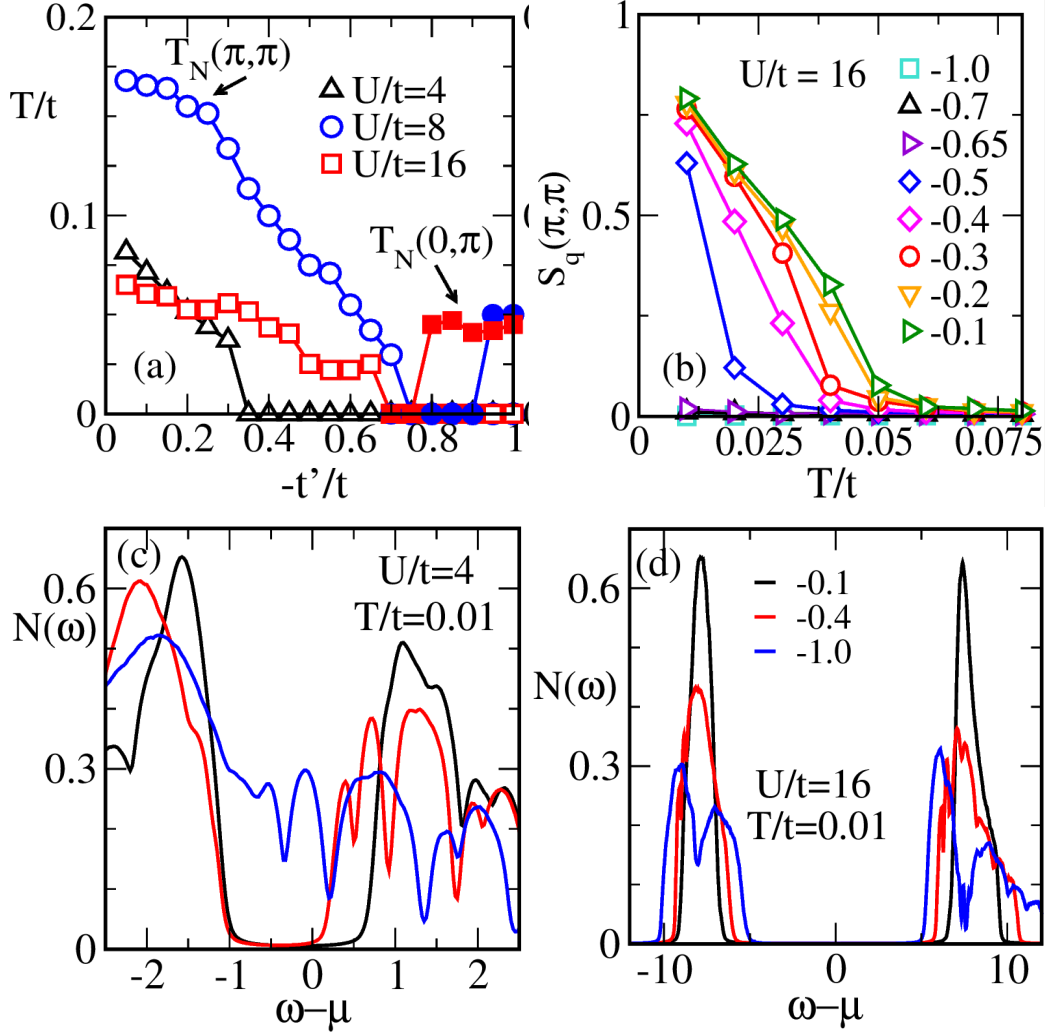


Figure 2.13: MCMF results for $t' \neq 0$. Panel (a) contains the evolution of T_N with increasing t'/t for different values of U/t and for $\mathbf{q} = (\pi, \pi)$ and $\mathbf{q} = (0, \pi)$. The case $\mathbf{q} = (\pi, 0)$ is identical to $\mathbf{q} = (0, \pi)$ and it is not shown. Panel (b) displays the typical $S(\pi, \pi)$ for $U/t = 16$ at various t'/t , as indicated. Panels (c) and (d) show $N(\omega)$ for $U/t = 4$ and $U/t = 16$ respectively, at different values of t'/t .

the DOS for $U/t = 4$. At $t'/t \sim 0.35$, there is an insulator-to-metal transition accompanying the magnetic to PM transition. For $U/t = 8$ and 16 , there is a similar loss of the $\mathbf{q} = (\pi, \pi)$ magnetic order with increasing t'/t . The critical t'/t needed shows nonmonotonic dependence on U/t similar to that of T_N with varying U/t . The collapse of the $\mathbf{q} = (\pi, \pi)$ order with increasing t'/t is shown in Fig. 2.13b for $U/t = 16$.

In Fig. 2.13d, the DOS for $U/t = 16$ is presented. Clearly, the gap in $N(\omega)$ changes only slightly with t'/t varying from 0 to 1. A similar evolution occurs for $U/t = 8$ as well. The gap survives because there is a transition from $\mathbf{q} = (\pi, \pi)$ to a linear combination of the $\mathbf{q} = (0, \pi)/\mathbf{q} = (\pi, 0)$. The locus of the peak of $\mathbf{q} = (0, \pi)$ is shown in Fig. 2.13d. In the region in between the two phases we only find a weak order difficult to distinguish from a PM state. Note that since this method reduces to the HF theory at $T = 0$, a spin-liquid phase cannot be captured within this approach due to the lack of quantum fluctuations; however, the MCMF method is able to suggest regions in parameter space where spin-liquid phases are possible.

In this chapter, a many-body technique that is intermediate between the canonical mean field Hartree-Fock approximation and the numerically exact determinant quantum Monte Carlo method has been discussed and tested for the case of the one-band Hubbard model. The thermal fluctuations that are properly considered in this method were shown to be sufficient to reproduce the expected up and down nonmonotonic behavior of the Néel temperature with increasing U/t at half filling, unveiling a normal-state regime where there are preformed local moments but no magnetic long-range order.

A necessary condition for this method to work properly is that the mean field approximation used (either the HF method employed here or some other mean field method) captures the essence of the ground-state magnetic, orbital, or even superconducting properties. After that step, the MCMF technique is expected to address reasonably well the temperature fluctuations and generation of short-range order near the critical temperature. There are no obvious restrictions in parameters such as couplings: if the mean field method works at a particular coupling, the MCMF will work as well varying the temperature. The coupling range where the method works best, in the sense of improving substantially over naive mean field finite-T approximations, is strong coupling where fluctuations start

developing when cooling down at temperatures much higher than the true long-range-order critical temperature. Our technique captures the regime where local moments are formed but they are coupled effectively only at short distances. In superconducting systems with strong attraction, the method would capture the formation of individual Cooper pairs upon cooling, followed at lower temperatures by the true superconducting state.

Another advantage of the MCMF method is that it can be applied to other Hubbard models that cannot be treated by DQMC due to the fermion sign problem. As in the case of the addition of realistic $t' < 0$ next-nearest-neighbor hopping amplitudes, where DQMC cannot reach the ordering temperature upon cooling because of the sign problem, the good performance of the MCMF remains unchanged with regard to the case $t' = 0$. Thus, examples where the MCMF approach can be applied include the one-orbital Hubbard model with hopping beyond nearest neighbors, as demonstrated here, or the multiorbital Hubbard models that are widely discussed for iron-based superconductors. With a good understanding of the one-orbital Hubbard model, we move toward application of MCMF to a more realistic two-orbital Hubbard model applied to iron-based superconductors.

Chapter 3

Two Orbital Model for Pnictides (MCMF)

This chapter is a modified version of Physical Review B **93**, 085144 (2016) [83].

3.1 Introduction

Several important materials, such as iron-based high critical temperature superconductors (FeSC) [50, 47, 27, 23], nickelates [129], cobaltites [128], manganites [24, 104], and many others, have several active orbitals. If electronic correlation effects are important, then multiorbital Hubbard models must be employed for their analysis. However, these models are difficult to study in two (2D) or three (3D) dimensions due to their complexity. While at particular electronic densities, such as integer fillings, zero temperature mean field (MF) approximations are often reliable, the phase diagrams of multiorbital Hubbard models varying *temperature* T are basically unknown. This is because thermal MF approximations provide qualitatively incorrect results at robust Hubbard repulsion U and intermediate temperature since they cannot generate local moments and short-range order. Moreover, “sign problems” complicate the application of quantum Monte Carlo (MC) when several orbitals are active [49]. Since not only U but also the Hund coupling J are important, it is imperative to apply alternative computational tools, even if crude, to study multiorbital

models at intermediate temperatures because the potential for new states in these systems is considerable.

In this chapter, the MCMF technique is applied for the first time to a 2D two-orbital Hubbard model, varying T and U . The unveiled phase diagram is rich, including a nematic phase above T_N [18, 37, 34, 35]. Even more importantly, here we report an unexpected novel regime, dubbed Orbital Selective Directional Conductor (OSDC), where a remarkable anisotropy in transport is observed, with one direction insulating and the other conducting, leading to a dimensional reduction from 2D to 1D. This dimensional reduction is different from that in $\text{Ti}_2\text{Ru}_2\text{O}_7$ [57] and $\text{BaCuSi}_2\text{O}_6$ [111] because they are insulating in all directions, and different from layered $\text{Sr}_3\text{Ru}_2\text{O}_7$ [13] because it requires a high magnetic field, and its resistivity is metallic in one direction but only flat with T in the other direction (not insulating). Our results are also different from layered oxides that are metallic in-plane but insulating out-of-plane [103], because their crystal structure already establishes an asymmetry. On the contrary, our 2D model is fully symmetric between the x and y directions but *spontaneously* becomes insulating in one direction and metallic in the other, without the help of the lattice, magnetic fields, or impurities.

3.2 Model and Method

Although five orbitals are needed for a faithful electronic description of FeSC, for simplicity we will focus on the two most important orbitals d_{xz} and d_{yz} [99, 79, 68, 88, 87]. The two-orbital Hubbard model studied here is defined as:

$$\begin{aligned}
H = & \sum_{\langle i,j \rangle, \alpha, \beta, \sigma} T_{\alpha, \beta}^{i,j} d_{i, \alpha, \sigma}^\dagger d_{j, \beta, \sigma} + U \sum_i n_{i, xz, \uparrow} n_{i, yz, \downarrow} + (U' - J/2) \sum_i n_{i, xz} n_{i, yz} \\
& - 2J \sum_i S_{i, xz}^z S_{i, yz}^z + J' \sum_i (d_{i, xz, \uparrow}^\dagger d_{i, xz, \downarrow}^\dagger d_{i, yz, \downarrow} d_{i, yz, \uparrow} + H.c.),
\end{aligned} \tag{3.1}$$

where $d_{i, \alpha, \sigma}^\dagger$ creates an electron at site i , orbital α (either xz or yz), and with spin projection σ . The number operator is $n_{i, \alpha, \sigma}$, $n_{i, \alpha} = \sum_\sigma n_{i, \alpha, \sigma}$, and $S_{i, \alpha}^z = (1/2)(n_{i, \alpha, \uparrow} - n_{i, \alpha, \downarrow})$. U , U' , J , and J' are the Kanamori parameters [48, 53]. The usual constraints $U' = U - 2J$ and $J = J'$

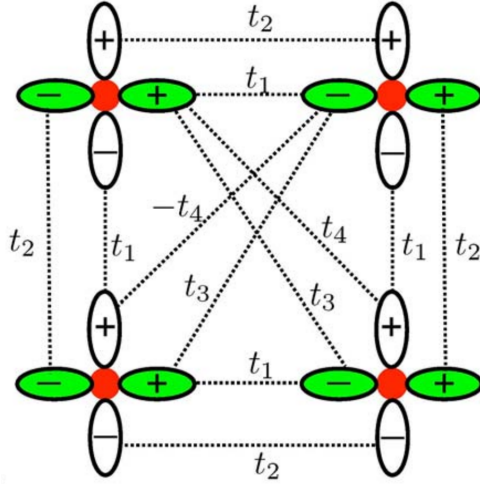


Figure 3.1: Schematic representation of the directions of electronic hopping for the two-orbital model on a square lattice. The green (white) shaded lobes are projection of orbital d_{xz} (d_{yz}) onto the xy plane. The values of hopping parameters are $t_1=1$, $t_2=-1.33$, and $t_3=t_4=-0.85$, which leads to a bandwidth $W=12t_1$ in the non-interacting limit. This schematic is reproduced from Ref. [99].

are assumed (in other portions of this thesis, the Hund coupling is called J_H). In the Hund term, only the Ising portion is kept because the expected magnetic order is collinear, most materials have an easy-axis, and it is technically simpler for this first study. The hopping parameters reproduce the Fermi surface of the undoped FeSC [99], but our conclusions could be realized as well in other materials with local tetragonal symmetry. The crystal location of the Se, As, or P atoms, used by electrons to tunnel from Fe to Fe, indicate that both nearest- (NN) and next-nearest-neighbor (NNN) hoppings are needed. The explicit hopping amplitudes are in [99] or in Eq. (14) of [79]. The NN sites hoppings t_1 and t_2 are only intraorbital (Fig. 3.1). Along the plaquette diagonals, the intraorbital (interorbital) hopping is t_3 (t_4) (Fig. 3.1). Their values are $t_1=1$, $t_2=-1.33$, and $t_3=t_4=-0.85$ and the bandwidth of this tight binding model is $W=12t_1$ [99]. Hereafter, t_1 will be denoted by t , and it will be the energy unit. To convert to eV, the *ab-initio* derived bandwidth for the d_{xz} - d_{yz} bands is $W \sim 1.8$ eV [4] (Note that the actual bandwidth is likely to be smaller as a result of a significant mass renormalization in the undoped FeSC materials.) The density is fixed to two electrons per site ($n = 2$).

Many-body techniques: MF approximations have already been applied to two-orbital Hubbard models at $T=0$ [68, 88, 87], showing several phases with increasing U/W : a

paramagnetic metal, a metal with $(\pi, 0)$ spin order, and an insulator also with $(\pi, 0)$ spin order. These previous $T=0$ studies and others [50, 47, 27, 23] showed that $J/U \sim 0.15 - 0.30$ is relevant for FeSC, and we will fix $J/U = 0.25$ in all the results below. Since FeSC materials vary substantially in their degree of electronic correlation, results will be presented varying the ratio U/W . Our main focus are the temperature effects since their influence on model Eq. (1) are unknown. Our study is performed in 2D, mainly on 32^2 lattices (Appendix B). The TCA traveling cluster size [81] is 6^2 . Via the parallelized version of TCA [81], lattices as large as 60^2 were reached. As shown in the Appendix B, mean field approach was chosen to be the Hartree approximation that works well for the half-filled one-orbital model (In the Hartree approximation the J' term does not contribute, but this term is only considered of marginal importance in Hubbard models).

3.3 Results

Our main results are in the phase diagram of model Eq. 3.1 at $n = 2$ (Fig. 3.2). It was constructed based on data for the two spin structure factors of relevance, $S(\pi, 0)$ and $S(0, \pi)$, the spin nematic order parameter Ψ_{Nem} [35, 34], their temperature derivatives, and the resistivity and density of states (DOS) (Appendix B). We also monitored local moment formation at intermediate temperature and large U/W [82] (Appendix B). This phase diagram is surprisingly rich because it contains *three* regimes with $(\pi, 0)$ long-range magnetic order [degenerate with $(0, \pi)$]. The insulator at large U/W is induced by the robust J that produces $S=1$ local spins interacting via a frustrated Heisenberg model known to have $(\pi, 0)$ - $(0, \pi)$ magnetic order. The other two states at intermediate couplings are more subtle. Their magnetic order arise from Fermi Surface nesting effects. However, the presence of *two*, instead of one, regions is unexpected. The first one, dubbed AF-M (antiferromagnetic metallic), is metallic in both directions, albeit anisotropic, as in FeSC experiments [18, 37]. But the second region, the OSDC, is metallic along the spin staggered direction but insulating along the spin uniform direction, leading to a surprising dimensional reduction at intermediate couplings.

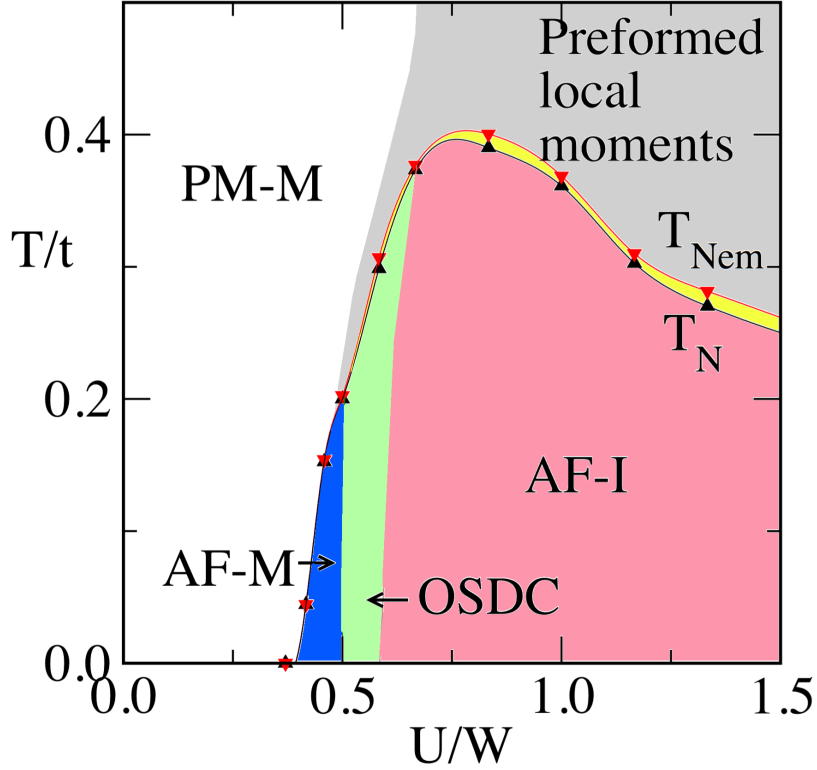


Figure 3.2: Phase diagram of the two-orbital Hubbard model Eq.(1) in the MCMF approximation and with hoppings from [99], at $J/U = 0.25$ and $n = 2$. Shown are results for a 32^2 lattice. Besides the weak coupling paramagnetic metal (PM-M) and the intermediate/large U/W region with “preformed local moments” (grey), other states were identified: (1) A $(\pi, 0)$ -spin-ordered metal, AF-M, where transport is anisotropic but metallic in both directions. (2) A novel $(\pi, 0)$ -spin-ordered regime, the OSDC, with metallic (insulating) behavior along the x (y) axis. (3) A $(\pi, 0)$ -spin-ordered insulator, AF-I, with a full gap. (4) A spin nematic regime above T_N . The AF-M and OSDC states break the same symmetries and, thus, no sharp distinction between them is expected but a rapid crossover. T_N has a non-monotonic behavior, maximizing close to the OSDC/AF-I boundary.

3.4 Magnetic phases

A typical magnetic order parameter OP displays a negative curvature increasing temperature, with a diverging slope at the critical temperature T_c in the bulk limit. Thus, for a finite system the temperature where the first derivative dOP/dT is maximized provides an estimation of T_c (here T_N and T_{Nem}). In addition, upon cooling T_{Nem} can also be estimated from the temperature T_{split} where $S(\pi, 0)$ and $S(0, \pi)$ split. While for a finite system T_{Nem} and T_{split} may be different, they should merge in the bulk limit. Typical results are in Fig. 3.3 for a 32^2 lattice. Our MC statistics and lattice sizes are sufficient to observe a robust order parameter behavior for $S(\pi, 0)$ and Ψ_{Nem} : nonzero at $T = 0$ and decreasing with increasing temperature with a negative curvature, with the exception of a small temperature window where the curvature is positive due to size effects. T_N and T_{Nem} can be estimated from the maximized derivative criterion (Fig. 3.3). Although close in temperature, systematically for all the couplings U/W with long range spin order at $T = 0$ and for all lattices, we find $T_{Nem} > T_N$ suggesting a small region of nematicity. We also found that T_{split} is always larger than T_{Nem} . The observed narrow window of nematicity in fact survives a finite-size scaling analysis (Fig. 3.4(a)). Remarkably, within the error bars the bulk limit extrapolated T_{Nem} and T_{split} leads to the same nematic critical temperature. For example, our results unveil a small nematic window of $0.0065t$ at $U/W = 1.16$. Such a fragile nematic phase, reported here for the first time in a Hubbard model, is compatible with previous studies using spin models [52, 136, 14], spin-fermion models [61], and with experiments [18, 37]. (General arguments [35, 34] suggest that either $T_N = T_{Nem}$ in a first-order transition, or $T_N < T_{Nem}$ with both transitions being second order. Since we do not observe indications of metastabilities in the MC time evolution, or in histograms of observables, our results are compatible with the existence of a nematic regime. The narrowness of the nematic regime is likely exaggerated by the $Z(2)$ nature of the Ising approximation used here for the Hund term).

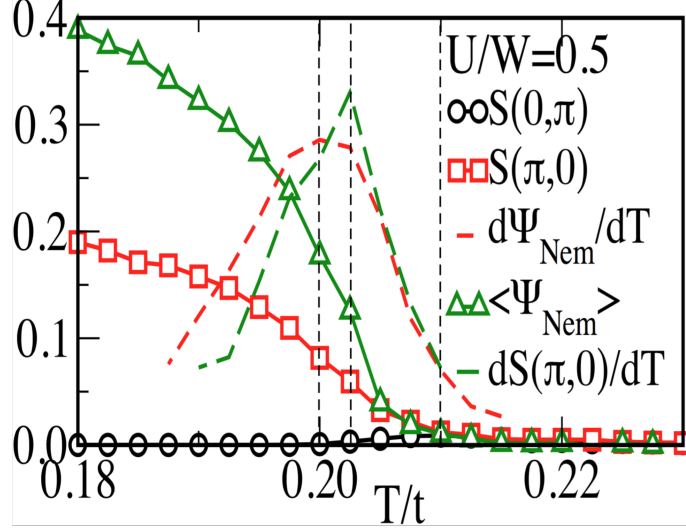


Figure 3.3: Spin structure factors and nematic order parameter at $U/W = 0.5$ (32^2 lattice; errors bars are the size of the points). Dashed curves show the corresponding numerical temperature derivatives. Vertical dashed lines (left to right) indicate the magnetic and nematic transition temperatures, from derivative maximization, and the temperature where $S(\pi, 0)$ - $S(0, \pi)$ split.

3.5 New State at Intermediate Coupling

As explained before it is surprising that there are three distinct regions below T_N : two metals (AF-M and OSDC) and one insulator (AF-I). The distinction between the two metals and the insulator can be understood via the DOS in Figs. 3.4(b,c). Panel (c) displays a canonical insulating behavior: in the temperature range shown, a pseudogap (PG) is observed in the local moments regime (At much larger temperatures $\sim U$ the PG disappears), transforming into a full gap at T_N . This is the AF-I state (Fig. 3.2). In panel (b), upon cooling toward T_N a PG opens because of Fermi surface nesting effects. But even at low temperatures, and independently analyzing the $T = 0$ Hartree equations, in both metallic regions the total DOS has a finite weight at the Fermi energy E_F .

