Theoretical Studies of Strong Correlations in Cuprates

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Abstract of Dissertation

This thesis presents work that has been done to describe the high temperature superconducting cuprates by going beyond first principles calculations using the Hubbard model with \((\pi, \pi)\) antiferromagnetic and BCS superconducting order. The unique approach here is to fit the Hubbard model to either first principles or experimental band structure and obtain electronic structure properties by self consisting the antiferromagnetic and superconducting gaps as well as self consistently computing self energy corrections. The self energy corrections are determined through the QPGW model which correctly describes the cuprates as having intermediate coupling of electrons. These methods of modeling cuprates are used to describe the experimental results of spectroscopies such as photoemission and scanning tunneling spectroscopy. The first topic presented here studies the one, three, and four band versions of the Hubbard model. It is shown that, in the three and four band model, by fitting the tight binding parameters to first principles calculations and the antiferromagnetic gap to experimental gap measurements, cuprates should be described as charge transfer insulators instead of Mott insulators which is predicted by one band models. The one, three, and four band models with parameters fit to experiment predict a negative electronic compressibility. This phenomenon is shown to be due to a dramatic decrease in the Hubbard U with increased electron doping away from half filling. Switching focus to hole doped cuprates, the addition of superconducting order reveals a property known as Fermi surface free superconductivity. Fermi surface free superconductivity drives a topological transition from open to closed Fermi surfaces in the hole doped cuprates. STM measurements of the local density of states on hole doped cuprates show a gap filling rather than a gap closing with increased doping. A model of nanoscale phase separation in conjunction with a Coulomb gap which describes stripe pinning by impurities is shown to successfully model this gap filling. In a recent STM
study, unusual features were observed in the quasiparticle interference spectra of single layered bismuth cuprates, within the superconducting gap at the antinodes. To describe these feature a realistic model of quasiparticle interference is employed with self energy corrections which show quasiparticle weight being pushed into the antinodal regions when in the superconducting state. Overall, it is shown that the Hubbard model can be an effective tool in describing the cuprates and spectroscopies performed on them especially when using realistic bands from experiments or first principles along with self consistent order parameters and self energy corrections.
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2. *Fermi-Surface-Free Superconductivity in Underdoped (Bi,Pb)$_2$(Sr,La)$_2$CuO$_{6+\delta}$ (Bi2201)*  

3. *Nanoscale Phase Separation in Deeply Underdoped Bi$_2$Sr$_2$CuO$_{6+\delta}$ and Ca$_2$CuO$_2$Cl$_2*  


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Chapter 1

Introduction

Since the discovery of cuprate superconductors in 1986 [1] there has been great activity in developing a microscopic theory for high temperature superconductivity (HTS). The origins of HTS remain a mystery despite the extensive work done to find a solution. The reason for this is the complexity of the cuprate phase diagram. Describing electron interactions and their dispersion is key to understanding how and why HTS materials act as they do. The cuprates are transition metal oxides that at half filling are magnetic insulators displaying long range antiferromagnetic (AFM) order. For electron doped materials a Hubbard model with AFM and superconducting (SC) order is an appropriate description [2, 3, 4], where the AFM order for hole doped systems extends out to around 19 percent doping. In the low doping regime there exists a pseudogap which persists to higher doping, even intersecting the superconducting phase. At high dopings the cuprates appear to behave as Fermi liquids.
The pseudogap, which in hole doped cuprates exists simultaneously with the superconducting gap, remains one of the biggest puzzles in HTS. There are many different explanations for the pseudogap which can be put into two general categories. The first is that the pseudogap is superconducting in origin. This is explained by preformed Cooper pairs being created at a temperature $T^*$ above the critical temperature for superconductivity $T_c$. Superconductivity is not present in this temperature range due to phase fluctuations being too strong to support superconducting order. Experiments have shown a spectroscopic signature of preformed pairs [5]. In the second category the pseudogap is not considered to be of superconducting origin. Models such as AFM, stripe, or charge ordered phases are used to describe the pseudogap as a non-superconducting competing phase.

In the phase diagram of cuprates, the superconducting phase appears away from half filling, for both hole and electron doping. From the dopings at which it appears, the superconducting state persists in temperature to some critical temperatures, $T_c$, above which the order disappears. These critical temperatures form a dome in phase space called the superconducting dome. In the hole doping region of the superconducting dome at temperatures below $T_c$ the superconducting order and pseudogap exist simultaneously, giving rise to a two gap scenario. It is not entirely agreed upon as to how critical temperatures, $T^*$ and $T_c$, of the two gaps progress with doping. It is not clear if the two gaps merge gradually towards the overdoped (OD) end of the superconducting dome, or if the two gaps become equal at optimal doping (OPT) with the pseudogap becoming smaller than the superconducting gap at higher doping, or if the pseudo and superconducting
gaps terminate abruptly at optimal doping and only the superconducting gap persists to the OD regime [6]. In theoretical studies of Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ (Bi2212) a two gap scenario has been identified in a tight-binding Hubbard model with AFM and SC order [7].

In a specific doping range the band edge of the upper magnetic band (UMB) is in close proximity to the Fermi energy and thus to the superconducting gap region. The superconducting gap then acts to symmetrize the band edge around the Fermi energy, effectively creating two gaps which are both superconducting in nature but also depend on the AFM properties of the system. Many experiments have measured these two gaps [6, 8]. The compiled set of data in reference [6] for Bi2212, YBa$_2$Cu$_3$O$_{7-\delta}$ (Y123), Tl$_2$Ba$_2$CuO$_{6+\delta}$ (Tl201), and HgBa$_2$CuO$_{4+\delta}$ (Hg1201) shows a universal two gap scenario for these materials which seems to agree with the gradually merging two gap picture. However, for single layered Bismuth compounds Pb$_{0.55}$Bi$_{1.5}$Sr$_{1.6}$La$_{0.4}$CuO$_{6+\delta}$ (Pb-Bi2201) He et al. [9] show in angle resolved photoemission spectroscopy (ARPES) studies that the two gaps converge near the Van Hove singularity (VHS) which is at a much higher doping than Sawatzky’s universal two gap scenario [6].

There are many models which promise predictive powers for current and new HTS materials. Density Functional Theory (DFT) is used to derive the electronic structure of materials from first principles and can be coupled with other theories and approximations to improve its overall accuracy in specific correlated materials. The most recent developments in DFT have been in the strong coupling regime. There are a wide variety of methods starting with the Local Density Approximation (LDA) and being improved upon with such techniques as LDA+U (Hubbard U) and LDA+DMFT (Dynamic Mean Field
Theory). Other theories such as the GW theory [10] have the potential to provide electron self-energies in the intermediate coupling regime with full momentum dependence whereas DMFT is usually restricted to a self-energy uniform in momentum space. Even the full GW theory can quickly become computationally intensive, and approximations like $G_0W_0$ [10] are often deployed to lighten the computational load by using only the first iteration of a full GW calculation compromising accuracy for speed. A technique which is reasonably fast and accurate is the $G_ZW_Z$ method, also known as the Quasi-Particle GW (QPGW) method [11, 12].

A good deal of the physics of the cuprates has been shown to be captured within the framework of a tight-binding Hubbard Hamiltonian with co-existing AFM and SC order [2, 3, 4, 13]. The tight binding parameters are obtained by fitting to first principles DFT bands. The AFM order is taken to be a commensurate $(\pi, \pi)$ order and the SC order is given by a Bardeen-Cooper-Schrieffer (BCS) type d-wave superconducting (dSC) pairing. The resulting Green’s function is a 4X4 tensor. Three susceptibilities can then be calculated: charge, and spin transverse and longitudinal, as first derived by Schrieffer, Wen, and Zhang [14]. The Random Phase Approximation (RPA) is invoked, accounting for spin density wave (SDW) excitations in the longitudinal spin susceptibility. Vertex corrections in $G_ZW_Z$ are where the QPGW method differs from other implementations of the GW scheme. The approximation used here is that the vertex correction is considered a constant, $1/Z$, where $Z$ is an effective renormalization factor for the bands. $Z$ is calculated self-consistently such that the renormalized bands match the coherent bands in the low energy region of the self-energy corrected spectral weight [11, 12].
The work presented here aims to describe properties of the cuprates through models guided by results from experimental spectroscopies. The workhorse of this research is the tight binding Hubbard model with AFM+SC order. The second chapter is, therefore, dedicated to the details of the Hubbard model. Two applications of the Hubbard model are also addressed. Driven by experimental ARPES results [15] that put cuprates in the category of charge transfer insulators (CTI), a multiband Hubbard model is presented with a set of parameters which describe experiment and put cuprates clearly into the CTI class. The second application shows how a Hubbard model with a doping dependent on-site energy can produce a negative electronic compressibility (NEC) which implies that cuprates can exhibit a NEC as well. This study is inspired by ARPES studies on iridates which exhibit NEC [16].

Chapter three is motivated by simulations [17] of the iron based family of high temperature superconductors known as pnictides. The results of reference [17] report that even in the absence of a Fermi surface, a superconducting gap can arise due to the symmeterization of the spectral weight around the Fermi energy. This effect is call Fermi-surface-free superconductivity (FS-free SC) and, in the pnictide material where this can occur, is theoretically shown to increase the superconducting critical temperature. This chapter shows that FS-free SC can be seen in a tight binding Hubbard model of the cuprates with BCS superconductivity included. Furthermore, When AFM order is included, the FS-free SC produces a two gap scenario which gives insight into the observed spectral weight loss in experiments [5]. It also shows two topological transitions with doping for hole doped cuprates, analogous to those found in electron doped cuprates [3]. This picture
also fits well with the concept of a “trisected dome” [18]. The chapter summarizes the work published in reference [19].

The fourth chapter describes scanning tunneling microscopy (STM) experiments [20, 21, 22] which show a gap filling rather than a gap closing in the density of states of cuprates when hole doped. In this chapter a model is presented which explains the gap filling by considering the density of states (DOS) observed by STM to be a weighted superposition of two electronic end phases of different effective dopings. These phase domains are on the order of nanometers in size such that the overall charge neutrality of the system is preserved. This phenomenon is known as nanoscale phase separation (NPS). The DOS of the end phases are modeled using the tight binding Hubbard model. For this model, self energy corrections are applied through the QPGW method [12]. Further details are included such as impurity pinning by stripes which are modeled as a Coulomb gap [23] which describes the low energy features seen in the DOS of the cuprate \( \text{Ca}_2\text{CuO}_2\text{Cl}_2 \) (CCOC) [22]. The chapter summarizes the work published in reference [24].

In the fifth chapter, a phenomenon known as quasiparticle interference (QPI) is studied. QPI occurs when quasiparticles scatter off of impurities in a system and interfere with each other. This phenomenon is observed in cuprates because of the numerous impurity sites present in doped systems. STM studies can observe QPI by Fourier transforming the real space data in order to quantify the periodicity of the charge density structures in their measurements. STM measurements [25, 26] on Bi2201 have shown a transition of the QPI pattern from the standard octet picture at low doping to a state with strong QPI response in the antinodal region within the superconducting state. This chapter attempts
to model this behavior with self energy corrections applied to a T-matrix formalism [27] for computing QPI. Three types of self energy are compared. These self energies are the Dynes self-energy [28], pairbreaking self energy [29], and QPGW self energy [12].

The type of scatterer used for the model is also considered and the results for different scattering types and self energies are compared.

The appendices focus on the technical aspects of the research presented here. Appendix A presents a derivation of the QPGW method. In this section the temperature and momentum dependence of the QPGW method are discussed as well as the importance of these properties in extending mode coupling calculations [30] which can be used to help understand the phase diagram of the cuprates. Appendix B presents a detailed derivation of the multiband susceptibility including the RPA corrections.

Overall, this thesis presents a study of the cuprates using a mostly one band tight binding Hubbard model with $(\pi, \pi)$-AFM and SC order. The unique aspects of this research are how the model is used to interpret ARPES and STM results as well as using the QPGW method which assumes the cuprates are in the intermediate coupling regime.
Chapter 2

The Hubbard Model

Introduction

The Hubbard model was originally developed by John Hubbard in 1963 [31] in order to describe the interactions of electrons in a solid and is a good approximation for how electrons interact in a periodic potential. Since the Hubbard model’s inception it has been studied extensively and is now often used as a model for high temperature superconductors such as the cuprates and other correlated materials. The Hubbard model can only be solved exactly in one dimension. Therefore, a large number of approximate calculations have been studied. When solving problems in the framework of many-body perturbation theory, the mean-field solution is a good starting point. Because the cuprates exhibit antiferromagnetism, the mean-field approximation will be used to investigate magnetic order. The magnetic order often creates a band gap and the turning on or off of order
with changing model parameters, such as doping, creates a metal insulator transition. This feature happens to be a characteristic of the cuprates and led to the extensive use of the Hubbard model in studying cuprates.

The original Hubbard model considered only an atom with a single electronic orbital. In this model, $U$, the Hubbard potential, describes the Coulomb repulsion between electrons of opposite spins attempting to occupy the same lattice site. The Hubbard Hamiltonian, for a single band, written in momentum space is given by

$$H = \sum_{\mathbf{k},\sigma}(\epsilon_\mathbf{k} - \epsilon_F)c^\dagger_{\mathbf{k},\sigma}c_{\mathbf{k},\sigma} + U\sum_{\mathbf{k},\mathbf{k}'}c^\dagger_{\mathbf{k}+\mathbf{Q},\uparrow}c^\dagger_{\mathbf{k}'-\mathbf{Q},\downarrow}c_{\mathbf{k}',\downarrow}c_{\mathbf{k},\uparrow},$$

(2.1)

where $c^\dagger_{\mathbf{k},\sigma}$ ($c_{\mathbf{k},\sigma}$) are the creation (annihilation) operators for an electron at momentum $\mathbf{k}$ with spin $\sigma$, $\epsilon_\mathbf{k}$ is the corresponding non-interacting energy level, $\epsilon_F$ is the Fermi energy, and $\mathbf{Q} = (\pi, \pi)$. The band dispersion of the non-interacting system, $\epsilon_\mathbf{k}$, is modeled in the tight binding form as

$$\epsilon_\mathbf{k} = -2t[c_x(a) + c_y(a)]$$

$$- 4t'c_x(a)c_y(a) - 2t''[c_x(2a) + c_y(2a)]$$

$$- 4t'''[c_x(2a)c_y(a) + c_y(2a)c_x(a)],$$

(2.2)

in terms of the tight binding parameters $t^i$. Here, $c_i(aa) = \cos(\alpha k_i a)$, and $a$ is the lattice constant.

The interaction in the Hubbard model is given by the on-site repulsion of particles of
different spin on the same lattice site where the on-site repulsion energy is given by $U$.

For interactions between electrons of different band character other on-site interactions can be added which will be discussed in appendix B on multiband susceptibility. In this chapter, the system will consist of only one type of atom with one type of orbital.

This chapter will show how to apply mean field theory to the Hubbard interaction term in real space and momentum space for commensurate ($\pi, \pi$)-AFM order and for superconducting order. The two interactions will be combined to form an extended Hubbard model with superconducting order. General schemes for self consistent order parameters will be derived and the specific case of a self consistent superconducting and AFM gap will be derived analytically. This will culminate in the discussion of multiband Hubbard models and how they can predict cuprates to be in the charge transfer insulator regime as well as a discussion on how one, three and four band models can show negative electronic compressibility.

**The One Band Hubbard Model in Real Space**

The simplest version of the Hubbard model is the one band Hubbard model. When the model is considered in real space, the mean field approximation clearly shows antiferromagnetism and provides a picture of the concept of magnetic order parameters. A magnetic order parameter quantifies how much of the system is currently in a magnetically ordered state such as paramagnetism, ferromagnetism, or antiferromagnetism.
In real space, the interaction term of the Hubbard Hamiltonian is

\[ I = U \sum_i n_{i\uparrow} n_{i\downarrow} \]  

(2.3)

where \( n_{i\sigma} = c_{i\sigma}^\dagger c_{i\sigma} \), and \( i \) is the lattice site index. The Hubbard model, with interactions given by equation 2.3, has not been solved exactly for a system of dimension greater than one. In order to make this model solvable, the mean field approximation will be used. The mean field approximation takes a many body problem, where interactions between each particle must be considered, and assumes that a single particle "feels" all of the many interactions as a single average potential field. This approximation is made by investigating the number operator, \( n_{i\sigma} \), and small fluctuations around its average, \( \delta_\sigma \), such that

\[ n_{i\sigma} = \langle n_{i\sigma} \rangle + (n_{i\sigma} - \langle n_{i\sigma} \rangle) \]
\[ = \langle n_{i\sigma} \rangle + \delta_\sigma. \]  

(2.4)

Substituting this into \( n_{i\uparrow} n_{i\downarrow} \),

\[ n_{i\uparrow} n_{i\downarrow} = (\langle n_{i\uparrow} \rangle + \delta_\uparrow) (\langle n_{i\downarrow} \rangle + \delta_\downarrow) \]
\[ = \langle n_{i\uparrow} \rangle n_{i\downarrow} + n_{i\uparrow} \langle n_{i\downarrow} \rangle - \langle n_{i\uparrow} \rangle \langle n_{i\downarrow} \rangle, \]  

(2.5)

where it is assumed that \( \delta_\uparrow \delta_\downarrow \approx 0 \) due to fluctuations being small.

With the mean field approximation, the system can be discussed in terms of the average
number of electrons of either spin up \( \langle n_{i\uparrow} \rangle \) or spin down \( \langle n_{i\downarrow} \rangle \) on the lattice site \( i \). A simple path forward is to assume that the electrons will order antiferromagnetically where electrons on neighboring lattice sites have opposite spins. The square lattice can now be considered as two sub-lattices whose unit cells are twice the size of the original unit cell. One sub-lattice has predominantly spin up electrons and the other predominantly spin down electrons. The average number of electrons of spin \( \sigma \) on site \( i \) is equal to half of the total number of electrons, \( n \), plus the average magnetic moment, \( m \), for the up spin sublattice or minus the average magnetic moment for the down spin sublattice such that

\[
\langle n_{i\sigma} \rangle = \frac{n}{2} \pm m
\] (2.6)

the expression for the average magnetic moment can be obtained by noting that the total number of electrons is

\[
n = \langle n_{i\uparrow} + n_{i\downarrow} \rangle
\] (2.7)

such that

\[
m = \frac{1}{2} \left( \langle n_{i\uparrow} \rangle - \langle n_{i\downarrow} \rangle \right).
\] (2.8)

It is important to note that \( n \) and \( m \) are independent of the lattice site index \( i \). Doing this means that it is assumed that there is no pile up of charge. This means that the system is homogeneous in charge and there is only spin order. This model therefore describes
low energy physics associated with magnetism.

Using equation 2.6, equation 2.5 can be written as

\[ n_{\uparrow_i n_{\downarrow_i}} = \frac{n}{2} (n_{\uparrow_i} + n_{\downarrow_i}) + m (n_{\uparrow_i} - n_{\downarrow_i}) - \frac{n^2}{4} - m^2. \]  

(2.9)

This results in an interaction term

\[ I = \frac{U n}{2} \sum_i (n_{\uparrow_i} + n_{\downarrow_i}) + U m \sum_i (n_{\uparrow_i} - n_{\downarrow_i}) \]  

(2.10)

where the constant terms are dropped due to their being a constant energy shift. This type of order is called \((\pi, \pi)\) antiferromagnetic \((\pi, \pi)\)-AFM order because the periodicity of the antiferromagnetic order in momentum space is equal to \((\pi, \pi)\). This results in two distinct bands; one from the spin up sublattice and one from the spin down. These bands mix and split, due to the onsite interaction, and create a gap called the antiferromagnetic gap. The band at the higher energy is known as the upper magnetic band (UMB) and the band at the lower energy is known as the lower magnetic band (LMB). The following section will explore this model in momentum space which will be useful when approaching the problem of multiband susceptibilities.

**Mean Field Interactions and \((\pi, \pi)\)-AFM Order**

The same procedure for the mean-field approximation in real space can be carried out in momentum space. The advantage of doing this is that when moving to multiband
models it is easier to visualize the inter-band magnetizations as well as deal with the self consisting of the order parameters. To begin, the interaction can be written in momentum space as

$$I = \frac{U}{2} \sum_{\sigma} \sum_{k_1 k_2 k_3 k_4} c_{k_1 \sigma}^{\dagger} c_{k_2 \sigma} c_{k_3 \bar{\sigma}}^{\dagger} c_{k_4 \bar{\sigma}} \delta_{k_1 + k_3, k_2 + k_4},$$

(2.11)

where $\bar{\sigma}$ is the opposite spin to $\sigma$. This is the same interaction stated earlier in equation 2.3, but Fourier transformed into momentum space. Here, the two scatterers must have the total momentum coming into the system equal to the total momentum leaving the system, $k_1 + k_3 = k_2 + k_4$ and the momentum transfer must be equal and opposite $k_1 - k_2 = q$ and $k_3 - k_4 = -q$. These conditions eliminate one of the momentum space sums. The remaining three sums can be relabeled as sums over $k$, $k'$ and $q$. At this point, no restrictions have been put on the value of the momentum transfer $q$. Therefore, one obtains the relationship between $k$ and $k'$ as follows,

$$k_1 = k_2 + q, \quad k_2 = k, \quad k_4 = k_3 + q, \quad k_3 = k'.$$

(2.12)

The Hubbard U generates scattering between $k$ and $k + q$, for arbitrary $q$. In the mean-field approximation, it is assumed that one $q$ is dominant, and the matrix which couples $k$, $k + q$, etc. is diagonalized. For cuprates at half filling, it is known that $q = Q \equiv (\pi, \pi)$ represents the ground state of the system near half filling, so only this case will be
discussed. Therefore the interaction can be split up into two parts as,

\[ I = \frac{U}{2} \sum_{\sigma} \sum_{kk'} \left( c_{k\sigma}\bar{c}_{k'\sigma}c_{k'\bar{\sigma}}c_{k\bar{\sigma}} + c_{k+Q\sigma}\bar{c}_{k'\sigma}c_{k'\bar{\sigma}}c_{k\bar{\sigma}} + c_{k\sigma}\bar{c}_{k'\bar{\sigma}}c_{k'\sigma}c_{k\bar{\sigma}} + Q_{\sigma}\bar{c}_{k\sigma}c_{k\bar{\sigma}}c_{k'\sigma}c_{k'\bar{\sigma}} + c_{k\sigma}\bar{c}_{k'\bar{\sigma}}c_{k'\sigma}c_{k\bar{\sigma}} + Q_{\bar{\sigma}}\bar{c}_{k\sigma}c_{k\bar{\sigma}}c_{k'\sigma}c_{k'\bar{\sigma}} \right) . \]  

(2.13)

By restricting \( q \), the periodicity of the ordered state which can emerge from the unordered state is restricted to two values \( Q \) and 0. This is reasonable because the ground state of the system will be given by the ordered or unordered state with the lowest free energy. This happens to be antiferromagnetic order with \( Q = (\pi, \pi) \) periodicity for much of the cuprate phase diagram. The \( q = 0 \) order represents the ferromagnetic state.