What is then the difference between AF-M and OSDC? Their physical distinction is illustrated in Fig. 3.5 (a) where the resistivity ρ vs. T is presented at three values of U/W , corresponding to the three low-temperature regions of Fig. 3.2. ρ is calculated from the optical conductivity $\sigma(\omega)$, integrating in a narrow range near $\omega = 0$ and then inverting [54] (Appendix B). At $U/W = 0.417$, ρ in the y spin uniform direction is larger than in the x spin

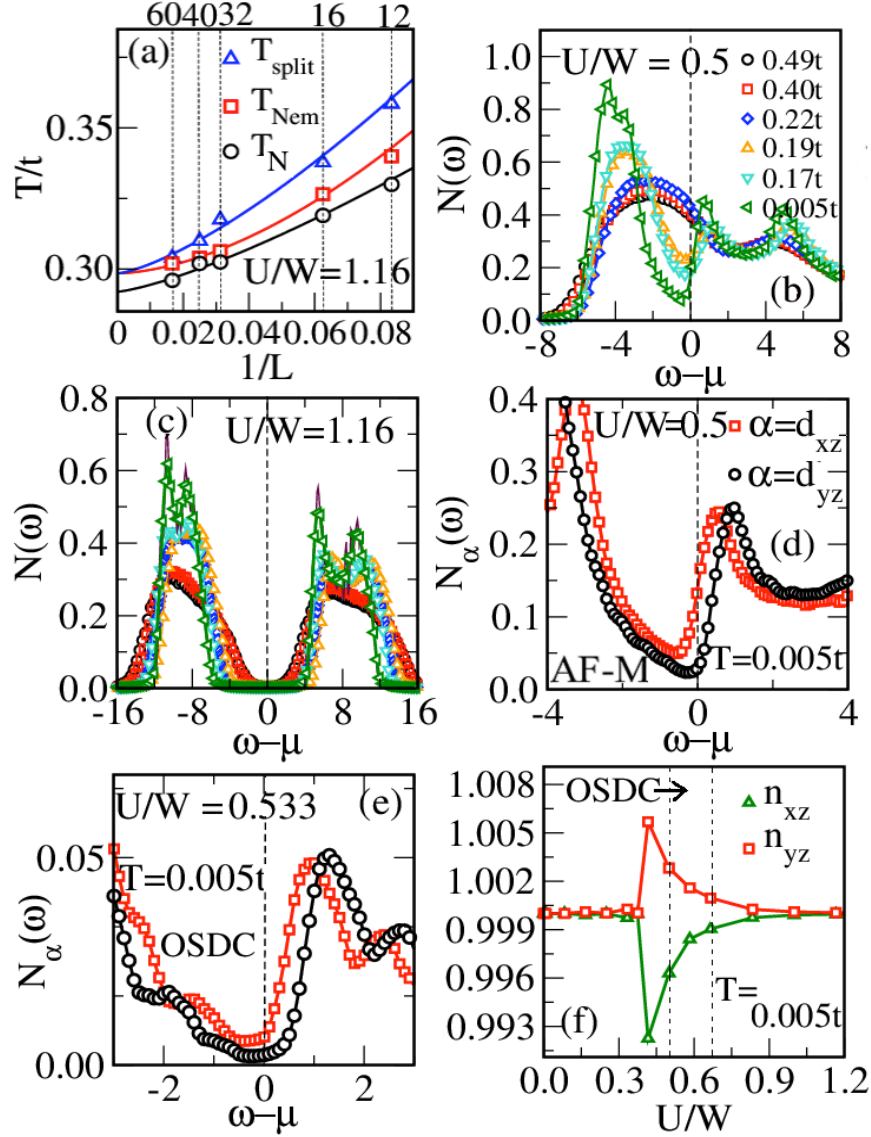


Figure 3.4: (a) Finite-size scaling analysis for T_N , T_{Nem} , and T_{split} at $U/W = 1.16$, using $L = 12, \dots, 60$ ($L \times L$) lattices plotted vs. $1/L$ (values at top). Data is fit with a scaling function $T_{ord}(L) = T_{ord}^{bulk} + b(1/L)^{(1/\nu)}$, where T_{ord}^{bulk} , b , and ν are independent fitting parameters for the three data sets. From the fit we obtain $T_N^{bulk} = 0.2919t$ and $T_{Nem}^{bulk} = T_{split}^{bulk} = 0.2984t$, with an error $0.0001t$. (b) Density of states $N(\omega)$ at $U/W=0.5$ (μ denotes E_F). A pseudogap develops below T_{Nem} that deepens with reducing temperature but never becomes a full gap. On the other hand, in panel (c) at $U/W=1.16$ a clear gap develops upon cooling. (d) contains the orbital-resolved DOS at $T=0.005t$ and $U/W=0.5$. The C_4 spontaneous symmetry breaking makes the (nonzero) population of the two orbitals different at E_F [19]. (e) Same as (d), but at $U/W=0.533$ in the OSDC regime where $N_{yz}(\omega = \mu) \ll N_{xz}(\omega = \mu)$. (f) The orbital-resolved total occupation n_{xz} and n_{yz} shown at low T vs. U/W . Dashed lines indicate the OSDC regime. Panels (b-f) were obtained using 32^2 lattices.

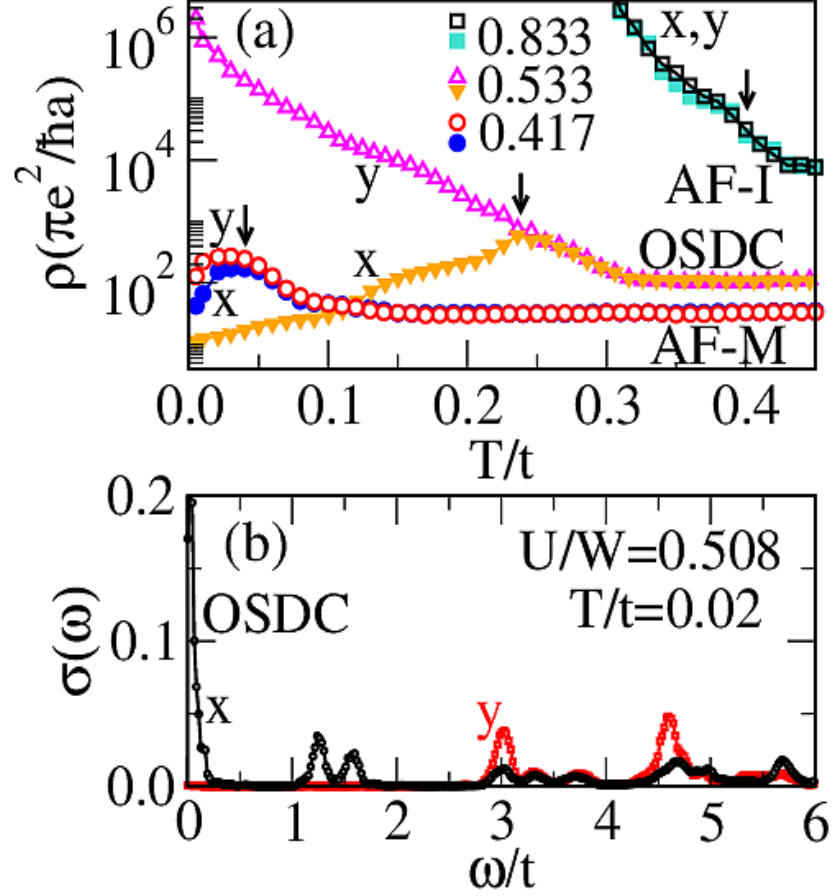


Figure 3.5: (a) Resistivity along the x spin staggered (solid symbols) and y spin uniform (open symbols) directions at several U/W 's, illustrating the transport properties of the AF-M, OSDC, and AF-I regions of Fig. 3.2. Arrows indicate T_N for each case. (b) Optical conductivity in the OSDC with electric fields along the y and x directions. In both panels a 16^2 lattice is used.

staggered direction, as in [61, 144, 60] and experiments [18, 37]. This is understood from the orbital resolved DOS of Fig. 3.4(d): in the magnetic $(\pi, 0)$ state that breaks rotational invariance, near E_F the orbital d_{yz} , related to conduction in the spin uniform direction, is more suppressed than d_{xz} , related to conduction in the spin staggered direction. But since both orbitals have a sizable DOS weight at the E_F , both directions are metallic (Experiments affected primarily by the vicinity of E_F may suggest strong orbital order, but the ω -integral of the orbital-resolved DOS, i.e. the orbital population n_{xz} and n_{yz} , is only different by 0.5. We also monitored $\Delta_{orb} = n_{xz} - n_{yz}$ vs. T . The results (not shown) indicate that upon cooling from high T , Δ_{orb} remains smaller than 0.001 at $U/W = 1.16$ until T_{Nem} is reached, eventually converging, as T is reduced, to the results derived from Fig. 3.4(f). Within our MC accuracy we conclude that in our model there is no additional orbital-order critical temperature above T_{Nem} .) At large $U/W = 1.0$, the DOS Fig. 3.4(c) displays a sharp gap at low temperature, and both directions must be insulating.

The interpolation between small and large U/W unveils a surprise: at intermediate couplings such as $U/W = 0.533$ the spin staggered direction remains metallic, but the spin uniform direction becomes *insulating* (Fig. 3.5 (a)). Intuitively, this is because the interpolation between the DOS of Fig. 3.4(d), where both orbitals have nonzero weight at E_F , and (c), where both orbitals have negligible weight at E_F , is not smooth. Instead, there is an intermediate coupling range where the d_{yz} weight at E_F is almost negligible while that of d_{xz} is still finite (Fig. 3.4(e)). Since the d_{yz} weight is very small but not zero, strictly speaking it cannot be used as a sharp order parameter: AF-M and OSDC are likely analytically connected because they break the same symmetries. However, our study shows that the orbital population difference at E_F in the OSDC is sufficiently large to induce one-dimensional transport. Results for the full $\sigma(\omega)$ (see Fig. 3.5 (b)) show that at $\omega \sim 0$ only one orbital dominates. Moreover, from n_{xz} and n_{yz} (Fig. 3.4 (f)) note that at large U/W both orbital populations converge to one since J is large, while in the PM-M regime they are also one by symmetry. However, in the OSDC region *both* n_{xz} and n_{yz} are different from one (and different among themselves because C_4 is spontaneously broken): the OSDC regime is not the same as an Orbital Selective Mott Phase (OSMP), where one orbital has population exactly one [5, 40]. Another interesting observation is that for both

AF-M and OSDC there is an insulating region $d\rho/dT < 0$ in both directions immediately above T_N due to the opening of the pseudogap in the local moments region and concomitant coexisting patches of $(\pi,0)$ and $(0,\pi)$ order [60], in agreement with spin fermion studies [60] and experiments [18, 37].

In conclusion, the phase diagram of a layered two-orbitals Hubbard model was studied with emphasis on temperature effects. We report a novel intermediate coupling U region, the OSDC, that is conducting in one direction via the d_{xz} orbitals, but insulating in the other because the associated d_{yz} orbitals have nearly vanishing weight at E_F . Although our $T = 0$ calculations do not include quantum fluctuations, the OSDC starts at relatively high temperatures $\sim T_N$ and for this reason our approach emphasizing thermal effects is sufficient. Moreover, we tested that using other hoppings, such as those in [20], the OSDC is also found. Experimentally, materials of the FeSC family are the most likely to realize the OSDC, and a mixing in the chemical formula of As, associated with weak coupling, and Se, associated with strong coupling, may be needed. But the OSDC could be realized in other layered materials where a transition metal atom M is coordinated with four ligand atoms X, establishing MX_4 tetrahedral cages with near degenerate d_{xz} and d_{yz} orbitals, and where a magnetic state that breaks lattice rotational invariance is stabilized.

Chapter 4

Magnetic properties and pairing tendencies of the iron-based superconducting ladder BaFe_2S_3 (DMRG)

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4.1 Introduction

Spin-1/2 Cu-oxide two-leg ladders have been extensively studied in cuprates because of their unusual spin gap, induced by the ladder geometry [26, 25, 22]. In those previous theoretical studies, it was possible to employ a variety of accurate many-body techniques for ladders, showing agreement between theoretical predictions and experimental results, which has provided considerable support to the notion that superconductivity in cuprates originates in AFM fluctuations.

Recently, an unexpected experimental result has been reported using BaFe_2S_3 [122, 141], that has the two-leg ladder structure. This material was found to become superconducting at a pressure above 10 GPa with an optimal critical temperature $T_c = 24$ K. The same material but at ambient pressure, is a Mott insulator with the same magnetic order as

KFe₂Se₃ namely involving FM rung and AFM leg spin correlations with a critical temperature ~ 120 K, according to power neutron diffraction studies [122]. These discoveries unveiled the first iron-based superconductor that does not rely on a square lattice structure of irons, opening an intriguing avenue of research similar to the one opened with the discovery of superconductivity in Cu oxide ladders in the context of the cuprates.

This chapter introduces a two-orbital Hubbard model for a two-leg ladder of BaFe₂S₃, based on *ab initio* calculations that are carried out by our collaborations. This model is subsequently solved numerically using the density matrix renormalization group (DMRG) technique [133, 109]. Our main results are two folded. First, we show that at half-filling with two electrons per iron, and using clusters as large as 16×2 , there is robust evidence for the same magnetic order found experimentally involving FM rungs and AFM leg correlations. This magnetic state becomes robust at intermediate and strong Hubbard couplings, in agreement with the growing perception that these materials are not in the weak coupling regime. Second, we assume that in the experiments [122] the high pressure alters the band structure in such a manner that the individual ladders become hole doped, although the insulator-superconductor transition could also be bandwidth-controlled [122]. In the Cu-oxide based ladders studied some years ago, experiments showed [98] that indeed pressure alters the amount of mobile electrons residing in the two-leg ladders in such a manner that the superconducting state is reached effectively by hole doping of the ladders. Here we simply assumed that a similar physics occurs in the iron-based ladders and focus on their hole doping. Although following our work, density function theory (DFT) calculations suggest that iron ladders are electron doped by charge transfer from sulfur atoms at finite pressures [145]. Studying the cases of one, two, and four holes, we have found pairing tendencies when using an 8×2 cluster in the strong coupling regime $U/W \simeq 2$, W being the tight binding electronic bandwidth. The complexity of the Hamiltonian with two active orbitals and a tight-binding term that must include plaquette diagonals hoppings renders the DMRG calculation so computing time demanding that a confirmation of the pairing tendency beyond 8×2 is not possible at present with the DMRG technique and our available computer resources. Nevertheless, the pairing indications we have observed are promising and suggestive that the theoretical study of iron-based two-leg ladders may illuminate the understanding of

iron-based superconductors using many-body techniques beyond the diagrammatic random phase approximation [89]. The organization of this chapter is as follows. Section 4.2 provides details of the *ab initio* calculations. Section 4.3 contains the actual model used, many-body technique, and observables studied. Section 4.4 presents our main results, organized separately for zero, one, and two holes, the latter including binding energies. Finally, Sec. 4.5 contains our main conclusions.

4.2 *Ab Initio* Calculations

This section presents the details of the derivation of the multiorbital Hubbard model for the BaFe₂S₃ ladder from first principles, to be used later in Sections 4.3 and 4.4. Following the procedure described in Ref. [6], first a calculation is performed based on the generalized gradient approximation with the QUANTUM ESPRESSO package [43]. There we employed the exchange-correlation functional proposed by Perdew, Burke and Ernzerhof [95], a plane-wave basis set with a cutoff energy of 40 Ry, and an 8×8×8 **k**-mesh for the first Brillouin zone (BZ). As for the lattice constants, we used the experimental values $a = 8.78$ Å, $b = 11.23$ Å, $c = 5.29$ Å for the ambient pressure case and reduced them by 4.0%, 8.0%, and 3.4%, respectively, for pressure $P = 12.4$ GPa [46]. The space group of the system is $Cmcm$, and the atomic positions of Ba(4c), Fe(8e), S(4c), and S(8g) are (0.0, 0.686, 0.25), (0.154, 0.0, 0.0), (0.0, 0.116, 0.25), and (0.208, 0.378, 0.25), respectively [46]. Because the magnetic properties will be considered when we solve the effective two-orbital Hubbard model in the following sections, magnetism was not included in the derivation of the model from first principles [121].

After this initial setup, we constructed two Wannier functions for each Fe atom in the unit cell using the WANNIER90 package [80]. One resulting Wannier orbital mainly consists of the standard $d_{x^2-y^2}$ orbital (orbital *a* below) while the other one is primarily made from the standard d_{xz} orbital (orbital *b* below). The two Wannier orbitals employed here do not have a high symmetry, because the $d_{x^2-y^2}$ or d_{xz} orbitals significantly hybridize with other *d*-orbitals or S-*p* orbitals. In particular, for the latter Wannier orbital there is a substantial contribution of the canonical d_{xy} orbital. To construct explicitly these orbitals, we have

to disentangle their complicated electronic structure. In order to preserve accurately the properties of the low-energy band dispersion, we introduced a “frozen” energy window [117] as large as (-0.3 eV, 0.2 eV) with respect to E_F , on top of the ordinary “global” energy window (-1.2 eV, 1.5 eV).

After we constructed an 8 band model, we further simplified this model by unfolding the BZ along the k_z direction. Namely, by introducing a local gauge transformation for one of the two Wannier orbitals to change its sign, we can expand the band dispersion from Γ to Z in the original BZ, and construct a 4 band model [6].

Finally, we want to derive an effective ladder model from the 4-band model, namely we wish to arrive to a model restricted to a two-leg ladder. One possibility is to neglect all inter-ladder electron hopping transfers and just focus on the intra-ladder transfers. However, the bandwidth in the k_x - k_y plane is not necessarily small (as large as 500 meV at maximum). Thus, in the present study the effect of inter-ladder transfers is taken into account by considering their average, *i.e.*, we construct our ladder model from the 4-band Hamiltonian by considering the case $k_x=k_y=0$.

4.3 Model and Method

This section explicitly provides the multiorbital Hubbard model derived by the procedure explained before, while Section IV will present the magnetic properties and pairing tendencies of BaFe₂S₃. The model studied here breaks up into kinetic energy and interaction terms: $H = H_K + H_{int}$. The tight-binding kinetic energy portion is

$$H_K = \sum_{\substack{i\sigma \\ \gamma\gamma'\vec{\alpha}}} t_{\gamma\gamma'}^{\vec{\alpha}} (c_{i\sigma\gamma}^\dagger c_{i+\vec{\alpha}\sigma\gamma'} + H.c.) + \sum_{i\gamma\sigma} \Delta_\gamma n_{i\gamma\sigma}, \quad (4.1)$$

where the first term represents the hopping of an electron from site i of a two-leg ladder and orbital γ to site $i + \vec{\alpha}$ and orbital γ' . The vector $\vec{\alpha}$ indicates the many different directions possible for the electronic hopping, as shown in the ladder sketch Fig. 4.1. We use a two orbital model where we label the down-folded [6] orbitals as a and b (*i.e.* γ and γ' are restricted to a and b). Δ_γ represents the crystal-field splitting of orbital γ . There are two

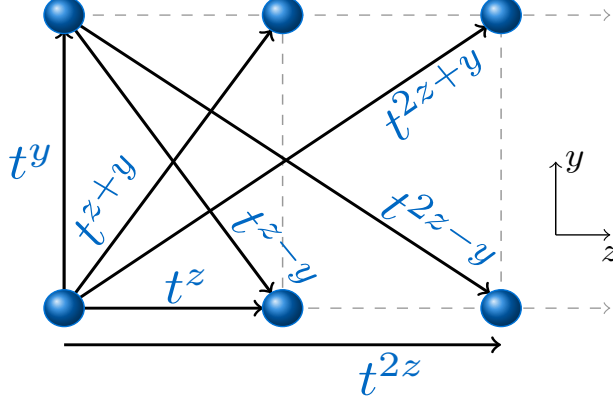


Figure 4.1: Schematic representation of the directions of electronic hopping for the two-leg ladder model considered here. The legs of the ladder are arranged in the z -direction while the rungs are in the y -direction. The hoppings to next-nearest neighbor rungs (*i.e.* $t^{2z}, t^{2z-y}, t^{2z+y}$) are dubbed the “long-range hoppings”. In total, there are seven different hopping directions shown.

sets of hopping parameters obtained from fitting the *ab initio* down-folded band structure calculations at different pressures. The crystal fields at $P = 0.0$ GPa are $\Delta_a = 0.308$ and $\Delta_b = -0.229$ (eV units used from now on) while the associated hopping amplitudes are

$$\begin{aligned}
 t^z &= \begin{bmatrix} -0.215 & -0.149 \\ +0.149 & +0.153 \end{bmatrix}, & t^y &= \begin{bmatrix} -0.012 & 0.000 \\ 0.000 & +0.153 \end{bmatrix}, \\
 t^{z+y} = t^{z-y} &= \begin{bmatrix} +0.075 & +0.174 \\ -0.174 & +0.083 \end{bmatrix}, & t^{2z} &= \begin{bmatrix} -0.137 & +0.004 \\ -0.004 & +0.037 \end{bmatrix}, \\
 t^{2z+y} = t^{2z-y} &= \begin{bmatrix} -0.007 & +0.016 \\ -0.016 & -0.041 \end{bmatrix},
 \end{aligned} \tag{4.2}$$

while the crystal fields at $P = 12.36$ GPa are $\Delta_a = 0.423$ and $\Delta_b = -0.314$, with associated hoppings amplitudes

$$\begin{aligned}
t^z &= \begin{bmatrix} -0.334 & -0.177 \\ +0.177 & +0.212 \end{bmatrix}, & t^y &= \begin{bmatrix} -0.024 & 0.000 \\ 0.000 & +0.216 \end{bmatrix}, \\
t^{z+y} = t^{z-y} &= \begin{bmatrix} +0.085 & +0.216 \\ -0.216 & +0.109 \end{bmatrix}, & t^{2z} &= \begin{bmatrix} -0.171 & -0.011 \\ +0.011 & +0.035 \end{bmatrix}, \\
t^{2z+y} = t^{2z-y} &= \begin{bmatrix} 0.000 & +0.042 \\ -0.042 & -0.044 \end{bmatrix}.
\end{aligned} \tag{4.3}$$

Figure 4.2 (a,b) show the non-interacting single particle spectrum, calculated to illustrate the band structure at both $P = 0.0$ and $P = 12.36$ GPa using all the hoppings in Eqs. 4.2 and 4.3 (“long range hoppings”). In Fig. 4.3 (a,b) similar results are presented but using only hoppings up to nearest neighbor rungs (“short range hoppings”). The band structures in both cases are similar, however, some discrepancies occur. For instance at the edges, such as $k_z=0$ and π , the short range hoppings present degeneracies (or near degeneracies) that are split in the long range case. It is unclear if these details are significant or not, and without performing the DMRG calculations in both cases explicitly this issue cannot be answered conclusively. Here, I simply wish to alert the readers of these small differences for completeness.

The electronic interaction portion of the Hamiltonian

$$\begin{aligned}
H_{int} &= U \sum_{i\gamma} n_{i\uparrow\gamma} n_{i\downarrow\gamma} + (U' - \frac{J}{2}) \sum_{\substack{i \\ \gamma < \gamma'}} n_{i\gamma} n_{i\gamma'} \\
&\quad - 2J \sum_{\substack{i \\ \gamma < \gamma'}} \mathbf{S}_{i\gamma} \cdot \mathbf{S}_{i\gamma'} + J \sum_{\substack{i \\ \gamma < \gamma'}} (P_{i\gamma}^\dagger P_{i\gamma'} + \text{h.c.}),
\end{aligned} \tag{4.4}$$

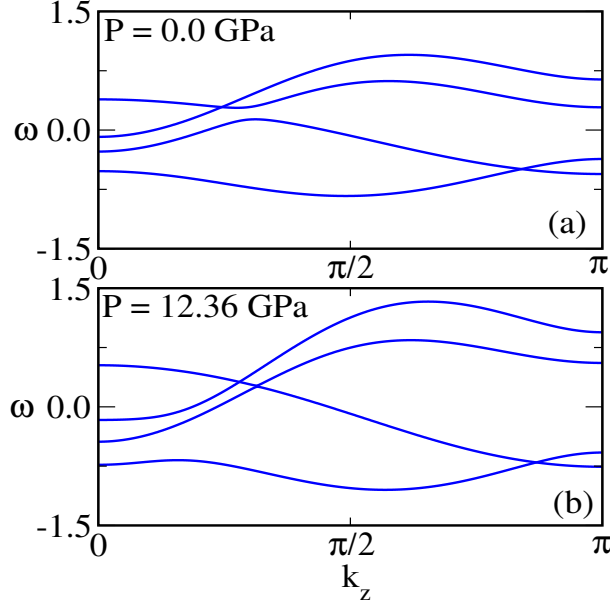


Figure 4.2: Tight-binding band structure ($U/W = 0.0$) involving hoppings up to the next-nearest neighbor rungs (*i.e.* long-range hoppings) for pressures of (a) 0.0 and (b) 12.36 GPa. The chemical potential is at zero energy for half-filling.

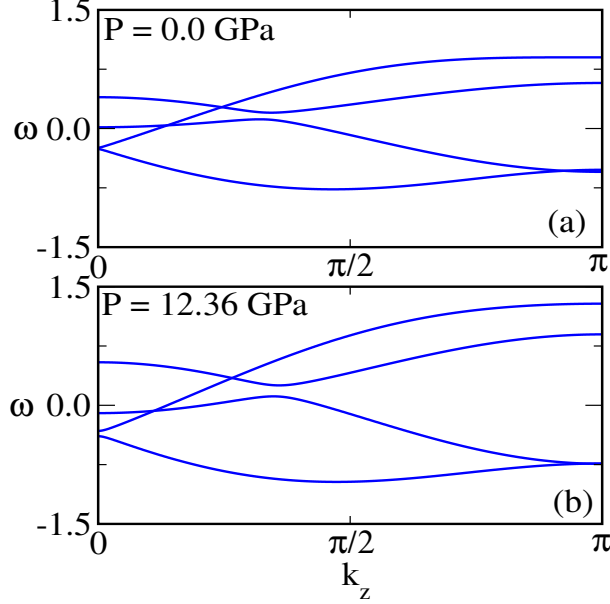


Figure 4.3: Tight-binding band structure ($U/W = 0.0$) involving hoppings only up to the nearest neighbor rungs (*i.e.* short-range hoppings) for pressures of (a) 0.0 and (b) 12.36 GPa. The chemical potential is at zero energy for half-filling.

contains the standard intra-orbital Hubbard repulsion U and the Hund's coupling J . The operator $\mathbf{S}_{i\gamma}$ ($n_{i\gamma}$) is the total spin (electronic density) for orbital γ at site i . $P_{i\gamma}^\dagger$ ($P_{i\gamma}$) are the pair creation (annihilation) operators. The standard relation $U' = U - 2J$ is assumed where we will fix the $J/U = 0.25$ and vary U as an independent parameter. The operators are defined in terms of the creation and annihilation fermion operators as

$$\mathbf{S}_{i\gamma} = \sum_{\sigma\sigma'} c_{i\sigma\gamma}^\dagger \sigma_{\sigma\sigma'} c_{i\sigma'\gamma}, \quad (4.5)$$

$n_{i\sigma\gamma} = c_{i\sigma\gamma}^\dagger c_{i\sigma\gamma}$, and $P_{i\gamma} = c_{i\downarrow\gamma} c_{i\uparrow\gamma}$. The half-filling electronic density corresponds to two electrons per site.

We use the ground state density matrix renormalization group (DMRG) technique with open boundary conditions in order to study the BaFe_2S_3 ladder using the two-orbital Hubbard model previously defined. DMRG grows the lattice by adding sites in a “snake-like” geometry. We have studied in detail a ladder size of 8×2 with up to four holes doped over a half-filled system. Calculations involving 12×2 and 16×2 ladders are also presented at half-filling. Keeping up to 800 states, the typical value of the discarded weight (truncation error) is of the order of 10^{-5} for the doping cases studied. Within this level of error, observables are converged. For the two holes case we can reach a similar accuracy only for 8×2 lattices, and for this reason our study of binding energies is restricted to those sizes. With typical computer resources, 8×2 ladder simulations with $m = 800$ states require 3-4 days at half-filling. The two holes doped case needs more than a week, even if using only short-range hoppings. The use of the long-range hoppings substantially increases the time required for convergence. This is because of three different reasons. First, the most time consuming part of the DMRG process is computing the Hamiltonian connections, and with long-range hoppings one has to sum a large number of terms in the Hamiltonian. Second, at fixed on-site interactions (U and J) and fixed density, the difficulty of the DMRG scales exponentially with the number of connections between system and environment, when a lattice is split in the middle. Third, our on-site Hilbert space is large due to the presence of two orbitals. In fact, the system we have studied can be translated into a one-dimensional one-orbital Hubbard model with hoppings up to 12^{th} neighbors. This illustrates the substantial numerical effort

presented here at the limit of what can be done with modern many-body computational techniques. We also want to remark that perhaps more modern versions of DMRG, such as those involving matrix product operators, may alleviate the effort needed in the present problem. In fact, recently $S = 1/2$ ladders including dipolar interactions were studied with up to 400 rungs with this method [70].

We will present a variety of charge and magnetic observables for doping of up to four holes on the half-filled system. The average occupation number of each orbital is

$$\langle n_\gamma \rangle = \frac{1}{N} \sum_{i,\sigma} \langle n_{i\sigma\gamma} \rangle. \quad (4.6)$$

We also calculate the spin-spin correlations by using the Fourier transform of the real space $\langle \mathbf{S}_i \cdot \mathbf{S}_j \rangle$,

$$S(k_z, k_y) = \frac{1}{N^2} \sum_{i,j} e^{-i\vec{k} \cdot \vec{r}_{ij}} \langle \mathbf{S}_i \cdot \mathbf{S}_j \rangle, \quad (4.7)$$

where $\mathbf{S}_i = \sum_\gamma \mathbf{S}_{i\gamma}$ (sum over the orbitals). Below, this spin structure factor will carry a subindex “L” or “S” depending on whether in the Hamiltonian the long-range or short-range hoppings are used, respectively.

To explore pairing tendencies, we study the binding energy of a pair of holes defined as [21]

$$\Delta E = E(N - 2) + E(N) - 2E(N - 1), \quad (4.8)$$

where $E(M)$ is the ground state energy of the model with a total of M electrons ($M = N$ is half-filling). If the particles minimize their energy by creating a bound state then ΔE is negative; if the holes become two independent particles this corresponds to zero binding energy in the bulk limit. In the case where the particles do not bind, this quantity is positive for finite systems.

To study the effects of holes on the magnetic correlations, we define a projector $P_{h\gamma}(i)$ at site i such that it projects out the portion of the ground-state in which site i and orbital γ is occupied [134]:

$$P_{h\gamma}(i) = c_{i\downarrow\gamma}^\dagger c_{i\downarrow\gamma} c_{i\uparrow\gamma}^\dagger c_{i\uparrow\gamma}. \quad (4.9)$$

In order to work in the Hilbert space corresponding to N_h number of holes at specific locations, we apply a product of projectors onto the ground state with N_h holes, $P_{h\gamma} = P_{h\gamma}(i_1)P_{h\gamma}(i_2)\dots P_{h\gamma}(i_{N_h})$, where i is the site to be projected while respecting the fermionic normal ordering ($i_1 < i_2 < \dots < i_{N_h}$). For example, $P_{ha} = P_{ha}(6)P_{ha}(8)$ projects out the occupied part of the ground state on orbital a at sites 6 and 8. In fact, for most results shown below, we only apply the projector onto orbital a in order to observe the corresponding local spin-spin correlations $\langle \psi | \mathbf{S}_{ia} \cdot \mathbf{S}_{ja} P_{ha} | \psi \rangle / \langle \psi | P_{ha} | \psi \rangle$, where the maximum possible magnitude of the correlations is $3/4$.

4.4 Results

This section presents our main results. We start with the half-filled case that should be contrasted with the experimental data for the two-leg BaFe_2S_3 at pressures where magnetic order was reported. *The magnetic order observed experimentally emerges very clearly from our calculations.* We then proceed to the addition of holes, under the assumption that the high pressure used experimentally moves bands around in such a manner that the two-leg ladders become effectively doped. Our main result is that indications of pairing are found in small systems, opening the possibility that indeed superconducting tendencies may be present in the models studied here.

4.4.1 Half-Filling

Figure 4.4 (a) shows the electronic population of the two orbitals calculated via DMRG as a function of U/W , for the two electrons per site, half filling case. Because of the crystal field splitting that locates orbital b approximately 0.7 eV below orbital a , in the weak coupling regime orbital b is considerably more populated. As the energy penalization for double occupancy increases with increasing U/W , eventually at $U/W \sim 1$ both orbitals become effectively singly occupied. These orbital populations are robust varying the lattice size and also using either the “short” or “long” version of the hopping amplitudes, as shown in Fig. 4.4 (a).

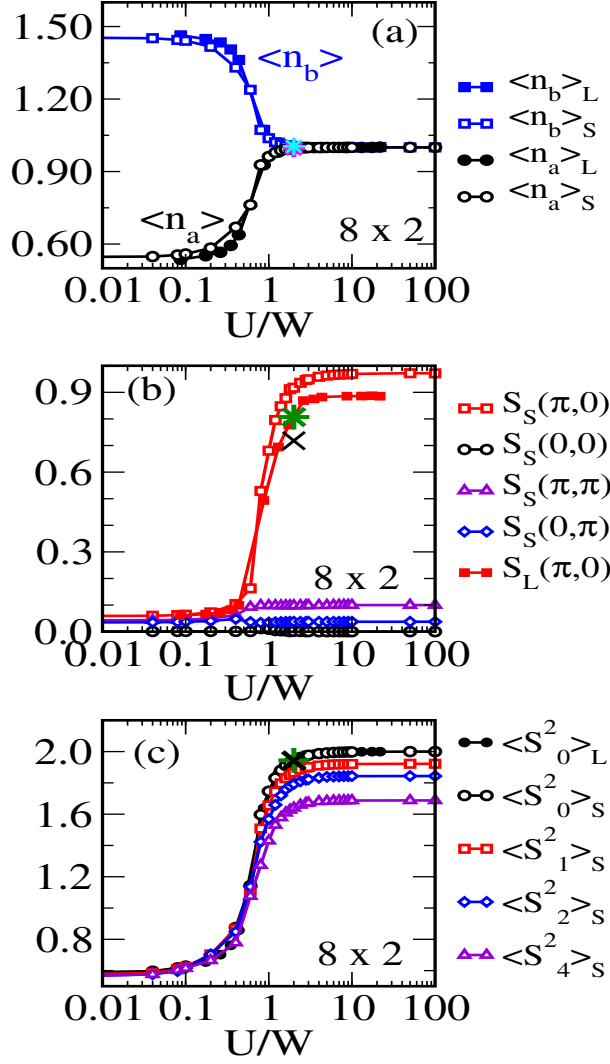


Figure 4.4: Charge and magnetic properties of a half-filled 8×2 ladder at $P = 12.36$ GPa (Eq. 4.3) studied with DMRG. Full (empty) points with subscript “L” (“S”) correspond to using long-range (short-range) hoppings. All results are at $J/U = 0.25$. (a) Average orbital occupation vs. U/W . Black (blue) color is for orbital a (b). The stars at $U/W = 2.0$ indicate results using a 12×2 lattice [cyan (magenta) for orbital b (a)]. Very similar results were obtained for a 16×2 lattice (not shown). (b) Fourier transform of the spin-spin correlations (*i.e.* spin structure factor) at representative wavevectors, indicating the dominance of $(\pi, 0)$ (see text). At $U/W = 2.0$, $S_S(\pi, 0)$ was also calculated using 12×2 (green star) and 16×2 (black X) lattices. $S_S(\pi, 0)$ slightly decreases with system size because the one dimensional nature of the lattice prevents long-range magnetic order. (c) $\langle S^2 \rangle$ vs. U/W , averaged over all sites. The subindexes 0, 1, 2, and 4 are the number of holes away from half-filling. The green star and black X are as in (b), suggesting small size effects. The convergence to 2 with increasing U/W at half-filling denotes a convergence to spin $S = 1$, as expected because J increases proportional to U . $\langle S^2 \rangle$ slightly decreases with increasing number of holes because of dilution effects.

Figure 4.4 (b) presents the spin structure factor at various wavevectors as a function of U/W . The wavevector $(\pi, 0)$ clearly dominates, particularly in the regime of intermediate and strong coupling. $S_S(\pi, 0)$ (see definition in caption of Fig. 4) starts growing already at $U/W \simeq 0.4$ even before the full moments are developed, an intermediate coupling regime that several investigations assign to the iron based superconductors [27]. Once again, these results are robust increasing the lattice size and using either “short” or “long” hopping amplitudes. The small decrease of $S_S(\pi, 0)$ in Fig. 4.4 (b) with increasing clusters from 8×2 to 16×2 is reasonable because a true long-range order is not expected in one dimension, but a slow power-law decay should instead prevail.

The dominance of the $(\pi, 0)$ magnetic order is in excellent agreement with neutron experiments for BaFe_2S_3 [122]. In our model, this magnetic order dominance arises primarily from the comparable strength of the hopping amplitudes t^{y+z} along the diagonal of the elementary plaquettes contrasted with those along the nearest-neighbor sites along the rungs and legs. This comparable strength originates in the location of sulphur, that acts as a bridge between irons, up and down the middle of the ladder plaquettes. This is also the same reason for the dominance of the (degenerate) $(\pi, 0)$ and $(0, \pi)$ wavevectors in planar geometries, at intermediate and strong couplings.

In two-leg ladders the explicit breaking of the lattice rotational invariance renders $(\pi, 0)$ and $(0, \pi)$ no longer degenerate. But why $(\pi, 0)$ dominates over $(0, \pi)$ according to the DMRG calculations? A possible simple explanation is the following. Consider a classical J_1 - J_2 spin model for spins of magnitude 1, where J_1 is the antiferromagnetic Heisenberg coupling for nearest-neighbors spins both along the rungs and legs, while J_2 is the antiferromagnetic coupling along the plaquette diagonals. The energy of the $(\pi, 0)$ state is always smaller than the energy of the $(0, \pi)$ because in $(\pi, 0)$ each spin always has two nearest-neighbors AFM links, while in $(0, \pi)$ there is only one nearest-neighbor AFM link. While at small J_2/J_1 the (π, π) order dominates as expected, a level crossing to $(\pi, 0)$ eventually occurs at $J_2/J_1 = 0.5$. This also provides a possible rationale for why (π, π) rather than $(0, \pi)$ appears to be the subdominant order in Fig. 4.4 (b): in these two-orbital Hubbard models for two-leg ladder materials the ratio J_2/J_1 between the effective Heisenberg couplings in strong coupling must be between 0.5 and 1.0.

Figure 4.4 (c) plots the spin squared expectation value as a function of U/W , showing the formation of local moments. The upturn with increasing U/W occurs at values similar to those where $S(\pi, 0)$ starts growing. Eventually at strong coupling, $U/W > 1$, the spins are fully developed and they acquire their maximum value $S = 1$ *i.e.* a magnetic moment $2.0 \mu_B$. Neutron scattering experiments at ambient pressure [122] report a moment of $1.2 \mu_B$ ($S \sim 0.6$), which we find at $U/W \simeq 0.5$; first principles predict a value of $2.0 \mu_B$ ($S \sim 1.0$) at the same pressure [121]. Note that neutron scattering may be capturing a moment that is time averaged, thus reducing its value, and other techniques should be used to find the actual instantaneous spin [12, 44]. Also note that comparing magnetic moment results of a two orbital model vs. calculations and experiments involving five orbitals is difficult. Regardless, intermediate to strong coupling is the physically relevant regime in this model from the magnetic moment perspective.

4.4.2 One Hole Doped

Figure 4.5 shows results for the case of one hole doped into the half-filled system. Panel (a) displays the population of each orbital. As at half-filling, the crystal field splitting induces a large difference at weak coupling between the two orbitals. However, it is curious to observe that in the strong coupling regime the hole is still almost entirely located at orbital a , in spite of the presence of a gap induced by the repulsion U . Nevertheless, since the U is the same for both orbitals, the only asymmetry between the orbitals is the original crystal field splitting that, therefore, must be inducing the asymmetric population with holes of the a orbital.

This strong-coupling Wannier orbital population, where one orbital is locked at one electron/site and the other at less than one electron/site, corresponds to an orbital selective Mott phase (OSMP) [102, 41, 62]. In this context orbital b provides localized spins $S = 1/2$, that are in interaction with delocalized carriers at orbital a . The physics of the OSMP state suggests that this state, if realized in the present two-leg ladders, may have exotic transport properties that include a very small quasiparticle weight.

Figure 4.5 (b) plots the spin structure factor. As in the case of half-filling, clearly the wavevector $(\pi, 0)$ dominates starting at $U/W \sim 0.4$, and irrespective of using “short” or

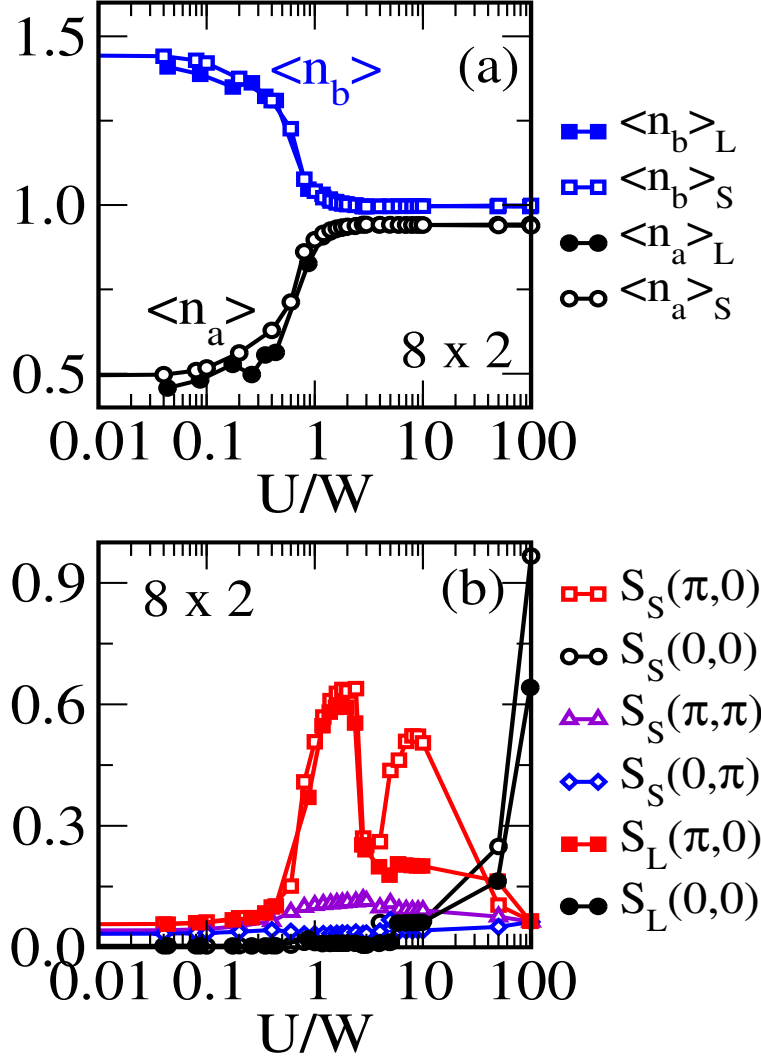


Figure 4.5: Charge and magnetic properties of an 8×2 ladder doped with one hole, at $P = 12.36$ GPa (Eq. 4.3) and studied with DMRG. Full (empty) points with subscript “L” (“S”) correspond to using long-range (short-range) hoppings. All results are at $J/U = 0.25$. (a) Average orbital occupancy vs. U/W . Black (blue) color is for orbital a (b). At small U/W the crystal field creates a substantial difference in the populations. At large U/W the b orbital converges approximately to one electron/site, while the a orbital contains most of the doped hole. The results are approximately the same for L and S hoppings. (b) Fourier transform of the spin-spin correlations (*i.e.* spin structure factor) at various representative wavevectors, indicating the dominance of $(\pi, 0)$. At very large U/W the one hole state becomes ferromagnetic due to double exchange tendencies, as discussed in the text.

“long” range hoppings. The dip at $U/W \sim 3$ is unexpected and it may reflect on how the hole scrambles the original magnetic order as the size of the spin distortion around the hole changes with U/W . This spin scrambling effect can be better visualized in Fig. 4.6 where from the entire wave function of the one-hole state, a projection is made for the case where the hole is located at the sites indicated. While far from the projected hole the spin order is basically unchanged from the half-filled $(\pi, 0)$ pattern, in the vicinity of the hole there is an inevitable scrambling effect that broadens the $(\pi, 0)$ peak. This shift of weight away from π along the leg direction is exemplified by the antiferromagnetic coupling “across the hole” involving *e.g.* spins 4 and 8 on the 8×2 lattice that otherwise should be ferromagnetically coupled. The 12×2 results in the same panel indicate very small size effects. This across-the-hole AFM coupling has been observed in the $t - J$ model context before [74, 72, 73], and it is considered a precursor of spin-charge separation at least at short distances. In fact, the exact ground state of the $U = \infty$ one-orbital Hubbard model in one dimension presents an exact decoupling between spin and charge with AFM couplings across all holes [90].

We warn the readers that there are some qualitative differences between the cases of short and long range hoppings. For instance in Fig. 4.5 (b) $S_S(\pi, 0)$ has a “second peak” at $U/W \sim 10$ which is suppressed in $S_L(\pi, 0)$. We do not know the qualitative reasons for this difference. However, in the important region of pair binding $U/W \sim 2$, to be described later in the text, both short and long range hoppings give very similar results.

An interesting observation from Fig. 4.5 (b) is that at very large U/W eventually the one hole state becomes ferromagnetic since $S(0, 0)$ dominates. In multiorbital systems, especially in cases where some degrees of freedom are localized and others itinerant as it occurs in this model, double exchange mechanisms can favor ferromagnetic tendencies as it occurs in manganites [24]. In the large U/W regime, the effective Heisenberg couplings J_1 and J_2 are very small since they are inversely proportional to U , while the Hund coupling being fixed to $J/U = 0.25$ is very large. Such a regime is clearly favorable for double exchange tendencies, as shown by the DMRG results. This also indicates that ferromagnetic states are close in parameter space to the realistic regimes for iron superconductors, a conclusion that also emerged from previous investigations [66, 65, 69].

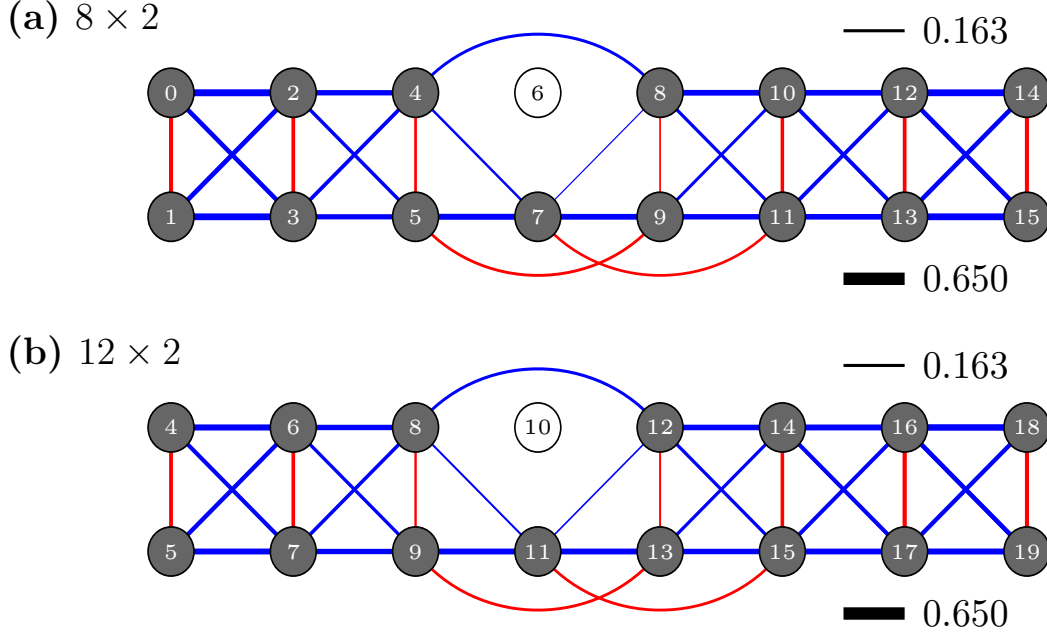


Figure 4.6: Results obtained from the wave function of a dynamical hole at $P = 12.36$ GPa (Eq. 4.3), using $U/W = 2$ and $J/U = 0.25$, for the case when the hole is projected on orbital a at the location denoted by the white circles (see Eq. 4.9). (a) are results for the 8×2 lattice and (b) are for the 12×2 lattice, showing that size effects are small. The thickness of the lines is linearly proportional to the magnitude of the spin-spin correlations involving orbital a . These correlations between spins at sites m and j are defined as $\langle \psi | \mathbf{S}_{ma} \cdot \mathbf{S}_{ja} P_{ha}(i) | \psi \rangle / \langle \psi | P_{ha}(i) | \psi \rangle$, where $P_{ha}(i)$ was defined in the text. Blue denotes antiferromagnetic correlation while red is ferromagnetic. In (a) $i = 6$, and in (b) $i = 10$. Magnetic correlations away from the hole are very similar to those in the undoped case. In both panels (a) and (b) in the vicinity of the hole a weak antiferromagnetic correlation between spins “across the hole” location can be clearly observed. For a discussion see the text.