The mean field approximation can be performed in momentum space the same way it was performed in real space in the previous section. The difference amounts to changing the index from \( i \) to \( k \). The approximation is made by investigating the number operator, \( n_{k\sigma} \), and small fluctuations around its average, \( \delta_{\sigma} \), such that

\[ n_{k\sigma} = \langle n_{k\sigma} \rangle + (n_{k\sigma} - \langle n_{k\sigma} \rangle) = \langle n_{k\sigma} \rangle + \delta_{\sigma} . \]  

(2.14)

Substituting this into \( n_{k\sigma}n_{k\bar{\sigma}} \),

\[ n_{k\sigma}n_{k\bar{\sigma}} = (\langle n_{k\sigma} \rangle + \delta_{\sigma})(\langle n_{k\bar{\sigma}} \rangle + \delta_{\bar{\sigma}}) = \langle n_{k\sigma} \rangle n_{k\bar{\sigma}} + \langle n_{k\bar{\sigma}} \rangle n_{k\sigma} - \langle n_{k\sigma} \rangle \langle n_{k\bar{\sigma}} \rangle , \]  

(2.15)
where it is assumed that $\delta^\alpha_\sigma \delta^\alpha_\bar{\sigma} \approx 0$ due to fluctuations being small. By choosing the proper momentum values for equations 2.15, mean field expressions for the first term,

$$
c_{k\sigma} c_{k\sigma} c_{k'\bar{\sigma}} c_{k'\bar{\sigma}} = \langle c_{k\sigma} c_{k\sigma} \rangle c_{k'\bar{\sigma}} c_{k'\bar{\sigma}} + \langle c_{k'\bar{\sigma}} c_{k'\bar{\sigma}} \rangle c_{k\sigma} c_{k\sigma} - \langle c_{k\sigma} c_{k\sigma} \rangle \langle c_{k'\bar{\sigma}} c_{k'\bar{\sigma}} \rangle, \quad (2.16)
$$

and second term,

$$
c_{k+Q_\sigma} c_{k+Q_\sigma} c_{k'\bar{\sigma}} c_{k'\bar{\sigma}} = \langle c_{k+Q_\sigma} c_{k+Q_\sigma} \rangle c_{k'\bar{\sigma}} c_{k'\bar{\sigma}} + \langle c_{k'\bar{\sigma}} c_{k'\bar{\sigma}} c_{k+Q_\sigma} c_{k+Q_\sigma} \rangle - \langle c_{k+Q_\sigma} c_{k+Q_\sigma} \rangle \langle c_{k'\bar{\sigma}} c_{k'\bar{\sigma}} c_{k+Q_\sigma} c_{k+Q_\sigma} \rangle, \quad (2.17)
$$

of equation 2.13 are obtained. By substituting these expressions back into the equation 2.13, the mean field interaction becomes

$$
I = \frac{U}{2} \sum_\sigma \sum_{kk'} \langle c_{k\sigma} c_{k\sigma} \rangle c_{k'\bar{\sigma}} c_{k'\bar{\sigma}} + \langle c_{k'\bar{\sigma}} c_{k'\bar{\sigma}} \rangle c_{k\sigma} c_{k\sigma} \\
+ \langle c_{k+Q_\sigma} c_{k+Q_\sigma} \rangle c_{k'\bar{\sigma}} c_{k'\bar{\sigma}} + \langle c_{k'\bar{\sigma}} c_{k'\bar{\sigma}} c_{k+Q_\sigma} c_{k+Q_\sigma} \rangle \langle c_{k+Q_\sigma} c_{k+Q_\sigma} \rangle \langle c_{k'\bar{\sigma}} c_{k'\bar{\sigma}} c_{k+Q_\sigma} c_{k+Q_\sigma} \rangle. \quad (2.18)
$$

The mean field interaction can be written as a matrix and the problem can be solved through matrix diagonalization.
The quantities in brackets in the mean field interaction have physical meaning which can be illuminated by defining the charge and longitudinal-spin densities as

\[
\langle \rho(q) \rangle = \sum_k \langle \left( c_{k+q\uparrow}^\dagger c_{k+q\downarrow}^\dagger \right) \begin{pmatrix} c_{k\uparrow} \\ c_{k\downarrow} \end{pmatrix} \rangle \\
= \sum_k \langle c_{k+q\uparrow}^\dagger c_{k\uparrow} \rangle + \langle c_{k+q\downarrow}^\dagger c_{k\downarrow} \rangle \\
= N_e \delta_{q,0} \tag{2.19}
\]

and

\[
\langle S^z(q) \rangle = \frac{1}{2} \sum_k \langle \left( c_{k+q\uparrow}^\dagger c_{k\uparrow}^\dagger \right) \sigma^z \begin{pmatrix} c_{k\uparrow} \\ c_{k\downarrow} \end{pmatrix} \rangle \\
= \frac{1}{2} \sum_k \left[ \langle c_{k+q\uparrow}^\dagger c_{k\uparrow} \rangle - \langle c_{k+q\downarrow}^\dagger c_{k\downarrow} \rangle \right]. \tag{2.20}
\]

respectively, where \( \mathbb{I} \) is the 2X2 identity matrix and \( \sigma^z \) is the Pauli spin matrix in the \( z \)-direction. At this point it is necessary to make a few assumptions about the system. First, it is assumed that the charge distribution is homogeneous by writing \( \delta_{q,0} \) in the charge density equation. Secondly, it is assumed that the system will have spin order with \( q = Q = (\pi, \pi) \). Therefore,

\[
\langle \rho(Q) \rangle = \sum_k \langle c_{k+Q\uparrow}^\dagger c_{k\uparrow} \rangle + \langle c_{k+Q\downarrow}^\dagger c_{k\downarrow} \rangle \\
= 0 \tag{2.21}
\]
which implies,

\[ \langle c_{k+Q\uparrow}^\dagger c_{k\uparrow} \rangle = -\langle c_{k+Q\downarrow}^\dagger c_{k\downarrow} \rangle. \]  (2.22)

Because the above quantity is hermitian, it is also true that

\[ \langle c_{k+Q\sigma}^\dagger c_{k\sigma} \rangle^\dagger = \langle c_{k\sigma}^\dagger c_{k+Q\sigma} \rangle. \]  (2.23)

Using the relation in equation 2.22, \( \langle S_z(q) \rangle \), for \( q = Q \), becomes

\[ \langle S_z(Q) \rangle = \frac{1}{2} \sum_k \left[ \langle c_{k+Q\uparrow}^\dagger c_{k\uparrow} \rangle - \langle c_{k+Q\downarrow}^\dagger c_{k\downarrow} \rangle \right] \]

\[ = \sum_k \langle c_{k+Q\uparrow}^\dagger c_{k\uparrow} \rangle. \]  (2.24)

At this point, it is convenient to define the magnetic order parameter, \( m \), and the electron density, \( n_\sigma \), as

\[ m = \sum_k \langle c_{k+Q\uparrow}^\dagger c_{k\uparrow} \rangle = -\sum_k \langle c_{k+Q\downarrow}^\dagger c_{k\downarrow} \rangle \]

\[ n_\sigma = \sum_k \langle c_{k\sigma}^\dagger c_{k\sigma} \rangle, \]  (2.25)
which can be used to simplify the equation for the interaction,

\[ I = \frac{U}{2} \sum_{kk'} (c_{k\uparrow} \c^\dagger_{k'} c_{k'} \downarrow c_{k'} \downarrow + c_{k\downarrow} \c^\dagger_{k'} c_{k'} \uparrow c_{k'} \uparrow + c_{k\downarrow} \c^\dagger_{k'} c_{k'} \uparrow c_{k'} \uparrow + c_{k\uparrow} \c^\dagger_{k'} c_{k'} \uparrow c_{k'} \uparrow ) c_{k\downarrow} c_{k\downarrow} \\
+ (c_{k+Q\downarrow} c_{k\uparrow} c_{k+Q\uparrow} + c_{k\downarrow} \c^\dagger_{k'} c_{k'} c_{k'} \downarrow c_{k'} \downarrow + (c_{k\uparrow} c_{k\downarrow} + c_{k\downarrow} c_{k\uparrow}) c_{k+Q\downarrow} c_{k\uparrow} \\
+ (c_{k+Q\downarrow} c_{k\uparrow} c_{k+Q\uparrow} + c_{k\downarrow} \c^\dagger_{k'} c_{k'} c_{k'} \uparrow c_{k'} \uparrow ) c_{k+Q\downarrow} c_{k\uparrow} \\
\] 

\[ = \frac{U}{2} \sum_{k} 2n_{\uparrow} c_{k\downarrow} c_{k\downarrow} + 2n_{\downarrow} c_{k\uparrow} c_{k\uparrow} \\
+ m c_{k+Q\downarrow} c_{k+Q\uparrow} - m c_{k+Q\uparrow} c_{k+Q\downarrow} - m c_{k\uparrow} c_{k\downarrow} - m c_{k\downarrow} c_{k\uparrow} \\
= \frac{U}{2} \sum_{k} 2n_{\uparrow} c_{k\downarrow} c_{k\downarrow} + 2n_{\downarrow} c_{k\uparrow} c_{k\uparrow} \\
+ m c_{k+Q\downarrow} c_{k+Q\uparrow} - m c_{k+Q\uparrow} c_{k+Q\downarrow} - m c_{k\uparrow} c_{k\downarrow} - m c_{k\downarrow} c_{k\uparrow} \tag{2.26} \]

The introduction of a non-zero transfer momentum in the scattering process results in a modification of the Brillouin zone (BZ). In the paramagnetic (PM) state each k-point in the BZ is unique, however the non-zero transfer momentum creates double counting for sums over the full PM BZ. This now signals that the PM BZ is no longer the smallest zone with unique k’s. This is because the real space unit cell must grow in size to accommodate periodic modulation of the AFM order parameter. When the real space unit cell grows, the BZ, in reciprocal space, must shrink. Because \( Q = (\pi, \pi) \) was assumed, the BZ must be shrunk to the AFM zone. To explicitly remove this double counting, the sum over the reduce AFM zone will be denoted as \( \sum_{k} \). By converting the sum, the interaction can be
written as

\[
I = \frac{U}{2} \sum'_{k} \left[ 2n_{\uparrow} \left( c_{k\downarrow}^{\dagger}c_{k\downarrow} + c_{k+Q\downarrow}^{\dagger}c_{k+Q\downarrow} \right) + 2n_{\downarrow} \left( c_{k\uparrow}^{\dagger}c_{k\uparrow} + c_{k+Q\uparrow}^{\dagger}c_{k+Q\uparrow} \right) \right]
+ m \left( c_{k\downarrow}^{\dagger}c_{k\downarrow} + c_{k+Q\downarrow}^{\dagger}c_{k\downarrow} \right)
- m \left( c_{k\uparrow}^{\dagger}c_{k\uparrow} + c_{k+Q\uparrow}^{\dagger}c_{k\uparrow} \right)
+ m \left( c_{k\downarrow}^{\dagger}c_{k\downarrow} + c_{k\uparrow}^{\dagger}c_{k\uparrow} \right) \right].
\] (2.27)

Where \( k + 2Q = k \) since \( 2Q = (2\pi, 2\pi) \) and \( k \) is \( 2\pi \) periodic (given a lattice length of 1).

Combining the terms gives

\[
I = \sum_{k} \left[ Un_{\uparrow} \left( c_{k\downarrow}^{\dagger}c_{k\downarrow} + c_{k+Q\downarrow}^{\dagger}c_{k+Q\downarrow} \right) + Un_{\downarrow} \left( c_{k\uparrow}^{\dagger}c_{k\uparrow} + c_{k+Q\uparrow}^{\dagger}c_{k+Q\uparrow} \right) \right]
+ \frac{U}{2} \left( m + m \right) \left( c_{k\downarrow}^{\dagger}c_{k\downarrow} + c_{k+Q\downarrow}^{\dagger}c_{k\downarrow} \right)
- \frac{U}{2} \left( m + m \right) \left( c_{k\uparrow}^{\dagger}c_{k\uparrow} + c_{k+Q\uparrow}^{\dagger}c_{k\uparrow} \right)
= \sum_{k} \left[ Un_{\uparrow} \left( c_{k\downarrow}^{\dagger}c_{k\downarrow} + c_{k+Q\downarrow}^{\dagger}c_{k+Q\downarrow} \right) + Un_{\downarrow} \left( c_{k\uparrow}^{\dagger}c_{k\uparrow} + c_{k+Q\uparrow}^{\dagger}c_{k+Q\uparrow} \right) \right]
+ \Delta \left( c_{k\downarrow}^{\dagger}c_{k+Q\downarrow} + c_{k+Q\downarrow}^{\dagger}c_{k\downarrow} \right)
- \Delta \left( c_{k\uparrow}^{\dagger}c_{k+Q\uparrow} + c_{k+Q\uparrow}^{\dagger}c_{k\uparrow} \right) .
\] (2.28)
Where \( \Delta = \frac{U}{2} (m + m^\dagger) = URe(m) \). The resulting Hamiltonian for the Hubbard model in the mean field approximation with \( q = (\pi, \pi) \) AFM order is given by

\[
\mathcal{H}_k = \sum_\sigma \left[ (\epsilon_{k\sigma} - \epsilon_F) c_{k\sigma}^\dagger c_{k\sigma} + (\epsilon_{k+Q\sigma} - \epsilon_F) c_{k+Q\sigma}^\dagger c_{k+Q\sigma} \right] \\
+ \sum_\sigma U n_{\sigma} c_{k\sigma}^\dagger c_{k\sigma} + U n_{\sigma} c_{k+Q\sigma}^\dagger c_{k+Q\sigma} \\
+ \sum \Delta \left( c_{k\downarrow}^\dagger c_{k+Q\downarrow} + c_{k+Q\downarrow}^\dagger c_{k\downarrow} \right) - \Delta \left( c_{k+Q\uparrow}^\dagger c_{k\uparrow} + c_{k\uparrow}^\dagger c_{k+Q\uparrow} \right),
\]

(2.29)

where \( \epsilon_{k\sigma} \) are the energy levels of the non-interacting system. This Hamiltonian can also be written in matrix form as

\[
H_k = \begin{pmatrix}
H_{0\uparrow}(k) + U n_{\downarrow} & -\Delta & 0 & 0 \\
-\Delta & H_{0\uparrow}(k + Q) + U n_{\downarrow} & 0 & 0 \\
0 & 0 & H_{0\downarrow}(k) + U n_{\uparrow} & \Delta \\
0 & 0 & \Delta & H_{0\downarrow}(k + Q) + U n_{\uparrow}
\end{pmatrix},
\]

(2.30)

where \( H_{0\sigma}(k) = (\epsilon_{k\sigma} - \epsilon_F) \). The AFM order leaves the up and down spins degenerate such that the Hamiltonian is diagonal in spin, allowing a compact version of the Hamiltonian to be written as

\[
H_{k\sigma} = \begin{pmatrix}
H_{0\sigma}(k) + U n_{\bar{\sigma}} & \text{sign}(\bar{\sigma}) \Delta \\
\text{sign}(\bar{\sigma}) \Delta & H_{0\sigma}(k + Q) + U n_{\bar{\sigma}}
\end{pmatrix},
\]

(2.31)
where the basis is $\Psi^\dagger = (c_{k\sigma}^\dagger, c_{k+Q\sigma}^\dagger)$. The off diagonal terms in this Hamiltonian split the resulting bands by an amount given by $\Delta$.

Hidden in this formalism are self consistent equations for the order parameters $n_\sigma$ and $m$. To perform this self consistency the expectation values must be expanded in terms of the diagonalized system. The creation and annihilation operators in the diagonalized system are given by $\gamma_{kn}^\dagger$ and $\gamma_{kn}$ respectively and are related to the electron creation and annihilation operators by

$$c_{k\sigma} = \sum_n V_{(\sigma)n}^k \gamma_{kn} \quad \text{and} \quad c_{k\sigma}^\dagger = \sum_n \gamma_{kn}^\dagger (V_{(\sigma)n}^k)^\dagger,$$

(2.32)

Where $V_{(\sigma)n}^k$ are the eigenvectors of $H_{k\sigma}$. Using these definitions in equation 2.25, $n$ can be rewritten as

$$n_\sigma = \sum_k \langle c_{k\sigma}^\dagger c_{k\sigma} \rangle$$

(2.33)

$$= \sum_{nm} \sum_k \langle \gamma_{k\sigma n}^\dagger (V_{n}^k)^\dagger V_{m}^k \gamma_{k\sigma m} \rangle$$

(2.34)

$$= \sum_{nm} \sum_k \left( (V_{n}^k)^\dagger V_{m}^k + (V_{n}^{k+Q})^\dagger V_{m}^{k+Q} \right) \langle \gamma_{k\sigma n}^\dagger \gamma_{k\sigma m} \rangle$$

(2.35)

$$= \sum_{nm} \sum_k \left( (V_{n}^k)^\dagger V_{n}^k + (V_{n}^{k+Q})^\dagger V_{n}^{k+Q} \right) \langle \gamma_{k\sigma n}^\dagger \gamma_{k\sigma n} \rangle$$

(2.36)

$$= \sum_{n} \sum_k \left( (V_{n}^k)^\dagger V_{n}^k + (V_{n}^{k+Q})^\dagger V_{n}^{k+Q} \right) f(\epsilon_{k\sigma n}),$$

(2.37)
and likewise for \( m \),

\[
m = \sum_n \sum_k \left( (V_n^{k+Q})^\dagger V_n^k + (V_n^k)^\dagger V_n^{k+Q} \right) f(\epsilon_{k\sigma n}),
\]

(2.38)

where if one is summing these terms over the whole PM zone, then to remove double counting one removes the second \( V^\dagger V \) terms from each expectation value. With these self consistent equations for the order parameters of the system, \( U \) becomes the only free parameter outside of those used to construct the bare Hamiltonian.

**Superconducting Order**

Another important interaction for studying cuprates and other superconducting materials is the superconducting interaction. Superconductivity can be added to the Hubbard model and the interaction can be treated in the mean field approximation just as AFM order was in the previous section. The superconducting interaction is given by

\[
I_{SC} = \sum_\sigma \sum_{kk'} V_{SC}(k,k') c_{k\sigma}^\dagger c_{-k\bar{\sigma}}^\dagger c_{-k'\bar{\sigma}} c_{k'\sigma},
\]

(2.39)

Where \( V_{SC}(k,k') \) is the superconducting potential. It is important to note that while the AFM solution is a valid mean field solution of the Hubbard model, this model for superconductivity does not specify the origin of the superconducting potential and may be considered more of a toy model. The mean field approximation for the creation and annihilation operators is done, once again, by studying small fluctuation around the
average value such that

$$c_{k\sigma}^\dagger c_{-k\bar{\sigma}} = \langle c_{k\sigma}^\dagger c_{-k\bar{\sigma}} \rangle + \left( c_{k\sigma}^\dagger c_{-k\bar{\sigma}} - \langle c_{k\sigma}^\dagger c_{-k\bar{\sigma}} \rangle \right),$$

(2.40)

and

$$c_{-k'\bar{\sigma}} c_{k'\sigma} = \langle c_{-k'\bar{\sigma}} c_{k'\sigma} \rangle + \left( c_{-k'\bar{\sigma}} c_{k'\sigma} - \langle c_{-k'\bar{\sigma}} c_{k'\sigma} \rangle \right).$$

(2.41)

Using the two relationships above, and only keeping terms up to first order in fluctuations, gives,

$$c_{k\sigma}^\dagger c_{-k\bar{\sigma}} c_{-k'\bar{\sigma}} c_{k'\sigma} = -\langle c_{k\sigma}^\dagger c_{-k\bar{\sigma}} \rangle \langle c_{-k'\bar{\sigma}} c_{k'\sigma} \rangle + \langle c_{k\sigma}^\dagger c_{-k\bar{\sigma}} \rangle c_{-k'\bar{\sigma}} c_{k'\sigma} + c_{k\sigma}^\dagger c_{-k\bar{\sigma}} \langle c_{-k'\bar{\sigma}} c_{k'\sigma} \rangle.$$  

(2.42)

The resulting mean field superconducting interaction is

$$I_{SC} = \sum_{\sigma} \sum_{kk'} V_{SC} (k, k') \left[ \langle c_{k\sigma}^\dagger c_{-k\bar{\sigma}}^\dagger \rangle c_{-k'\bar{\sigma}} c_{k'\sigma} + c_{k\sigma}^\dagger c_{-k\bar{\sigma}} \langle c_{-k'\bar{\sigma}} c_{k'\sigma} \rangle \right]$$

$$= \sum_{\sigma} \sum_{k} \left[ \Delta_{k}^c_{-k\bar{\sigma}} c_{k\sigma} + \Delta_{k}^c_{k\sigma} c_{-k\bar{\sigma}}^\dagger \right],$$

(2.43)

where the superconducting gap is given by $\Delta_k = \sum_{k'} V_{SC} (k, k') \langle c_{-k'\bar{\sigma}} c_{k'\sigma} \rangle$. In most situations, like the cuprates, where the superconducting gap is known to have $d$-wave symmetry, it is convenient to have $V_{SC} (k, k') = V_{SC}$ such that the superconducting potential is momentum independent and to apply the $d$-wave form factor, $g_k = \left( \cos (k_x) - \cos (k_y) \right) / 2$, directly to the superconducting gap such that $\Delta_k = \Delta_0 g_k$. 

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Combining Superconducting and \((\pi, \pi)\)-AFM Order

Introducing superconducting order into the Hubbard model with \((\pi, \pi)\)-AFM order, equation 2.43 must be expressed as a sum over the reduced BZ as

\[
I_{SC} = \sum_{\sigma} \sum_{k} \Delta^*_k c_{-k\sigma} c_{k\sigma} + \Delta_k c_{k\sigma}^\dagger c_{-k\sigma}^\dagger + \Delta^*_k (k+Q) c_{-(k+Q)\sigma} c_{k+Q\sigma} + \Delta_k (k+Q) c_{k+Q\sigma}^\dagger c_{-(k+Q)\sigma}^\dagger.
\]  

(2.44)

Adding this interaction to the Hamiltonian in equation 2.31 requires extending the basis to \(\Psi^\dagger = \left( c_{k\sigma}^\dagger, c_{k+Q\sigma}^\dagger, c_{-k\sigma}, c_{-(k+Q)\sigma} \right)\). The Hamiltonian with AFM+SC order now becomes,

\[
H_{k\sigma} = \begin{bmatrix}
H_{0\sigma}(k) + Un_{\bar{\sigma}} & sign(\bar{\sigma})\Delta & \Delta_k & 0 \\
sign(\bar{\sigma})\Delta & H_{0\sigma}(k+Q) + Un_{\bar{\sigma}} & 0 & \Delta_{k+Q} \\
\Delta_k & 0 & H_{0\sigma}(k) + Un_{\bar{\sigma}} & sign(\bar{\sigma})\Delta \\
0 & \Delta^*_k + Q & sign(\bar{\sigma})\Delta & H_{0\sigma}(k+Q) + Un_{\bar{\sigma}} \\
\end{bmatrix}
\]

(2.45)

Self Consistent Order Parameters

Self consistent equations for the order parameters for superconductivity and AFM order can be analytically derived by diagonalization of the combined Hamiltonian given in equation 2.45. Because the matrix is 4X4, diagonalization is not a trivial task. Therefore,
it is important to begin by diagonalizing the 2X2 Hamiltonian with only AFM order. The AFM ordered Hamiltonian found in Equation 2.31 has the form

$$H = \begin{pmatrix} a & b \\ b & c \end{pmatrix}. \quad (2.46)$$

The eigenvalue problem $HX = X\Lambda$, where $X$ is the matrix of eigenvectors and $\Lambda$ is a diagonal matrix whose elements are the eigenvalues $\lambda^\pm = \frac{a+c}{2} \pm \lambda_0$ with $\lambda_0 = \sqrt{\left(\frac{a-c}{2}\right)^2 + b^2}$, must be solved in order to diagonalize $H$. This is done by solving

$$\begin{pmatrix} a - \lambda^\pm & b \\ b & c - \lambda^\pm \end{pmatrix} \begin{pmatrix} x^\pm \\ y^\pm \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \end{pmatrix}, \quad (2.47)$$

for the components of the eigenvector matrix $X$ given by $x^\pm$ and $y^\pm$. Equation 2.47 can be written in terms of $a$, $c$, and $\lambda_0$ as

$$\begin{pmatrix} \pm \left[ \lambda_0 \mp \left( \frac{a-c}{2} \right) \right] \\ \pm \left[ \lambda_0 \mp \left( \frac{a-c}{2} \right) \right] \end{pmatrix} \begin{pmatrix} \sqrt{\lambda_0 - \left( \frac{a-c}{2} \right) \left[ \lambda_0 + \left( \frac{a-c}{2} \right) \right]} \\ \pm \left[ \lambda_0 \mp \left( \frac{a-c}{2} \right) \right] \end{pmatrix} \begin{pmatrix} x^\pm \\ y^\pm \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \end{pmatrix}, \quad (2.48)$$

which gives the properly normalized eigenvectors

$$\begin{pmatrix} x^\pm \\ y^\pm \end{pmatrix} = \frac{1}{\sqrt{2}} \begin{pmatrix} \pm \sqrt{1 \mp \left( \frac{a-c}{2\lambda_0} \right)} \\ \sqrt{1 \pm \left( \frac{a-c}{2\lambda_0} \right)} \end{pmatrix}. \quad (2.49)$$
These results are the general solution to the eigenvalue problem of a matrix of the form in equation 2.46. For the system with only AFM order, \( a = \xi_k, b = \Delta, \text{ and } c = \xi_{k+Q} \)
where \( \xi_k = \epsilon_k - \epsilon_F, \xi^\pm_k = (\xi_k \pm \xi_{k+Q})/2, E_{0k} = \xi^+_k + E_{0k}, \text{ and } E_{0k} = \sqrt{(\xi^-_k)^2 + (\Delta)^2}. \)
The AFM order only Hamiltonian, given in equation 2.31 will be denoted as \( H_{k\sigma}^{AFM} \). Also, the eigenvector matrix is \( X = V_{2^2}^{AFM} \) and is given by

\[
V_{2^2}^{AFM} = \begin{bmatrix}
\alpha & -\beta \\
\beta & \alpha
\end{bmatrix}
= \frac{1}{\sqrt{2}} \begin{bmatrix}
\sqrt{1 + (\xi^-_k/E_{0k})} & -\sqrt{1 - (\xi^-_k/E_{0k})} \\
\sqrt{1 - (\xi^-_k/E_{0k})} & \sqrt{1 + (\xi^-_k/E_{0k})}
\end{bmatrix}.
\] (2.50)

such that \((V_{2^2}^{AFM})^{-1} H_{k\sigma}^{AFM} V_{2^2}^{AFM} = E_{k}^{AFM}\) where

\[
E_{k}^{AFM} = \begin{bmatrix}
E^+_0 & 0 \\
0 & E^-_0
\end{bmatrix}.
\] (2.51)