4.4.3 Two Holes Doped

The results for two doped holes shown in Fig. 4.7 continue the trends observed before for one hole. Panel (a) shows the Wannier orbital populations as a function of U/W . As for one hole, at large U/W the orbital b population remains locked at one electron/site, while the two holes almost entirely reside at orbital a . This confirms the tendency towards an OSMP state with doping. With regards to the spin magnetic order, panel (b), the $(\pi, 0)$ order still dominates in the broad region between $U/W = 0.4$ and 10, but the spin order scrambling caused by the mobile holes reduces the intensity of $S(\pi, 0)$ as expected. In addition, the tendency towards ferromagnetism triggered by double exchange continues at very large U/W .

4.4.4 Binding Energy

After calculating the ground state energies for the N , $N - 1$ (1 hole), and $N - 2$ (2 holes) subspaces, we can also calculate the binding energy ΔE previously defined. The remarkable result shown in Fig. 4.8 (a) is that for the 8×2 cluster this quantity becomes negative between $U/W \sim 1.5$ and $U/W \sim 4.5$. This is a broad region, in spite of the perceived narrowness in panel (a) because of the logarithmic scale used. In this regime the spins are already well developed and near saturation as it was shown in Fig. 4.4 (c). Considering that holes are located at orbital a , this surprising result brings similarities with negative binding energies found in one-orbital models for the cuprates, such as the $t - J$ model [21]. In fact, the crude rationale for binding based on the “number of broken AFM links” may apply here as well [21]. In this context, binding occurs because each hole damages the AFM spin state, and the manner to minimize the size of that distorted magnetic background is by bringing the holes together. It is also interesting that exact results obtained via the Lanczos method applied to a very small 3×2 lattice produce a profile for the binding energy, shown in Fig. 4.8 (b), that qualitatively resembles panel (a) suggesting that size effects are mild. Alas, as already explained, we have not been able to reach sufficient accuracy in the two holes sector to confirm the pairing tendencies of Fig. 4.8 (a) with larger lattices, thus our pairing analysis below is restricted to the 8×2 cluster

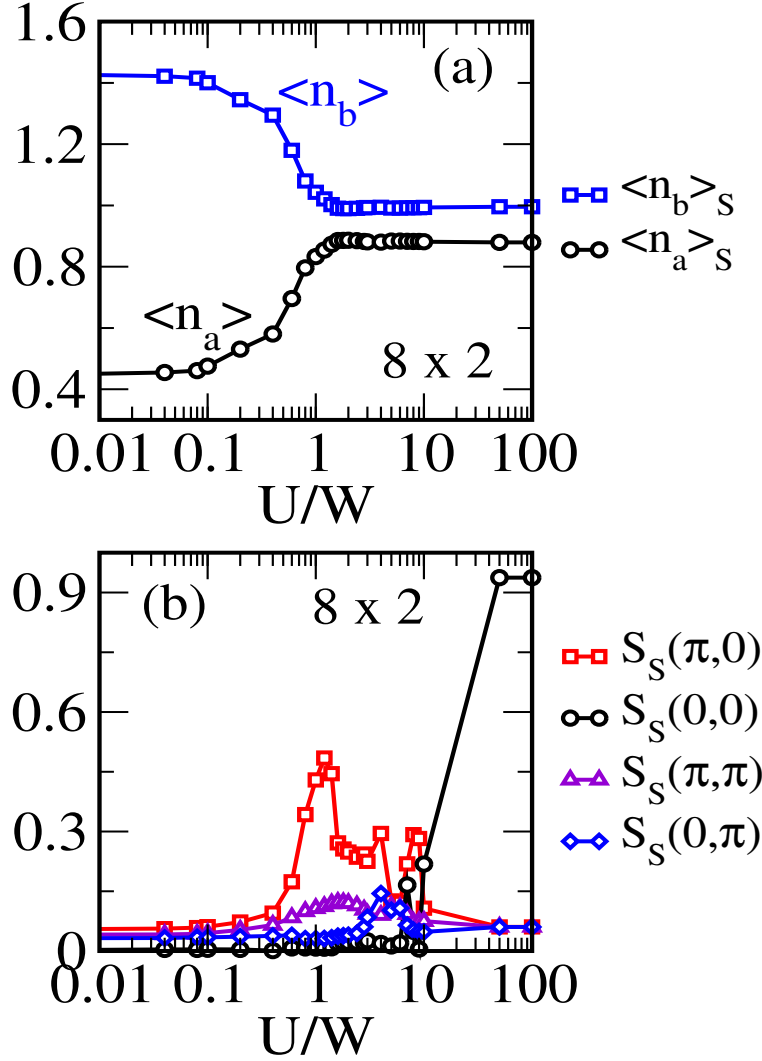


Figure 4.7: Charge and magnetic properties of an 8×2 ladder doped with two holes, at $P = 12.36$ GPa (Eq. 4.3), using short range (“S”) hoppings, and studied with DMRG. All results are at $J/U = 0.25$. (a) Average orbital occupancy vs. U/W . Black color is for orbital a and blue for orbital b . At small U/W the crystal field creates a substantial difference in the populations. At large U/W , the b orbital converges approximately to one electron/site, while the a orbital contains most of the doped holes. (b) Fourier transform of the spin-spin correlations (*i.e.* spin structure factor) at representative wavevectors, indicating the dominance of $(\pi, 0)$ at intermediate/large U/W . At very large $U/W > 10$ the two holes state becomes ferromagnetic due to double exchange tendencies, as discussed in the text and as for one hole.

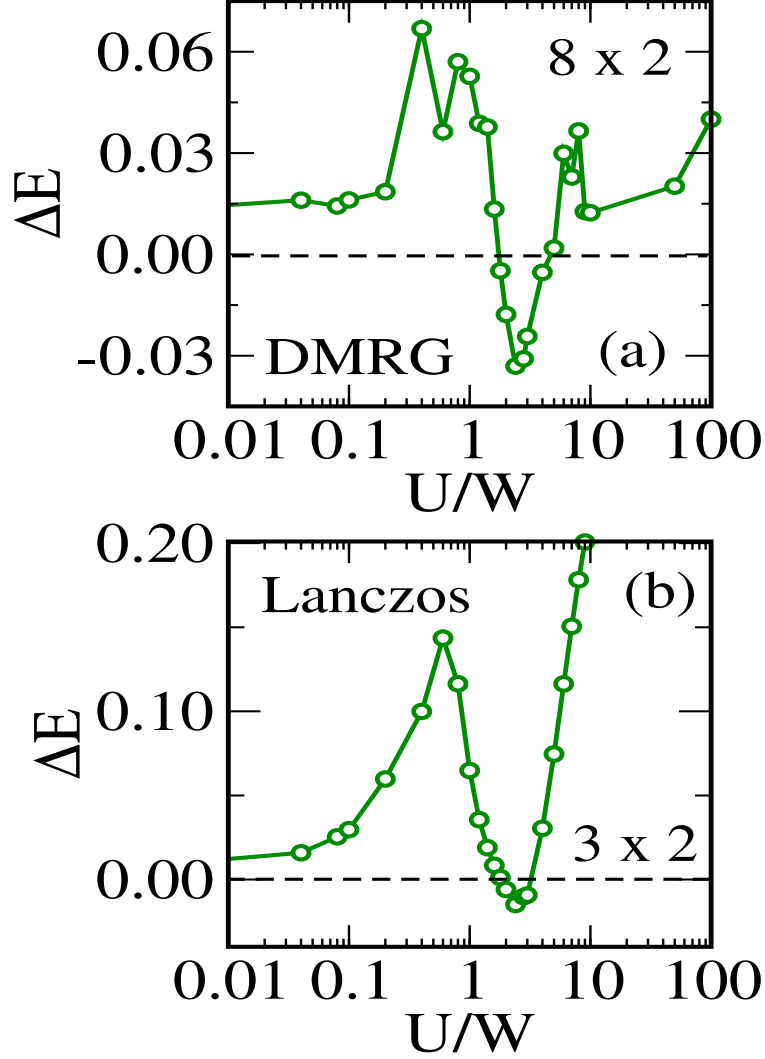


Figure 4.8: Binding energy vs. U/W calculated using (a) DMRG for an 8×2 ladder and (b) Lanczos for a 3×2 ladder. In both cases, we observe a non-monotonic up-down-up behavior where the minimum of the binding energy can be found at $U/W \sim 2$. Since this minimum of ΔE is negative, panel (a) suggests binding of holes between $U/W \sim 1.5$ and $U/W \sim 4.5$. The results in both panels were obtained at $P = 12.36$ GPa (Eq. 4.3), using short-range hoppings, and $J/U = 0.25$.

The results in figure 4.8 (a) suggesting pairing in a region of parameter space brings analogies with the negative binding energies reported before in Kondo lattice models for heavy fermions [138]. For instance in Fig. 2 (b) of Ref. [138] a negative ΔE is reported using up to 32×2 lattices. Even the pair-pair correlation functions of Fig. 3 (b) of Ref. [138] (unfortunately not within the reach of the present study that uses the full two-orbital Hubbard model) suggest a dominant pairing tendency in the doped Kondo lattice on two-leg ladders. Perhaps having holes primarily at orbital a (as shown before), while orbital b remains singly occupied at strong coupling, effectively transforms our model into a Kondo lattice model.

To further test the pairing implication of finding a negative binding energy in Fig. 4.8, we have also analyzed the real space distribution of holes in the doped system. In Fig. 4.9 (a), the electronic density is shown for orbital a , where the holes are mostly located, for each of the 16 sites of the 8×2 lattice at coupling $U/W = 2$ where ΔE is negative. For N electrons, *i.e.* half-filled, the electronic density is basically uniform. In the case of $N - 1$ electrons, *i.e.* one hole, this hole is located in the middle of the cluster as expected for a system with open boundary conditions. For the case of two holes corresponding to $N - 2$ electrons, these two holes are also located near the center of the cluster but in a tight manner compatible with pairing. The most important result is for the case of four holes, corresponding to $N - 4$ electrons, since Fig. 4.9 (a) indicates the presence of *two* minima in the electronic density, a result compatible with the presence of two hole pairs, as opposed to a single broad minimum which would indicate independent holes or four minima which would signal a charge density wave of holes. There are also no indications of phase separation. The results in Fig. 4.9 (b) for orbital b simply mirror those of orbital a but with a far more suppressed hole density.

Figure 4.10 (a) illustrates the internal structure of the hole pairs that we have found using the 8×2 cluster. This figure is based on the wave function of two holes, with one of the holes projected to site “9” which is on “Leg-1”. This panel shows that the second hole is primarily located on the other leg, *i.e.* “Leg-0”, mainly at the sites either in the same rung as “9” or diagonally across the plaquettes. Projecting now the two holes to those particular locations, these two dominant “plaquette diagonal” and “rung” states for the pair of holes are shown in Figs. 4.10 (b) and (c), respectively. Similarly as for the case of one hole, there is

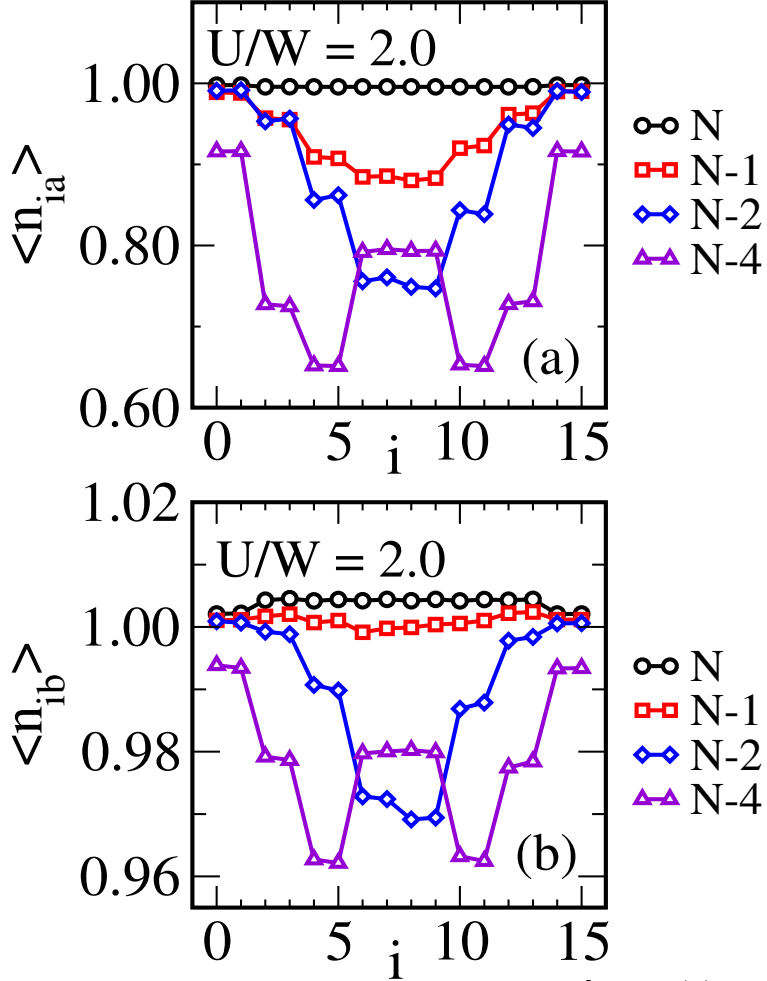


Figure 4.9: Real space electronic density of each orbital [panel (a) is for orbital a , while panel (b) for orbital b] using an 8×2 ladder, $U/W = 2.0$, $J/U = 0.25$, short-range hoppings, and at $P = 12.36$ GPa (Eq. 4.3). Results are shown for half-filling (N electrons), one hole ($N - 1$), two holes ($N - 2$), and four holes ($N - 4$) as a function of the position “ i ” (see Fig. 4.6 (a) for the site labeling convention). The most striking result corresponds to four holes where the presence of two minima is indicative of hole pairing. Doping of four holes reduces the orbital a electron density by approximately 25%, while orbital b has a charge depletion of only $\sim 3\%$, illustrating again that holes mainly reside at orbital a .

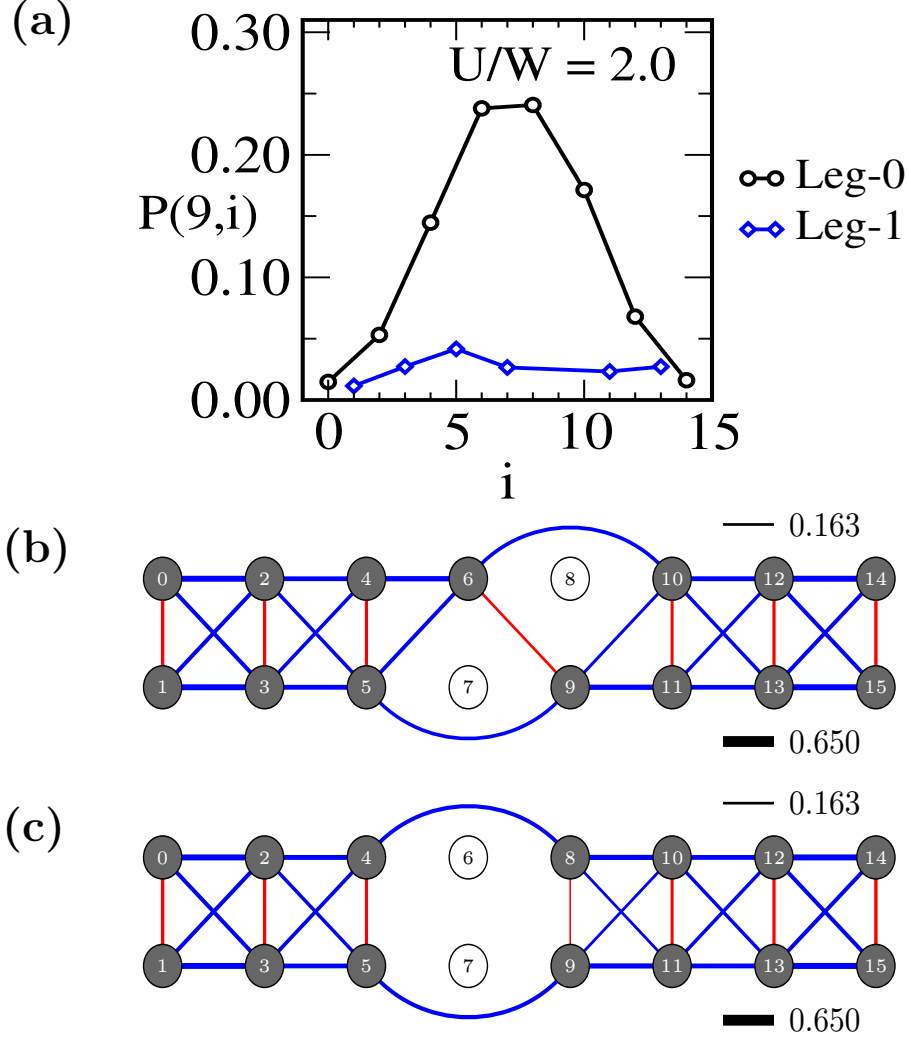


Figure 4.10: (a) For the case of the two holes ground state on an 8×2 ladder, this panel shows the probability of a hole to be located at a site “ i ” assuming the other hole is fixed at site 9 of the bottom leg (Leg-1). Results are normalized to one. Since sites are labeled with a snake-like geometry where site zero starts from the upper leg (Leg-0), this panel indicates that the two holes in the bound state are primarily located in different legs. See Fig. 4.6 (a) for site labelling details. (b,c) Results from the DMRG ground state wave function of two holes on an 8×2 cluster, using $U/W = 2.0$ (binding region) and $J/U = 0.25$, and with short-range hoppings at $P = 12.36$ GPa (Eq. 4.3). Shown are spin-spin correlations for the case where the two holes are projected to be at the white circles, *i.e.* (b) along a plaquette diagonal and (c) along a rung. These are the two most dominant configurations in the hole pair. The spin-spin magnetic correlations are defined as $\langle \psi | \mathbf{S}_{ma} \cdot \mathbf{S}_{na} P_{ha}(i) P_{ha}(j) | \psi \rangle / \langle \psi | P_{ha}(i) P_{ha}(j) | \psi \rangle$, involving only orbital a because it is the primary location for the doped holes. In (b) $i = 7$ and $j = 8$ and in (c) $i = 6$ and $j = 7$. Blue (red) lines are AFM (FM) bonds. For all hole configurations, the $(\pi, 0)$ magnetic order is substantially distorted only near the holes. Note also the presence of “across the hole” AFM correlations in both panels.

a notorious “across the hole” antiferromagnetic coupling between spins that otherwise should be ferromagnetically coupled in the undoped system. This AFM correlation facilitates the movement of the hole. Note also that if in panel (b) the hole located at “8” and the spin at “6” are interchanged, as it would happen via the action of electronic hopping and assuming that the AFM and FM bonds remain the same as if they were elastic bands, then panel (c) is obtained. In fact, this panel (c) has an AFM across-the-hole coupling between “4” and “8” and a FM coupling between “8” and “9” that was originally a FM coupling along the diagonal from “6” to “9” in panel (b). Then panels (b) and (c) are compatible with one another with regards to hole pairing: the two holes are oscillating in different legs close to one another due to an attraction created by the antiferromagnetic background.

4.5 Conclusions

In this chapter, we have presented the first study of a realistic (derived from first principles) electronic model Hamiltonian for the two-leg ladder compound BaFe_2S_3 that was recently shown to become superconducting at high pressure [122, 141]. The model has two orbitals and electronic hoppings beyond nearest neighbor iron sites, rendering its study difficult even with the powerful DMRG method. For this reason our analysis has been restricted to relatively small clusters. Nevertheless, we have been able to extract interesting information from the model that is in good agreement with experiments. For example, the parent compound has magnetic order involving ferromagnetic rungs that are coupled antiferromagnetically along the legs, as found in neutron scattering experiments [122]. In the strong coupling limit, this order emerges from the competition between antiferromagnetic Heisenberg couplings along rungs and legs and along the diagonals of the plaquettes. With hole doping, we observed that only one of the two Wannier orbitals used here becomes populated. This indicates a tendency towards effective models involving a combination of itinerant and localized orbitals, as in the context of an orbital selective Mott phase. Even more exciting, we have found that at strong coupling and using an 8×2 cluster with two holes, there are indications of hole pair formation induced by antiferromagnetism. While this result must be confirmed using larger systems and more DMRG states, a challenging

task, it suggests that this type of two-orbital models contains the essence of the mechanism for superconductivity in iron-based two-leg ladders, a mechanism that could be similar to that in layered systems. As a consequence, our present effort paves the way and motivates further studies in this context. We believe that the theoretical and experimental analysis of iron-based two-leg ladders may prove to be as interesting and illuminating as the early studies in copper oxide two-leg ladders were for cuprate physics, providing a novel playground in the context of iron-based high- T_c superconductivity.

Chapter 5

Pairing Tendencies in a Two-orbital Hubbard Model in One Dimension

This chapter is a modified version of Physical Review B **96**, 024520 (2017) [94].

5.1 Introduction

In this chapter, we study a two-orbital model that is mathematically similar to that used before for BaFe_2S_3 , but now defined simply on a *chain* as opposed to a two-leg ladder using DMRG. When the binding energy is calculated vs. U/W the result, to be shown below, is similar to that found in the case of the two-leg ladder, with binding observed at intermediate couplings and with a shape of the binding energy curve vs. U/W , resembling that previously reported. Due to this similarity, it is reasonable to believe that common physics causes the pairing tendencies both in ladders and in chains as long as two orbitals are active. The advantage of using chains, of course, is that much longer systems can be analyzed thus reducing size effects. For all these reasons, in this chapter a systematic study of a two-orbital model defined on chains is presented, with emphasis on pairing and superconducting tendencies. The analysis is presented in a systematic manner, varying the many couplings and electronic densities and even further boosting pairing tendencies by introducing extra Heisenberg interactions. Overall, our analysis concludes that it is the Hund coupling J_H that primarily drives the pairing tendencies, supplemented by AFM tendencies between the

effective $S = 1$ spins of the undoped sites. As explained below, it is known that there are unrealistic ranges of couplings where J_H explicitly boosts hole attraction. What is remarkable of our results described here is that *similar tendencies survive into the realistic regime $J_H/U = 0.25$, where the model is not explicitly attractive* because of the competition between J_H with the interorbital repulsion U' . These promising results are preliminary steps towards a clarification of the origin of pairing in iron-based superconductors, but more work is needed to establish definitely that pairing of electronic origin is active in two-orbital Hubbard models.

The organization of this chapter is as follows. Section 5.2 provides details of the model, technique used, and observables measured. Section 5.3 contains the main results, addressing both magnetic and pairing properties of the model under scrutiny. Section 5.4 contains our main discussion and conclusions.

5.2 Model and Method

The multiorbital Hubbard model used in this chapter is defined as

$$\begin{aligned}
H = & -t \sum_{\substack{\langle ij \rangle \\ \gamma \sigma}} (c_{i\gamma\sigma}^\dagger c_{j\gamma\sigma} + h.c.) + U \sum_{i\gamma} n_{i\gamma\uparrow} n_{i\gamma\downarrow} \\
& + (U' - \frac{J_H}{2}) \sum_{\substack{i \\ \gamma < \gamma'}} n_{i\gamma} n_{i\gamma'} - 2J_H \sum_{\substack{i \\ \gamma < \gamma'}} \mathbf{S}_{i\gamma} \cdot \mathbf{S}_{i\gamma'} \\
& + J_H \sum_{\substack{i \\ \gamma < \gamma'}} (P_{i\gamma}^\dagger P_{i\gamma'} + h.c.),
\end{aligned} \tag{5.1}$$

where $c_{i\gamma\sigma}^\dagger$ ($c_{i\gamma\sigma}$) creates (annihilates) an electron at site i of a chain, with orbital γ (either a or b), and spin projection along the z -axis σ . The first term represents the kinetic energy of the electrons. Note that for simplicity, the 2×2 hopping matrix is the unit matrix *i.e.* only hoppings between the same orbitals are allowed. Although our overarching goal is the understanding of iron-based superconductors, these hoppings are not intended to represent the tunneling amplitudes of any particular material but they are chosen for simplicity. The second is the standard on-site Hubbard repulsion U between spins $\uparrow$ and $\downarrow$ electrons. The

third term is the repulsion between electrons at different orbitals. As shown in previous chapter, besides the canonical U' repulsion the coupling strength affecting this term contains a contribution regulated by the Hund coupling J_H . Fourth is the portion that explicitly shows the ferromagnetic character of the Hund interaction. The last term is the pair hopping. The number operator is defined as $n_{i\sigma\gamma} = c_{i\sigma\gamma}^\dagger c_{i\sigma\gamma}$ and the pair operator as $P_{i\gamma} = c_{i\gamma\uparrow} c_{i\gamma\downarrow}$. The standard relation $U' = U - 2J_H$ is assumed. While many of the results are for $J_H/U = 0.25$, considered realistic and used in the previous publication for ladders [93], in some of the results below the Hund coupling is varied. The bandwidth corresponding to the kinetic energy portion is $W = 4t$ and the Hubbard strength will be provided primarily as U/W . The hopping is the unit of energy $t = 1.0$, unless stated otherwise.

To obtain our results we use the DMRG technique with open boundary conditions with focus on the ground-state of the two-orbital chain, employing at least 1600 states. Most of the results are for a 32 site two-orbital chain, unless stated otherwise, while some of the results were confirmed using up to 64 sites. Truncation error remain below $\sim 10^{-6}$ for all of our results.

We have measured several observables. The binding energy that is an indicator for pairing tendencies [21] is defined as

$$\Delta E = E_N + E_{N-2} - 2E_{N-1}, \quad (5.2)$$

where E_N , E_{N-1} , and E_{N-2} are the total ground state energy of the half-filled, 1-hole doped, and 2-hole doped systems. Here, $N = 2L$ with L the length of the chain. The real space charge and spin correlations are

$$N(R) = \frac{1}{N_R} \sum_{|i-j|=R} [\langle n_i n_j \rangle - \langle n_i \rangle \langle n_j \rangle], \quad (5.3)$$

$$Sp(R) = \frac{1}{N_R} \sum_{|i-j|=R} \langle \mathbf{S}_i \cdot \mathbf{S}_j \rangle, \quad (5.4)$$

where N_R is the number of neighbours at distance R from site i (namely, averages over pairs of sites at equal distance are performed). The Fourier transform of $Sp(R)$ is the spin structure factor $Sp(k)$

To study the effects of holes on the magnetic correlations, we define a projector operator $\mathcal{P}_{i\gamma}$ that projects out the portion of the ground state where site i and orbital γ are occupied [134]:

$$\mathcal{P}_{i\gamma} = c_{i\gamma\uparrow}^\dagger c_{i\gamma\uparrow} c_{i\gamma\downarrow}^\dagger c_{i\gamma\downarrow}. \quad (5.5)$$

To work in the Hilbert space corresponding to n_h number of holes at specific locations, we apply a product of projectors onto the ground state with n_h holes. For example, $\mathcal{P}_{6a}\mathcal{P}_{9b}|\psi_{N-2}\rangle$ projects out the occupied part of the two-hole ground state on orbital a at site 6, and on orbital b at site 9. We also calculate the local spin-spin correlations $\langle\psi|\mathbf{S}_{i\gamma} \cdot \mathbf{S}_{j\gamma'}\mathcal{P}|\psi\rangle/\langle\psi|\mathcal{P}|\psi\rangle$, where the maximum possible magnitude of the correlations is $3/4$.

There are many possible superconducting pair correlations that one can explore for this system. Due to the local inter- and intra-orbital Coulomb repulsion, on-site pairing is not expected to dominate [21]. Thus, pairing operators for two electrons at nearest-neighbor (NN) sites i and $i+1$ will be considered in analogy with the approach taken in other purely electronic models where magnetic properties trigger pairing.

An intraorbital nearest neighbor pairing operators is

$$\Delta_{nn,-}^{\gamma\gamma\dagger}(i) = c_{i,\gamma,\uparrow}^\dagger c_{i+1,\gamma,\downarrow}^\dagger - c_{i,\gamma,\downarrow}^\dagger c_{i+1,\gamma,\uparrow}^\dagger, \quad (5.6)$$

which creates a pair of electrons at nearest neighboring sites i and $i+1$ in orbital γ and forming a spin singlet. The corresponding pairing operator for the case in which the two electrons form a spin triplet is given by

$$\Delta_{nn,+}^{\gamma\gamma\dagger}(i) = c_{i,\gamma,\uparrow}^\dagger c_{i+1,\gamma,\downarrow}^\dagger + c_{i,\gamma,\downarrow}^\dagger c_{i+1,\gamma,\uparrow}^\dagger. \quad (5.7)$$

Since the Hamiltonian is invariant under an orbital exchange only orbital symmetric combinations of the intraorbital pairing correlations have to be considered. An interorbital

nearest neighbor pairing operator is given by

$$\Delta_{nn,-}^{ab\dagger}(i) = c_{i,a,\uparrow}^\dagger c_{i+1,b,\downarrow}^\dagger - c_{i,a,\downarrow}^\dagger c_{i+1,b,\uparrow}^\dagger, \quad (5.8)$$

which creates two electrons at site i and orbital a and site $i+1$ and orbital b , forming a spin singlet (the orbital exchanged pairing operator is identical due to the orbital symmetry). The interorbital pairing operator that creates the electrons in a triplet state is given by

$$\Delta_{nn,+}^{ab\dagger}(i) = c_{i,a,\uparrow}^\dagger c_{i+1,b,\downarrow}^\dagger + c_{i,a,\downarrow}^\dagger c_{i+1,b,\uparrow}^\dagger. \quad (5.9)$$

The pair-pair correlations are given by

$$\mathcal{O}_{nn,\pm}^{\gamma\gamma'}(R) = \frac{1}{2N_R} \sum_i \langle \Delta_{nn,\pm}^{\gamma\gamma'\dagger}(i) \Delta_{nn,\pm}^{\gamma\gamma'}(i+R) \rangle, \quad (5.10)$$

where $\pm$ indicates if the pair is a spin triplet or singlet, and γ and γ' indicate orbitals a or b . Since the results explicitly presented in this manuscript are for the interorbital spin singlet $\mathcal{O}_{nn,-}^{ab}(R)$ and triplet correlations $\mathcal{O}_{nn,+}^{ab}(R)$, below we will use the notation $\mathcal{O}_{nn,-}^{ab}(R) \equiv S_{nn}^{ab}(R)$ and $\mathcal{O}_{nn,+}^{ab}(R) \equiv T_{nn}^{ab}(R)$, respectively. Analogous on-site pairing operators were also considered but their correlations always decayed faster than the dominant NN sites pair-pair correlation ($S_{nn}^{ab}(R)$ as shown below). For this reason, the actual expressions for on-site operators are not provided explicitly.

We have measured other observables as well. For example, by averaging the pair correlations over a finite intermediate portion of the chain we can reduce short distance effects, that sometimes lead to believe that pairing is dominant even if the long distance tail is small, and also to reduce boundary effects caused by the open boundary conditions. Here we define the pairing strength as

$$\bar{D} = \sum_{R=7}^{12} |S_{nn}^{ab}(R)|, \quad (5.11)$$

where we have used the spin-singlet nearest-neighbor combination explicitly because it will be shown below that it is dominant in our study.

5.3 Results

In this section, the main results will be described. The language to be used below should always be considered in the framework of one dimensional systems where long-range order is not possible. For example, expressions such as “staggered AFM order” indicate that staggered spin arrangements decay the slowest with distance as compared with other patterns, but eventually all correlation functions decay to zero with increasing distance in one dimension with short-range interactions.

As expressed before, we remind the readers that experimentally in the Fe-ladders superconductivity appeared with increasing pressure, not with explicit hole doping. However, similarly as in the case of the Cu-ladders, it is believed that pressure may lead to a rearrangement of charge, particularly with regards to the average number of electrons at the iron atoms. This perception is supported by recent *ab-initio* calculations [146]. As a consequence, in our effort described below, we will search for pairing indications by doping with holes the half-filled system, rather than modelling pressure directly.

5.3.1 Magnetic order and local moments

Let us start our computational analysis of the two-orbital Hubbard model defined in the previous Section by focusing on the magnetic order. Figure 5.1 contains the spin structure factor at $U/W = 1.60$, a coupling strength of much importance for pairing as shown below, for different number of holes. For the case of half-filling, $N = 64$ electrons for the 32-sites chain of focus in Fig. 5.1, the spin order is clearly of the staggered antiferromagnetic (AFM) form as expected. In this regime of Hubbard couplings the local spin at every site is already well developed and close to the spin-1 limit, as shown in panel (a) of Fig. 5.2 for most Hund couplings studied, with the exception of $J_H = 0$. Thus, the AFM correlations are compatible with the spin correlations of a Haldane spin-1 chain. Our study of a two-orbital Hubbard model, instead of a Heisenberg model, involves energy scales much higher than those typical of the integer-spin chains and for this reason we will not focus on subtle issues such as spin-gaps in the system. Panel (b) of Fig. 5.2 shows that robust AFM correlations develop

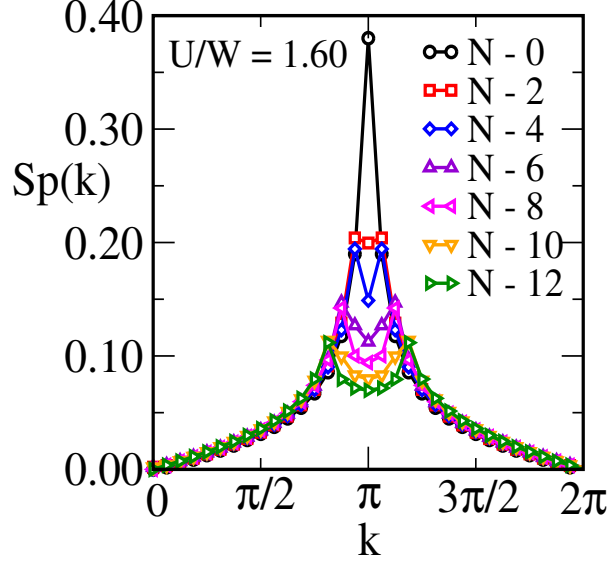


Figure 5.1: Spin structure factor vs. wavevector k along the chain direction. Results correspond to $U/W = 1.60$, $J_H/U = 0.25$, various hole dopings as indicated, and employing a 32 sites chain ($N = 64$) and the DMRG technique. In this figure, and others not shown at several values of U/W , the peak at $k = \pi$ denoting staggered order at half-filling becomes incommensurate upon hole doping.

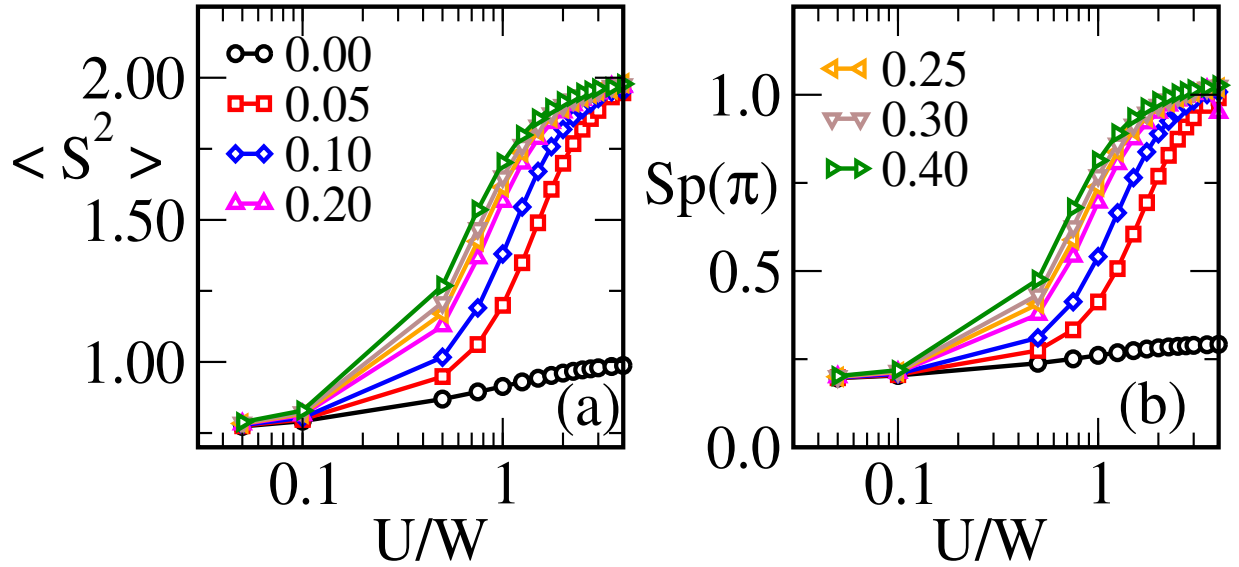


Figure 5.2: (a) Spin squared $\langle S^2 \rangle$ and (b) staggered spin structure factor $Sp(\pi)$ vs. U/W for a half-filled 8-sites system using various Hund's coupling (J_H/U) as indicated. At zero J_H/U , local moments are not developed up to large U/W and therefore there is no robust magnetic ordering.

at least at short distances, with the exception of $J_H = 0$, together with the development of the spin-1 moments at every site.

As the doping of holes increases, Fig. 5.1 illustrates that spin incommensurate (IC) correlations develop smoothly. While this spin IC order is compatible with spin excitations from the band dispersion in the kinetic energy portion of the model, note that $U/W = 1.60$ is already in intermediate coupling. Analysis of the spin-spin correlations “across holes” [74, 72, 73], to be shown in more detail below, indicate that the spins tend to arrange and couple in a manner qualitatively compatible with the exact results for the one-orbital Hubbard model at $U = \infty$ [90]. This arrangement is the most optimal to favor simultaneously the hole mobility and spin correlations, and it is qualitatively different from the analysis based on Fermi surface characteristics.

5.3.2 Hole pairs and their internal structure

The main result of this chapter is that the model studied here presents a regime of hole pair binding that correlates with robust pair-pair correlations in a spin-singlet channel, as will be described below. Figure 5.3 shows the binding energy ΔE , as defined in Eq. 5.2, for the case of two holes added to half-filling, varying U/W at a fixed Hund coupling $J_H/U = 0.25$. Starting approximately at $U/W \sim 0.6$ and up to $U/W \sim 3.0$, the binding energy is negative indicative of the formation of a bound state of two holes. Considering recent developments in the study of iron-based superconductors [27] this regime of U/W is realistic. Moreover, the Hund coupling value is also in a reasonable range for pnictides and chalcogenides that are well known for having a robust Hund-driven physics. The results in Fig. 5.3 were obtained using a 16-sites chain but they appear robust varying the length of the system. For instance, approximately at the minimum of the curve at $U/W = 1.60$ results for 48 sites are only slightly more negative than for 16 sites. Figure 5.4 contains a size scaling analysis of binding at $U/W = 1.60$ illustrating this conclusion. Our best efforts indicate that size effects are small and moreover with increasing chain length the binding magnitude slightly increases in absolute value. Thus, the bulk-limit binding energy at $U/W = 1.60$ appears to be close to -0.13 in hopping units.

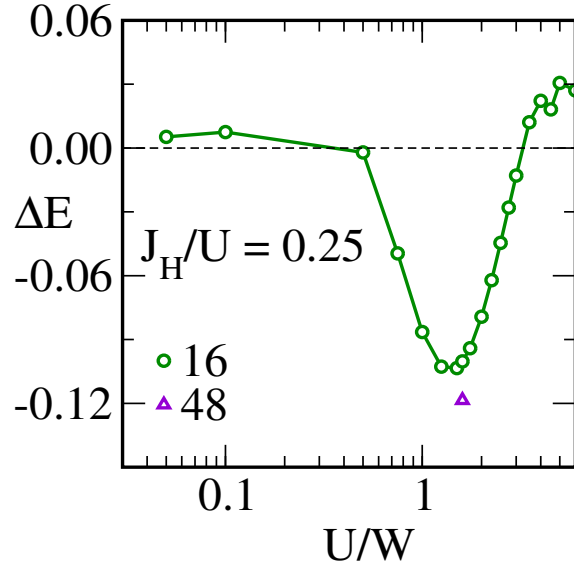


Figure 5.3: Binding energy (ΔE) vs. U/W at a fixed $J_H/U = 0.25$, and using a 16-sites chain. For intermediate interaction strength, there is a wide range with negative binding energy indicating a region where holes pair. Recent results on 64 sites are very similar to the 48 sites results shown.

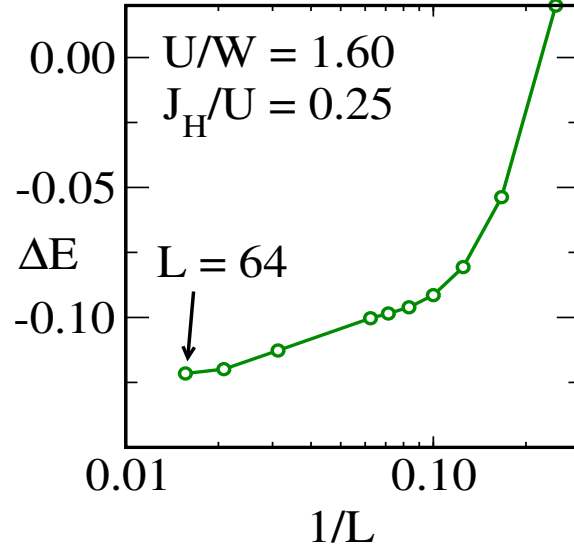


Figure 5.4: Scaling of ΔE vs. inverse chain length ($1/L$) at coupling $U/W = 1.60$ where there is robust negative binding. In the bulk limit, ΔE remains negative according to the extrapolation of these results.

Besides the surprising result that binding is possible even in the presence of a strong Hubbard U repulsion, it is interesting to remark the similarity of Fig. 5.3 with the binding results found before in the context of two-leg ladders (see Fig. 8 of [93]). In both cases, ladders (short sizes were studied in [93]) and chains, ΔE starts positive with increasing U/W , drops to negative at intermediate couplings where it remains into the strong coupling regime, and then it becomes positive again at abnormally large U/W . Note that the region where ΔE is positive is not important: in the bulk limit the energy of two holes that do not form a bound state should converge to the energy of two independent holes, rendering ΔE equal to zero. But the negative region of ΔE is physically realistic and representative of pair formation: two holes lower the energy of the system by being close to each other.

Figure 5.5 provides the electronic density using a 32-sites chain, corresponding to the orbital a (the results for b are identical, because the model is invariant if a and b are exchanged). For half-filling, the density is virtually equal to one at all sites. What is interesting is that for 2 holes, there is only 1 minimum indicative of the existence of a hole pair. For 4 holes there are 2 minima, for 6 holes 3 minima, and for 8 holes 4 minima. All these results are at least compatible with the existence of hole pair formation, as the binding energy indicates.

What is the internal structure of this pair? In Fig. 5.6 the probability of finding the second hole is shown when the first hole is projected to be at site 16, namely at the center of the 32-sites chain used, and at orbital a . It is clear that the probability for the second hole is the largest close to the first projected hole, compatible with pairing. Moreover, the second hole is primarily at orbital b if the first is at orbital a . Thus, the pairs unveiled here involve holes primarily located at different orbitals. This will be shown below to be compatible with the pair-pair correlation that is the most dominant in many portions of the phase diagram. Figure 5.6 also has prominent sharp peaks located at nearest-neighbors sites. Thus, the dominant hole configuration in the pair is that of holes separated by just one lattice spacing, located at different orbitals.

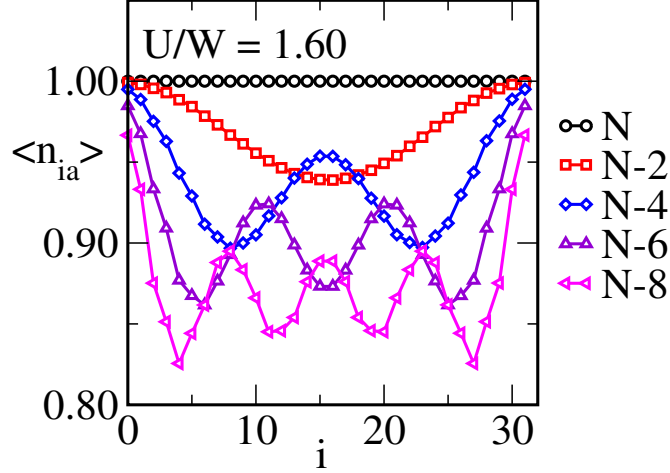


Figure 5.5: Local charge density of orbital a at $U/W = 1.60$, *i.e.* in the binding region, using a 32-sites chain and $J_H/U = 0.25$. For a fixed number of holes n_h , we find $n_h/2$ number of “dips” in the charge density. Note that the charge density profile of orbital b is the same as that of orbital a , by symmetry.

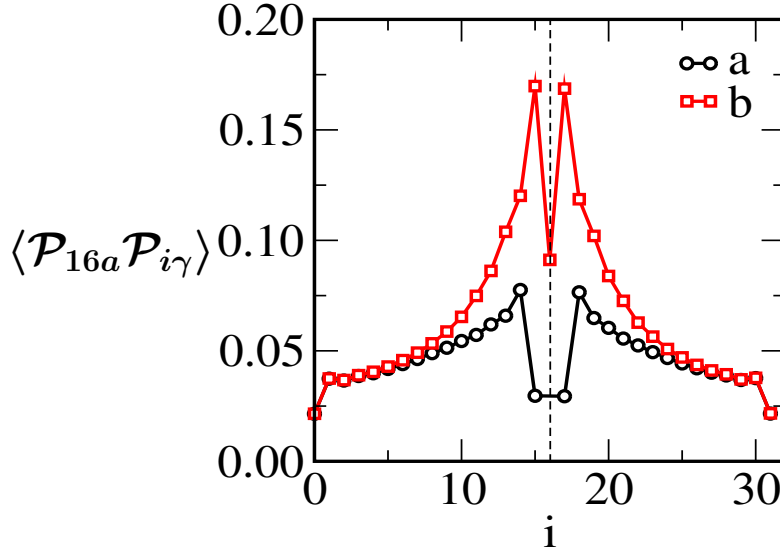


Figure 5.6: Probability of finding a hole at site i and orbital γ given that the other hole is projected from the two-holes ground state to be at site 16, orbital a , of a two-orbital chain with 32 sites, at $U/W = 1.60$ and $J_H/U = 0.25$. The result is normalized to $\langle \mathcal{P}_{16a} \rangle$. We find the largest probability of the non-projected hole to be in the other orbital b and at the neighboring site. In the ladder analogy of the two-orbital chain (see Fig. 5.7), these dominant pairs are equivalent to pairs along the diagonal of effective plaquettes.

Projecting now one or two holes to particular locations and analyzing the spin-spin correlations in that framework leads to interesting conclusions. The results are shown in Fig. 5.7. First, note that once the two-orbital chain results are displayed representing each of the two orbitals by a chain, then this illustrates that two-orbital chains can be mapped formally into a special case of one-orbital two-leg ladders. This is interesting in several respects, but here we wish to emphasize the resemblance, once again, with the previously published results for two-orbital ladders [93]. Consider panel (a) for one hole: here the rungs of the effective ladder are ferromagnetic and the legs are AFM. Thus, once the results are plotted as in Fig. 5.7 the magnetic order resembles the “rung FM - leg AFM” of BaFe_2S_3 as reported in [93]. Also the AFM spin-spin correlation “across the hole” observed in early studies of models for cuprates [74, 72, 73] and also found more recently in models for iron-based ladders [93] is present in panel (a). From the spin perspective, “across the hole” AFM correlations are effectively equivalent to dropping sites of the chain, explaining the spin IC tendency in Fig. 5.1 with increasing doping.