The full AFM+SC ordered Hamiltonian from equation 2.45 can be diagonalized by first writing it as a block matrix

\[
H_{k\sigma} = \begin{bmatrix}
H_{k\sigma}^{AFM} & D \\
D & -H_{k\sigma}^{AFM}
\end{bmatrix}
\] (2.52)

where

\[
D = \begin{bmatrix}
\Delta_k & 0 \\
0 & \Delta_{k+Q}
\end{bmatrix}.
\] (2.53)
The strategy now is to rotate $H_{\kappa\sigma}$ such that all of its blocks are diagonal and therefore commute with each other. This can be done with the matrix

$$V_{AFM}^{4\times4} = \begin{bmatrix} V_{2\times2}^{AFM} & 0 \\ 0 & V_{2\times2}^{AFM} \end{bmatrix} = \begin{bmatrix} \alpha & -\beta & 0 & 0 \\ \beta & \alpha & 0 & 0 \\ 0 & 0 & \alpha & -\beta \\ 0 & 0 & \beta & \alpha \end{bmatrix} \quad (2.54)$$

and

$$\left(V_{AFM}^{4\times4}\right)^{-1} = \begin{bmatrix} (V_{2\times2}^{AFM})^{-1} & 0 \\ 0 & (V_{2\times2}^{AFM})^{-1} \end{bmatrix} = \begin{bmatrix} \alpha & \beta & 0 & 0 \\ -\beta & \alpha & 0 & 0 \\ 0 & 0 & \alpha & \beta \\ 0 & 0 & -\beta & \alpha \end{bmatrix} \quad (2.55)$$

such that

$$\left(V_{AFM}^{4\times4}\right)^{-1} H_{\kappa\sigma} V_{AFM}^{4\times4} = \begin{bmatrix} E_{AFM}^{\kappa} & D \\ D & -E_{AFM}^{\kappa} \end{bmatrix}. \quad (2.56)$$

This form can be easily diagonalized because all of the block diagonal elements commute with each other. Equation 2.56 also has the same form as equation 2.46 with $c = -a$. Therefore the eigenvalues are

$$E_{k} = \pm \sqrt{(E_{0k})^{2} + \Delta_{k}^{2}}. \quad (2.57)$$
and the eigenvectors are

\[
V^{SC}_{4X4} = \begin{bmatrix}
u_k & -v_k \\
v_k & u_k \\
v_k & -u_k \\
v_k & u_k \\
\end{bmatrix} = \begin{bmatrix}
u_k^+ & 0 & -v_k^+ & 0 \\
0 & u_k^- & 0 & -v_k^- \\
v_k^+ & 0 & u_k^+ & 0 \\
0 & v_k^- & 0 & u_k^- \\
\end{bmatrix}, \quad (2.58)
\]

\[
(V^{SC}_{4X4})^{-1} = \begin{bmatrix}
u_k & v_k \\
v_k & u_k \\
v_k & u_k \\
v_k & -u_k \\
\end{bmatrix} = \begin{bmatrix}
u_k^+ & 0 & v_k^+ & 0 \\
0 & u_k^- & 0 & v_k^- \\
v_k^+ & 0 & u_k^+ & 0 \\
0 & -v_k^- & 0 & u_k^- \\
\end{bmatrix}. \quad (2.59)
\]

The coherence factors which make up the individual components of the eigenvector matrix are

\[
u_k = \sqrt{\left[1 + (\xi_k^+ + E_{0k})/E_k\right]/2},
\]

\[
v_k = \sqrt{\left[1 - (\xi_k^+ + E_{0k})/E_k\right]/2}. \quad (2.60)
\]
The Hamiltonian can now be diagonalized using \((V_{4\times4}^{SC})^{-1} (V_{4\times4}^{AFM})^{-1} H_{\kappa \sigma} V_{4\times4}^{AFM} V_{4\times4}^{SC}\) where the unitary matrix which diagonalizes the Hamiltonian is given by

\[
V = V_{4\times4}^{AFM} V_{4\times4}^{SC} = \begin{bmatrix}
\alpha_k u_k^+ & -\beta_k u_k^- & -\alpha_k v_k^+ & \beta_k v_k^- \\
\beta_k u_k^+ & \alpha_k u_k^- & -\beta_k v_k^+ & -\alpha_k v_k^- \\
\alpha_k v_k^+ & -\beta_k v_k^- & \alpha_k u_k^+ & -\beta_k u_k^- \\
\beta_k v_k^+ & \alpha_k v_k^- & \beta_k u_k^+ & \alpha_k u_k^-
\end{bmatrix}
\]  

(2.61)

Equation 2.32 shows how the quasiparticle creation and annihilation operators transform into the electron creation and annihilation operators through \(V\). Because the components of \(V\) are known explicitly, the expectation values \(\langle c_k c^\dagger_{k+Q}\rangle\) and \(\langle c_{k\sigma} c_{-k\sigma}\rangle\) can be expressed in terms of the components of \(V\). Using equation 2.38,

\[
\langle c_k c^\dagger_{k+Q}\rangle = \alpha_k u_k^+ \beta_k u_k^- f (E_k^+) - \alpha_k u_k^- \beta_k u_k^+ f (E_k^-) + \alpha_k v_k^+ \beta_k v_k^- f (-E_k^+) - \alpha_k v_k^- \beta_k v_k^+ f (-E_k^-) \\
= \alpha_k \beta_k \left\{ (u_k^+)^2 f (E_k^+) - (u_k^-)^2 f (E_k^-) + (v_k^+)^2 \left[ 1 - f (E_k^+) \right] - (v_k^-)^2 \left[ 1 - f (E_k^-) \right] \right\} \\
= \alpha_k \beta_k \left\{ (v_k^+)^2 - (v_k^-)^2 \right\} + \left\{ (u_k^+)^2 - (v_k^+)^2 \right\} f (E_k^+) - \left\{ (u_k^-)^2 - (v_k^-)^2 \right\} f (E_k^-)
\]

(2.62)

and

\[
\langle c_k c_{-k}\rangle = \alpha_k u_k^+ \alpha_k v_k^- f (E_k^+) + \beta_k u_k^- \beta_k v_k^+ f (E_k^-) - \alpha_k v_k^+ \alpha_k u_k^- f (-E_k^+) - \beta_k v_k^- \beta_k u_k^+ f (-E_k^-) \\
= -\alpha_k^2 u_k^+ v_k^+ \left[ 1 - 2 f (E_k^+) \right] - \beta_k^2 u_k^- v_k^- \left[ 1 - 2 f (E_k^-) \right],
\]

(2.63)
where \( f(E) \) is the Fermi function. These equations can be simplified further by using the following identities associated with the coherence factors and the Fermi function:

\[
\alpha_k^2 - \beta_k^2 = \xi_k^- / E_{0k}, ~~~~~~~~~~~~ (2.64)
\]

\[
2\alpha_k \beta_k = \Delta / E_{0k}, ~~~~~~~~~~~~ (2.65)
\]

\[
(u_k)^2 - (v_k)^2 = E_{0k} / E_k, ~~~~~~~~~~~~ (2.66)
\]

\[
2u_k v_k = \Delta_k / E_k, ~~~~~~~~~~~~ (2.67)
\]

\[
1 - 2f(x) = \tanh (\beta x / 2). ~~~~~~~~~~~~ (2.68)
\]

Where \( \beta = 1/k_B T \) with \( T \) being the temperature of the system and \( k_B \) being the Boltzmann constant. Using these identities gives

\[
\sum_k \langle c_k c_{k+\mathbf{Q}} \rangle = \frac{1}{N} \sum_k \frac{\Delta}{4E_{0k}} \left[ \frac{E_{0k}^+}{E_k^+} \tanh (\beta E_k^+ / 2) - \frac{E_{0k}^-}{E_k^-} \tanh (\beta E_k^- / 2) \right] = \Delta / U, ~~~~ (2.69)
\]

and

\[
\sum_k \langle c_k c_{-k} \rangle = -V_{SC} \Delta_0 \sum_k \left[ \frac{\alpha_k^2}{2E_k^+} \tanh(\beta E_k^+ / 2) + \frac{\beta_k^2}{2E_k^-} \tanh(\beta E_k^- / 2) \right] = \Delta_0 / V_{SC}. ~~~~ (2.70)
\]

These equations can be solved selfconsistently for the superconducting and antiferomagnetic gaps simultaneously. This is an example of competing order parameters because these two equations are coupled and therefore depend on each other.
Charge Transfer Insulators in Three and Four Band Models of Cuprates

The following is based on work done in an unpublished paper by Hsin Lin, R. S. Markiewicz, Chris Lane, Peter Mistark, and A. Bansil titled Zaanen-Sawatzky-Allen physics in multiband models of cuprates. This work provides the background for modeling negative compressibility in the cuprates as well as the multiband susceptibility which will be discussed latter in this document.

The main advantage of using the Hubbard Model is that it correctly predicts the insulating state of some materials when first principles caculations, such as DFT, fail to do so. This is because the onsite interaction term opens a gap in the quasiparticle spectrum called a Mott gap [32, 33, 34]. In a single band model, onsite interactions create long range magnetic order which results in an enlarged unit cell. This breaks the degeneracy between electrons on different atomic sites resulting in extra bands due to the induced order. These effects are the same in multiband models where the unordered unit cell has multiple atomic sites. Here, a Mott gap can form between bands of the same atomic character, or a charge transfer gap can form between bands of different atomic character. Multiband models have the advantage that they can predict charge transfer insulators whereas a single band model cannot. A theoretical model of transition-metal compounds by Zaanen, Sawatzky, and Allen [35] shows bands of oxygen character lying within the gap between the bands of predominantly copper character. Because cuprates fall in the
category of transition-metal oxides, they should also follow the Zaanen-Sawatzky-Allen physics and should be described as charge transfer insulators.

Multiband models of cuprates have been proven to be useful in modeling resonant inelastic x-ray spectroscopy (RIXS) [36] and tunneling studies [37] because they show spectroscopic features which are due to oxygen orbitals. Because of the inclusion of both copper and oxygen, and potentially more, atoms, multiband models can answer the question of whether cuprates are Mott insulators or charge transfer insulators. A large difference in onsite energies between copper and oxygen was found by Hybertsen, Schulter, and Christensen (HSC) [38] when fitting multiband models directly to first principles calculations. It is now apparent that their result puts cuprates in the Mott insulator class. More recently Kent et al. [39] used first principles and Monte-Carlo techniques to show that the copper-oxygen hybridization is in fact much smaller than predicted by HSC. Experimental evidence showing cuprates to be charge transfer insulators was first given in the results of Armitage et al. [15]. The remaining question is can we find a set of parameters in a multiband Hubbard model which explains experiments by putting cuprates into the charge transfer insulator class.

Here the approach to solving this problem will be to make the gap in the multiband models match that of the single band model. This scheme is reasonable due to the success of the one band model in the intermediate coupling regime where the low temperature gap always matches experimental results. The first model to be studied is the Emery model
with onsite Hubbard repulsion,

\[
H_3 = \sum_j \Delta d_0 d_j^\dagger d_j + \sum_{\langle ij \rangle} t_{dp} (d_j^\dagger p_i + (c.c.) + \sum_{\langle ii' \rangle} t_{pp} (p_i^\dagger p_{i'} + (c.c.)) + \sum_j U n_{dj\uparrow} n_{dj\downarrow} + \sum_i U_p n_{pi\uparrow} n_{pi\downarrow}. \tag{2.71}
\]

This model describes the Cu-d_{x^2-y^2}, O-p_x, and O-p_y orbitals resulting in three fundamental bands. The Hamiltonian is written in real space with the sums over \( j, i, \) and \( i' \) denoting lattice sites. Sums over \( \langle ij \rangle \) denote that sites \( i \) and \( j \) must be nearest neighbors.

The reason for having \( i \) and \( i' \) in the \( t_{pp} \) term is because the nearest oxygen neighbor of a O-p_x orbital will be a O-p_y orbital and vice versa. The creation and annihilation operators for the Cu-d_{x^2-y^2} electrons are \( d_i^\dagger \) and \( d_i \) respectively. The creation and annihilation operators for the oxygen p electrons are \( p_i^\dagger \) and \( p_i \) respectively where the location of the orbital in the unit cell determines whether the electron has O-p_x or O-p_y symmetry. The copper-oxygen splitting parameter is \( \Delta d_0 \). This term provides a rigid shift of the copper d bands with respect to the oxygen p bands. \( t_{dp} \) describes the hopping from copper to oxygen orbitals, \( t_{pp} \) represents the hopping between oxygen orbitals, \( U \) is the onsite repulsion energy for copper sites, and \( U_p \) is the onsite repulsion energy for oxygen sites. The number operators for copper and oxygen orbitals with spin \( \sigma \) are \( n_{dj\sigma} \) and \( n_{pi\sigma} \) respectively.

The Hamiltonian can be solved in a mean field sense as outlined in section 2.3. Even though this Hamiltonian is in real space, the mean field procedure is the same where the number operator is expanded to first order in small fluctuations around the average
number density. The Hamiltonian, with no order, can be written as a matrix in the basis
\[ \Psi^\dagger = \left( p^\dagger_{x,a} , p^\dagger_{y,a} , d^\dagger_{x^2-y^2,a} \right) , \]
where the subscript \( a \) represents the lattice site and \( x, y, \) and \( x^2 - y^2 \) represent the orbital type. The Hamiltonian then looks like
\[
H_3 = \begin{pmatrix}
0 & 4t_{pp} \sin(k_x/2) \sin(k_y/2) & -i2t_{pd} \sin(k_x/2) \\
4t_{pp} \sin(k_x/2) \sin(k_y/2) & 0 & i2t_{pd} \sin(k_y/2) \\
i2t_{pd} \sin(k_x/2) & -i2t_{pd} \sin(k_y/2) & \Delta_{d0}
\end{pmatrix} .
\] (2.72)

When \((\pi, \pi)\)-AFM order is added to the system, the unit cell doubles such that each orbital can have a type denoted \( a \) or \( b \) depending on if they are on the spin up or down sub-lattice.

The Hamiltonian with \((\pi, \pi)\)-AFM order in the mean field approximation can be written in matrix form within the expanded basis \( \Psi^\dagger = \left( p^\dagger_{x,a} , p^\dagger_{x,b} , p^\dagger_{y,a} , p^\dagger_{y,b} , d^\dagger_{x^2-y^2,a} , d^\dagger_{x^2-y^2,b} \right) \)
\[ H_3 = \begin{bmatrix}  
-\langle m_p \rangle U_p + \langle n_p \rangle U_p/2 & 0 & g_0 \\
0 & \langle m_p \rangle U_p + \langle n_p \rangle U_p/2 & g_1 \\
g_0^\dagger & g_1^\dagger & -\langle m_p \rangle U_p + \langle n_p \rangle U_p/2 \\
g_1^\dagger & g_0^\dagger & 0 \\
d_0^\dagger & d_1^\dagger & d_2^\dagger \\
d_1^\dagger & d_0^\dagger & d_3^\dagger \\
g_1 & d_0 & d_1 \\
g_0 & d_1 & d_0 \\
0 & d_2 & d_3 \\
\langle m_p \rangle U_p + \langle n_p \rangle U_p/2 & d_3 & d_2 \\
d_3^\dagger & \Delta_{d0} - \langle m_d \rangle U + \langle n_d \rangle U/2 & 0 \\
d_2^\dagger & 0 & \Delta_{d0} + \langle m_d \rangle U + \langle n_d \rangle U/2 
\end{bmatrix} \]
where,

\[
g_1 = -t_{pp} \exp(i(k_x/2 + k_y/2)) - t_{pp} \exp(i(-k_x/2 - k_y/2))
\]

\[
g_0 = t_{pp} \exp(i(k_x/2 - k_y/2)) + t_{pp} \exp(i(-k_x/2 + k_y/2))
\]

\[
d_0 = t_{dp} \exp(-ik_x/2)
\]

\[
d_1 = -t_{dp} \exp(i k_x/2) = -d_0^\dagger
\]

\[
d_2 = -t_{dp} \exp(-i k_y/2) = -d_3^\dagger
\]

\[
d_3 = t_{dp} \exp(i k_y/2)
\]

Because the zero of energy for the system is arbitrary, a constant value can be added or subtracted from the diagonal elements of the Hamiltonian matrix. It is convenient to subtract \( \langle n_p \rangle U_p/2 \) from the Hamiltonian to leave only the magnetic terms on the diagonal elements associated with the oxygen \( p \) orbitals. This results in a renormalized copper-oxygen splitting parameter

\[
\Delta_d = \Delta_{d0} + \langle n_d \rangle U/2 - \langle n_p \rangle U_p/2.
\]

(2.74)

\( \Delta_d \) is analogous to the previously mentioned difference in onsite energies between copper and oxygen extracted from HSC [38] and Kent et al. [39]. A large \( \Delta_d \) puts cuprates in the Mott insulator regime whereas a small \( \Delta_d \) puts them in the charge transfer insulator regime. \( \Delta_d \) is therefore a useful indicator of Mott vs charge transfer insulator systems.
The average particle density and magnetic order parameter for the copper $d$ orbitals is given by

$$\langle n_d \rangle = \left( \frac{n_{d\uparrow} + n_{d\downarrow}}{2} \right)$$

$$\langle m_d \rangle = \left( \frac{n_{d\uparrow} - n_{d\downarrow}}{2} \right).$$

To obtain an expression for the average particle density of the oxygen $p$ orbitals in terms of the doping $x$ and $\langle n_d \rangle$ the total electron density of the system is taken to be $\langle n_d \rangle + \langle n_{px} \rangle + \langle n_{py} \rangle = \langle n_d \rangle + 2 \langle n_p \rangle = 5 + x$, where $x$ can range from zero to one. This is because there are three atoms per unit cell and each can contain two electrons, one spin up and one spin down, for a total of six possible electrons. Solving this equation gives

$$\langle n_p \rangle = \left[ 5 + x - \langle n_d \rangle \right] / 2.$$ 

It is reasonable to assume that the magnetic order on the oxygen is frustrated such that

$$\langle m_p \rangle = 0.$$ 

The cuprate which will be studied here is the electron doped $\text{Nd}_{2-x}\text{Ce}_x\text{CuO}_4$ (NCCO). Previous studies using a one band Hubbard model for NCCO have been able to reproduce the experimental gap and the appearance of hole pockets at higher electron doping [40]. The tight binding parameters are determined by fits to first principles calculations. In order to obtain the correct experimental gap with the three band model, the value of $U$
can be adjusted until the self consistent gap matches the gap in the one band model.

<table>
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<tr>
<th>(eV)</th>
<th>HSC</th>
<th>$x = 0.0$</th>
<th>$x = 0.04$</th>
<th>$x = 0.10$</th>
<th>$x = 0.15$</th>
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<td>0.8</td>
<td>0.8</td>
<td>0.8</td>
</tr>
<tr>
<td>$t_{pp}$</td>
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<td>-0.4</td>
<td>-0.4</td>
<td>-0.4</td>
<td>-0.4</td>
</tr>
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<td>-0.755</td>
<td>-0.755</td>
<td>-0.755</td>
<td>-0.755</td>
</tr>
<tr>
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<td>-0.23</td>
<td>-0.25</td>
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<td>7.00</td>
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<td>5.65</td>
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<td>5</td>
<td>5</td>
<td>5</td>
<td>5</td>
</tr>
<tr>
<td>$\langle m_d \rangle$</td>
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<td>0.26</td>
<td>0.12</td>
<td>0.06</td>
<td></td>
</tr>
<tr>
<td>$\langle n_d \rangle$</td>
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<td>1.444</td>
<td>1.643</td>
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</tr>
</tbody>
</table>

Table 2.1: Table of values for the tight-binding parameters, $U$, $U_p$, and the resulting gaps, magnetization, and number of d-electrons for the three band model of NCCO at multiple dopings.

The calculations done here give a $\Delta_d$ which is negative and an order of magnitude smaller than that in the HSC results. This puts the cuprates in the charge transfer regime. This can be seen by comparing the atomic character of the DOS as shown in figure 2.1. The UMB is of primarily copper character whereas the LMB is primarily oxygen character. This is indicative of a charge transfer insulator because a gap has opened between electrons of different atomic character. This is in agreement with the results of Armitage et al. [15].

The one and three band models both fail to predict the gap as a function of doping if the half filling parameters are held constant and the gap is determined self consistently. This is because the gap closes with doping faster in experiments than can be described by a constant $U$. If $U$ is allowed to vary with doping while all other parameters are held constant this problem is resolved.

The doping dependence of the bands, shown in figure 2.1, shows electron pockets near $(\pi, 0)$ and $(0, \pi)$ at low doping that evolve into the paramagnetic Fermi surface as the gap collapses near $x = 0.15$. Experimental results show that in NCCO a hole pocket emerges
Figure 2.1: (top left frame) Energy vs. DOS for the three band model of NCCO at half filling. Blue shading represents the total DOS and red shading shows the portion of the DOS with copper character. (other frames) Three band model band structures for increasing electron doping and decreasing U.
before the gap collapse [15]. The three band model fails to capture the emergence of hole pockets. This is surprising, because the one band model does describe the emergence of the hole pockets [40]. The reason for this is because the one band model includes next nearest neighbor hopping on copper where the three band model only considers nearest neighbor hopping from copper to oxygen. The multiband analogy to the next nearest neighbor hopping is hopping from a copper $d$-orbital to a copper $s$-orbital [41]. This can be modeled by using a four band model with hopping to copper $s$-orbitals.

The four band model is represented by the following Hamiltonian,

$$
H_4 = \sum_j \Delta_s s_j^\dagger s_j + \sum_j \Delta_{d0} d_j^\dagger d_j + \sum_{<ij>} t_{dp}(d_j^\dagger p_i + (c.c.)) + \sum_{<ij>} t_{sp}(s_j^\dagger p_i + (c.c.))
+ \sum_j U n_{dj\uparrow} n_{dj\downarrow} + \sum_i U_p n_{pi\uparrow} n_{pi\downarrow},
$$

where the notation is the same as in equation 2.71 with the inclusion of the copper $4s$-orbital creation and annihilation operator given by $s_i^\dagger$ and $s_i$ respectively. In the four band model the $t_{pp}$ term is not needed because an indirect oxygen-oxygen hopping arises from the $4s$-orbital. Just as with the three band model, the four band model can be
written as a matrix, in the basis given by \( \Psi^\dagger = \left( s^\dagger_a, p^\dagger_{x,a}, p^\dagger_{y,a}, d^\dagger_{x^2-y^2,a} \right) \), as

\[
H_4 = \begin{pmatrix}
\Delta_s & -i2t_{ps} \sin(k_x/2) & -i2t_{ps} \sin(k_y/2) & 0 \\
-i2t_{ps} \sin(k_x/2) & 0 & 4t_{pp} \sin(k_x/2) \sin(k_y/2) & -i2t_{pd} \sin(k_x/2) \\
i2t_{ps} \sin(k_y/2) & 4t_{pp} \sin(k_x/2) \sin(k_y/2) & 0 & i2t_{pd} \sin(k_y/2) \\
0 & i2t_{pd} \sin(k_x/2) & -i2t_{pd} \sin(k_y/2) & \Delta_0
\end{pmatrix}.
\]

(2.80)

And once again the Hamiltonian with \((\pi, \pi)\)-AFM order in the mean field approximation can be written in matrix form within the expanded basis.
\[ \Psi^\dagger = \left( s^\dagger_a, s^\dagger_b, p^\dagger_{x,a}, p^\dagger_{x,b}, p^\dagger_{y,a}, p^\dagger_{y,b}, d^\dagger_{x_2-y^2,a}, d^\dagger_{x_2-y^2,b} \right) \] as

\[
H_4 = \begin{pmatrix}
\Delta_s & 0 & s_0 & s_1 \\
0 & \Delta_s & s_1 & s_0 \\
s_0^\dagger & s_1^\dagger & -m_p U_p + U_p n_p/2 & 0 \\
s_1^\dagger & s_0^\dagger & 0 & +m_p U_p + U_p n_p/2 \\
s_2^\dagger & s_3^\dagger & g_0^\dagger & g_1^\dagger \\
s_3^\dagger & s_2^\dagger & g_1^\dagger & g_0^\dagger \\
0 & 0 & d_0^\dagger & d_1^\dagger \\
0 & 0 & d_1^\dagger & d_0^\dagger \\
s_2 & s_3 & 0 & 0 \\
s_3 & s_2 & 0 & 0 \\
g_0 & g_1 & d_0 & d_1 \\
g_1 & g_0 & d_1 & d_0 \\
-m_p U_p + U_p n_p/2 & 0 & d_2 & d_3 \\
0 & +m_p U_p + U_p n_p/2 & d_3 & d_2 \\
d_2^\dagger & d_3^\dagger & \Delta_0 - m_d U_d + U_d n_d/2 & 0 \\
d_3^\dagger & d_2^\dagger & 0 & \Delta_0 + m_d U_d + U_d n_d/2
\end{pmatrix},
\]

(2.81)
with,

\[ g_1 = -t_{pp} \exp(i(k_x/2 + k_y/2)) - t_{pp} \exp(i(-k_x/2 - k_y/2)) \]

\[ g_0 = t_{pp} \exp(i(k_x/2 - k_y/2)) + t_{pp} \exp(i(-k_x/2 + k_y/2)) \]

\[ d_0 = t_{dp} \exp(-i k_x/2) \]

\[ d_1 = -t_{dp} \exp(i k_x/2) \]

\[ d_2 = -t_{dp} \exp(-i k_y/2) \]

\[ d_3 = t_{dp} \exp(i k_y/2) \]

\[ s_0 = -t_{sp} \exp(i k_x/2) \]

\[ s_1 = t_{sp} \exp(-i k_x/2) \]

\[ s_2 = -t_{sp} \exp(i k_y/2) \]

\[ s_3 = t_{sp} \exp(-i k_y/2). \]

The four band model maintains many of the same properties of the three band model which can be seen in figure 2.2 and table 2.2. At half filling the gap can be fit to the one band model and experimental results in the same way the three band was. This results in a charge transfer type insulator because the gap lies between bands of copper character above and primarily oxygen character below the Fermi level. The doping dependence still requires a variable \( U \) in order to match the gap closing at \( x = 0.15 \). The advantage of the four band model is that it successfully predicts the emergence of hole pockets around \((\pi/2, \pi/2)\).
Figure 2.2: (top left frame) Energy vs. DOS for the four band model of NCCO at half filling. Blue shading represents the total DOS and red shading shows the portion of the DOS with copper character. (other frames) Four band model band structures for increasing electron doping and decreasing $U$. 

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Table 2.2: Table of values for the tight-binding parameters, $U$, $U_p$, and the resulting gaps, magnetization, and number of d-electrons for the four band model of NCCO at multiple dopings.

<table>
<thead>
<tr>
<th>(eV)</th>
<th>$x=0.0$</th>
<th>0.04</th>
<th>0.10</th>
<th>0.15</th>
</tr>
</thead>
<tbody>
<tr>
<td>$t_{dp}$</td>
<td>0.8</td>
<td>0.8</td>
<td>0.8</td>
<td>0.8</td>
</tr>
<tr>
<td>$t_{sp}$</td>
<td>1.2</td>
<td>1.2</td>
<td>1.2</td>
<td>1.2</td>
</tr>
<tr>
<td>$\Delta_{d0}$</td>
<td>-0.755</td>
<td>-0.755</td>
<td>-0.755</td>
<td>-0.755</td>
</tr>
<tr>
<td>$\Delta_s$</td>
<td>10</td>
<td>10</td>
<td>10</td>
<td>10</td>
</tr>
<tr>
<td>$U$</td>
<td>6.63</td>
<td>6.23</td>
<td>4.83</td>
<td>4.53</td>
</tr>
<tr>
<td>$U_p$</td>
<td>5</td>
<td>5</td>
<td>5</td>
<td>5</td>
</tr>
<tr>
<td>$m_d$</td>
<td>0.35</td>
<td>0.30</td>
<td>0.14</td>
<td>0.07</td>
</tr>
<tr>
<td>$n_d$</td>
<td>1.29</td>
<td>1.37</td>
<td>1.60</td>
<td>1.67</td>
</tr>
</tbody>
</table>

**Negative Electronic Compressibility**

Compressibility, in thermodynamics, is a quantity that describes how the volume of a material changes when pressure is applied. Likewise, in quantum mechanical systems of electrons, the electronic compressibility becomes an important quantity. The electronic compressibility is defined as $(1/n^2) \partial n/\partial \mu$, where $n$ is the electron density and $\mu$ is the chemical potential of the system. When the electronic compressibility is positive, as electrons are added to the system the chemical potential will increase. This is analogous to a filling a cup with water; as water is added to the cup the water level rises. Now, if the electronic compressibility is negative, the chemical potential will decrease as electrons are added to the system. A *negative* electronic compressibility (NEC) is quite unexpected, just as it is unexpected for the water level in a cup to decrease when water is poured into it. This anomaly called NEC has, however, previously be predicted [42] and observed at the boundary between some two dimensional materials [43, 44, 45, 46, 47, 48]. In three dimensional materials NEC has been observed in the iridate compounds [16]. The iridates
are similar to the cuprates in that they are transition metal oxides with a tetrahedral oxygen layer. Both systems are also insulators at half filling due to electronic correlation effects. Because of the similarities it is reasonable to expect to find a NEC regime in the cuprates as well. The one, three and four band models of cuprates presented thus far in this chapter can be used to theoretically investigate NEC in cuprates and to describe its origins.

In this study, negative compressibility and whether it exists within a Hubbard model with \((\pi, \pi)-\text{AFM} \ [40, 49, 50]\) is investigated. The electron doped cuprate system NCCO can be modeled with tight binding parameters determined through a fit to the corresponding DFT calculations [50]. These parameters are \(t = 0.42\text{eV}, t' = -0.1\text{eV}, t'' = 0.06\text{eV},\) and \(t''' = 0.008\text{eV}.\)

![Figure 2.3:](image)

**Figure 2.3:** (a) Fermi energy vs electron doping. (b) \(U/t\) exponential fit of data from reference [40] (c) staggered magnetization vs electron doping.
Within the one band model, the key ingredient for NEC is the significant change of $U$ with doping, Ref. [40]. This change is associated with how at half filling the material is an insulator, so $U$ is unscreened, while for a 2D system the susceptibility has a step at the band onset, which translates into a large and weakly doping-dependent screening starting at $x = 0^+$. Here, the data from Ref. [40] is fit to a decaying exponential, $U/t = a_1 e^{-x/x_0} + a_2$, where $x$ is the doping, $a_1 = 3.0707$, $a_2 = 2.9587$, and $x_0 = 0.0646$. Figure 2.3(b) shows $U(x)$, and 2.3(c) the self consistent value of the staggered magnetization $m$, which is equivalent to the magnetic order parameter of equation 2.25.

Given the doping dependence of $U$ and $m$, the resulting Fermi energy, $\epsilon_F$, is plotted in Fig. 2.3(a). While $\epsilon_F$ increases relative to the bottom of the UMB, the UMB moves rapidly to lower energies as the AFM gap collapses with doping due to the rapid decrease in $U$. This leads to a doping range of NEC because the UMB is decreasing (relative to the LMB) faster then the Fermi energy is increasing (relative to the UMB). The Fermi energy shows a decrease until $x = 0.11$ where it begins to increase linearly.

If a constant value for the Hubbard $U$ is assumed this effect would not be observed as seen in the red, blue and magenta curves of figure 2.4(a). Thus a doping-independent $U$ will not produce a fast enough gap collapse to out-pace the rise in energy due to adding electrons. These results show that a doping dependent $U$ is essential to observing a decreasing Fermi energy in a one band model of NCCO. The rapid drop in $U$, together with the closing of the gap as the UMB starts to fill, conspire to produce a NEC.

An important parameter controlling the NEC is the shift of the VHS away from half
Figure 2.4: Black dashed curve same as figure 2.3. Other curves show the effects of a constant $U$ of varying magnitude.

Figure 2.5: Black curve shows the (a) Fermi energy vs electron doping and (b) staggered magnetization for the full set of NCCO tight binding parameters. Blue curve has parameters $t', t'', t'''$ equal to zero. Both curves use a doping dependent $U$ as in figure 2.3(b).
Figure 2.6: The colored surface shows the Fermi energy as a function of $t'/t$ and electron doping where $t = 0.42\,\text{eV}$, $t''/t'$ is fixed, $t''' = 0.008\,\text{eV}$, and $U$ is the same as in figure 2.3(b). The shaded region in the $t'/t$-doping plane shows the region where negative compressibility is observed.

filling. This feature is controlled by the hopping parameters $t'$ and $t''$. The blue curve in figure 2.5(a) shows the results of setting $t' = t'' = 0$. Figure 2.6 shows the doping dependence of the Fermi energy, as $t'$ is varied, keeping the ratio of $t''/t'$ at a constant value of -0.6. NEC is largest and persists over the widest doping range for $t' = 0$ where the VHS crosses the Fermi level at $x = 0$. For larger $t'$ NEC weakens and persists over a smaller doping range, until NEC vanishes at $x = 0$ for $t' = -0.5\,\text{eV}$.

When $t'$ and $t''$ are zero the half filled bands become electron-hole symmetric about the Fermi energy where the VHS is located. Introducing AFM order opens a gap as seen in the
DOS of the blue curve in figure 2.7(a). When $t'$ is turned on, the electron-hole symmetry is broken, pushing the VHS into the hole doped region, and the DOS becomes the black curve in figure 2.7(a). Figure 2.7(b) shows the number of electrons in the system. When $t' = 0$ (blue curve) adding an electron will increase the Fermi energy a very small amount compared to adding an electron when $t' \neq 0$ (black curve). Therefore, for small $t'$, doping near half filling has a smaller effect on the Fermi energy than in systems with a larger $t'$ allowing the gap closing to dominate the shift in Fermi energy to higher electron doping.

![Figure 2.7](image)

**Figure 2.7:** (a) Black curve shows the DOS for the full set of NCCO tight binding parameters. Blue curve has parameters $t'$ and $t''$ equal to zero. (b) Average number of electrons per lattice site minus one obtained from the DOS of the corresponding color in (a). Where the curves intersect the gray dashed line gives their corresponding shift in Fermi energy at one percent doping.
It is important to note that for hole doped cuprates this model predicts a doping dependence of the Fermi energy similar to what was presented here for the electron doped case except that the Fermi energy increases anomalously from half filling before returning to a regular decreasing trend with hole doping. One major difference which could obscure any anomalous doping dependence of the Fermi energy is the presence of nanoscale phase separation in the hole doped cuprates[24, 51].

Whereas a large doping dependence of $U$ ($\sim 50\%$) is necessary to capture the physics of the cuprates in the one band model, the change is only $\sim 20\%$ in three and four band models that explicitly consider the oxygens. This is because the doping dependence of $U$ is intended to simulate the presence of the oxygen (and perhaps other) atoms which screen $U$ at higher dopings. Moving towards more realistic models reduces the doping dependence of $U$ by explicitly including oxygen atoms. Despite the weaker doping dependence of $U$, NEC is still present in the three and four band models of NCCO. Using the three and four band models outlined in section 2.7, figure 2.8 shows the existence of negative compressibility derived from these multiband models. The change in Fermi energy in the NEC regime for the one band model is $\sim 0.3eV$, for the three band model is $\sim 1eV$ and for the four band model is $\sim 1.2eV$. Therefore, as the number of bands in the model increases, the doping dependence of $U$ decreases and the strength of the NEC increases. This shows that not only is NEC in cuprates due to the doping dependent screening of $U$, as shown in the one band model, but that it is linked to the influence of the oxygen atoms on the band structure.
Figure 2.8: The variation in the Fermi energy for the 3 (red) and 4 (blue) band models with respect to doping, which show the existence of NEC. It should be noted that there is only data available up to $x = 0.15$ where the electronic compressibility is still negative. It is expected that the electronic compressibility will become positive at higher doping.
Chapter 3

Fermi Surface Free Superconductivity

Introduction

ARPES studies on the pnictides have shown that the superconducting transition temperature $T_c$ depends sensitively on details of the band structure and Fermi surface [52]. In particular, proximity of the Fermi surface to a band edge and the associated Van Hove singularity (VHS) correlates with significantly enhanced $T_c$'s. An investigation of Ba$_{1-x}$K$_x$Fe$_2$As$_2$ [17] indicates that when a band edge approaches the Fermi energy, superconductivity can be observed even before the band crosses the Fermi energy. Bang [17] suggests that this *Fermi-surface-free superconductivity* (FS-free SC) is driven by the shadow bands resulting from the symmetrization of the spectral weight around the Fermi energy and the formation of a related shadow gap in the BCS theory of superconductivity.
With this information, a question becomes: Does this effect also arise in the single-band case of the hole-doped cuprates when the band is split by magnetic order? To answer this question inspiration comes from the change in Fermi surface topology of the electron-doped cuprates. In the electron doped system it is well known that the \((\pi, \pi)\)-AFM order can induce two distinct topological transitions (TT’s) with doping \([3, 40]\). At half-filling, the AFM order splits the band into upper and lower magnetic bands (U/LMBs), and low electron doping moves the Fermi energy into the bottom of the UMB. As doping increases, the LMB moves up in energy and eventually crosses the Fermi energy leading to the emergence of hole pockets around \((\pi/2, \pi/2)\). This is the first topological transition (TT1) in this system. The second topological transition (TT2) occurs at higher doping when the electron and hole pockets merge into the single large Fermi surface of the paramagnetic state.

In this chapter a transition similar to TT1 will be shown to occur in hole doped cuprates such as \((\text{Bi,Pb})_2(\text{Sr,La})_2\text{CuO}_6+\delta\) (Bi2201) \([5]\). This transition is however different in that the first holes now enter the LMB, so that the transition occurs when the UMB moves down in energy and crosses the Fermi energy, introducing electron pockets around \((\pi, 0)\) \([53, 54]\). Thermopower studies \([55, 56]\) have suggested the existence of electron pockets appearing in hole-doped cuprates in the underdoped regime. Also, the remarkable finding of the ARPES experiment of Kondo et al. \([5]\) is that there is spectral weight in the \((\pi, 0)\) region, starting at low doping, suggestive of the appearance of an electron pocket at \((\pi, 0)\). To preserve the analogy with TT1 in the electron doped cuprates \([3, 40]\), a \((\pi, \pi)\)-AFM model will be used to investigate the emergence of electron pockets in Bi2201.
and Bi2212 [7], although a resonant-valence-bond spin-liquid model [57] (YRZ) would yield similar results [55]. Near this transition evidence for FS-free SC is found, which is consistent with many recent experiments [55, 56, 58, 59, 60] suggesting that FS-free SC in Bi2201 may be a general property of the cuprates. The material in this chapter is a summary of the work done in reference [19].

Model

The analysis presented here is based on a one band mean-field Hubbard model with competing ($\pi, \pi$)-AFM and d-wave superconducting orders, which has been invoked previously in connection with electron-doped cuprates [2]. The model is described in detail in chapter 2. The hopping parameters used here are based on photoemission experiment [9], for which the VHS in the AFM+SC system is found around $x = 0.37$.

For a given doping $x$, $\epsilon_F$ and the staggered magnetization, $S$, are determined self-consistently by using Luttinger’s theorem to obtain $\epsilon_F$ from $x$, and $S$. The results are shown in figure 3.4. The Hubbard $U(x)$ is taken as a screened Coulomb potential, which has been studied extensively [40, 61, 62]. Figure 3.4(b) shows the doping dependence of $U(x)$. For this study the data for the effective $U/t$ calculated in reference [40] were fit to a decaying exponential $U/t = a_1 e^{-x/x_0} + a_2$, where $a_1 = 4.6263$, $a_2 = 2.95$, and $x_0 = 0.045$. For SC order, it was assumed that $\Delta_0$ forms a parabolic dome in doping [6] with maximum at $x = 0.21$ based on fits of the Fermi surface to experiment [63]. The SC dome was taken to terminate at the VHS [64]. This gives a SC dome which starts at $x = 0.05$, peaks at $x = 0.21$, and terminates at $x = 0.37$. $V_{SC}$ can then be calculated
based on the given gap size. However, experimental data are often described in terms of a ‘universal superconducting dome’ (USD) with optimal $T_c$ at $x_{USD} = 0.16$ [65]. This is the case for the Bi2201 ARPES experiment [5] with which the analysis in figure 3.3 is compared. For this comparison two doping scales $x_{DFT}$ and $x_{USD}$ are defined. $x_{DFT}$ describes the doping determined from the current model and $x_{USD}$ describes doping obtained from the experimental data described in terms of the USD. The transformation of $x_{USD}$ to $x_{DFT}$ is given by $x_{DFT} = (32/22)x_{USD} - 0.022727$. $x_{DFT}$ is named as such because tight-binding parameters are often fit to DFT calculations, although here these are taken from fits to experimental data [9].

Results

To begin, how FS-free SC arises within this model will be analyzed by considering the hole doped Bi2201 system using the tight-binding parameters: $t = 0.22$eV, $t' = -0.034$eV, $t'' = 0.036$eV, $t''' = -0.007$eV, and $t'''' = 0$eV, as determined from fits to experimental data in reference [9]. The leftmost column of figure 3.1 shows how the DOS varies in the vicinity of TT1 as a function of hole-doping $x$. In the absence of SC order (red dashed curves), the bottom of the UMB shows up as a step increase in the DOS at an energy that decreases with doping, signaling the decrease of the AFM gap, $\Delta_{AFM}$. TT1 occurs at $x = 0.138$ in Bi2201 when the step edge crosses $\epsilon_F$. The transition would occur between the dopings shown in frames (d) and (g) of figure 3.1. In the central and right hand rows of figure 3.1, the DOS is separated into nodal and antinodal contributions, showing that the bottom of the UMB lies close to the antinodal direction $(\pi,0)$. The nodal region is
Figure 3.1: (a-i): DOS for the AFM+SC system (blue curves) and the AFM only system (red dashed curves). The dopings shown are $x = 0.12$ (a-c), $x = 0.13$ (d-f), and $x = 0.16$ (g-i). The first column (a,d,g) shows the full DOS. The second column (b,e,h) is the DOS calculated only in the nodal region. Similarly, the third column (c,f,i) is the DOS in the antinodal region. $\epsilon_F$ is defined to be the energy zero.
defined as the part of the first BZ contained in $0 < k_x < 3/4\pi, 0 < k_y < 3/4\pi$), while the antinodal region is the remainder of the first BZ.

When SC order is turned on, coherent peaks and a d-wave gap appear in the DOS (solid blue lines in figure 3.1). While the gap in the nodal region depends only weakly on doping, as seen in the central column in figure 3.1, evolution of the antinodal SC state with doping is more complex. At the highest doping, $x = 0.16$, there is a well-formed antinodal electron pocket with a conventional SC gap (bottom row of figure 3.1). Note however that when superconductivity turns on, the bottom of the UMB is shifted to lower energy. The SC gap is nearly symmetric, except for excess weight below $\epsilon_F$. At $x = 0.13$ (middle row), the situation is completely changed. The bottom of the UMB is no longer seen clearly, but the SC gap now has two components, an inner gap and an outer gap. Note that the gap asymmetry has now reversed, with more weight lying above $\epsilon_F$. The interpretation of these features can be clarified with reference to the $x = 0.12$ results in the top row of figure 3.1. Here the anisotropy is larger, indicating that the outer peak above $x = 0.13$ is derived from the bottom of the UMB. Decomposing the DOS, one sees that the inner gap arises from the nodal Fermi surface, figure 3.1(b,e), and the outer gap from the antinodal region, figure 3.1(c,f), even though the UMB would be entirely above the Fermi energy in the absence of SC order. This is the essence of the phenomenon of FS-free SC. In this case, FS-free SC exists at the antinodes simultaneously with a standard SC gap at the node resulting in the outer and inner gap seen in the DOS as discussed. Notably, here FS-free SC acts as a precursor to TT1.

Further insight is obtained from figure 3.2, which presents spectral weight maps for cuts
Figure 3.2: (a) Cut in momentum space from $k = (-0.2\pi, \pi)$ to $(0.2\pi, \pi)$ for the spectral weight in the AFM ordered system at $x = 0.12$. At this doping, (b) shows the same cut in the presence of AFM+SC order. (c) Antinodal DOS of the system in (a), red dashed curve, and (b), blue curve. The second row (d-f) is the same as the first row (a-b) except that this row refers to $x = 0.16$. Gray dashed lines mark $\epsilon_F$. Width of the green double arrow is proportional to the AN pair spectral weight, which is the gapped spectral weight for the SC ordered system.

along $k = (-0.2\pi, \pi)$ to $(0.2\pi, \pi)$ and compares them to the antinodal (AN) DOS. Frames (a) and (d) of figure 3.2 show the location of the UMB with only AFM order present for $x = 0.12$ and $x = 0.16$. The UMB crosses $\epsilon_F$ (gray dashed lines) as doping is increased. The bottom of the UMB is seen in the antinodal DOS, reproduced from figure 3.1 in figure 3.2 (c) and (f), as a step edge in the red-dashed curves. When SC order is turned on, spectral weights in figure 3.2(a) and (d) develop a gap around $\epsilon_F$, shown in figure 3.2(b) and (e), respectively, shown in the antinodal DOS as the blue curves in figure 3.2(c) and

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(f). To quantify the development of the AFM and SC orders, the *gapped spectral weight* is defined as the magnitude of the decrease in the antinodal DOS as temperature is lowered when the corresponding order turns on [5]. Thus, figure 3.2(f) shows the change in the antinodal DOS at $\epsilon_F$ between the AFM+SC and AFM only ordered states (indicated by a double green arrow). Figure 3.2(c) shows that the gapped spectral weight is zero before TT1 takes place, where the system is in the FS-free SC regime.

Gapped spectral weight is useful when comparing systems at different critical temperatures (or pressure, magnetic field or other parameters) involving phase transitions between states. In this connection, the current model is compared to a recent ARPES study of underdoped Bi2201 [5], where the relative strength of two different transitions (pseudogap and antinodal pairing) was determined as a function of doping and temperature [5]. It is interesting to compare their measure of the gapped spectral weight, as the depression of spectral weight at $\epsilon_F$, with the AFM+SC Hubbard model’s spectral weight difference between different ordered states. The Bi2201 ARPES experiment of reference [5] found that there was a depression of spectral weight at the Fermi energy, which began to decrease linearly with temperature ($T$) below the pseudogap temperature scale, $T^*(x)$. This was followed by a change in slope to a second value below a scale $T_{AN}(x)$, with $T^* > T_{AN} > T_c$, where $T_c$ is the SC critical temperature, as illustrated schematically in figure 3.3(c). $T_{AN}$ signals the onset of SC pair fluctuations in the antinodal region near $(\pi, 0)$, determined at the point on the Fermi surface nearest to the antinode. The depression of spectral weight which follows the linear trend down from $T^*$ is defined as the contribution to the gapped spectral weight due to the pseudogap, or the *pseudogap spectral weight*. The depression
of spectral weight which, beginning at $T_{AN}$, deviates from the linear trend is defined as the \textit{AN pair spectral weight}. In order to compare these results with the current model, an analysis is performed on the $T = 0$ limits of the theoretical measure of gapped spectral weight at the momentum point on the Fermi surface of the AFM system at $(\pi, 0)$ when no electron pocket is present, or closest to $(\pi, 0)$ when the pocket emerges. The calculated pseudogap spectral weight in this model is estimated as the change in spectral weight between the PM and AFM phase (defined as the AFM state at $T = 0$ with SC order suppressed). The AN pair spectral weight is defined as the difference in spectral weight, at the momentum points stated above, between the AFM state and the zero temperature system with AFM+SC order.