The results for two holes are equally interesting and also resemble those of previous investigations for real iron-based ladder models. Panel (b) contains the hole arrangement with the largest probability in the two-holes ground state. Similarly as in Fig. 10 of [93], the plaquette diagonal opposite to the projected holes is FM and the “across the hole” antiferromagnetism is robust. Panel (c) shows the case where the two holes are along the rung (*i.e.* on-site in the real chain). Panels (b) and (c) are smoothly connected: for instance by moving the electron at “17b” to “16b” in panel (c), panel (b) is recovered if the spin correlations follow as if they were “rubber bands” attached to the electrons. Previous studies in models for cuprates have unveiled similar physics.

Ending this subsection, we will discuss a subtle effect related with the spin quantum number of the two holes state. Employing Lanczos methods we studied the total spin of the two-holes state employing both periodic and open boundary conditions (PBC and OBC, respectively) using chains of length 4 and 6, at various Hund couplings and $U/W = 1.60$. The behavior is erratic: while for 6 sites and PBC the spin is always 0, for 4 sites with OBC it is always 1. The interpretation of these results is difficult because of the presence of the well-known edge states of Haldane chains when OBC are used. Considering the difficulty in

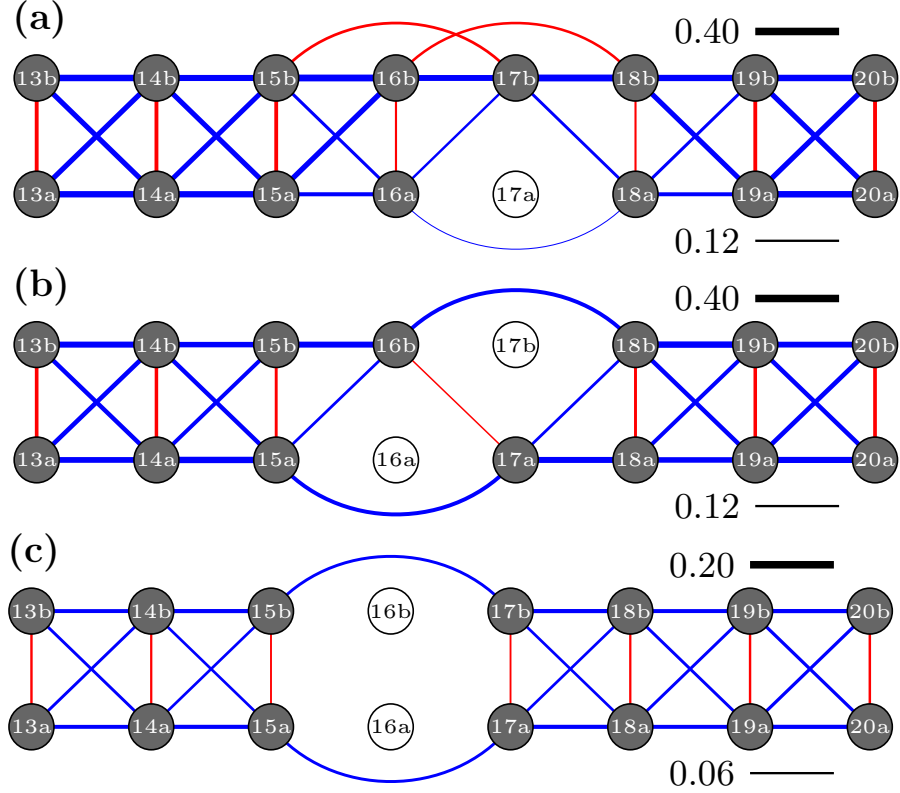


Figure 5.7: Structure of the spin-spin correlations for fixed projected arrangements of holes, using hole configurations with large weight in the ground states. (a) is the one-hole case; (b) corresponds to holes along the effective plaquette diagonals (largest weight for two holes), while (c) are for holes along the effective rung. All results are obtained using 32 sites with two-orbitals at fixed $U/W = 1.60$ and $J_H/U = 0.25$. Blue lines correspond to AFM bonds while red lines are for FM bonds. Away from the holes, the expected pattern of FM effective rungs and AFM legs is recovered. In the fixed hole configurations of panels (b) and (c), the normalized probability of the holes configuration is $\langle \mathcal{P}_{16a} \mathcal{P}_{17b} \rangle / \langle \mathcal{P}_{16a} \rangle = 0.169$ and $\langle \mathcal{P}_{16a} \mathcal{P}_{16b} \rangle / \langle \mathcal{P}_{16a} \rangle = 0.0912$, respectively. In all cases there is a prominent “across the hole” AFM correlation. The case of two holes located along the same “leg” of the effective ladder has much smaller weight in the two-hole ground state and it is not shown.

distinguishing between intrinsic spin quantum numbers of a hole pair vs. those at the edge of a Haldane chain, we do not investigate further this topic and below we study both singlet and triplet pair channels to find out which one dominates explicitly. The final result is that spin-singlet pairs are dominant suggesting that the spin 1 quantum number found for some two-holes chains originates in the edge states.

5.3.3 Pair-pair correlations and tendency to superconductivity

The existence of hole binding (negative binding energy) at half-filling is often a precursor of superconducting tendencies with increasing doping. For this reason, we have measured the pair-pair correlations in all the channels described in Sec. 5.2, and contrasted their behavior with increasing distance against density-density and spin-spin correlations to determine which channel dominates. Representative results are shown in Fig. 5.8. Panel (a) contains results for 2 holes. Here the pair-pair correlations are robust in the spin-singlet channel when involving different orbitals and using nearest-neighbor sites, in agreement with the analysis of the internal structure of the pair in the previous subsection. The analogous spin-triplet pair correlations decay much faster, while spin and charge correlations are in between. However, in spite of the robustness of the singlet pair correlations in (a), the ground state only has two holes and these results, while promising, may be anomalous. More standard and exciting are the results in panel (b) with 6 holes and a nominal hole doping $x = 6/96 = 0.0625$ ($N = 96$ for a half-filled 48-sites cluster). The same spin-singlet inter-orbital NN-sites pair correlation dominates here as well, as in (a). The decay with distance is similar as in the charge and spin channels but only if the maxima is used for the latter. If, instead, the minima in charge and spin correlations are included in finding the most optimal fits then pair-pair correlations dominate. Note that the prominent oscillations in charge and spin correlations have been often reported before (for recent state-of-the-art efforts see [28]), although their origin is not fully clear; for the pair correlations a smoother behavior is often observed, as we found. As the number of holes increases, then the superconducting tendencies remain robust but diminish compared with spin and charge. In panel (c) with $x = 8/96$, the pair-pair decay with distance approximately follows the average of spin and charge indicating that they compete, while in panel (d) with $x = 12/96$, pairing is already less robust than charge and

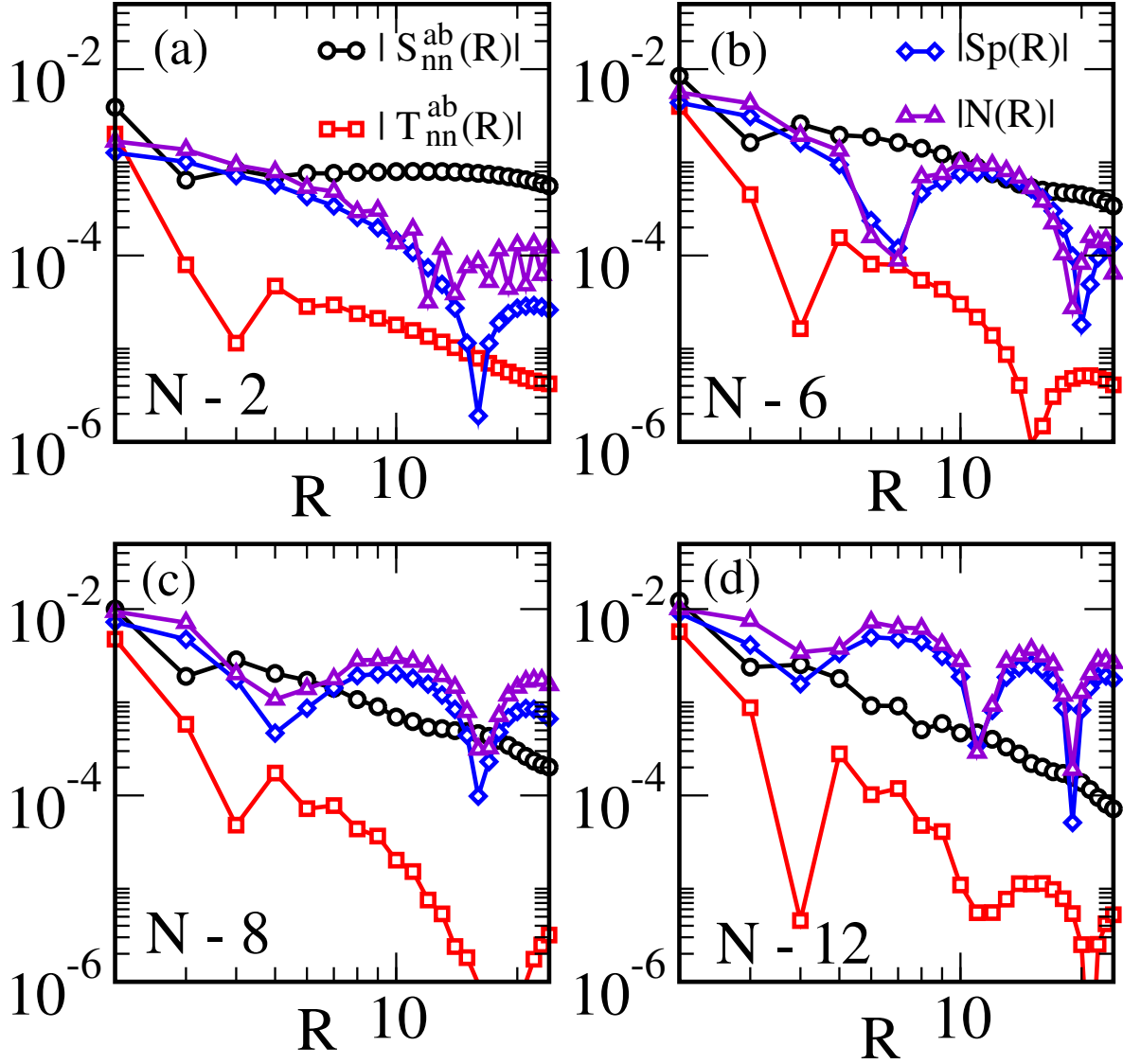


Figure 5.8: Real-space decay of the pair-pair (singlet and triplet), spin-spin, and charge-charge correlations involving nearest-neighbor sites at fixed $U/W = 1.60$ and $J_H/U = 0.25$, and using a 48-sites chain. (a) corresponds to two holes doping, (b) to six holes doping, (c) to eight holes doping, and (d) to twelve holes doping.

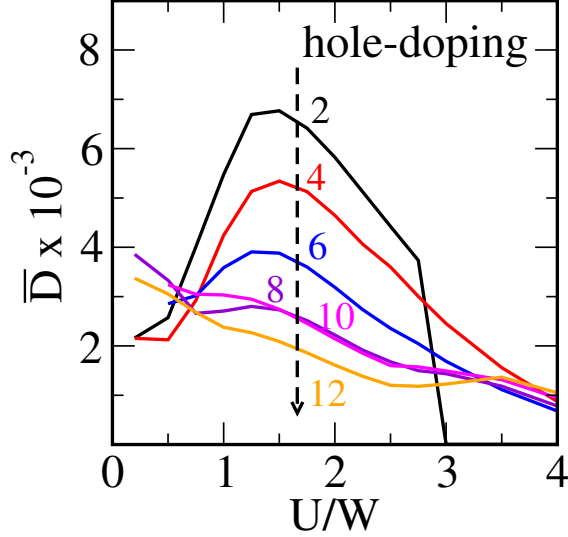


Figure 5.9: Pairing strength $\bar{D}$ (see Eq. 5.11) vs. U/W , parametric with number of holes. A 32-sites chain is used and $J_H/U = 0.25$.

spin channels. In summary, in a range of doping near half-filling and for the clusters that we studied, the superconducting correlations appear to dominate, or at the minimum decay at a similar rate as spin and charge. With increasing hole doping the importance of the pair-pair correlations diminishes.

In Fig. 5.9 we show the pairing strength $\bar{D}$, defined in Eq. 5.11, as an indicator of the robustness of pairing correlations varying U/W for various number of holes. Clearly it is the intermediate range of U/W where pairing dominates the most – as found in the hole binding analysis – and also when the numbers of holes is small. Note that the presence of robust superconducting correlations in Fig. 5.8 occurs in the region of pair binding shown in Fig. 5.3. Away from that region, for example at small Hubbard coupling such as $U/W = 0.2$ or at very large Hubbard coupling $U/W = 10$ pairing is not as robust as at intermediate couplings, as illustrated in Fig. 5.10. Thus, once again we arrive to the conclusion that the behavior of the binding energy and the pair correlation is compatible with one another.

For completeness, note that previous work also unveiled tendencies towards pairing in electronic two orbitals models but under rather different circumstances. (i) For instance, in [101] Kondo models for $\text{Y}_{2-x}\text{Ca}_x\text{BaNiO}_5$ were studied using Lanczos and DMRG techniques, supplemented by AFM Heisenberg J terms. The emphasis was on ferromagnetism and phase separation but tendencies towards hole binding were also briefly

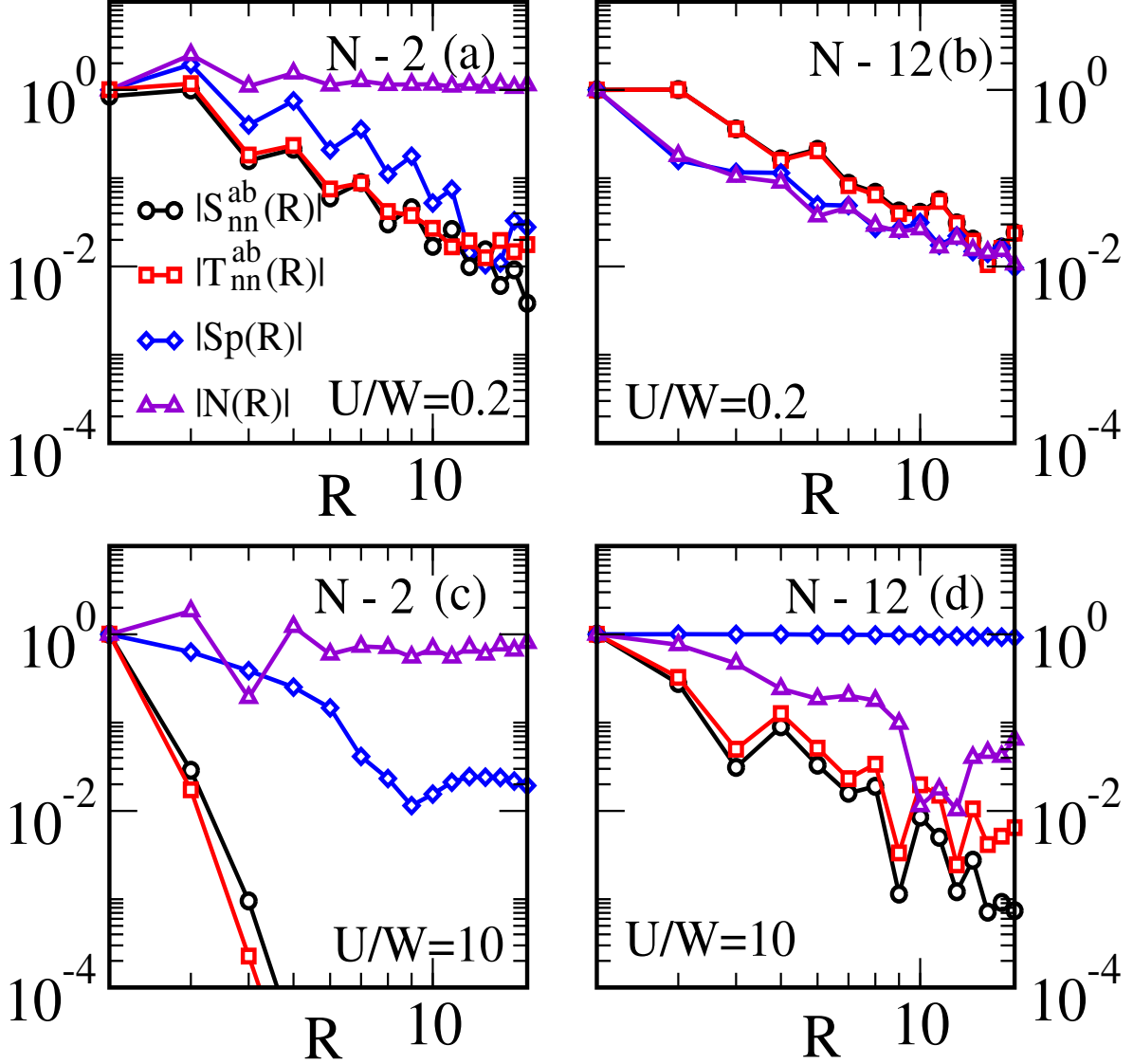


Figure 5.10: Real-space decay of the pair-pair (singlet and triplet), spin-spin, and charge-charge correlations for two and twelve holes at $U/W = 0.2$ and 10.0 , as indicated, and at $J_H/U = 0.25$ using a 32-sites chain. In all cases, the decay of the pairs is either faster or approximately equal to the spin and charge correlations. Thus, the pairing tendencies are robust at intermediate coupling, compatible with the conclusions regarding hole binding.

reported. The signal for binding was strongest at high hole concentration such as $x = 0.4$ and robust values of J of order one. No pair-pair correlations were calculated, nor competition triplet vs. singlet was studied. (ii) In [114] two one-orbital Hubbard chains coupled by an explicitly ferromagnetic Heisenberg interaction were studied via bosonization and DMRG/Lanczos methods. Regions of singlet and triplet superconductivity were reported, but note that this model has an explicit Heisenberg effective attraction, without a U' repulsion (similar to our previous effort [137] to be discussed below). The goal in [114] was to study the singlet vs. triplet competition in superconductivity, unlike our efforts that focus on unveiling pairing tendencies from a complete two-orbital Hubbard model that is explicitly repulsive. (iii) In [3] a two-orbital Hubbard model at $U = \infty$ was studied with emphasis on the influence of the Hund coupling. When U' was included, charge-density waves were reported to dominate, while in the absence of U' but with robust J_H then singlet or triplet pairing dominates. Our analysis, on the other hand, focuses on a finite intermediate U/W range where surprisingly we found that singlet pairing dominates even in the presence of a realistic $U' > J_H$. As $U/W \rightarrow \infty$, we found that hole binding no longer occurs, as shown in Fig. 5.3, and the charge or spin channels dominate over pairing (see panels (c,d) of Fig. 5.10) compatible with [3]. (iv) In [105] results compatible with ours were produced via the exact diagonalization of a PBC 6-sites chain with emphasis on Luttinger liquid parameters using a two-orbital Hubbard model with a robust band splitting. (v) In [143] using the statistically consistent Gutzwiller approximation for a square lattice, conclusions similar to ours were reached, reporting a stable spin-triplet s -wave superconducting state for a two orbital degenerate Hubbard model. This occurs, like in our case, even in the case $U' > J_H$ and near half-filling.

5.3.4 Role of Hund coupling and magnetic moments

The origin of the pairing tendencies unveiled here is subtle and in this subsection we report some observations to help clarify this matter. More work is needed to fully comprehend this hole pair formation, so ours are just the first steps in that direction. One important factor correlated with the pairing we are reporting is the presence of well-formed magnetic moments at every site. This is along the same direction as early studies of the $t - J$ model for

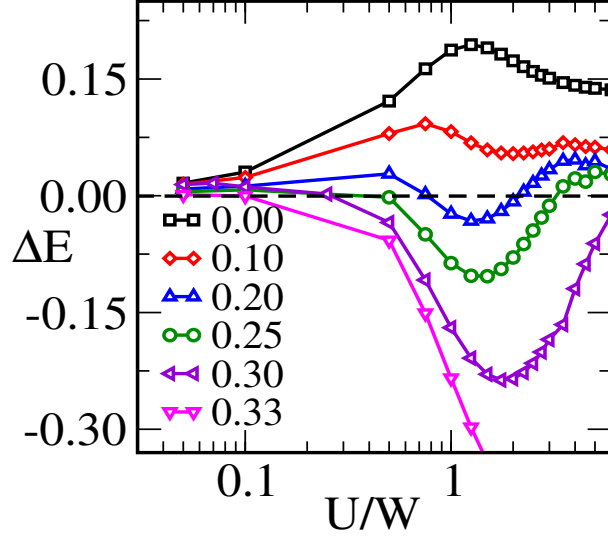


Figure 5.11: ΔE vs. U/W parametric with J_H/U (inset) using an 8-sites chain with two orbitals.

cuprates [21], where holes form bound states to reduce the damage that mobile holes induce in an otherwise optimal antiferromagnetic arrangement. Each hole alters the spin order in a finite region, and pairing of holes reduces the size of the regions where spins are not properly arranged. This simple and well known notion must be at least part of the explanation for our results because pairing in ΔE , as shown in Fig. 5.3, occurs in regions where moments are well formed, as indicated in Fig. 5.2.

In addition, we have observed that the Hund coupling in our model clearly is directly related to binding. Figure 5.11 shows the binding energy in a wide range of U/W parametric with J_H/U . At the smallest J_H/U shown, the binding energy is positive and pairs do not have a tendency to form. Consider now the special value $J_H/U = 1/3$. In this case $J_H = U'$ because of the relation $U = U' + 2J_H$. Thus, the natural repulsion U' for two electrons at different orbitals in the same lattice site is compensated by the natural tendency to bind induced by J_H . In fact for $J_H/U = 1/3$, and beyond *i.e.* $J_H/U > 1/3$, the binding energy ΔE is negative at all values of U/W .

The reader should note that the connection between the realistic regime $J_H/U < 1/3$ and the unphysical region $J_H/U > 1/3$ is non trivial. Naively, one may expect ΔE to be negative for all U/W for $J_H/U > 1/3$, and positive for all U/W for $J_H/U < 1/3$. However, the interpolation, while smooth, is more complex. Figure 5.11 shows that in the intermediate

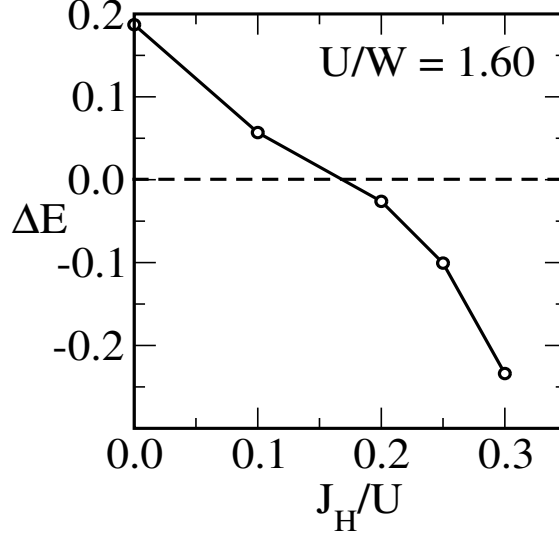


Figure 5.12: ΔE vs. J_H/U at fixed $U/W = 1.60$ using 16 sites with two-orbitals. ΔE becomes significantly more negative as J_H/U is increased.

U/W range, the binding is negative for $J_H/U = 0.20, 0.25$, and 0.30 , with a clear dip in the $U/W \sim 1 - 2$ range. This dip, being smoothly connected with the broad negative binding energy region of $J_H/U = 1/3$, must be caused by J_H attraction effects that somehow are not fully compensated by U' at intermediate couplings.

The “earlier” than anticipated attractive effects of J_H at intermediate U/W are explicitly illustrated in Fig. 5.12 where ΔE is shown at the optimal $U/W = 1.60$ of our focus, varying J_H/U . At least for the small system studied here, ΔE changes sign before $J_H/U = 0.2$ and it becomes increasingly negative with further increasing J_H . While the causal effect of J_H is clear, further work is needed to clarify how can this attraction overcome the U' repulsion in the intermediate coupling range. Moreover, the attraction channel favors spin-singlets involving different orbitals at nearest-neighbor sites. In fact, pairing in the spin-singlet inter-orbital NN-sites channels is enhanced as J_H increases as shown in panels (a,b) of Fig. 5.13. Thus, it is a subtle combination of the Hund interaction together with antiferromagnetic short-range order that induces singlet pairing in this one-dimensional multiorbital model. More work is needed to clarify this interesting effect.

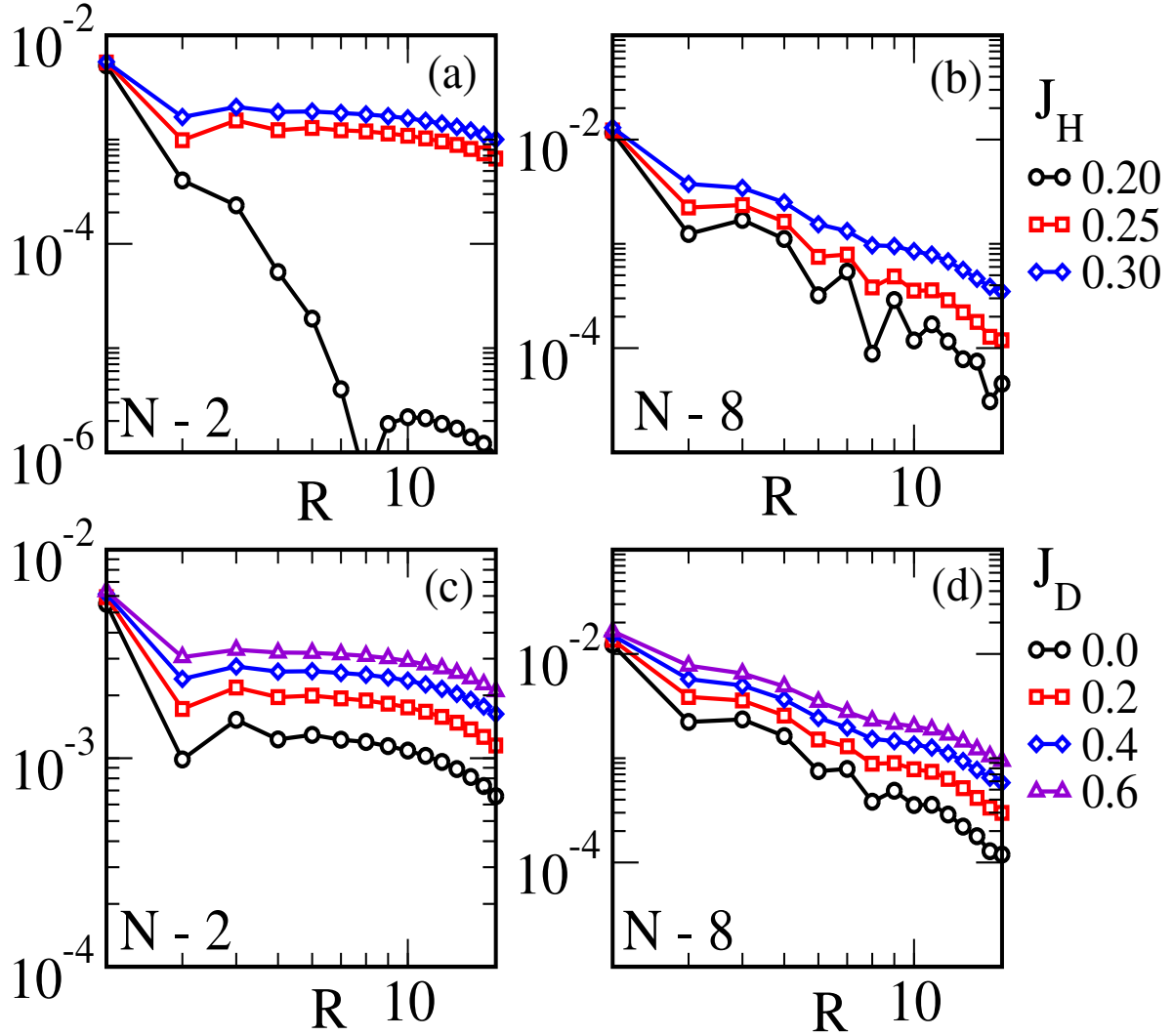


Figure 5.13: Decay of pair-pair singlet correlations S_{nn}^{ab} at $U/W = 1.60$, for various Hund couplings in panels (a) and (b), and for various diagonal AFM Heisenberg couplings J_D in panels (c) and (d). Shown are results for 2 and 8 holes, as indicated, using in all cases a 32-sites chain and $J_H/U = 0.25$. Clearly increasing both J_H and J_D magnify the pairing tendencies.

5.3.5 Influence of additional inter-orbital Heisenberg interactions

For completeness, we have also added an extra term to the Hamiltonian in order to boost pairing tendencies. This term is simply a Heisenberg spin-spin interaction with strength J_D defined as

$$H_D = J_D \sum_{\langle ij \rangle} \mathbf{S}_{ia} \cdot \mathbf{S}_{jb}. \quad (5.12)$$

The motivation for adding this term is two folded. First, it plays a role similar to that of “ J ” in the standard $t - J$ model, and we know that increasing J increases pairing tendencies [21]. Second, the new term links the spins of two electrons located at NN sites and at different orbitals (note orbital indexes in Eq. 5.12), resembling the structure of the pairs that we have found above. In agreement with these expectations indeed we have observed that pairing tendencies in the dominant spin-singlet NN-sites inter-orbital channel are enhanced as shown in panels (c) and (d) of Fig. 5.13. A similar analysis adding instead a NN Heisenberg coupling between electrons in the *same* orbital only showed minor changes in the decay of the correlations (not shown).

Clearly the electrons/holes in the dominant pairs have a preference to be in different orbitals. Future studies of superconductivity in the two-orbital Hubbard model analyzed here can benefit from enhanced pairing effects by including J_D .

5.4 Conclusions

In this chapter we have investigated the magnetic, hole pairing, and superconducting properties of a two-orbital Hubbard model defined on a chain. The primary motivation is the recent report of hole binding tendencies in a similar model but defined on a two-leg ladder geometry [93], motivated by the discovery of superconductivity under high pressure in the ladder compound BaFe_2S_3 [122]. In that previous computational effort, the binding tendency was found to be negative, thus signaling pairing, but the results could not be confirmed beyond small systems 2×8 . In addition pair-pair correlations were not measured

in that early effort. In the present work much longer chains can be studied and a variety of correlation functions were measured and their decay with distance compared to decide which is dominant. In the same spirit as in [93], here our search for superconducting tendencies was based on hole doping while the experimental setup relied on pressure. The *ab-initio* calculations in [146] justify our theoretical approach because they reported that pressure leads to modifications in the average electronic density at the iron atoms.

Our results are interesting for several reasons. The data reported here for the binding energy resemble those of the ladder, but on chain sizes up to 64 sites. Size scaling shows that the results survive the bulk limit. Qualitatively both for ladders and chains it is the intermediate region of U/W where binding does occur. Having almost saturated magnetic moments is important together with a robust Hund coupling. Neither very weak nor very strong U/W coupling seem suitable for pairing, a conceptually interesting result. The absence of pairing at very large U/W may be related with competing ferromagnetic tendencies when holes are added, as in double exchange models. This line of research is being investigated at present.

Moreover, by measuring pair-pair correlations in the spin singlet channel, and using pair operators involving different orbitals and nearest-neighbor sites, a region of hole density and couplings was identified where superconducting correlations decay slower, or at least at the same rate, than spin and charge correlations. Having different orbitals and nearest-neighbor sites is compatible with the internal structure of the pair.

By varying the Hund coupling into the region believed to be unphysical where J_H becomes as large as U' (this occurs at $J_H/U = 1/3$ if the standard relation $U = U' + 2J_H$ is assumed [137, 138]), then an unexpected smooth continuity was observed between $J_H/U > 1/3$, where binding occurs at all values of U/W because J_H becomes an effective attraction when it overcomes U' , and the region widely believed to be realistic $J_H/U \sim 0.25$. This smooth continuity occurs primarily at intermediate U/W couplings. Thus, for reasons that still need better clarification the effective J_H attraction at $J_H/U > 1/3$ can become operative even at smaller Hund couplings in a reduced U/W range. The chosen dominant channel involves holes in different orbitals, a spin-singlet combination, and nearest-neighbors sites.

The observation that pairing, charge, and spin correlations are sometimes of similar strength, as in panel (c) of Fig. 5.8 for $N = 8$ electrons (48 sites), suggests that future work should also address the possible formation of “pair density waves”. These are subtle broken-symmetry states that intertwine charge density waves, spin density waves, and superconducting orders. In this state the superconducting order parameter is spatially modulated in such a way that the uniform component is zero or very small, but it has a strong oscillatory component [9, 1, 38].

In summary, these results contribute towards understanding pairing tendencies in quasi one-dimensional iron-based superconductors. Binding was found to occur at intermediate couplings, a regime that previous studies showed to be realistic for chalcogenides [27, 23]. There is plenty of work ahead. While superconducting correlations already appear to dominate at low hole doping, these results must be confirmed using even longer chains. Moreover, although it seems clear that a robust Hund coupling and robust magnetic moments are needed, developing an even more detailed qualitative understanding of the origin of pairing is important.

Chapter 6

Summary

This dissertation presents many numerical results, using mean field theories and the exact numerical method DMRG, which reproduce the experimentally relevant properties of the iron-based superconductors. In Chapter 2, we present a numerical method that mixes the mean-field approximation with classical Monte Carlo simulations (MCMF). We showed that this method produces results that are qualitatively similar to the known results of one-orbital Hubbard model in two- and three-dimensions at finite temperature. In fact, we go beyond the limitations of Determinantal Quantum Monte-Carlo and show results of the $t - t'$ Hubbard model that contains the plaquette diagonal hoppings in a two-dimensional square lattice.

In Chapter 3, we apply MCMF to a two-orbital Hubbard that is more realistic for our understanding of properties of the Fe-based pnictide layer compounds. We capture many experimental results: 1) local moment formation at high temperature and $(\pi, 0)$ magnetic ordering below the Néel temperature (T_N), 2) magnetic nematic ordering above the T_N , and 3) resistivity anisotropy between the two real space dimensions. These listed results are experimentally known to be present in the pnictides. Additionally, we predict a phase that is an insulator in one direction and conducting in the other, indicating an effective dimensional reduction in real-space.

In Chapter 4, we apply an exact numerical method, density matrix renormalization group, at zero temperature to study a recently discovered quasi-one-dimensional Fe-based superconductor, BaFe_2S_3 , that forms iron ladders. We use DMRG to solve the two-orbital Hubbard model that is built using realistic parameters obtained from an *ab initio*

calculation. We find the experimentally relevant $(\pi, 0)$ magnetic ordering. Upon doping, we find tendencies towards superconductivity that naturally occurs from a repulsive model. We further demonstrate that, in real-space, the hole pairs are most probable to be arranged along the diagonals ladders.

In Chapter 5, we simplify the two-orbital Hubbard model of BaFe_2S_3 ladders to a two-orbital chain to understand the mechanism of hole-pairing in the multi-orbital systems. Using DMRG at zero temperature, we find that pairing tendencies are also present in chains. Upon hole-doping, we show that diagonally arranged hole pairs are most likely in the system. In fact, we demonstrate that diagonal singlet hole pairs are dominant in the ground-state with few holes. We also emphasize that the Hund coupling interactions are playing a pivotal role in stabilizing the superconducting order while Heisenberg interactions tend to stabilize the hole-pairs further.

In summary, this dissertation has emphasized that the Fe-based superconductors can be described by intermediate to large Hubbard interaction strength with a large Hund coupling. Using DMRG, we have shown that magnetic fluctuations, such as Hund coupling and Heisenberg Interactions, are playing a dominant role in the creation of robust Cooper pairs. This dissertation further adds to the evidence that superconductivity in the Fe-based SCS has its origin in the magnetic fluctuations that are driven by correlations.

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Appendices

Appendix A

One Orbital Model (MCMF)

Let us start the specific application of the ideas outlined in chapter 2 by considering the one-band Hubbard model defined below (in a standard notation):

$$H = H_o + H_1 = -t \sum_{\langle i,j \rangle, \sigma} c_{i,\sigma}^\dagger c_{j,\sigma} + U \sum_i n_{i,\uparrow} n_{i,\downarrow}. \quad (\text{A.1})$$

To set up the formalism, it is convenient to perform a rotationally invariant decoupling of the interaction term in the following manner [110]:

$$\begin{aligned} n_{i,\uparrow} n_{i,\downarrow} &= \frac{1}{4} (n_i)^2 - S_{iz}^2 \\ &= \frac{1}{4} (n_i)^2 - (\mathbf{S}_i \cdot \hat{\Omega}_i)^2. \end{aligned} \quad (\text{A.2})$$

Here, the spin operator is $\mathbf{S}_i = \frac{\hbar}{2} \sum_{\alpha,\beta} c_{i,\alpha}^\dagger \sigma_{\alpha,\beta} c_{i,\beta}$, $\hbar = 1$, $(\sigma_x, \sigma_y, \sigma_z)$ are the Pauli matrices, and $\hat{\Omega}_i$ is an arbitrary unit vector. In the previous identity, we have used the fact that $(\mathbf{S}_i \cdot \hat{\Omega}_i)^2 = (S_{i,x})^2 = (S_{i,y})^2 = (S_{i,z})^2$. The expression in the last line of Eq. (2) is rotationally invariant since it is in terms of the scalars n_i and the dot product between $\mathbf{S}_i$ and $\hat{\Omega}_i$. It should be noted that there are other possible decouplings, but the formula above is the only one whose saddle point leads to the correct Hartree-Fock equations after implementing a Hubbard-Stratonovich (HS) decomposition. Below we will use the notation followed in recent literature [29]. For the HS decomposition, let us start with the partition function $Z = \text{Tr } e^{-\beta H}$. Here the trace is over all particle numbers and site occupations. $\beta = 1/T$

, with k_B set to 1. We now divide the interval $[0, \beta]$ into M equally spaced slices, defined by $\beta = M\Delta\tau$, separated by $\Delta\tau$ and labeled from 1 to M . For large M , $\Delta\tau$ is a small parameter and allows us to employ the Suzuki-Trotter decomposition, so that we can write $e^{-\beta(H_0+H_1)} = (e^{-\Delta\tau H_0} e^{-\Delta\tau H_1})^M$ to first order in $\Delta\tau$. Then using Eq. (2) and the Hubbard-Stratonovich identity, $e^{-\Delta\tau U \sum_i [(1/4)(n_i^2) - (\mathbf{S}_i \cdot \hat{\Omega}_i)^2]}$, for a generic time slice ' l ', can be shown to be proportional to

$$\int d\phi_i(l) d\Delta_i(l) d^2\Omega_i(l) \propto e^{-\Delta\tau [\sum_i (\phi_i(l)^2/U + i\phi_i(l)n_i + \Delta_i(l)^2/U - \Delta_i(l)\hat{\Omega}_i(l) \cdot \mathbf{S}_i)]}. \quad (\text{A.3})$$

Here we have introduced two auxiliary fields, $\phi_i(l)$ which couples to the charge density, and $\Delta_i(l)$ that couples to the spin density. We note that the integration over the unit vector $\hat{\Omega}_i(l)$ at every site shows the SU(2) invariance explicitly. We further combine the product $\Delta_i(l)\hat{\Omega}_i(l)$ into a new vector auxiliary field, $\mathbf{m}_i(l)$, at every site. This nomenclature is used from now on. Using the above decoupling for the quartic term in the expression of the partition function, we can write

$$Z = \text{const} \times \text{Tr} \prod_{l=M}^1 \int d\phi_i(l) d^3m_i(l) e^{-\Delta\tau [-t \sum_{\langle i,j \rangle, \sigma} c_{i,\sigma}^\dagger c_{j,\sigma} + \sum_i [\phi_i(l)^2/U + i\phi_i(l)n_i + \mathbf{m}_i(l)^2/U - 2\mathbf{m}_i(l) \cdot \mathbf{S}_i]]} \quad (\text{A.4})$$

In the above, trace "Tr" is over all particle numbers and site occupations as before. The continuous integrals are over the auxiliary fields, $\{\phi_i(l), \mathbf{m}_i(l)\}$ at every site, and the argument l denotes the imaginary time slice label. The product over l from M to 1 implies time ordered products over time slices, with the earlier times appearing to the right. Finally, the $d^3m_i(l)$ in the integral implies integration over the amplitude and orientation of vector auxiliary fields, $\mathbf{m}_i(l)$.

This allows us to identify an effective Hamiltonian H_{eff} in which fermions couple to auxiliary fields fluctuating in both space and (imaginary) time. Typically this is the starting point of quantum Monte Carlo (QMC) approaches. However, for reasons discussed in chapter 2, we take a different route by making the following approximations: (i) We drop the τ dependence of the HS auxiliary fields and (ii) we use the saddle point value $i\phi_i = \frac{U}{2} \langle n_i \rangle$. This allows us to extract the following effective Hamiltonian (H_{eff}) where the fermions

couple to the static HS field $\mathbf{m}_i$ and to the average local charge density:

$$H_{eff} = H_o - \mu \sum_i n_i + \sum_i \frac{U}{2} \langle n_i \rangle n_i - \sum_i \mathbf{m}_i \cdot \sigma_i + \frac{1}{U} \sum_i \mathbf{m}_i^2 - \frac{U}{4} \sum_i \langle n_i \rangle^2. \quad (\text{A.5})$$

Here, H_o contains the fermionic kinetic energy. The redefinition $\mathbf{m}_i \rightarrow \frac{U}{2} \mathbf{m}_i$ allows us to arrive at the final form of the effective Hamiltonian:

$$H_{eff} = H_o + \frac{U}{2} \sum_i (\langle n_i \rangle n_i - \mathbf{m}_i \cdot \sigma_i) + \frac{U}{4} \sum_i (\mathbf{m}_i^2 - \langle n_i \rangle^2) - \mu \sum_i n_i, \quad (\text{A.6})$$

which is our effective model belonging to the spin-fermion family.

It should be noted that H_{eff} coincides with the mean field Hamiltonian at $T = 0$, where $\mathbf{m}_i$ has the interpretation of the local magnetization. As discussed in chapter 2, to study the model at finite temperature, we simulate H_{eff} by sampling the $\mathbf{m}_i$ fields via a classical Monte Carlo (MC) procedure [76, 126, 124]. The main result of the present effort will arise when these MC results at finite temperature are compared against DQMC results. It will be demonstrated that retaining thermal fluctuations in the fields $\mathbf{m}_i$ leads to results well beyond simple Hartree-Fock mean field calculations at finite temperature T and, more importantly, in good qualitative and sometimes quantitative agreement with DQMC.

While the HS fields are treated via MC methods, the quadratic fermionic sector still needs to be handled numerically. The simplest and most widely employed method, starting with efforts in the manganite community to study the double exchange model [142], is simply to carry out an exact diagonalization (ED) of the fermions in a fixed classical $\mathbf{m}_i$ background, employing library subroutines. The $\mathbf{m}_i$ variables are then updated with a standard classical MC procedure where updates are accepted/rejected using the Metropolis algorithm. At a fixed temperature, this process is repeated until a thermalized regime is reached where observables can be measured.

Similarly as with the majority of techniques dealing with strongly correlated electrons, here there is no small parameter controlling the approximation. In particular there is no rigorous proof of convergence or bounded errors. However, as long as the mean field approximation employed as the starting point of the approximation (in the present example

the HartreeFock method) captures the essence of the ground state, then it is reasonable to assume that the present method will treat correctly the thermal fluctuations and associated short-range order tendencies above the critical temperature.

In the present work, we consider the case of half filling, where the total density is fixed by adjusting the chemical potential μ . The Hamiltonian is studied on cubic lattices with 4^3 to 16^3 sites and with periodic boundary conditions. The magnetic structure factors $S(q)$ are used to perform finite lattice size scaling to extract thermodynamic Néel temperatures in three dimensions. Results obtained on two-dimensional clusters will also be shown. All parameters are specified in units of the hopping t . In practice a total of 4000 MC system sweeps were performed: 2000 were used to thermalize the system, while the rest were used for calculating observables. A MC system sweep consists of sequentially visiting every lattice site and updating the local $\mathbf{m}_i$ vector followed by the fermionic ED, and then accepting/rejecting the proposed local field change following the Metropolis algorithm. The local density $\langle n_i \rangle$ is computed from the eigenvectors after each diagonalization. In our calculation, we start the simulation at high temperature with a random configuration of $\mathbf{m}_i$ variables and then cool down to lower temperatures. To study the formation of local moments, as explained below, we start the MC runs at $T/t = 100$ and cool down in steps of $\Delta T/t = 1$ up to $T/t = 1$. From $T/t = 1$ to 0.1 , we use a step size of $0.1t$. Below this temperature, specifically from $0.1t$ to $0.005t$, we reduce further the interval and use $\Delta T/t = 0.05$. This slow process allows us to avoid metastable states or obtaining results that depend substantially on the initial conditions of the calculation.

To characterize the Hubbard model, a number of observables are computed during the MC procedure. In particular, we calculate the density of states (DOS), $N(\omega) = \sum_m \delta(\omega - \omega_m)$, where ω_m are the eigenvalues of the fermionic sector and the summation runs up to $2N^3$, i.e., the total number of eigenvalues of a N^3 system with spin. $N(\omega)$ is calculated by implementing the usual Lorentzian representation of the δ function. The broadening needed to obtain $N(\omega)$ from the Lorentzians is $\sim BW/2N^2$, where BW is the fermionic bandwidth at $U = 0$. Numerically for the 4^3 system, the broadening is about $0.09t$. Two hundred $N(\omega)$ samples are obtained from the 2000 measurement system sweeps at every temperature. We discard 10 MC steps between measurements to reduce self-correlations in the data. The 200

$N(\omega)$ samples are used to obtain the thermally averaged $\langle N(\omega) \rangle_T$ at a fixed temperature. These are further averaged over data obtained from 10 to 20 independent runs with different random number seeds.

Information regarding the Néel AFM order expected at half filling is obtained from the magnetic structure factor,

$$S(\mathbf{q}) = \frac{1}{N^2} \sum_{i,j} e^{i\mathbf{q} \cdot (\mathbf{r}_i - \mathbf{r}_j)} \langle \mathbf{S}_i \cdot \mathbf{S}_j \rangle, \quad (\text{A.7})$$

where $\mathbf{q} = \{\pi, \pi, \pi\}$ is the wave vector of interest. The spins $\mathbf{S}_i$ are constructed from the eigenvectors of the equilibrated configurations.

We also calculate the real-space correlation function between the $\mathbf{S}_i$ vectors. This correlation function is defined as

$$C(|\mathbf{r}|) = \frac{1}{P} \sum_{|\mathbf{r}|=|\mathbf{i}-\mathbf{j}|, a} (-1)^{|\mathbf{i}-\mathbf{j}|} \langle \mathbf{S}_i^a \cdot \mathbf{S}_j^a \rangle, \quad (\text{A.8})$$

In $C(|\mathbf{r}|)$ the summation runs over all P pairs of sites at a distance $|\mathbf{r}|$ and is normalized accordingly. The sum over a runs over the three directions x, y and z .

The distribution of the magnitude of $|\mathbf{m}_i|$ on the lattice is measured by the distribution function $P_q(|m|)$. This is defined as $P_q(|m|) = \sum_i \delta(|m| - |\mathbf{m}_i|)$. For computational purposes, a Lorentzian representation with suitable broadening is used. $S(\mathbf{q})$, $P_q(|m|)$, and $C(|\mathbf{r}|)$ are also averaged in the same manner as described for $N(\omega)$. We also compute the specific heat $C_v(U, T) = \frac{dE(U, T)}{dT}$ by numerically differentiating the average energy with respect to temperature. Other observables that we measured are presented in Appendix B.

Appendix B

Two-Orbital Model for Pnictides (MCMF)

A Details of the MC simulation and observables measured

The lattices used all have periodic boundary conditions. In our calculations, a total of 4000 MC system sweeps were typically performed: 2000 to thermalize the system, and the rest for calculating observables. A MC system sweep consists of sequentially visiting every lattice site and updating the local auxiliary fields followed by the fermionic diagonalization or TCA procedure to accept/reject via the Metropolis algorithm. We start the simulation at high temperature with a random configuration of auxiliary fields and then slowly cool down to lower temperatures to avoid being trapped in metastable states. The MC runs start at $T = 1.0t$, which corresponds to about 1000 K and cool down in steps of $\Delta T = 0.01t$ up to $T = 0.005t$. This slow process allows us to obtain reliable results independent of the initial conditions of the calculation.