The model predicts that at low doping the dominant gapped spectral weight is predominantly associated with the pseudogap spectral weight (blue curve in figure 3.3(a)), but this weight drops suddenly as AN pair spectral weight (green curve in figure 3.3(a)) turns on near $TT1$ ($x_{TT1} \approx 0.13$), in good agreement with the aforementioned estimates of $TT1$. Referring to the doping scale at the top of figure 3.3, $TT1$ has been found in Bi2201 from thermopower [55] near $x = 0.166$ (green dashed line in figure 3.3), and in STM [66] (orange dot-dashed line) near $x = 0.19$. The experimental data [5] display similar steps in both pseudogap (blue symbols in figure 3.3(b)) and AN pair spectral weight (green symbols in figure 3.3(b)) near the same doping, arrow in figure 3.3(b). However, the size of the step is much smaller, and the AN pair spectral weight remains small in most of the underdoped (UD) regime, then increases sharply to a peak slightly above optimal-doping (OPT), decreasing finally in the overdoped (OD) regime. This second
Figure 3.3: Theoretical (a) and experimental [5] (b) pseudogap spectral weight (blue curve in (a) and triangles in (b)), and AN pair weight in Bi2201 (green curve in (a) and triangles in (b)). The red dashed curve shows the SC dome, $T_c(x)$, with temperature on the right hand vertical axis. The values for $T_c$ are estimated as $\Delta_{SC} = 5k_BT_c$ [6] and the SC dome is assumed to be parabolic, given by $\Delta_0 = 0.01637[1 - 39.0625(0.21 - x_{DFT})^2]$. Vertical lines spanning (a) and (b) represent the beginning of TT1 as determined in this work (black dotted line), thermopower [55] (green dashed line), and STM [63] (orange dot-dashed line) experiments. The black arrow in (b) points to the onset of AN weight in experimental data. Light blue and green dot-dashed lines in (b) represent model data in (a) shifted by $x_{USD} = 0.0903$ and scaled by 5/30. (c) Schematic representation of the experimental data form Kondo et. al. [5], showing spectral weight differences as a function of temperature at an arbitrary doping. This illustrates how the zero temperature experimental data in (b) were determined by Kondo et. al. [5]. The blue and green areas represent pseudogap and AN spectral weight, respectively. Blue and green double-headed arrows to the left of the vertical axis show the zero temperature magnitude of the pseudogap and AN spectral weight differences, respectively. Black arrows indicate the onset of temperature scales $T_{AN}$ and $T^*$. 
transition seen in ARPES [5] may be associated with a different topological transition, possibly related to a competing charge density wave (CDW) order not captured by the present model [58, 59, 60, 63, 67, 68, 69, 70, 71].

**Figure 3.4:** (a) Self-consistent values of $\Delta_{AF}$ as a function of doping for a system with AF order only (gray) or with combined SC+AF order (blue). The red curve shows the SC gap with the scale on the right hand vertical axis. The black dashed line indicates TT1 for our model at $x_{DFT} = 0.138$. (b) $U/t$ fit (blue curve) to the results from reference [40] (green circles) as a function of doping and $V/t$ (red curve) calculated with equation (5) from the assumed SC dome. For the present analysis we are only interested in dopings greater than $x = 0.05$, where the fit is quite good. The orange and light blue dashed curves in (a) and (b) represent the same quantities as their red and blue, solid lined counterparts, respectively, except that the doping dependence of $V$ is assumed linear and $\Delta_{SC}$ and $S$ are calculated using Eqs. 5 and 6. This shows that a large potential $V$ is needed for SC order to be sustained to dopings well below the TT1.
It is interesting to examine how TT1 modifies other properties of the cuprates, leading to possible experimental signatures. Figure 3.4(a) shows that the self-consistent $\Delta_{AF}$ drops sharply across the transition (vertical dashed line) as the electron pocket opens up. Note that in order to reproduce the experimental SC dome in the low-doping regime, the interaction parameter $V_{SC}$ in figure 3.4(b) must increase rapidly with underdoping below TT1. While a strong increase of the pairing potential near half-filling has been predicted [72], the dashed line in figure 3.4(b) indicates the effects of a more modest increase in $V_{SC}$. $T_c$ now decreases very rapidly below the TT1, figure 3.4(a), but there is still a range of FS-free SC in figure 3.2(c). To explain the lower part of the experimental SC dome in this scenario, one would postulate that the uniform AFM+SC phase becomes unstable to nanoscale phase separation (NPS) [24, 51, 73], which is sensitive to impurities. It could thus lead to the observed low-energy spin-glass phase and to the opening of an additional nodal gap [24]. Termination of this NPS at TT1 suggests that at this doping SC order stabilizes the associated $(\pi, \pi)$-AFM order.

**Conclusion**

The results presented here fit reasonably within the complex picture of pseudogap phenomena in the cuprates. Our analysis indicates that YBCO would harbor four distinct doping regimes within the SC dome. The commensurate $(\pi, \pi)$-AFM order at very low doping crosses over to a regime of spin-density wave (SDW)/stripe order, then to a regime of CDW order, and finally to a Fermi-liquid regime. Within each regime there may be
further T-dependent phase-mixing, with nearly pure phases occurring only at a few special dopings. The Hubbard model with AFM+SC order is designed for the SDW regime, and it appears that the sudden onset of SC order at TT1 stabilizes this doping and drives the NPS at lower dopings [24].

It has been shown that FS-free SC, previously observed in the pnictides [17, 52], can also occur in hole-doped cuprates. This occurs near the doping at which the topology of the Fermi surface changes as an electron pocket appears in the antinodal region, similar to the case of electron doped cuprates [3, 40]. The resulting spectral weight loss in the SC state is similar to that found in the related ARPES measurements [5]. The results presented in this chapter provide evidence for the presence of two topological transitions under the SC dome in Bi2201, consistent with the picture of a trisected dome in Bi2212 [18].
Chapter 4

Nanoscale Phase Separation

Introduction

Phase separation is a phenomenon where a uniform system separates into multiple distinct phases such as a solution separating into multiple immiscible liquids. A common example of this is the separation of oil and water. Phase separation can also occur in solids, as phase separated crystal states, or in systems of electrons where the electrons are spatially separated into domains of different ordered phases or dopings. When the size of the domains of a phase separated system of electrons is on the order of the size of the unit cell the system is said to have nanoscale phase separation (NPS).

NPS differs from macroscopic phase separation in that in the macroscopic case only two electronic phases are involved, so that properties such as the AFM and SC gaps will be doping independent. This is because when doping a macroscopically phase separated
system the majority of the domains are isolated from the neighboring domains such that the neighboring domains have little effect on each other. In the NPS scenario, the individual domains are so small that their properties will evolve with doping in each domain type due to the proximity of other domains\cite{74, 75}. Examples of this can be seen in the cuprates. In oxygen-doped La$_2$CuO$_{4+\delta}$, the excess oxygen remains mobile down to below room temperature. This results in a macroscopic phase separation over a wide doping range where the SC transition temperature $T_c$ has a fixed value close to that at optimal doping, while the AFM Néel temperature remains nearly unchanged from its value at zero doping\cite{76, 77, 78}. In contrast, in Sr-doped LSCO, the Sr ions are immobile, and a macroscopic phase separation is replaced by spin-and-charge stripes\cite{79}, which can be a form of NPS, while $T_c$ varies smoothly with $x$. The key difference in these examples is the mobility of the ions. If the positive ions are fixed in the lattice then the electrons cannot form macroscopic domains without breaking the charge neutrality of the system. However, if the electrons phase separate on the length scale of the unit cell the system as a whole can remain charge neutral.

In this chapter the question of how the Mott gap evolves away from half filling in a correlated system will be presented. The answer to this question will come in the form of a simple model of NPS after showing that a tight binding Hubbard model in a system of uniform density fails to reproduce experimental results. The model will be made more realistic by including QPGW self energy corrections and using the intermediate coupling scheme. Finally, near the Fermi energy, a Coulomb gap, applied to the NPS model, will be shown to qualitatively describe the soft gap seen in STM measurements of the DOS.
This shows that the soft gap is due to stripe pinning by impurities.

**Doping a half-filled Mott insulator**

How a correlated material evolves from being a Mott insulator into a high-$T_c$ superconductor remains a highly contentious issue nearly three decades after the discovery of the cuprates with important implications for the underlying mechanism of superconductivity. A major puzzle concerns what happens to the 2 eV gap present in the half-filled Mott insulator when it is doped. This question is explored in reference [24] and this chapter will elaborate on the results of that paper.

Experimental data in the deeply underdoped regime could help discriminate between different theoretical scenarios. However, each type of experiment has its limitations. Photoemission is unable to probe the Mott gap as it only sees the filled states. STM finds only a $\sim 100$ meV pseudogap [22] without revealing how this gap is connected to the 2 eV optical gap at half-filling. RIXS provides evidence of gap collapse, but since it measures a joint density of states, the analysis is model dependent [36]. Very recent STM data from CCOC [20] and Bi2201 [21] give insight into the question of doping evolution. These data show the presence of a large gap at half-filling, comparable to the optical gap. Remarkably, the gap in the STM spectra neither remains unchanged nor shrinks with doping, but instead it fills in. This observed behavior is not consistent with a uniform doping scenario. A possible explanation is provided by a model involving competing magnetic orders in which there can be a phase separation between the undoped insulator and an incommensurate magnetic phase near 1/8 doping [51]. Since the positive ions are fixed in
the lattice, this electronic phase separation cannot be macroscopic, but must be a NPS, possibly in the form of stripes.

Various theoretical models make clear predictions as to the doping dependence of Mott insulators. In strong coupling theories, such as the $t - J$ or the $U \to \infty$ Hubbard model, the gap remains large, but there is an anomalous spectral weight transfer (ASWT) from UMB to the LMB[80]. To understand ASWT, consider a half filled lattice with one electron per lattice site. If an electron is removed from the system there will exist a completely vacant lattice site. Given that this is a magnetically ordered system with an UMB and a LMB, the Fermi energy will now intersect the LMB because the system is now hole doped. Naively, one would now expect a single state of the LMB to lie above the Fermi energy. However, the vacant lattice site now contains two holes which is a more favorable energy state than having a single hole on the lattice site. In fact, the hole that was present before the removal of the electron will now move down in energy into the LMB such that there are two states in the LMB which lie above the Fermi energy when one electron is removed. This anomalously fast transfer of spectral weight from the UMB to the LMB describes ASWT. An effect of the ASWT in the strong coupling theories is that the width of the LMB gradually increases from $\sim 2J$ to $\sim 8t$ as doping changes from $x = 0$ to $x = 1$. For the smaller-$U$ Hubbard models, the ASWT is actually faster, as electrons can lower their kinetic energy by hopping through occupied states. In the intermediate coupling models[12], this is associated with a decrease of the magnetic gap with doping.[49]

The role of NPS or stripe physics near a Mott transition has been discussed often in
the literature\cite{79, 81, 82}. NPS bears a resemblance to the strong-coupling effects, which give rise to ASWT with doping in the cuprates\cite{80}. In an intermediate coupling model with \((\pi, \pi)\)-AFM order, the Mott gap is modeled as an AFM gap. In the presence of NPS, adding a hole creates a region of higher doping, where the AFM gap is considerably smaller, so that the second hole is shifted to a much lower energy just as in ASWT (see Fig. 4.2). Finally, in the strong coupling regime, there is a tendency for atoms with two holes to cluster (to increase the kinetic energy without introducing a \(U\)-penalty), thus providing an additional link with NPS.

**Model**

It is reasonable to start exploring the low doping physics of cuprates with a one band tight binding model. To do this, the band dispersion is taken from DFT calculations, renormalized to account for the effects of electronic correlations\cite{49, 50}. The bands are fit to a tight binding model and the magnetic interactions are obtained self consistently as described in chapter 2. Tight binding parameters for CCOC were approximated by the tight binding parameters for Bi2212. These tight binding parameters were determined through a fit to the corresponding DFT calculations \cite{83, 84, 85, 86} which are \(t = 0.36\text{eV}, t' = -0.1\text{eV}, t'' = 0.035\text{eV},\) and \(t''' = 0.01\text{eV}\). All tight-binding parameters are taken to be doping independent as in a rigid band model, which is expected to be a reasonable approximation for doping away from the cuprate planes. The tight binding parameters for Bi2201, which are taken from an experimental fit \cite{9} are \(t = 0.22\text{eV}, t' = -0.034315\text{eV}, t'' = 0.035977\text{eV}, t''' = -0.0071637\text{eV}.\)
Figure 4.1: (a-d) DOS for uniformly doped Bi2201 with AFM+SC orders at four different dopings $x$. Fermi energy is defined to be zero. The dip near zero energy in (a) is due to the SC gap. (e) Two gap scenario, showing self-consistent values of $\Delta_{AFM}$ (solid blue line) and $\Delta_0$ (dashed red line) vs doping. The $\Delta_0$ curve is scaled up by 50 for clarity.
With undoped CCOC tight binding parameters, the one band Hubbard model predicts a large, $\sim 2$ eV gap at half filling, consistent with experiment [20] as seen in figure 4.2(c). As a note, for this chapter the AFM gap term will be given by $\Delta_{AFM}$ and $\Delta$ will be used later for the Coulomb gap. The results show that twice the AFM gap parameter, $2\Delta_{AFM} = 2.76eV$, is approximately equal to the energy difference between the top of the LMB and the VHS of the UMB. The same model reproduces the experimental dispersions and density of states (DOS) for $x \geq 0.1$, in both Bi2201 [19] and Bi2212 [7].

Despite the success of the simple tight binding Hubbard model at half filling for these materials, the model fails to capture the doping dependence away from half filling. STM experiments on deeply underdoped CCOC and Bi2201 reveal a doping evolution, where a strong growth of in-gap states in the local DOS is observed in the spectra. This gap filling is in contrast to the results of doping in the tight binding Hubbard model, as seen for Bi2201 in figure 4.2. These results show a gap closing that leaves only a small gap at higher doping.

The concept of NPS can be used in order to explain this gap filling. There exists reasonably good evidence that cuprates can exhibit NPS due to sources of charge inhomogeneity. First, a charge-density wave phase has been reported in a number of cuprates [68, 87, 88, 89, 90, 91, 92]. Then, STM studies find patches of varying local density, which are mainly correlated with oxygen vacancies in the Bi-cuprates [93]. Finally, a phase separation has been predicted in Bi-cuprates, most notably between the insulating phase in the undoped compound and at a doping around $x = 0.125$, [51]. While NPS can have different spatial patterns, including stripes, dots, or polaronic stripes seem to be favored in $La_{2-x}Sr_xCuO_4$. 73
Figure 4.2: DOS for the AFM system in the presence of a NPS for $x_{av} = 0.03$ (a) and 0.08 (b) for Bi2201. Parameters used are $x_0=0.0$, $x_1=0.09$, with $Z=0.9$ and $Z=0.7$ respectively. $\Delta=153\text{meV}$ in (a) and 20\text{meV} in (b). (c) Calculated DOS at half-filling (solid blue line) for CCOC compared to the corresponding STM data [20] (black curve with noise). (d) Calculated DOS for the AFM system with NPS in CCOC (solid blue line) compared to the corresponding experimental data [20] (solid red line with noise). STM intensities in (c,d) are scaled to match the VHS below the Fermi energy. Here, $x_0=0.0$, $x_1=0.09$, with $Z=1$ and $Z=0.5$ respectively, and $\Delta=82.4\text{meV}$ with $x_{av} = 0.03$.

(LSCO) [79, 82, 94, 95] and Bi2201 [96] as they provide a natural explanation for the Yamada plot [97]. The two domains of this stripe like NPS will be represented by two separate dopings of the tight binding Hubbard model which will be called end phases. The end phases will be given by the half filled state and a doping at the end of the gap filling regime. The total DOS will be approximately given by the superposition of the DOSs of the two end phases.
To model the DOS end phases more realistically, the QPGW method outlined in appendix A will be used in combination with the NPS model. Figure 4.2 displays the DOS calculated in the NPS model for Bi2201 and CCOC within the QPGW scheme. The dopings for CCOC correspond to the STM data of Ref. [20], which is plotted along with the calculated curves in frames (c) and (d), assuming that each patch contains a stripe-like mix of two phases. For Bi2201, in figures 4.2(a,b), the two stable phases are a state with $x_0 = 0.0$, $U/t = 7.576$, and $Z = 0.9$, and the other with $x_1 = 0.09$, $U/t = 3.576$, and $Z = 0.7$. Linear combinations of these DOSs are then generated to represent an average doping, $x_{av}$, where $x_{av} = 0.03$ in figure 4.2(a) and 0.08 in figure 4.2(b). For CCOC, figures 4.2(c,d), the stable phases have $x_0 = 0.0$ with $U/t = 8.5$, $Z = 1$ and $x_1 = 0.09$ with $U/t = 3.3$, $Z = 0.5$. In this model, $x_0$ corresponds to half-filling and gives the experimentally observed large gap, $x_1$ is taken to be the doping which reproduces the DOS of the in-gap states, and $Z$ is determined self consistently within the QPGW scheme [12]. For Bi2201 the effective $U/t$ calculated in reference [40] was used and fit to a decaying exponential $U/t = a_1 e^{-x/b} + a_2$, where $a_1 = 4.626$, $a_2 = 2.95$, and $b = 0.045$. These are the same values for $U/t$ as used in the previous two chapters. The resulting self consistent values of $\Delta_{AFM}$ are shown in figure 4.1(e). For CCOC, values of $U$ are chosen such that the self consistent $\Delta_{AFM}$s give the DOSs that match the experimental large gap and in-gap states in reference [20] for $x_0$ and $x_1$, respectively. This simple picture is seen in figure 4.2 to reproduce the effect of gap-filling as a function of doping, rather than the gap-closing shown in figure 4.1. In figures 4.2(c,d) the model calculations are in good agreement with the corresponding experimental data represented by the solid black and
red lines. The model captures both the gap edge and the DOS peak, and, for the doped sample, the in-gap DOS. Note that at energies $< -1 \text{ eV}$ or $> 3 \text{ eV}$ there is additional DOS weight associated with bands not included in the present modeling.

The Coulomb Gap

The NPS model, outlined in the previous section, captured experimental DOS over a wide range of energy. In this section the NPS model will be applied to the details of the experimental DOS from reference [22] in a small energy range around the Fermi energy. Figure 4.3 compares the model calculations with data from reference [22] over a narrow energy range. It is important to recognize here that at low energies the system undergoes a metal-insulator transition associated with a nodal gap [18, 22, 98, 99, 100]. Within the present model, this gap arises on the charged stripes, which are regions of higher doping, and increases as the doping decreases and the stripes separate further. Assuming this to be an effect of stripe pinning by impurities, the gap is modeled as a Coulomb gap [23], as has been suggested previously [99, 101]. The Coulomb gap is a soft gap arising from the Coulomb interaction of particles on impurity sites, and its effect on the DOS, in a two-dimensional system, can be calculated self consistently via the following equation [23]:

$$g(\omega) = \Delta exp \left( -\int_0^\infty \frac{g(\omega')d\omega'}{(\omega + \omega')^2} \right), \quad (4.1)$$
where \( g(\omega) \) is the DOS at frequency \( \omega \), and \( \Delta \) is the width of the Coulomb gap. In this case, \( g(\omega) \) is multiplied by the DOS obtained from a QPGW computation with AFM order to simulate the presence of a Coulomb gap. Figure 4.3 shows that the resulting DOS reproduces the characteristic features of the STM data quite well, including the peak above the Fermi energy due to the bottom of the UMB in the \( x_1 \) phase, the peak below the Fermi energy from the LMB in the \( x_0 \) phase, and the soft gap with zero states at the Fermi energy due to the Coulomb gap. Table 4.1 gives details of the parameters used for the model results (dashed curves) in figure 4.3 from top down. In order to minimize the number of parameters, we estimate the energy of the Coulomb gap from the experimental data. The onset temperature, \( T_0 \), of the Coulomb gap was derived from transport measurements given in figure 4(c) of reference [99], and used to estimate the size of the gap assuming \( 2\Delta/k_B T_0 = 4 \). Gap values for CCOC, determined in this way, are listed in Table 4.1 and figure 4.3.

**Table 4.1:** Value of Coulomb gap, \( \Delta \), and the corresponding average doping values, \( x_{av} \), used in calculating the DOSs of CCOC shown in figure 4.3. The calculated curves from top to bottom in figure 4.3 correspond to DOS indices 1 through 9 in this table.

<table>
<thead>
<tr>
<th>DOS index</th>
<th>( \Delta ) (meV)</th>
<th>( x_{av} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>44.5</td>
<td>0.060</td>
</tr>
<tr>
<td>2</td>
<td>50.8</td>
<td>0.055</td>
</tr>
<tr>
<td>3</td>
<td>57.2</td>
<td>0.050</td>
</tr>
<tr>
<td>4</td>
<td>63.5</td>
<td>0.045</td>
</tr>
<tr>
<td>5</td>
<td>69.8</td>
<td>0.040</td>
</tr>
<tr>
<td>6</td>
<td>76.1</td>
<td>0.035</td>
</tr>
<tr>
<td>7</td>
<td>82.4</td>
<td>0.030</td>
</tr>
<tr>
<td>8</td>
<td>88.8</td>
<td>0.025</td>
</tr>
<tr>
<td>9</td>
<td>95.1</td>
<td>0.020</td>
</tr>
</tbody>
</table>

The Coulomb gap is the natural result of pinning of stripe order by impurities. Therefore,
it exists over the doping range, \(0.00 \leq x_{av} \leq 0.09\), where NPS has been modeled as a stripe order. As \(x_{av}\) moves away from half-filling, the magnitude of the Coulomb gap decreases to account for the shrinking width of magnetic stripes. If the Coulomb gap is ignored, spectral weight will be present at the Fermi energy as seen in the DOS of figure 4.1(b-d) resulting in a significant deviation from experimental results at low energies \([20, 22]\).

Notably, the experimental data in figure 4.3 show that the soft gap closes at a point at the Fermi energy, whereas the calculations presented here yield a broad minimum which is symmetric around the Fermi energy. The simple model of DOS introduced in this study, however, is only meant to capture global features of a NPS with a Coulomb gap. A more sophisticated model should take into account the local topology of the NPS phase, and the details of the associated phase separation. For instance, in the low-doping regime, the doped-phase could take the form of islands or stripes, and these details will modify the nature of the resulting Coulomb gap. It may also be necessary to include disorder effects (see figure 65 of reference \([82]\)).

**Conclusion**

In conclusion, the gap-filling with doping, rather than gap-closing, observed experimentally in STM studies of deeply underdoped CCOC and Bi2201 is naturally understood in terms of a NPS\([51]\). Local stripes would be strongly pinned by impurities, explaining the occurrence of a nodal gap, a metal-insulator transition, and spin-glass-like phenomena found in underdoped cuprates. The NPS model also predicts\([51]\) the coexistence of
Figure 4.3: Calculated DOSs including Coulomb gap for modeling a NPS in an AF system (dashed lines) compared to the corresponding STM data from reference [22] (solid lines). The number next to a theoretical curve indicates the DOS index of that curve associated with table 4.1. The curves are shifted vertically for clarity. Different experimental curves correspond to DOS measured on different patches in a single CCOC sample.

($\pi, \pi$) AF order and an incommensurate SDW phase, as has been observed recently in LSCO[102].
Chapter 5

Quasiparticle Interference Models

Introduction

Scanning tunneling microscopy (STM) is an important tool for understanding and investigating the properties of condensed matter systems. STM is a real space probe of the electrons on the surface of a material. A STM works by bringing a metal tip atomically close to the surface of a sample material. A bias voltage is then applied between the tip and the sample causing electrons in the sample to tunnel into the tip. This results in a measurable current through the STM tip. The tip can be translated across the surface of the sample to produce a real space map of conductance. Varying the bias voltage with the tip held at a single location on the sample gives the current, $I$, as a function of voltage, $V$. The derivative $dI/dV$ is proportional to the DOS. Measuring $dI/dV$ at a
single location produces a local density of states (LDOS) which can change depending on the location of the tip on the sample.