In our finite systems, there is no energy difference between the $(\pi, 0)$ and $(0, \pi)$ magnetic states. As a consequence, MC simulations that start at high temperature in a random state for the auxiliary fields may end up in $(\pi, 0)$ or $(0, \pi)$ with equal chance upon cooling. In

practice, we simply discarded all cooling down MC processes that led to a $(0, \pi)$ state at low temperatures.

The antiferromagnetic order is studied via the spin structure factors,

$$S(\mathbf{q}) = \frac{1}{L^4} \sum_{i,j} e^{i\mathbf{q} \cdot (\mathbf{r}_i - \mathbf{r}_j)} \langle S_i^z S_j^z \rangle. \quad (\text{B.1})$$

The two wavevectors of interest in FeSC are $\mathbf{q} = (\pi, 0)$ and $(0, \pi)$. The expectation value is generated by using the eigenvectors of the MC equilibrated configurations. For the study of the nematic regime above T_N , we compute the nematic order parameter

$$\Psi_{Nem} = \frac{1}{2L^2} \sum_{i,\pm} (S_i^z S_{i\pm\hat{y}}^z - S_i^z S_{i\pm\hat{x}}^z), \quad (\text{B.2})$$

where $\hat{x}$ and $\hat{y}$ are unit vectors connecting site i with its nearest neighbors. The $\pm$ summation is over all nearest neighbors, and $\langle \Psi_{Nem} \rangle > 0$ in the $(\pi, 0)$ magnetic phase. To better locate critical temperatures, we also calculate numerically the magnetic ($\chi_S = \partial S(\mathbf{q})/\partial T$) and nematic ($\chi_{Nem} = \partial \Psi_{Nem}/\partial T$) susceptibilities.

In this work, we have also evaluated the orbital resolved density of states (DOS), $N_\alpha(\omega) = \sum_m |\langle \xi_{m,\alpha} | \psi \rangle|^2 \delta(\omega - \omega_m)$, where ω_m are the eigenvalues of the fermionic sector and the summation runs up to $2L^2$, i.e. the total number of eigenvalues of a L^2 system with spin. $|\langle \xi_{m,\alpha} | \psi \rangle|^2$ is the weight of the m^{th} eigenstate for orbital α in the state $|\psi\rangle$. $N_\alpha(\omega)$ is calculated by implementing the usual Lorentzian representation of the δ function. The broadening needed to obtain $N(\omega)$ from the Lorentzians is $\sim W/2L^2$, where W is the fermionic bandwidth at $U = 0$. Finally, $N(\omega)$, the total DOS, is the sum of the different orbital densities of states. Numerically, e.g., for the 8^2 system the broadening is about $0.09t$. Two hundred $N_\alpha(\omega)$ samples are obtained from the 2000 measurement system sweeps at every temperature. We discard 10 MC steps between measurements to reduce self-correlations in the data. The 200 $N_\alpha(\omega)$ samples are used to obtain the thermally averaged $\langle N_\alpha(\omega) \rangle_T$ at a fixed temperature. These are further averaged over data obtained from 10 independent runs with different random number seeds. A very similar procedure is followed to calculate the optical conductivity, that involves matrix elements of the current operator.

To determine the crossover temperature between the weak coupling paramagnetic state and the regime with preformed local moments above the magnetic order, we compute the specific heat, $C_v(U, J, T) = \frac{dE(U, J, T)}{dT}$, by numerically differentiating the average energy with respect to temperature, as well as the orbital resolved double occupation $\langle n_{\alpha, \uparrow} n_{\alpha, \downarrow} \rangle$. The details of the procedure we followed is presented in our earlier work on the one-orbital Hubbard model [82], but, briefly, $C_v(U, J, T)$ has a high-temperature peak that corresponds to local moment formation that can be tracked varying U . In addition, the double occupation has to be below a cutoff for the system to have local moments (see [82]). This is of particular importance at small U values because $C_v(U, J, T)$ can have a considerable contribution from the electronic delocalization and in this regime it cannot be used to track local moment formation.

B Test of the Technique in the Hartree Approximation

The many-body technique used in this thesis was already introduced and tested in [82] of the main text. However, since an easy axis is present in most materials and considering that empirically the Monte Carlo convergence is improved under such circumstances (colloquially, Ising is easier than Heisenberg), in this project it was decided to use the Ising approximation in the Hund term. In the language of the one-orbital Hubbard model that corresponds to using the Hartree approximation instead of the Hartree-Fock approximation employed before. This requires, then, a test of the Hartree assumption for the three-dimensional one-orbital standard Hubbard model at half-filling. There is no need to provide explicitly the Hamiltonian for such well known model, thus we move immediately to discuss the results, which are provided in Fig. B.1.

The results shown in Fig. B.1 are encouraging. The critical temperatures found with the MC-MF technique capture the “up and down” behavior of T_N with increasing U/t and they converge close to the expected scaling at large U/t . Moreover, in the entire range of U/t investigated the MC-MF results are close to those of quantum Monte Carlo (with the largest discrepancy being about 20%). The successful test presented in Fig. B.1 suggests that the MC-MF technique captures the essence of the one-orbital problem, not only qualitatively

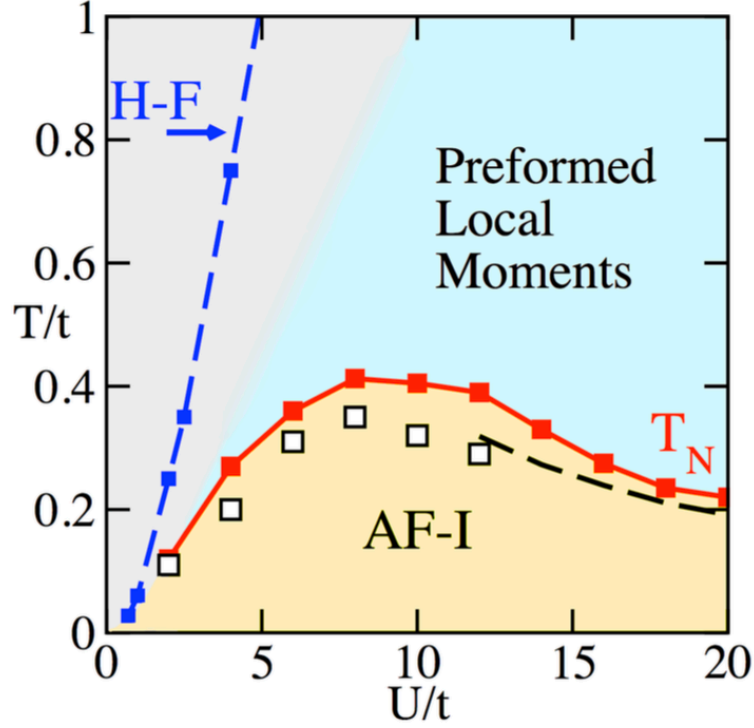


Figure B.1: The Hubbard U/t vs. temperature T/t phase diagram corresponding to the one-orbital Hubbard model in three dimensions using the Hartree approximation in the MC-MF technique. T_N denotes the Néel critical temperature where antiferromagnetism with (π, π, π) staggered magnetic order develops (region denoted as AF-I) according to measurements of the spin structure factor and the spin-spin correlation functions. The MC-MF results (red filled squares) are compared against data obtained using the Determinant Quantum Monte Carlo [white squares, reproduced from R. Staudt *et. al*, Eur. Phys. J. **B** 17, 411 (2000)]. The lattice size used in the MC-MF method is 4^3 . The expected $4t^2/U$ behavior at large U/t is indicated by the black dashed line. The more crude mean-field Hartree Fock approximation results are denoted by “H-F” (blue dashed line): it incorrectly predicts the growth of the critical temperature with increasing U/t . In the light blue region measurements of the spin square operator and the double occupancy indicate the presence of a local moment, as discussed before in [82] of the main text.

but also quantitatively. This gives us confidence that the results for two orbitals in the main text, that have not been studied before in the literature, are reliable.

C Optical Conductivity

To illustrate the physics of the three different states reported in the two-orbital Hubbard Model, the optical conductivity was calculated. The results are in Fig. B.2. Panel (a) is in the AF-M regime: while the $(\pi, 0)$ magnetic order breaks the symmetry between the x and y directions, the difference is not dramatic and leads to both directions being metallic. Panel (c) corresponds to the Mott insulating regime: here a gap is present both when the electric field points along the x and y directions. The first excitations occur at the scale of the Hund coupling, along the x spin staggered direction. The most novel result is shown in panel (b), already presented in the main text and reproduced here for the benefit of the readers, corresponding to the new OSDC region: here at $\omega/t \sim 0$ there is a finite weight in the x direction but negligible weight in the y direction, compatible with the calculation of the resistivity shown in the main text.

Some subtle technical details are worth discussing, particularly with regards to the window in ω , around zero, used to define the resistivity. Consider a L^2 lattice. Its associated mean level spacing is roughly estimated as $s = W/(4L^2)$ where W is the bandwidth. In practice the individual δ -functions broadening used in the conductivity calculation is s times a factor, which it has been chosen to be 4 in our calculations. As example, for a 20^2 system $s = 12/(4 \times 20^2) = 0.0075$, and the broadening used is then 0.03. The integration range is decided as follows: the smallest frequency (the starting ω) is chosen to be at least one order of magnitude smaller than s . The frequency increment is chosen to be $s/10$. The data of these 10 frequency points is used for the integration and the outcome is ascribed as the average conductivity for the frequency value at the lower end of the interval. In the example given above, then the frequency step is 0.003. The integration is performed over 10 frequency steps. The calculation is started at an initial frequency of $0.0001t$.

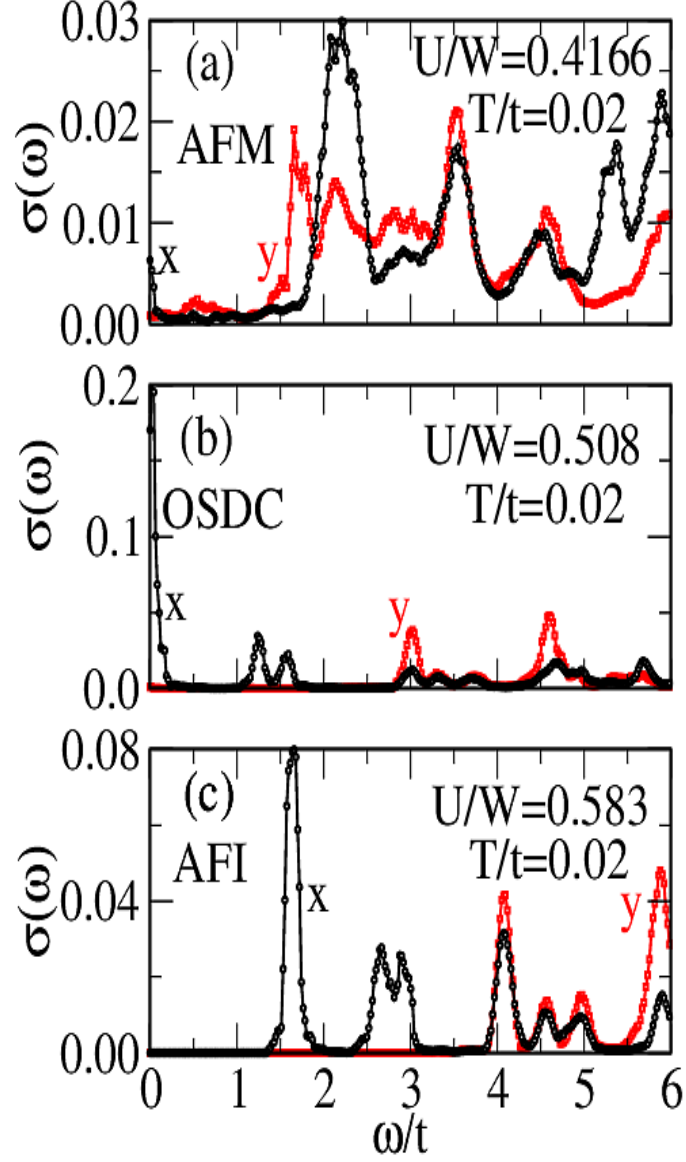


Figure B.2: Optical conductivity of the 2D two-orbital model used in this study at the temperatures and couplings U/W indicated. Panel (a) is in the AF-M regime; panel (b) in the OSDC regime; and panel (c) in the AF-I state. All data are using a 16^2 system.

D Other results used to construct the magnetic phase diagram

The analysis presented in Fig. 3.3 of the main text is just an example of the detailed computational study carried out at various values of U/W and used to construct the phase diagram of Fig. 3.2. In Fig. B.3 this substantial effort is illustrated further by providing data corresponding to other lattice sizes and couplings as well. The information gathered from these efforts for T_N , T_{Nem} , and T_{split} were used for the finite-size scaling analysis of Fig. 3.4(a).

E Mean field formalism

For completeness, here details of the mean-field approximation are provided. In general, the multiorbital Hubbard model shown in the main text can be written as follows:

$$H = H_o + H_{int} = \sum_{\langle i,j \rangle, \alpha, \beta, \sigma} T_{\alpha, \beta}^{i,j} d_{i, \alpha, \sigma}^\dagger d_{j, \beta, \sigma} + \sum_{i, \sigma, \sigma'} \sum_{\alpha, \alpha', \beta, \beta'} U_{\sigma, \sigma'}(\alpha, \alpha', \beta, \beta') d_{i, \alpha, \sigma}^\dagger d_{i, \alpha', \sigma'}^\dagger d_{i, \beta', \sigma'} d_{i, \beta, \sigma}. \quad (\text{B.3})$$

H_o (H_{int}) in the kinetic (interaction) term. $d_{i, \beta, \sigma}^\dagger$ creates an electron at site i^{th} , orbital β , and with spin projection σ . The onsite interaction is:

$$\begin{aligned} U_{\sigma, \sigma'}(\alpha, \alpha', \beta, \beta') &= \frac{U}{2} \delta_{-\sigma, \sigma'} \delta_{\alpha, \alpha'} \delta_{\alpha \beta} \delta_{\alpha \beta'} \\ &+ \frac{U'}{2} (1 - \delta_{\alpha \alpha'}) \delta_{\alpha \beta} \delta_{\alpha' \beta'} \\ &+ \frac{J}{2} (1 - \delta_{\alpha \alpha'}) \delta_{\alpha \beta'} \delta_{\alpha' \beta} \\ &+ \frac{J'}{2} \delta_{\alpha \alpha'} \delta_{\beta \beta'} (1 - \delta_{\sigma \sigma'}) (1 - \delta_{\alpha \beta}). \end{aligned} \quad (\text{B.4})$$

To derive the MF Hamiltonian we follow the treatment in *Quantum Theory of Finite Systems*, by Blaizot, J.-P. & Ripka, G., The MIT Press (1985). The advantage of this

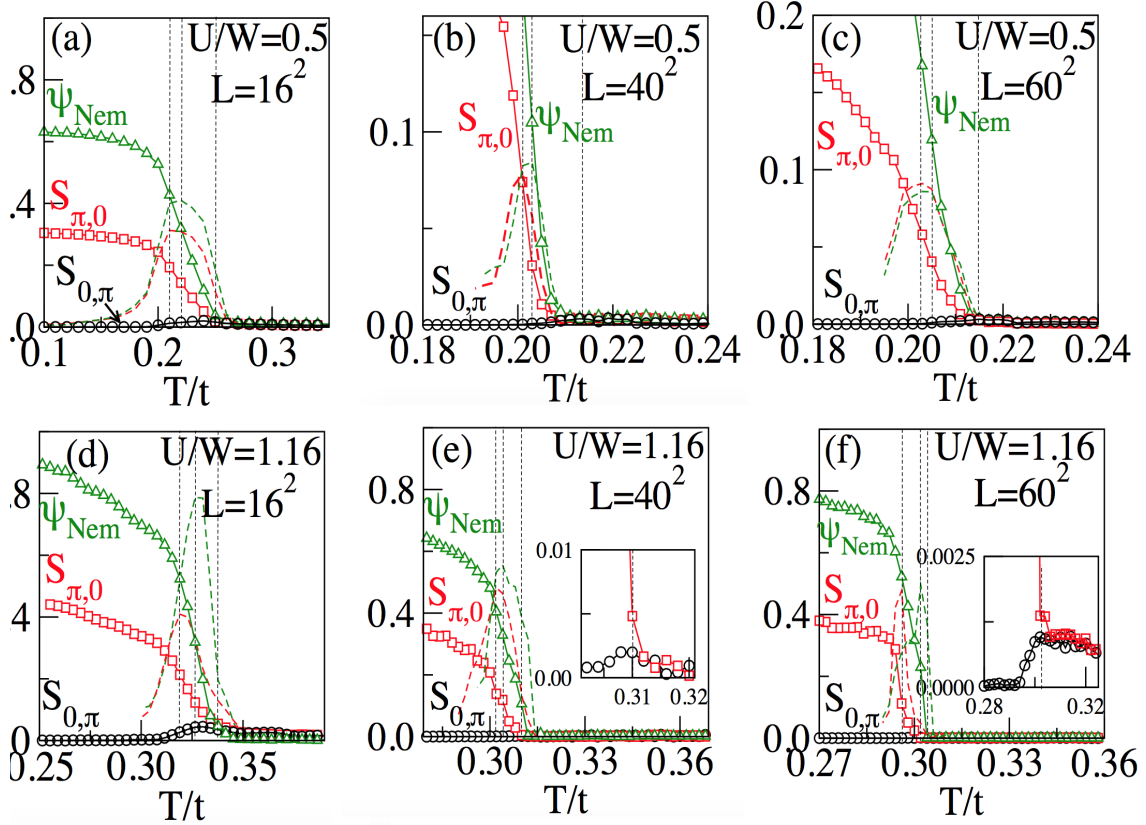


Figure B.3: The $(\pi, 0)$ and $(0, \pi)$ magnetic structure factors, and the nematic order parameter (Ψ_{Nem}) vs. temperature for different lattice sizes, at the couplings indicated. The red and green dashed lines without symbols are the temperature derivatives (numerically obtained) of the magnetic structure factor $S(\pi, 0)$ and Ψ_{Nem} with the same color coding as those order parameters. The insets in (e) and (f) are magnified versions of the data in the main panel showing the splitting between the $(\pi, 0)$ and $(0, \pi)$ magnetic structure factors. In all panels the dashed vertical lines (black) indicate, from left to right, the location of the peak of the derivatives of $S(\pi, 0)$ and Ψ_{Nem} , as well as the temperature where the $(\pi, 0)$, $(0, \pi)$ magnetic structure factors split, respectively.

approach is that one can derive a single general expression for the mean field parameters, for any number of orbitals. This expression can be easily coded in, thus avoiding the need to derive all possible mean field decouplings by hand. For this purpose we introduce the following notation:

$$\rho_{i,j,\alpha,\beta} = \langle \alpha | \rho_{i,j} | \beta \rangle = \langle d_{j,\beta}^\dagger d_{i,\alpha} \rangle, \quad (\text{B.5})$$

where $\rho_{i,j,\alpha,\beta}$ are elements of the single particle density matrix. We now make the Hartree-Fock approximation assumption: the state of the system can be represented by a single Slater determinant, $|\Psi\rangle$. By using the Wick's theorem, we can then write down the expectation value of H in $|\Psi\rangle$, denoted by $E[\rho]$, as:

$$\begin{aligned} E[\rho] = & \sum_{\langle i,j \rangle, \alpha, \beta, \sigma} T_{\alpha, \beta}^{i,j} \langle \beta, \sigma | \rho_{i,j} | \alpha, \sigma \rangle + \\ & \sum_{i, \sigma, \sigma'} \sum_{\alpha, \alpha', \beta, \beta'} U_{\sigma, \sigma'}(\alpha, \alpha', \beta, \beta') \times \\ & [\langle \beta, \sigma | \rho_{i,i} | \alpha, \sigma \rangle \langle \beta', \sigma' | \rho_{i,i} | \alpha', \sigma' \rangle \\ & - \langle \beta', \sigma' | \rho_{i,i} | \alpha, \sigma \rangle \langle \beta, \sigma | \rho_{i,i} | \alpha', \sigma' \rangle]. \end{aligned} \quad (\text{B.6})$$

We now take the derivative of $E[\rho]$ with respect to a generic density matrix element, $\langle \bar{\alpha}, \bar{\sigma} | \rho_{i,i} | \bar{\beta}, \bar{\sigma}' \rangle$, to get an explicit formula for Hartree-Fock mean field parameters, $\langle \bar{\beta}, \bar{\sigma}' | h | \bar{\alpha}, \bar{\sigma} \rangle$. Running over all values of orbitals and spin in $\langle \bar{\alpha}, \bar{\sigma} | \rho_{i,i} | \bar{\beta}, \bar{\sigma}' \rangle$, for taking the derivatives, generates all possible mean field decouplings in the Hartree-Fock channel. Thus, the general formula is as follows:

$$\begin{aligned} \langle \bar{\beta}, \bar{\sigma}' | h | \bar{\alpha}, \bar{\sigma} \rangle = & \sum_{\alpha', \beta'} \left(\sum_{\sigma'} \right. \\ & [U_{\bar{\sigma}\sigma'}(\bar{\beta}, \alpha', \bar{\alpha}, \beta') \delta_{\bar{\sigma}\bar{\sigma}'} + U_{\sigma'\bar{\sigma}}(\alpha', \bar{\beta}, \beta', \bar{\alpha}) \delta_{\bar{\sigma}\bar{\sigma}'}] \times \\ & \langle \beta', \sigma' | \rho_{i,i} | \alpha', \sigma' \rangle \\ & - [U_{\bar{\sigma}'\bar{\sigma}}(\bar{\beta}, \alpha', \beta', \bar{\alpha}) + U_{\bar{\sigma}\bar{\sigma}'}(\alpha', \bar{\beta}, \bar{\alpha}, \beta')] \times \\ & \left. \langle \beta', \bar{\sigma}' | \rho_{i,i} | \alpha', \bar{\sigma} \rangle \right). \end{aligned} \quad (\text{B.7})$$

Finally, the full Hartree-Fock Hamiltonian is given by:

$$\begin{aligned}
h_{MF} = & \sum_{\langle i,j \rangle, \alpha, \beta, \sigma} T_{\alpha, \beta}^{i,j} d_{i, \alpha, \sigma}^{\dagger} d_{j, \beta, \sigma} \\
& + \sum_{i, \bar{\alpha}, \bar{\sigma}} \sum_{\bar{\beta}, \bar{\sigma}'} \langle \bar{\beta}, \bar{\sigma}' | h | \bar{\alpha}, \bar{\sigma} \rangle d_{i, \bar{\alpha}, \bar{\sigma}}^{\dagger} d_{i, \bar{\beta}, \bar{\sigma}'} \\
& - \sum_{i, \sigma, \sigma'} \sum_{\alpha, \alpha', \beta, \beta'} U_{\sigma, \sigma'}(\alpha, \alpha', \beta, \beta') \times \\
& (\langle \beta, \sigma | \rho_{i,i} | \alpha, \sigma \rangle \langle \beta', \sigma' | \rho_{i,i} | \alpha', \sigma' \rangle \\
& - \langle \beta', \sigma' | \rho_{i,i} | \alpha, \sigma \rangle \langle \beta, \sigma | \rho_{i,i} | \alpha', \sigma' \rangle).
\end{aligned} \tag{B.8}$$

Vita

Niravkumar Patel was born in Gujarat, India, to parents Dipak and Sunita Patel. As an undergraduate student, he first attended the East Arkansas Community College from 2007 to 2008, later transferring to the University of Central Arkansas from 2009 to 2012 where he earned a Bachelors of Science in Physics. After graduation, Niravkumar was accepted to the Doctoral program at the University of Tennessee in Knoxville, Tennessee. While a graduate student, Niravkumar worked as a Graduate Research Assistant for Dr. Elbio Dagotto. During this time, he focused on the numerical study of iron based superconductors using Monte Carlo mean field and density matrix renormalization group methods. The results of his graduate research led to a total of nine publications, and three APS march meeting presentations. He is scheduled to defend his Ph.D. in May 2018, and move in the Fall 2018 to Ohio State University to continue his scientific career as postdoctoral assistant.