Fourier-transform scanning tunneling spectroscopy (FT-STS) is an analysis technique for extracting momentum space information from real space STM conductance maps. FT-STS is performed by taking the two dimensional Fourier transform of the STM conductance map revealing the momentum dependence of the LDOS. Ref. [103] explains that momentum space variations in the LDOS are due to quasiparticle scattering off of impurities. The LDOS is proportional to the real space wave function

\[
\rho(r, \omega) = \sum_k |\psi_k(r)|^2 \delta(\omega - \epsilon(k)), \quad (5.1)
\]

where, \( r \) is the real space vector, \( \omega \) is the frequency, \( k \) is the momentum space index, \( \psi_k(r) \) is the real space wave function, and \( \epsilon(k) \) is the band dispersion. In a metal the wave functions are given by Bloch wavefunctions of the form \( \psi_k(r + R) = e^{ik\cdot R} \psi_k(r) \) where \( R \) is the lattice vector. Using the Bloch wavefunction in equation 5.1 the LDOS becomes independent of \( k \) between lattice sites. The LDOS becomes momentum dependent when impurities are added to the system. Impurities mix quasiparticle states at \( k_1 \) and \( k_2 \) leaving a \( k \) dependence in equation 5.1.

The FT-STS images contain a significant amount of information about the material’s electronic and physical structure. LDOS modulations which are non-dispersive can signify types of order in the system such as checkerboards, charge density waves, and fluctuating
stripes [104, 105, 106, 107, 108]. The dispersive modulations are often related to interference of quasiparticles in the system. This phenomenon is called quasiparticle interference (QPI) and the FT-STS images containing QPI will be referred to as QPI images or patterns. In this chapter QPI images will be modeled using a scattering T-matrix formalism and self-energy corrections in order to explain antinodal states appearing at energies lower than the superconducting gap in experimental QPI images.

STM experiments have shown that QPI images in Bi2201 [25, 26] exhibit a topological transition from a small-to-large Fermi surface. This is inferred due to the emergence with increased hole doping of QPI patterns in the antinodal region at energies inside of the superconducting gap. These patterns have been called the “antinodal triplet QPI”. The goal of the work presented here is to model these patterns using the Hubbard model with self energy corrections to investigate correlation effects. The standard octet model for QPI, which the antinodal triplet QPI diverges from, will be outlined first. Next, a realistic model for impurity scattering, known as the Maltseva-Coleman T-matrix model [27], will be presented. Self energy corrections will then be applied to the T-matrix model. Three different types of self energy will be compared. The Dynes self energy [28] is entirely imaginary and therefore only effects the broadening of the spectral weight and in turn QPI. In superconductors, pair-breaking scattering, which breaks superconducting pairs, can be modeled as a self energy effect [29]. This pair-breaking self energy goes beyond the simple Dynes self energy and takes into account the superconducting properties of the system. The most sophisticated self energy that will be used here is the QPGW self energy. These corrections should include pair breaking effects as well as other electron
correlations in the system. These models for QPI will be compared with each other and experimental results to give insight into the causes of the observed antinodal triplet QPI.

Octet Model

QPI can be understood most simply as elastic scattering between quasiparticle states with the most probable values of \( k \). STM has found that in the superconducting state of cuprates QPI presents an octet of dispersive bright spots. These bright spots can be explained using the band structure and a scattering T-matrix formalism [109]. In the superconducting state, cuprates exhibit a d-wave superconducting gap, which has been discussed in chapter 2 on the Hubbard model. A simple picture of what the superconducting state in cuprates looks like starts by describing the superconducting gap.

The gap is said to be d-wave because, in momentum space, it has the same symmetry that the electron d-orbitals do in real space. The d-wave gap has been observed experimentally [110] and when modeling cuprates in the superconducting state one adds a momentum dependent gap of the form

\[
\Delta_k = \frac{\Delta_0}{2} \left( \cos (k_x) + \cos (k_y) \right).
\] (5.2)

Applying superconducting order to a one band tight binding model the following Hamiltonian is obtained:

\[
H = \sum_{k\sigma} \epsilon_k c_{k\sigma}^{\dagger} c_{k\sigma} - \sum_k \left( \Delta_k c_{k\uparrow}^{\dagger} c_{-k\downarrow} + \Delta_k^{\ast} c_{k\downarrow}^{\dagger} c_{-k\uparrow} \right),
\] (5.3)
where $\epsilon_k$ is the bare tight-binding dispersion, $c_{k\sigma}^\dagger(c_{k\sigma})$ is the electron creation (annihilation) operator at momentum $k$ and with spin $\sigma$. The eigenvalues of this Hamiltonian are

$$E(k) = \pm \sqrt{\epsilon_k^2 + \Delta_k^2}. \quad (5.4)$$

$E(k)$ describes the dispersion of quasiparticles in this system. The scattering of quasiparticles off one another is largest between states in momentum space which have the largest LDOS. A simple way to identify which states have the largest LDOS is by inspecting the contours of constant energy (CCE). Figure 5.1 shows an example of some of these contours generated by $E(k)$. First, the LDOS is proportional to

$$\int_{E(k)=\omega} |\nabla_k E(k)|^{-1} dk. \quad (5.5)$$

This indicates that points along the path of lowest slope along the energy contours (where CCE are farthest apart) will give the largest LDOS at that particular energy cut. The lowest energy contour in the superconducting state is given by four symmetric points along the nodes. The nodes are diagonal lines across the square Brillouin zone where $\Delta_k$ is equal to zero. As energy increases the contours evolve from points at zero energy, to closed arcs dubbed “banana-shaped” contours. It is at the tips of these closed arcs, which happen to follow the line which is defined by the Fermi surface in the normal state, where the LDOS is largest. At the energy of the superconducting gap the closed contours just touch the edge of the Brillouin zone. At energies outside of the superconducting gap the closed arcs become open lines and this analysis is no longer valid.
Figure 5.1: black lines show the CCE of $E(k)$ from equation 5.4. These contours have the characteristic closed banana shape until they touch the Brillouin zone boundary and become open contours. The tips of the bananas are the location of the LDOS maximum for that specific energy cut and follow the red curve which is the paramagnetic Fermi surface given by $\epsilon_k$. 
Now that the maximum LDOS has been identified, the main features in the QPI patterns can be described. Scattering between states with the largest LDOS should produce the most probable scattered states. The momentum vector of an elastically scattered state is given by the difference of each state’s momentum vector. This results in a vector which connects the two states being scattered and describes the momentum of the scattered state. At a given energy cut there are eight LDOS maxima in the Brillouin zone. These correspond to the tips of the banana-shaped contours. Each tip state can scatter off of itself (zero momentum transfer) and the other seven tip states resulting in a total of eight unique scattering states. Due to the four fold symmetry of momentum space, the QPI pattern will show 32 dispersive, bright points.

This is known as the octet model and is a good first step in predicting QPI patterns in the superconducting state of cuprates. This simple picture gets complicated by order parameters which compete with superconductivity, the pseudogap, and static features in the QPI. A more detailed model, T-matrix scattering, can be used to describe the effects of these features on QPI patterns and will be discussed in the following sections.

**Maltseva-Coleman T-matrix model**

To realistically model QPI a unique theoretical technique developed by Maltseva and Coleman [27] which considers fluctuations in the LDOS will be used. Fluctuations are important because experiments consider the ratio of conductance maps at positive and negative bias voltages to eliminate noise in the Fourier transformed image. This ratio is
known as a Z-map and is given by

\[ Z(r, V) = \frac{dI/dV(r, +V)}{dI/dV(r, -V)} = \frac{\rho(r, +V)}{\rho(r, -V)}. \]  

To include fluctuations ref. [27] writes the LDOS as

\[ \rho(r, \omega) = \rho_0(\omega) + \delta\rho(r, \omega) \]

where \( \rho_0(\omega) \) is a reference LDOS which is independent of position and \( \delta\rho(r, \omega) \) are the fluctuations representing the deviation from \( \rho_0(\omega) \). Equation 5.6 becomes

\[ Z(r, V) = \frac{\rho_0(\omega) + \delta\rho(r, \omega)}{\rho_0(\omega) - \delta\rho(r, \omega)}. \]  

For small fluctuations, such that \( \delta\rho(r, \pm V) \ll \rho_0(\pm V) \), \( Z(r, V) \) is approximately

\[ Z(r, V) = Z_0(V) \left[ 1 + \frac{\delta\rho(r, +V)}{\rho_0(\omega)} - \frac{\delta\rho(r, -V)}{\rho_0(\omega)} \right]. \]  

QPI is a momentum space picture, therefore a Fourier transform must be applied to equation 5.8, yielding

\[ Z(q, V) = Z_0(V) (2\pi)^2 \delta^2(q) + Z_0(V) \left[ \frac{\delta\rho(q, +V)}{\rho_0(\omega)} - \frac{\delta\rho(q, -V)}{\rho_0(\omega)} \right]. \]  

For \( q \neq 0 \) the first term in equation 5.9 is zero leaving only the second term. The second term in equation 5.9 can be written in terms of even and odd fluctuations \( \delta\rho^{even(odd)}(q, \omega) = \)
\[(\delta \rho (q, \omega) \pm \delta \rho (q, -\omega)) / 2\] to take the form

\[
Z(q \neq 0, V) = Z_0(V) \left\{ \delta \rho_{\text{even}}(q, V) \left[ \frac{1}{\rho_0(V^+)} - \frac{1}{\rho_0(V^-)} \right] \right.
+ \delta \rho_{\text{odd}}(q, V) \left[ \frac{1}{\rho_0(V^+)} + \frac{1}{\rho_0(V^-)} \right] \right\}.
\] (5.10)

Equation 5.10 reveals that if \(\rho_0(\omega)\) is particle-hole symmetric (i.e. symmetric in \(V\)) then the even fluctuation term becomes zero and the Z-map is proportional to the odd fluctuations. Likewise, if the system deviates from particle-hole symmetry even fluctuations start to effect the Z-map. In the case of particle-hole symmetry, equation 5.10 becomes

\[
Z(q \neq 0, V) = Z_0(V) \frac{2}{\rho_0(V)} \delta \rho_{\text{odd}}(q, V).
\] (5.11)

At this stage it is important to show how the fluctuations are calculated such that equation 5.8 can be simplified further. Here, the vector space is defined by the Balian-Werthamer spinor[111]

\[
\Psi(r, \tau) = \left( \begin{array}{c} 
\psi_\uparrow(r, \tau) \\
\psi_\downarrow(r, \tau) \\
\psi_\downarrow^\dagger(r, \tau) \\
-\psi_\uparrow^\dagger(r, \tau) \end{array} \right).
\] (5.12)

The Green’s function, in the basis defined by equation 5.12, is given by

\[
G(r', r, \tau) = -\left\langle T_\tau \Psi(r', \tau) \Psi_\dagger(r, 0) \right\rangle.
\] (5.13)
The LDOS is related to the greens function by

\[ \rho(r, \omega) = \frac{1}{\pi} \text{Im} \text{Tr} \left[ \frac{1 + \tau_3}{2} G(r, r, \omega - i\delta) \right]. \]  

(5.14)

The trace of the quantity in square brackets is a convenient way of representing the sum of electron pairs of the same spin.

The LDOS can be broken down into even and odd functions in \( \omega \) as,

\[ \rho^{\text{even}}(r, \omega) = \rho(r, \omega) + \rho(r, -\omega) = \frac{1}{2\pi} \text{Im} \text{Tr} \left[ G(r, r, \omega - i\delta) \right] \]  

(5.15)

\[ \rho^{\text{odd}}(r, \omega) = \rho(r, \omega) - \rho(r, -\omega) = \frac{1}{2\pi} \text{Im} \text{Tr} \left[ \tau_3 G(r, r, \omega - i\delta) \right]. \]  

(5.16)

Fourier transforming these equations gives the momentum space representations of the even and odd LDOS functions. The even function, for example, becomes

\[ \rho^{\text{even}}(q, \omega) = \frac{1}{2\pi} \int dr e^{i r \cdot q} \rho^{\text{even}}(r, \omega) \]

\[ = \frac{1}{2\pi} \int dr e^{i r \cdot q} \frac{1}{2\pi} \text{Im} \text{Tr} \left[ G(r, r, \omega - i\delta) \right] \]

\[ = \frac{1}{2\pi} \int \int dr dk dk' e^{i r \cdot q - i r \cdot k'} \frac{1}{2\pi} \text{Im} \text{Tr} \left[ G(k, k', \omega - i\delta) \right] \]

\[ = \int \int dk dk' \delta(q - k - k') \frac{1}{2\pi} \text{Im} \text{Tr} \left[ G(k, k', \omega - i\delta) \right] \]

\[ = \frac{1}{2\pi} \int \text{Im} \text{Tr} \left[ G(k, q - k, \omega - i\delta) \right] dk . \]  

(5.17)
The fluctuations in the Greens function are assumed to be proportional to the fluctuations in the LDOS. The even and odd fluctuations are given by

$$\delta \rho^{\text{even}}(q, \omega) = \frac{1}{2\pi} Im \text{Tr} \left[ \int \delta G(k, q-k, \omega-i\delta) \, dk \right] \quad (5.18)$$

$$\delta \rho^{\text{odd}}(q, \omega) = \frac{1}{2\pi} Im \text{Tr} \left[ \int \tau_3 \delta G(k, q-k, \omega-i\delta) \, dk \right]. \quad (5.19)$$

The fluctuations of the Green’s function are found using the T-matrix expansion in terms of the bare Green’s function, $G(k, \omega)$ and the scattering T-matrix, $\hat{t}(k, k')$. The T-matrix expansion of the Green’s function is

$$G(k, k', \omega) = G(k, \omega) + G(k, \omega) \hat{t}(k, k') G(k', \omega)$$

$$= G(k, \omega) + \delta G(k, k', \omega). \quad (5.20)$$

The above equation defines the Green’s function fluctuations in terms of the scattering T-matrix. Using this definition in equations 5.18 and 5.19 gives

$$\delta \rho^{\text{even}}(q, \omega) = \frac{1}{2\pi} Im \text{Tr} \left[ \int G(k, \omega-i\delta) \hat{t}(k, q-k) G(q-k, \omega-i\delta) \, dk \right] \quad (5.21)$$

$$\delta \rho^{\text{odd}}(q, \omega) = \frac{1}{2\pi} Im \text{Tr} \left[ \int \tau_3 G(k, \omega-i\delta) \hat{t}(k, q-k) G(q-k, \omega-i\delta) \, dk \right] \quad (5.22)$$

With these equations the even and odd fluctuations can be calculated given the bare Green’s function and the form of the scattering T-matrix.
This formalism was developed such that the coherence factors, which modify the qualitative picture of the octet model, can be extracted and used to describe magnetic field dependent effects in QPI measurements [112]. In a d-wave superconductor the superconducting gap is anisotropic such that the superconducting order parameter picks up a phase in different regions of momentum space. By applying a magnetic field, vorticies are induced in the superconductor. The vorticies become the impurity which quasiparticles will scatter off of. When quasiparticles scatter between states in momentum space with superconducting order parameters of different phases in an applied magnetic field, their QPI intensity is modified by that phase. This is known as a coherence factor. The coherence factors can be studied experimentally by applying a magnetic field and comparing the relative QPI intensities to the zero field QPI intensities. The paper by Maltseva and Coleman [27] continues the above derivation to extract these coherence factors. Because magnetic field effects will not be considered here, the derivation presented in this chapter will stop at equation 5.22 which gives a realistic model for the Z-maps.

The T-matrix

The key ingredient in preforming realistic QPI calculations is the scattering T-matrix. The T-matrix can be expanded in terms of the scattering potential $\hat{U}(k, k')$ as follows

$$
\hat{t}(k, k') = \hat{U}(k, k') + \sum_{k''} \hat{U}(k, k'') G(k'', \omega') \hat{U}(k'', k') + \ldots
$$

$$
= \hat{U}(k, k') + \sum_{k''} \hat{U}(k, k'') G(k'', \omega') \hat{t}(k'', k').
$$

(5.23)
In the first Born approximation \( \hat{t}(k, k') = \hat{U}(k, k') \). This approximation is good for weak scatterers.

**Table 5.1: T-matrix of scatterers to first order**

<table>
<thead>
<tr>
<th>Scatterer</th>
<th>( t(k, k') )</th>
</tr>
</thead>
<tbody>
<tr>
<td>scalar</td>
<td>( \tau_3 )</td>
</tr>
<tr>
<td>Andreev</td>
<td>( \tau_1 )</td>
</tr>
<tr>
<td>magnetic</td>
<td>( \sigma \cdot m )</td>
</tr>
</tbody>
</table>

The three types of scatterers used in this work are scalar, Andreev and magnetic. The form of the T-matrix associated with these scattering types is listed in table 5.1. Scalar and Andreev scattering mix electron and hole states in equations 5.21, and 5.22. In contrast, magnetic scattering mixes only up and down spin components which, to first order, results in an LDOS of zero in a non-magnetic system.

In order to see the effects of magnetic scatterers in an antiferromagnetic system the expansion must be carried out to second order in the Born approximation. The resulting magnetic scattering T-matrix is

\[
\hat{t}(k, k') = \hat{U}(k, k') + \sum_{k''} \sigma \cdot m G(k'', \omega') \sigma \cdot m.
\]

(5.24)

The sum over intermediate states produces a non-zero QPI pattern.

**The Antinodal Triplet QPI**

The antinodal triplet QPI was observed in experimental STM measurements of Bi2201 [25, 26] as three distinct lines in constant energy cuts near the antinodal region close to
at energies less than the superconducting gap was quite unexpected. The uniqueness of this feature came from its large momentum value and that it turned on inside the superconducting dome with increased hole doping. These properties can be explained by a suppressed superconducting coherence due to a competition between superconductivity and the pseudogap phase. The onset of the antinodal triplet QPI with increased hole doping appeared to be associated with a topological transition from a small-to-large Fermi surface, signifying a transition out of the pseudogap phase.

ARPES measurements define the onset of the pseudogap phase by the shape of the Fermi surface. In the paramagnetic state, there is a single, fully connected Fermi surface leaving no abrupt truncations within the extended Brillouin zone. This is called a closed Fermi surface. Upon transitioning to the pseudogap phase the closed Fermi surface is replaced by “Fermi arcs”. Fermi arcs are lines, defined by the electronic spectral weight at the Fermi energy, that appear disconnected in momentum space. Luttinger’s theorem states that the volume enclosed by the Fermi surface is directly proportional to the system’s particle density. In the case of the Fermi arcs, there is no way to define an enclosed volume because the arcs truncate abruptly. Quantum oscillation measurements suggest that these Fermi arcs could be one side of a Fermi pocket [113]. A Fermi pocket is a closed Fermi surface within a single Brillouin zone. When states at energies below the Fermi energy are enclosed by the Fermi pocket, this is called an electron pocket. Conversely, if states above the Fermi energy are enclosed by the Fermi pocket, it is known as a hole pocket.
There is a long history of STM identifying periodic structure in cuprates. A first step to identifying the antinodal triplet feature is to see how it compares to previously studied periodic features. A main feature of the antinodal triplet is that it is non-dispersive and appears at low energy (less than the pseudogap energy) and large momenta (nearly \((0, \frac{2\pi}{a})\); the reciprocal lattice vector). This property rules out static checkerboard, charge density wave, smectic, and fluctuating stripe orders as being the cause of the antinodal triplet. The static checkerboard \([104, 106]\), though non-dispersive, has a much lower momentum vector (around \((0, \frac{\pi}{4a})\)). The same situation occurs in charge-density-wave order, which can explain the origin of the static checkerboard \([105]\). Other non-dispersive orders like smectic \([108]\) order and fluctuating stripes \([107]\) appear at a momentum vector of \((0, \frac{3\pi}{2a})\) with energies around that of the pseudogap.

The goal of the rest of this chapter will be to describe the antinodal triplet QPI using the T-matrix formalism presented in sections 5.3 and 5.4. The unique approach of this method is the inclusion of self energy corrections using Dynes self-energy, pair breaking self-energy, and the QPGW method. These results will be compared to experimental data which were performed on four samples of hole doped Bi2201 \([25]\). These samples were designated by their superconducting critical temperature, \(T_c\), and location in doping relative to the doping with maximum \(T_c\). From lowest to highest doping, these samples were labeled as underdoped at \(T_c=25\)K, underdoped at \(T_c=32\)K, optimally doped at \(T_c=35\)K, and overdoped at \(T_c=15\)K (UD25K, UD32K, OPT35K, and OD15K).
Dynes Self-Energy

Here, the previously laid out T-matrix formalism, in conjunction with self-energy calculations on a one band \((\pi, \pi)\)-AFM+SC tight binding model, described in chapter 2, will be used to explain STM measurements [25] on Bi2201 at dopings and temperatures inside of the superconducting dome. The tight-binding parameters used here are \(t=0.22\text{eV},\)
\(t'=0.34315\text{eV},\)
\(t''=0.035977\text{eV},\)
\(t'''=-0.0071637\text{eV}.\) These parameters can be found in the supplementary material of reference [25].

The first step to modeling the QPI is to define the Green’s function. A BCS Hamiltonian with a \(d\)-wave gap, \(\Delta_k = \Delta_0 (\cos(k_x) - \cos(k_y))/2,\) where \(\Delta_0\) is the maximum gap value, is assumed. The resulting Greens function is a tensor

\[
G_k(i\omega_n) = [i\omega_n - \epsilon_k \tau_3 - \Delta_k \tau_1]^{-1}
\]

(5.25)

The quantity \(\epsilon_k\) is the normal state dispersion which is given by the tight-binding model found in chapter 2. Using the bare Greens function, which will be referred to as \(G_0\), would lead to a situation with antinodal spectral weight appearing only when \(\omega \simeq \Delta_0\). Observation of the antinodal features at low energies \(\omega \ll \Delta_0\) suggests that there is considerable broadening of the spectral weight due to the finite lifetimes of the quasiparticles.

To model the broadening of the spectral weight, the Dynes self energy was used [28]. The Dynes model is often used to describe the broadening seen in the spectral weight of ARPES measurements[114, 115]. This model includes a purely imaginary self energy
Figure 5.2: (a-d) represent samples UD25, UD32, OPT 35, and OD15, respectively along with the Dynes pairbreaking model. Each image is divided into three sections. Upper left: model QPI with pairbreaking $\Gamma$. Lower left: model QPI with no self energy corrections. Right: experimental QPI data. The color scale for the theoretical images is the same for (a-c), while the scale for (d) is 9.09 times larger. This is due to the proximity of OD15K to the Van Hove singularity which leads to an increased spectral weight. It is clear that when the self energy corrections are included the antinodes become more apparent. The black dashed lines trace out the fermi-surface lines of the model and are reflected onto the right hand panel.
to describe pairbreaking, which amounts to adding an imaginary quantity to the energy term in the BCS form of the density of states$[28]$. To apply this to equation 5.25 it is necessary to modify the Green’s function with a self energy term. To do this, a bare Green’s function, $G_0$, like the one in equation 5.25, is applied to Dyson’s equation for the self energy corrected Green’s function,

$$G = \frac{G_0}{1 - \Sigma G_0},$$

(5.26)

where $\Sigma$ is the self energy which, for the Dynes model, is entirely imaginary and diagonal with equal diagonal elements, $i\Gamma$.

Figure 5.2 compares the T-matrix QPI model with and without pairbreaking to experiment. What is plotted is the amplitude of $\delta\rho^{odd}(q, \omega)$. The T-matrix for a scalar scatterer, given by $\hat{t}(q, k) = \tau_3$ $[27]$, is used to compare to the STM Z-maps. In figure 5.2 the images are arranged in order of increasing hole doping from left to right. Each image shows the magnitude of $\delta\rho^{odd}$ using the bare Greens function in the bottom left, $\delta\rho^{odd}$ using the Dynes self energy in the top left and the experimental Z-map to the right. It is apparent that only with the inclusion of broadening from the Dynes self energy do the QPI calculations show the antinodal triplet features seen in the experimental data. Table 5.2 gives the parameters used in these calculations. It should be noted that the value for $\Gamma$ of 50meV is larger that that used in ARPES experimental analyses $[115]$. This is because reference $[115]$ uses cuts in the ARPES spectra which eliminate considerable inhomogeneous broadening. Therefore, it is expected that a larger broadening is necessary to
model STM results. The values for $\Gamma$ used here are necessary to shift antinodal features to the low experimental energies, given the observed superconducting gaps.

Table 5.2: Parameters for the QPI model

<table>
<thead>
<tr>
<th>Sample</th>
<th>$d$-wave gap (meV)</th>
<th>$\Gamma$(meV)</th>
<th>$\omega$(meV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>UD25K</td>
<td>20</td>
<td>50</td>
<td>9</td>
</tr>
<tr>
<td>UD32K</td>
<td>20</td>
<td>50</td>
<td>5</td>
</tr>
<tr>
<td>OPT35K</td>
<td>20</td>
<td>50</td>
<td>5</td>
</tr>
<tr>
<td>OD15K</td>
<td>9</td>
<td>10</td>
<td>3</td>
</tr>
</tbody>
</table>

Pair Breaking Self-Energy

The Dynes self-energy corrections are a good starting point to show that self-energy is a key component to modeling the antinodal triplet QPI. To understand the physics of what is causing these states, and therefore self-energy, the effects of quasiparticle pairbreaking were investigated. The antinodal triplet QPI was modeled as a form of QPI where pairbreaking is large enough to weaken the octet model structure, leaving behind a convoluted Fermi surface map. The Fermi surfaces of the dopings analyzed are those of the paramagnetic system because there is a transition for a Fermi surface with pockets to a large Fermi surface. This transition is discussed in detail in chapter 3. The effects on the QPI of a two-pocket scenario would not look very different from the large Fermi surface scenario as long as the residual magnetic gap is not too large. Superconductivity with a d-wave gap $\Delta_k = \Delta_0 (\cos(k_x) - \cos(k_y))/2$, where $\Delta_0$ is the maximum gap value, was included and the QPI was modeled as the autocorrelation of the spectral weight [109]. By itself, this would lead to the conventional octet model, with antinodal spectral weight appearing only when $\omega \simeq \Delta_0$. Since the antinodal features are seen for $\omega << \Delta_0$, it is
assumed that there is considerable pair-breaking \cite{115}. There are two kinds of scattering in a superconductor; pair-breaking interactions which cause scattering in a superconductor even at low temperatures, and non-pairbreaking scattering of quasiparticles at finite temperatures which dominate the normal-state scattering, but which obey Anderson’s theorem and so have no effect on superconductivity at T=0.

The self consistent equations which give the pair-breaking self-energy corrections to the bare Matsubara frequency, \(i\omega_n\), and the bare superconducting gap, \(\Delta_k\), are as follows and can be found in appendix A of reference \cite{29}:

\[
i\tilde{\omega}_n = i\omega_n + \frac{\Sigma_1 i\tilde{\omega}_n}{\sqrt{\tilde{\omega}_n^2 + \tilde{\Delta}_k^2}} + \Sigma_2 uf_2(u), \tag{5.27}\]

\[
\tilde{\Delta}_k = \Delta_k + \frac{\Sigma_1 \tilde{\Delta}_k}{\sqrt{\tilde{\omega}_n^2 + \tilde{\Delta}_k^2}} + \Sigma_2 uf_2(u). \tag{5.28}\]

Here, \(i\tilde{\omega}_n\) and \(\tilde{\Delta}_k\) are the renormalized Matsubara frequency and superconducting gap respectively. \(\Sigma_1\) controls the magnitude of scattering in regions outside of the superconducting gap. Conversely, \(\Sigma_2\) controls the magnitude of scattering in regions inside of the gap. This can be seen by looking at the square root in the denominator of the second term in both of the above self consistent equations. \(i\tilde{\omega}_n\) is a Matsubara frequency and when squared and analytically continued it presents a negative quantity. The square root term will therefore result in an imaginary number when \(\tilde{\omega}_n^2\) is greater than \(\tilde{\Delta}_k^2\). The imaginary part of \(\tilde{\omega}_n\) describes how much scattering is present. Therefore, the second term in the self consistent equation only contributes to scattering, and therefore broadening the
The quantities $u$ and $f_2(u)$ must also be found self consistently. They act to modulate the $\Sigma_2$ term in the equations for $i\tilde{\omega}_n$ and $\tilde{\Delta}_k$ and are given by the following equations.

\begin{align}
    u &= \frac{i\tilde{\omega}_n}{\tilde{\Delta}_k}, \\
    f_2(u) &= \int \frac{d\Omega}{4\pi N(0)} \frac{N_k(0)}{\sqrt{\phi_k^2 - u^2}},
\end{align}

where $N(0)$ is the density of states at the fermi-energy, $N_k(0)$ is the partial density of states at the Fermi energy in the $k$ direction and $\phi_k = \Delta_k/\Delta_0$. The integral is performed over the Fermi surface. The solution for $u$ using all of these equations is

\begin{equation}
    u = \frac{i\tilde{\omega}_n}{\Delta_0 - \Sigma_2 f_2(u)}
\end{equation}

Figure 5.3(a-d) shows data for the four samples UD25, UD32, OPT35, and OD15, respectively. The upper panel in each frame (a-d) is the model QPI with pair breaking self-energy included. The lower panel does not have pair breaking corrections. It is most apparent in figure 5.3(b) and (c) that pair breaking pulls weight into the gap causing the antinodal fermi-surface lines to appear. As shown by the black dashed lines overlaying the QPI in figure 5.3 the Fermi surface lines show a good fit to the experimental data.
Figure 5.3: Samples UD25, UD32, OPT 35, and OD15, respectively along with the pair breaking self energy corrected tight-binding model. Each image is divided into three sections. Upper left: model QPI with pair breaking corrections with $\Sigma_1 = 20\,\text{meV}$, $\Sigma_2 = 3\,\text{meV}$ and $\omega = 5\,\text{meV}$ for all samples. Lower left: model QPI with no pair breaking corrections. Right: experimental QPI data.
QPGW Self-Energy and Realistic T-matrix Calculations

To go beyond the simple models presented above the QPGW self-energy is applied to the QPI calculations. The QPGW self-energy has the effect of pushing spectral weight into the superconducting gap through both the band renormalization (real part of the self-energy) and broadening (imaginary part of the self-energy). This differs from the Dynes self-energy which has only an imaginary part, and from the pair-breaking self-energy which has a smaller effect overall. One result of the QPGW self energy is that spectral weight is pushed into the superconducting gap, effectively filling in the gap. This causes the gap in the DOS to appear much smaller than is seen in experiment. This is due to the large broadening at energies lower than the superconducting gap. The gap filling cannot be a consequence of the real part of the self-energy. This is because when self-consisting the QPGW renormalization constant, $Z$, the low energy self-energy corrected spectral weight function (coherent bands) must match the $Z$ renormalized bare bands which must also match the experimental bands. Therefore, the coherent bands, before broadening, must match the experimental gap. This difficulty implies that, to resolve low energy effects, it may be necessary to self consist $\Delta_0$ along with $Z$ in a QPGW calculation with superconductivity.

In section 5.6 the T-matrix for scalar scattering was used. The calculations can also be expanded to include Andreev and second order magnetic scattering. The T-matrices for the three scatterers studied are listed in table 5.1. When the T-matrix is applied to the QPI calculations, scalar scatterers reduce the QPI weight along the antinodal direction.
whereas Andreev scatterers reduce the QPI weight along the nodal direction. The second order magnetic scattering T-matrix, when acting on a system with degenerate spin up and down states such that the system is paramagnetic, does not show such sharp regions of QPI weight reduction. Figure 5.4(a) summarizes the results of these calculations for tight binding parameters consistent with the OPT35K sample and figure 5.4(b) shows the experimental result [25] that the model hopes to explain.

Figure 5.4: (a) QPI images using odd fluctuations to simulate experimental Z-maps for OPT35K. Columns represent the type of self-energy used in the calculations and rows represent the type of T-matrix scatterer used. (b) Experimental data for OPT35 sample from reference [25]. (c) Calculation using Wannier orbitals from the paper by A. Kreisel et al. [116].
Discussion

In the UD25K sample, the octet model of QPI matches well with the measured QPI images. This is because the material is in the superconducting state and has not yet transitioned to having a large Fermi surface such that the qualitative picture of the octet model is still observed. In this case, the self-energy corrections are inconsequential and the main features are captured as in figure 5.2(a). At higher doping, when the system transitions to having a large Fermi surface, the antinodal triplet QPI appears in the QPI measurements. Referring to figures 5.2(b-d), the self-energy pushes weight into the antinodal region, giving rise to part of the antinodal triplet QPI which follows the shape of the Fermi surface. However, the relative intensity of the features in the modeled QPI are incorrect. Experimental results show that the antinodal triplet QPI is brighter than states between the node and antinode where intensity seems to fade. Conversely, our model shows very weak intensity in the antinodal region due to these states being populated by the slight broadening effect of the self energy. The most striking missing factor in the self energy corrected models for QPI is the part of the antinodal triplet QPI which is directly on the antinode, \((0,2\pi)\). This state is absent in scalar, Andreev, and magnetic scattering models and is not recovered by any form of self-energy used here.

The failure of these models to describe the experimental results suggests that a more sophisticated understanding of the electron wave functions is necessary. As it turns out, this has been done by A. Kreisel \textit{et al.} \cite{116}. Their approach was to expand on the method of Nieminen \textit{et al.} \cite{7}, who model real space STM images by using atomic orbitals, by using...
first principles DFT methods to calculate more realistic Wannier functions. The Wannier functions act as matrix elements in the real space Green’s function. A Fourier transformation of the spectral function, obtained from the Green’s function, reproduces the QPI structure seen in reference [26]. An image of their results can be seen in figure 5.4(c). This result indicates the importance of the band character on STM calculations. The models presented in this chapter, though sophisticated in terms of self-energy and T-matrix scattering, only accounted for the copper-d electrons whereas Wannier functions can be used to generate a simple one band tight binding model but the Wannier functions themselves still contain information about band character from all of the atoms in the unit cell.
Chapter 6

Summary, Extensions, and Outlook

Summary

This thesis has presented theoretical methods and results for going beyond first principles to understand correlated electron effects. The tight binding Hubbard model is the basis for these studies. It was shown that using a multiband Hubbard model, fit to experimental bands, puts the cuprates into the charge transfer insulator regime. Another study of the one, three, and four band Hubbard models showed that with a doping dependent Hubbard U these models of the cuprates can exhibit negative electronic compressibility. Fermi-surface free superconductivity is another unusual property which was predicted by studying the effects of $(\pi, \pi)$ antiferromagnetic order in competition with BCS superconducting order which drives the topological transition for hole doped cuprates.
The studies just mentioned all relied solely on the Hubbard model and exhibit the versatility of this model in describing the cuprates. In order to understand the electronic structure away from the Fermi surface, self energy corrections are required. Specifically, the QPGW model was used, which is an intermediate coupling model, and is the key to going beyond first principles for theoretical studies of the cuprates. QPGW and other self energies were shown to be useful in realistic modeling of QPI experiments. Also, appendix A shows how QPGW is derived and how including the correct terms for temperature dependent calculations may prove insightful to extending mode coupling calculations and understanding the pseudogap in cuprates.

Extensions

At the time of writing this document there are plans to extend the work presented here in order to explain the cuprate pseudogap and investigate materials beyond the cuprates. This section will highlight the current state of the work towards understanding these two topics.

The field of cuprates has a long history with many well established models, theories, and ideas. Many materials in the family of cuprates have been studied. What the different materials have in common is their phase diagram. An extension into categorizing types of cuprates comes in the form of analyzing, not the phase diagram, but the instabilities of the system. These instabilities can be derived from the susceptibility and categorized by the momentum transfer, $q$, of the electron-electron interaction. The key feature in an analysis like this is which state with momentum $q$ goes unstable first with decreasing temperature.
When these unstable states are plotted as a function of temperature and doping, patterns of instability orders emerge. These plots called fluctuation maps appear different between materials. However, parameterizing materials with a tight binding model allows cuprates to be grouped into reference families which share similar fluctuation maps. This concept of fluctuation maps and analyzing the system stability can become a useful new tool in studying the cuprates. This technique is described in full in reference [30]. When mode coupling corrections are applied to the Hubbard U, as discussed in Appendix A, and a temperature dependent study of QPGW self energy corrections are performed on a one band tight binding model, a pseudogap is formed around the Fermi level due entirely to the bosonic term of the self energy corrections. In preliminary calculations the spectral weight, corrected by the fully momentum dependent QPGW self energy, can show a momentum dependent gap which is stronger near \( q = (\pi, 0) \) and weaker along the diagonal from \( q = \Gamma \) to \( q = (\pi, \pi) \). The DOS in these calculations confirms the observation of a gap around the Fermi level by showing a significant drop in the DOS at the Fermi level. As the temperature is lowered from high temperatures, the Fermi surface transitions from a full Fermi surface to one with Fermi arcs near \( q = (\pi/2, \pi/2) \). This observation, in conjunction with analyzing the fluctuations of a system through the stability of the susceptibility, can lead to a prediction of the origin of the pseudogap in cuprates. \( T_{VHS} \) seems to signify the temperature scale associated with ordered states. The VHS represents a momentum space clustering of electronic states at a particular energy which can be associated with a temperature. \( T_{VHS} \) is defined by the difference in energy between the Fermi surface and the VHS divided by Boltzmann’s constant. In the
equation for the bare susceptibility, equation A.19, the difference of Fermi functions in the numerator represents an energy window which broadens with increasing temperature. The peak will not fully encompass states associated with the VHS until the temperature $T_{VHS}$ is reached. Because the VHS represents the energy with the highest number of states, bosonic modes associated with the VHS will dominate the susceptibility at and above $T_{VHS}$. Fluctuations driven by the electronic states at the VHS may result in the momentum dependent self energy, which renormalize the Fermi surface, creating Fermi arcs. This would imply that it is the VHS that defines the temperature where momentum dependent correlations turn on and cause Fermi arcs and the pseudogap.

An important condition for this result is that it is only achievable with a fully momentum dependent self energy. The momentum dependence of the self energy acts as a Fermi surface renormalization while the frequency dependence creates the gap at high temperature. If the self energy was assumed to be constant in momentum space then the Fermi surface would not be renormalized because there would be no mechanism for it to change shape. These initial observations give a promising start towards future research. Work is needed on this topic in order to correctly describe Fermi arcs and pseudogap physics.

The extension of QPGW to systems containing multiple orbitals and multiple atomic sites is important in order to study correlated materials beyond the cuprates. For example, pnictides are a popular material which fall into the class of high temperature superconductors. Unlike the cuprates, pnictides typically have five bands which cross the Fermi energy and can present nematic ordered states. Other systems such as iridates include
not only multiple bands, but also exhibit spin orbit coupling. Unlike the pnictides, iridates have not been shown to exhibit unconventional superconductivity. The formalism for the multiband susceptibility, which is an important piece of a multiband formalism for QPGW, is carried out in appendix B.

The study of systems with both correlation effects and spin orbit coupling is an emerging field of research which may have a profound impact on the understanding of the intermediate regime between Mott insulators and topological insulators. A review paper by Witczak-Krempa, Chen, Kim, and Balents [117] states that materials such as Weyl semimetals and Topological Mott insulators exist in this correlated spin orbit coupling regime. The iridates are materials which exist in this regime and because of this, interest in exploring their phase diagram has emerged. Doping iridates is still quite difficult experimentally, though doing so may produce properties such as a pseudogap and superconductivity. Furthermore, iridates might be able to produce quantum criticality in a strongly spin orbit coupled material (which would be unique in itself) and result in a superconducting topological insulator. A multiband QPGW model could be used to describe the competition between correlations and spin orbit coupling in such a material through self consisting the $Z$ parameter.

**Outlook**

In the future, this work will take the direction of momentum dependent self energy calculations and calculations on multiband systems. The momentum dependent self energies
have the unique ability to renormalize the Fermi surface which, as mentioned, can contribute to understanding the origin of the pseudogap in cuprates and potentially other unexplained aspects of electronic structure. The ability to use this technique on multi-band systems will greatly expand how correlated electron systems are understood by providing insight into when, how, and why intermediate coupling type physics appears in a variety of materials.
Bibliography


weight in doped strongly correlated systems. Physical Review Letters, 67(8):1035–


with special emphasis on pseudogaps and striped phases. Journal of Physics and
doi.org/10.1016/S0022-3697(97)00025-5. URL http://www.sciencedirect.com/
science/article/pii/S0022369797000255.


[84] Hsin Lin, S. Sahrkorpi, R. S. Markiewicz, and A. Bansil. Raising Bi-O Bands above

of electronic states in structurally disordered metals: Application to liquid Cu.

metallic glasses. Solid State Communications, 55(3):223 – 226, 1985. ISSN 0038-
sciencedirect.com/science/article/pii/0038109885907203.

[87] W. D. Wise, Kamalesh Chatterjee, M. C. Boyer, Takeshi Kondo, T. Takeuchi,
H. Ikuta, Zhijun Xu, Jinsheng Wen, G. D. Gu, Yayu Wang, and E. W. Hudson.
Imaging nanoscale Fermi-surface variations in an inhomogeneous superconductor.
Nature Physics, 5(3):213–216, March 2009. ISSN 1745-2473. doi:
10.1038/nphys1197. URL http://www.nature.com/nphys/journal/v5/n3/abs/
nphys1197.html.

[88] Tao Wu, Hadrien Mayaffre, Steffen Krämer, Mladen Horvatić, Claude Berthier,
W. N. Hardy, Ruixing Liang, D. A. Bonn, and Marc-Henri Julien. Magnetic-field-
induced charge-stripe order in the high-temperature superconductor YBa$_2$Cu$_3$O$_y$.


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Appendix A

Momentum Dependent QPGW

Introduction

The self energy of an electron in a condensed matter system describes how the dynamic properties of the electron are affected by electron-electron correlations. This effect becomes important in a system which has significant electron-electron correlations. When modeling correlated systems one must go beyond standard band theory and include self energy in order to describe many features seen in photoemission experiments [118]. Computationally, self energies are difficult to calculate. A full GW [10] type calculation quickly becomes too computationally intensive because of the iterative nature of GW. To improve computational speed often only the first iteration of the GW method is used. However, this can lead to self energies that are too strong or fail to model the system being studied. For modeling the electronic structure of the cuprates a new approximation to the GW method has been developed known as the quasi-particle GW (QPGW) model [12]. This model reduces the number of iterations necessary to get good self energies by approximating the vertex corrections as a constant renormalization.
The only essential parameters involved in the QPGW scheme are the material specific tight-binding band parameters and the value of the Hubbard $U$ at half-filling. Furthermore, the tight-binding band parameters can be fixed to reproduce the DFT band dispersions. The doping dependent electronic structure can then be determined via a self-consistently computed self-energy correction. A key feature of the scheme is that the low energy 'uncorrelated' DFT based states near the Fermi energy are renormalized and broadened due to charge and magnetic fluctuations in the presence of AFM and SC orders, in addition to the appearance of high energy features in the electronic spectrum, which reflect remnants of Mott physics. Notably, the theory predicts the value of the low-energy dispersion renormalization factor, $Z$, in a doping dependent manner. The renormalization factor $Z$ is determined self consistently within the QPGW scheme.

In this appendix the QPGW method will be derived and discussed. How QPGW can fit into a mode coupling formalism will also be shown as well as the importance of the intrinsic momentum dependence of the QPGW self energy in investigating pseudogap physics.

**Single Band QPGW**

The QPGW method can be most simply understood when applied to a single band model. In this case there will be no interactions between bands to consider. In systems like the cuprates, the electronic behavior is dominated by features close to the Fermi level. In the paramagnetic state of cuprates only one band crosses the Fermi level. This, along with being a correlated electron system, makes cuprates a perfect candidate for study with a one band QPGW method.

QPGW is rooted in the fundamentals of Landau-Fermi liquid theory (also known as Fermi liquid theory). Fermi liquid theory aims to understand an interacting system of fermions by first considering the non-interacting system and slowly turning on the interactions.
This adiabatic inclusion of the interactions evolves the ground state of the non-interacting system smoothly into the ground state of the interacting system. The particles in the interacting system are called quasiparticles. The quasiparticles maintain the spin, charge, and momentum of their non-interacting counterparts. However, dynamical properties, such as mass, become renormalized. This renormalization factor, $Z$, broadens the spectral weight associated with the quasiparticles, resulting in a finite lifetime. This implies that $Z$ can be expressed in terms of a self energy.

Relating self energy and $Z$ requires the self energy corrected Green’s function,

$$ G = G_0 + G_0 \Sigma G, $$

and Dyson’s equation for the Green’s function,

$$ G^{-1} = G_0^{-1} - \Sigma, $$

where $G_0$ is the bare Green’s function and $\Sigma$ is the self energy. Using the band dispersion, $E_k$, $G$, can be written in terms of the Matsubara frequency, $i\omega_n$, and momentum, $k$, as

$$ G(k, i\omega_n) = \frac{1}{i\omega_n - E_k - \Sigma(k, i\omega_n)}. $$

The retarded Green’s function is obtained by analytical continuation of $i\omega_n$ in equation A.3 resulting in

$$ G_{ret}(k, \omega) = \frac{1}{\omega + i\delta - E_k - \Sigma_{ret}(k, \omega)}. $$

The self energy is a complex function and can therefore be represented as $\Sigma_{ret}(k, \omega) = \Sigma'_{ret}(k, \omega) + i\Sigma''_{ret}(k, \omega)$. The spectral function is defined for the retarded Green’s function
as \( A(k, \omega) = -2Im \left[ G_{ret}(k, \omega) \right] \) which has the form,

\[
A(k, \omega) = \frac{-2 \Sigma''_{ret}(k, \omega)}{\left[ \omega - E_k - \Sigma'_{ret}(k, \omega) \right]^2 + \left[ \Sigma''_{ret}(k, \omega) \right]^2}, \tag{A.5}
\]

where \( \delta \) has been absorbed into \( \Sigma''_{ret}(k, \omega) \). The spectral function given in equation A.5 is a Lorentzian with width given by the imaginary part of the self energy. In the limit that \( \Sigma''_{ret}(k, \omega) \to 0 \) the spectral function becomes a delta function such that

\[
A(k, \omega) = 2\pi \delta \left( \omega - E_k - \Sigma'_{ret}(k, \omega) \right). \tag{A.6}
\]

The spectral function is non-zero when the argument of the delta function is zero. This condition is satisfied by the solution to the equation

\[
\xi_k - \mu = E_k + \Sigma'_{ret}(k, \xi_k - \mu). \tag{A.7}
\]

Because the argument of the delta function in equation A.6 is a function itself, the property \( \delta \left[ f(x) \right] = \frac{\delta(x-x_0)}{|df(x)/dx|_{x=x_0}} \), where \( f(x) \) is zero at \( x = x_0 \), can be used to simplify the expression for the spectral function. This results in a spectral function written as

\[
A(k, \omega) = \frac{2\pi \delta (\omega - \xi_k + \mu)}{\left| 1 - \frac{\partial}{\partial \omega} \Sigma_{ret}(k, \omega) \right|_{\omega=\xi_k-\mu}}, \tag{A.8}
\]

or more compactly as

\[
A(k, \omega) = 2\pi Z(k) \delta (\omega - \xi_k + \mu), \tag{A.9}
\]
where $Z(k)$ is the renormalization factor which is now seen to be related to the self energy by

$$Z(k) = \left[ 1 - \left. \frac{\partial}{\partial \omega} \Sigma_{\text{ret}}(k, \omega) \right|_{\omega = \xi_k - \mu} \right]^{-1}. \quad (A.10)$$

The relationship between $Z$ and self energy is used as an input for the GW scheme [10].

Using Ward's identity gives an expression for the vertex correction,

$$\Gamma(k) = \left| 1 - \left. \frac{\partial}{\partial \omega} \Sigma_{\text{ret}}(k, \omega) \right|_{\omega = \xi_k - \mu} \right|, \quad (A.11)$$

which implies,

$$\Gamma(k) = \frac{1}{Z(k)}. \quad (A.12)$$

This is the approximation which is fundamental to QPGW. It says that the vertex correction is a quantity which can be self consisted to determine the self energy within the GW scheme.

The GW scheme is used to evaluate the following diagram

$$W(q, i\xi_m)$$

$$G(k, i\omega_n) \quad G(k - q, i\omega_n - i\xi_m) \quad G(k, i\omega_n) \quad $$

Where $G(k, i\omega_n)$ is the electron propagator and $W(q, i\xi_m)$ is the dressed interaction. This
is known as a sunset diagram which describes how a fermion is dressed upon emitting and reabsorbing a bosonic interaction particle. The region of the diagram between the incoming and outgoing electron is called the self energy and can be evaluated by

$$\Sigma(k, i\omega_n) = -\frac{1}{\beta} \int \frac{d^3 q}{(2\pi)^3} \sum_{i\xi_m} G(k - q, i\omega_n - i\xi_m) \Gamma(k) W(q, i\xi_m)$$  \hspace{1cm} (A.14)

Where $\beta$ is the inverse temperature times the Boltzmann constant or $\beta = 1/k_B T$. The QPGW approximations, laid out above, can now be used to simplify equation A.14.

Solving for $\Sigma_{ret}$ in equation A.10 gives

$$\Sigma_{ret}(k, \omega) = \left[1 - Z(k)\right] \omega. \hspace{1cm} (A.15)$$

Because this will be used as the input self energy in the self consistent scheme, it is assumed that self energy has the same slope with respect to $\omega$ at the Fermi level as it does everywhere else. This approximation will be corrected by solving equation A.14.

The region in $\omega$ near the Fermi level where this approximation holds is known as the coherent region of the spectrum. Given this approximation it is plausible that the self energy before analytic continuation is given by

$$\Sigma(k, i\omega_n) = \left[1 - Z(k)\right] i\omega_n. \hspace{1cm} (A.16)$$

By applying this equation to equation A.3, an expression for the renormalized Green’s function is obtained. The Green’s function now has the form

$$G_Z(k, i\omega_n) = \frac{Z(k)}{i\omega_n - Z(k) E_k} \hspace{1cm} (A.17)$$

and will be used in evaluating equation A.14. This also shows that, in the QPGW approximation, $\xi_k - \mu$ from equation A.7 becomes the renormalized bands $Z(k) E_k$. 133
The next task is to determine the dressed interaction, $W(q, i\xi_m)$. The dressed interaction is given by an infinite series of polarization bubble diagrams which screen the bare interaction, $V(q)$. The equation resulting from these diagrams is given by

$$W(q, i\xi_m) = V(q) + V(q) \chi^{RPA}(q, i\xi_m) V(q). \quad (A.18)$$

$\chi^{RPA}(q, i\xi_m)$ represents the susceptibility calculated in the random phase approximation (RPA). In order to find the RPA susceptibility the bare susceptibility must be calculated. For the one band case, the bare susceptibility is given by

$$\chi^{(0)}(q, i\xi_m) = \frac{1}{\beta} \sum_{k,p,n} G(k, ip_n) G(k + q, i\xi_m + ip_n)$$

$$= - \sum_k \frac{f(E_k) - f(E_{k+q})}{i\xi_m + E_k - E_{k+q}} \quad (A.19)$$

where $f(E_k)$ is the Fermi function. There are three types of RPA susceptibility which can be obtained from the bare susceptibility. These are charge, spin longitudinal and spin transverse susceptibility. In the one band case, where there is no spin interaction, the charge RPA susceptibility is given by

$$\chi^{RPA}_{\rho\rho}(q, i\xi_m) = \chi^{(0)}(q, i\xi_m) \frac{1}{1 + U \chi^{(0)}(q, i\xi_m)}. \quad (A.20)$$

Here, it has been assumed that the only interaction in this system is the on-site Hubbard $U$. The spin transverse, $\chi^{RPA}_{+ -}(q, i\xi_m)$, and spin longitudinal, $\chi^{RPA}_{zz}(q, i\xi_m)$, RPA susceptibility are the same in the paramagnetic system and given by

$$\chi^{RPA}_{zz}(q, i\xi_m) = \chi^{RPA}_{+ -}(q, i\xi_m) = \frac{\chi^{(0)}(q, i\xi_m)}{1 - U \chi^{(0)}(q, i\xi_m)}. \quad (A.21)$$

The major difference here is that the denominator, called the Stoner denominator, can
become zero or go negative. When this happens it indicates an instability in the system, meaning that the interaction parameter, \( U \), has become large enough to drive a phase transition to an ordered state.

Just as with the Green’s function, to work within the framework of QPGW, the susceptibility must be renormalized by \( Z \). At this point, it is useful to approximate that \( Z \) is constant in momentum space. The goal of the \( Z \) renormalization factor is to bring the interactions from the weak coupling regime, where the vertex corrections are ignored, to the intermediate coupling regime where the bands get renormalized. Taking \( Z \) to be constant in momentum space is a simple approximation that seems to work well. The renormalization of the susceptibility is done by replacing the Green’s function in equation A.19 with the renormalized Green’s function from equation A.17. This results in a renormalized bare susceptibility

\[
\chi_Z^{(0)}(q, i\xi_m) = \frac{1}{\beta} \sum_{k,p_n} G_Z(k, ip_n) G_Z(k + q, i\xi_m + ip_n) \\
= -\sum_k Z^2 \frac{f(Z E_k) - f(Z E_{k+q})}{i\xi_m + Z E_k - Z E_{k+q}},
\]  

(A.22)

and the renormalized RPA susceptibilities

\[
\chi^{\text{RPA}}_{\rho\rho, Z}(q, i\xi_m) = \frac{\chi_Z^{(0)}(q, i\xi_m)}{1 + U \chi_Z^{(0)}(q, i\xi_m)}
\]  

(A.23)

and

\[
\chi^{\text{RPA}}_{zz, Z}(q, i\xi_m) = \chi^{\text{RPA}}_{+-, Z}(q, i\xi_m) = \frac{\chi_Z^{(0)}(q, i\xi_m)}{1 - U \chi_Z^{(0)}(q, i\xi_m)}
\]  

(A.24)

which can be used to determine the renormalized screened interaction

\[
W_Z(q, i\xi_m) = U + U \chi^{\text{RPA}}_{Z}(q, i\xi_m) U.
\]  

(A.25)
For $W_Z(q, i\xi_m)$, the bare interaction $V(q)$ has been replaced with $U$ because in the Hubbard model the only interaction in this system is on-site Hubbard repulsion. Also, in equation A.25, $\chi^{RPA}_Z(q, i\xi_m)$ is used to denote any of the three possible RPA susceptibilities.

Continuing the task of writing equation A.14 using the QPGW scheme can now be done by using the renormalized Green’s function and screened interaction. Plugging equations A.17 and A.25 into equation A.14 and replacing the vertex with $1/Z$ gives

$$\Sigma(k, i\omega_n) = -\frac{1}{\beta} \frac{1}{Z} \int \frac{d^3q}{(2\pi)^3} \sum_{i\xi_m} G_Z(k - q, i\omega_n - i\xi_m) U$$

$$- \frac{1}{\beta} U^2 Z \int \frac{d^3q}{(2\pi)^3} \sum_{i\xi_m} G_Z(k - q, i\omega_n - i\xi_m) \chi^{RPA}_Z(q, i\xi_m). \quad (A.26)$$

The first term is the self energy due to the bare interaction which is the self energy in the Hartree-Fock approximation denoted by $\Sigma_{HF}(k, i\omega_n)$. The Hartree-Fock term of the self energy only pushes the band structure around and contributes no broadening effects. This term is therefore taken care of by fitting the tight binding bands to experiment or first principles. The interesting physics comes from the second term in equation A.26 which will be called

$$\Sigma_{QPGW}(k, i\omega_n) = -\frac{1}{\beta} \frac{U^2}{Z} \int \frac{d^3q}{(2\pi)^3} \sum_{i\xi_m} G_Z(k - q, i\omega_n - i\xi_m) \chi^{RPA}_Z(q, i\xi_m). \quad (A.27)$$

At this point, further simplifications can be made. Specifically, the frequency sum over imaginary frequencies can be calculated, allowing for analytic continuation, using the spectral representation of $G_Z$ and $\chi^{RPA}_Z$. The spectral representations of these propagators
are

\[ G_Z (k - q, i\omega_n - i\xi_m) = \int_{-\infty}^{\infty} \frac{dx}{2\pi} A_Z (k - q, x) \frac{i\omega_n - i\xi_m - x}{2\pi i\omega_n - i\xi_m - x}, \]

\[ \chi^{RPA}_Z (q, i\xi_m) = \int_{-\infty}^{\infty} \frac{dy}{2\pi} B_Z (q, y) \frac{i\xi_m - y}{2\pi i\xi_m - y}. \]  

(A.28)

Substituting the spectral representations into equation A.27 gives

\[ \Sigma_{QPGW} (k, i\omega_n) = -\frac{1}{\beta} \frac{U^2}{Z} \int \frac{d^3 q}{(2\pi)^3} \sum_{i\xi_m} \int_{-\infty}^{\infty} \frac{dx}{2\pi} A_Z (k - q, x) \int_{-\infty}^{\infty} \frac{dy}{2\pi} B_Z (q, y) \]

\[ = -\frac{1}{\beta} \frac{U^2}{Z} \int \frac{d^3 q}{(2\pi)^3} \int_{-\infty}^{\infty} \frac{dx}{2\pi} A_Z (k - q, x) \int_{-\infty}^{\infty} \frac{dy}{2\pi} B_Z (q, y) \]

\[ \times \sum_{i\xi_m} \frac{1}{i\omega_n - i\xi_m - x} \frac{1}{i\xi_m - y}. \]  

(A.29)

Through standard techniques, the frequency sum over \( i\xi_m \) can be reduced to

\[ -\frac{1}{\beta} \sum_{i\xi_m} \frac{1}{i\omega_n - i\xi_m - x} \frac{1}{i\xi_m - y} = \frac{n_B (y) - n_B (i\omega_n - x)}{i\omega_n - y - x} \]

\[ = \frac{n_B (y) + f (-x)}{i\omega_n - y - x} \]  

(A.30)

where \( n_B (y) \) is the Bose function and using the identity \(-n_B (i\omega_n - x) = \frac{1}{e^{\beta x} - 1} = f (-x)\). This reduces the self energy equation to

\[ \Sigma_{QPGW} (k, i\omega_n) = \frac{U^2}{Z} \int \frac{d^3 q}{(2\pi)^3} \int_{-\infty}^{\infty} \frac{dx}{2\pi} A_Z (k - q, x) \int_{-\infty}^{\infty} \frac{dy}{2\pi} B_Z (q, y) \]

\[ \frac{n_B (y) + f (-x)}{i\omega_n - y - x}. \]  

(A.31)

There are two more simplifications left to be made. First, \( A_Z (k - q, x) \) is the spectral function of the \( Z \) renormalized Green’s function of equation A.17 and is given by

\[ A_Z (k - q, x) = 2\pi Z \delta (x - Z E_{k-q}). \]  

(A.32)
Because $A_Z(k - q, x)$ contains a delta function, it can be used to eliminate the integral over $x$ such that equation A.31 becomes

$$\Sigma_{QGW}(k, i\omega_n) = \frac{U^2}{Z} \int \frac{d^3q}{(2\pi)^3} Z \int_{-\infty}^{\infty} \frac{dy}{2\pi} B_Z(q, y) \frac{n_B(y) + f(-ZE_{k-q})}{i\omega_n - y - ZE_{k-q}}. \quad (A.33)$$

Similarly, $B_Z(k, y)$ can be replaced by

$$B_Z(q, y) = -2 \text{Im} \left[ \chi_{RPA}^Z(q, y) \right] \quad (A.34)$$

resulting in

$$\Sigma_{QGW}(k, i\omega_n) = -\frac{U^2}{Z} \int \frac{d^3q}{(2\pi)^3} Z \int_{-\infty}^{\infty} \frac{dy}{\pi} \text{Im} \left[ \chi_{RPA}^Z(q, y) \right] \frac{n_B(y) + f(-ZE_{k-q})}{i\omega_n - y - ZE_{k-q}}. \quad (A.35)$$

The second simplification is to reduce the integral over $y$ from $\int_{-\infty}^{\infty} \to \int_{0}^{\infty}$. This is done by splitting the $y$ integral into a term where $y \geq 0$ and a term where $y \leq 0$ such that

$$\Sigma_{QGW}(k, i\omega_n) = -\frac{U^2}{Z} \int \frac{d^3q}{(2\pi)^3} Z \left[ \int_{0}^{\infty} \frac{dy}{\pi} \text{Im} \left[ \chi_{RPA}^Z(q, y) \right] \frac{n_B(y) + f(-ZE_{k-q})}{i\omega_n - y - ZE_{k-q}} ight. \\
+ \left. \int_{-\infty}^{0} \frac{dy}{\pi} \text{Im} \left[ \chi_{RPA}^Z(q, -y) \right] \frac{n_B(-y) + f(-ZE_{k-q})}{i\omega_n + y - ZE_{k-q}} \right]. \quad (A.36)$$

Flipping the bounds of the second term and substituting $y \to -y$ gives

$$\Sigma_{QGW}(k, i\omega_n) = -\frac{U^2}{Z} \int \frac{d^3q}{(2\pi)^3} Z \left[ \int_{0}^{\infty} \frac{dy}{\pi} \text{Im} \left[ \chi_{RPA}^Z(q, y) \right] \frac{n_B(y) + f(-ZE_{k-q})}{i\omega_n - y - ZE_{k-q}} \right. \\
+ \left. \int_{0}^{\infty} \frac{dy}{\pi} \text{Im} \left[ \chi_{RPA}^Z(q, -y) \right] \frac{n_B(-y) + f(-ZE_{k-q})}{i\omega_n + y - ZE_{k-q}} \right]. \quad (A.37)$$

In order to recombine the two terms back into a single integral over $y$, which will now run from $0 \to \infty$, the property $\text{Im} \left[ \chi_{RPA}^Z(q, -y) \right] = -\text{Im} \left[ \chi_{RPA}^Z(-q, y) \right]$ is used. Likewise
inversion symmetry gives $\text{Im} \left[ \chi^{\text{RPA}}_Z(q, y) \right] = \text{Im} \left[ \chi^{\text{RPA}}_Z(-q, y) \right]$. Combining these two properties gives $\text{Im} \left[ \chi^{\text{RPA}}_Z(q, y) \right] = -\text{Im} \left[ \chi^{\text{RPA}}_Z(q, -y) \right]$. It is important to note that the inversion symmetry property is an assumption which may not hold for all systems. Nevertheless, if inversion symmetry is assumed the resulting QPGW self energy is

$$
\Sigma_{\text{QPGW}}(k, i\omega_n) = -\frac{U^2}{Z} \int \frac{d^3q}{(2\pi)^3} Z \int_0^\infty dy \frac{1}{\pi} \text{Im} \left[ \chi^{\text{RPA}}_Z(q, y) \right] \left[ \frac{n_B(y) + f(-ZE_{k-q})}{i\omega_n - y - ZE_{k-q}} - \frac{n_B(-y) + f(-ZE_{k-q})}{i\omega_n + y - ZE_{k-q}} \right].
$$

(A.38)

The above form for the QPGW self energy can now be used by self consistently determining $Z$ such that the $Z$-renormalized bands match the QPGW self energy corrected bands in a low energy window around the Fermi energy. This is the same as asking does the input self energy, the self energy in equation A.10, equal the output self energy, equation A.38, near the Fermi energy?

The equation for the QPGW self energy can be massaged further to separate the contributions of bosons and fermions. Using $f(-ZE_{k-q}) = 1 - f(ZE_{k-q})$ and $n_B(-y) = -[1 + n_B(y)]$ gives $n_B(-y) + 1 - f(ZE_{k-q}) = -[n_B(y) + f(ZE_{k-q})]$. With these results, equation A.38 simplifies to

$$
\Sigma_{\text{QPGW}}(k, i\omega_n) = -\frac{U^2}{Z} \int \frac{d^3q}{(2\pi)^3} Z \int_0^\infty dy \frac{1}{\pi} \text{Im} \left[ \chi^{\text{RPA}}_Z(q, y) \right] \left[ \frac{n_B(y) + 1 - f(ZE_{k-q})}{i\omega_n - y - ZE_{k-q}} + \frac{n_B(y) + f(ZE_{k-q})}{i\omega_n + y - ZE_{k-q}} \right].
$$

(A.39)

The quantity in brackets containing terms proportional to Bose and Fermi functions can be separated into the sum of the terms with a Fermi function factor

$$
g_F(k - q, y, i\omega_n) = \frac{1 - f(ZE_{k-q})}{i\omega_n - y - ZE_{k-q}} + \frac{f(ZE_{k-q})}{i\omega_n + y - ZE_{k-q}}.
$$

(A.40)
and the terms with a Bose function factor

\[ g_B (k - q, y, i\omega_n) = n_B (y) \left[ \frac{1}{i\omega_n - y - ZE_{k-q}} + \frac{1}{i\omega_n + y - ZE_{k-q}} \right] \]  

such that the QPGW self energy can be written more compactly as

\[ \Sigma_{QPGW} (k, i\omega_n) = -\frac{U^2}{Z} \int \frac{d^3 q}{(2\pi)^3} Z \int_0^\infty \frac{dy}{\pi} Im \left[ \chi^{RPA}_Z (q, y) \right] \times \left[ g_F (k - q, y, i\omega_n) + g_B (k - q, y, i\omega_n) \right] . \]  

This form shows that there are both boson and fermion contributions to the self energy. This is important because as temperature increases the boson contributions will increase causing an increase in the imaginary part of the self energy at the Fermi energy. The QPGW self energy can be summarized as

\[ \Sigma_{QPGW} (k, i\omega_n) = -\frac{U^2}{Z} \int \frac{d^3 q}{(2\pi)^3} Z \int_0^\infty \frac{dy}{\pi} Im \left[ \chi^{RPA}_Z (q, y) \right] \times \left[ g_F (k - q, y, i\omega_n) + g_B (k - q, y, i\omega_n) \right] , \]  

These are the equations used to perform QPGW calculations. It is important to note that the equations are explicitly momentum dependent and the Bose term \( g_B \) has a strong temperature dependence. These aspects of the theory will be the primary concern.
of the following investigations. Another aspect about the Bose term is that it can be approximated to be zero at low temperatures. In fact, the Bose term has been left out of the review paper on QPGW [12] based on the low temperature assumption.

Mode Coupling and Temperature Dependence

As outlined above, the QPGW formalism uses the RPA susceptibility. This is important because the RPA susceptibility captures the strong spin density wave (SDW) response of the system, when the Hubbard $U$ is large enough, which the bare susceptibility does not. When self energy is applied to the system, the result of the SDW is a large broadening of the spectral weight resulting in nearly vertical dispersions called waterfalls [119, 120, 121].

The state of the system is strongly dependent on the value of $U$. The paramagnetic system becomes unstable when the denominator in equation A.24, also known as the Stoner denominator, becomes zero or negative. The point at which this instability occurs can be described mathematically by the condition that $\chi^{(0)}_Z (\mathbf{q}, i\xi) = 1/U$. The dependence of the instability condition on the momentum vector $\mathbf{q}$ indicates that not all bosonic modes will become unstable at the same value of $U$. This leads to a competition between instabilities where the mode with the lowest free energy will be preferred. RPA treats this competition one mode at a time, in that there is only a single value of $\mathbf{q}$ in equation A.24.

When temperature dependence is involved strong correlation effects make it necessary to describe all of the modes from the beginning. This can be done by extending the RPA with a theory of mode coupling which effectively changes the value of $U$ [30].

In a single band model, the mode coupling susceptibility has the form

$$\chi (\mathbf{q}, i\omega_n) = \frac{\chi_0 (\mathbf{q}, i\omega_n)}{1 + \lambda - U\chi_0 (\mathbf{q}, i\omega_n)}. \quad (A.48)$$

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By factoring out $1 + \lambda = 1/\Gamma_{mc}$ from the denominator, the above equation can be represented as an RPA susceptibility with a renormalized magnitude and Hubbard $U$ as follows:

$$\chi(q, i\omega_n) = \frac{\Gamma_{mc} \chi_0(q, i\omega_n)}{1 - U\Gamma_{mc} \chi_0(q, i\omega_n)}.$$  \hspace{1cm} (A.49)

The renormalized Hubbard $U$ is defined as

$$U_{sp} \equiv \Gamma_{mc} U = \frac{U}{1 + \lambda}. \hspace{1cm} (A.50)$$

Formulating the mode coupling susceptibility as a renormalized RPA susceptibility allows it to be used in the QPGW formalism.

The mode coupling parameter $\lambda$, is given by a Matsubara sum over the mode coupling susceptibility. This results in a self consistent equation which can be simplified using a Hubbard-Stratonovich transformation such that

$$\lambda = \frac{A_0 T}{N} \sum_{q, \omega_n} \chi(q, i\omega_n)$$

$$= A_0 \int d^2 q \left( \frac{a}{2\pi} \right)^2 \int_0^\infty \frac{d\omega}{\pi} \coth \left( \frac{\omega}{2T} \right) \chi''(q, \omega + i\delta)$$

$$\approx \lambda_1 + \lambda_2 T, \hspace{1cm} (A.51)$$

where

$$\lambda_1 = \int d^2 q \left( \frac{a}{2\pi} \right)^2 \int_0^\infty \frac{d\omega}{\pi} \chi''(q, \omega), \hspace{1cm} (A.52)$$

$$\lambda_2 = \int d^2 q \left( \frac{a}{2\pi} \right)^2 \chi'(q, 0). \hspace{1cm} (A.53)$$

Equation A.51 is expanded to linear order in temperature $T$. The zeroth order term, $\lambda_1$,
can be ignored because it contributes a small, non-singular correction to $\lambda$ leaving the expression for $\lambda$ further simplified to $\lambda \approx \lambda_2 T$. With these equations $\lambda$ can be solved self consistently [30]. An example of the results of this mode coupling can be seen in LSCO. Here, LSCO is modeled using a tight binding model with $t = 0.4195\text{eV}$, $t'/t = -0.23$ and $U = 2\text{eV}$ at half filling. Because the goal is to be able to use the QPGW model with an appropriate value of $U_{sp}$ based on the mode coupling corrections, the bare susceptibility in equation A.48 is replaced with the renormalized bare susceptibility from equation A.22. The renormalization constant is taken to be $Z = 0.5$. The table A.1 gives the calculated mode coupling corrected $U_{sp}$s for different temperatures for half filled LSCO.

**Table A.1:** Results of mode coupling calculations done on a tight binding model for LSCO at half filling

<table>
<thead>
<tr>
<th>T(K)</th>
<th>$\lambda$</th>
<th>$U_{sp}(\text{eV})$</th>
<th>$U_{sp}/t$</th>
</tr>
</thead>
<tbody>
<tr>
<td>50</td>
<td>1.1598</td>
<td>0.926</td>
<td>2.207</td>
</tr>
<tr>
<td>100</td>
<td>1.1622</td>
<td>0.925</td>
<td>2.204</td>
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<td>1.1668</td>
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<td>500</td>
<td>1.1716</td>
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<td>1.1097</td>
<td>0.948</td>
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<td>1.0141</td>
<td>0.993</td>
<td>2.367</td>
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<tr>
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<td>1.1739</td>
<td>0.920</td>
<td>2.192</td>
</tr>
<tr>
<td>4000</td>
<td>1.5873</td>
<td>0.773</td>
<td>1.841</td>
</tr>
</tbody>
</table>

**Importance of Momentum Dependence**

The momentum dependence of the Green’s function, susceptibility, and self energy, and the ability to calculate them with high resolution, is important to understanding the phase diagram and the correlation strength of the models and materials being studied. Momentum dependent studies have the unique ability to predict the periodicity (given by the momentum vector $Q$) of the ordered state representing the primary instability of the system, as well as the Fermi surface renormalization. Studying the doping and temperature dependence of these properties along with the mode coupling theory allows for a better understanding of the phase diagram of correlated materials.

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The momentum dependence of the susceptibility is often ignored [122] but has been shown to be important in analyzing stability [30]. In this section, the susceptibility has been treated within the RPA. The system becomes unstable to magnetically ordered phase transitions when the denominator of equation A.21 is zero at zero frequency. This is known as the Stoner instability and is written as

$$U\chi^{(0)}(\mathbf{q}, 0) = 1.$$  \hspace{1cm} (A.54)

The susceptibility in the condition for the Stoner instability is taken at zero frequency, meaning that only elastic scattering is considered and that the susceptibility is entirely real. The momentum dependence of the susceptibility implies that some bosonic modes, given by the susceptibility, can go unstable before others with changing parameters such as $U$, doping and temperature. If the instability happens at a specific value of $Q = q$ then the ordered state will have a periodicity of $Q$ because $Q$ is the transfer momentum associated with the scattering process which creates the unstable bosonic modes.

One can analyze where instabilities will occur by studying the structure of the bare susceptibility. This can be understood in terms of “nesting”. “Nesting” is when the auto-correlation of a function becomes large due to the product of two high magnitude points in a function. If the function is two dimensional, curves of nesting features can be identified, and in three dimensions, one can even conceive of nesting surfaces. In the bare susceptibility at $\xi$ equals zero, Equation A.19 becomes a sum in frequency space over a momentum space auto-correlation of the Green’s function. Because of this, in practice, the susceptibility can be thought of as an auto-correlation, modified by coherence factors, of the Fermi surface.
Conclusion

Preliminary results of using mode coupling techniques on the one band tight binding model show a pseudogap being formed around the Fermi level due entirely to the bosonic term of the self energy corrections. The spectral weight, corrected by the fully momentum dependent QPGW self energy, shows a momentum dependent gap which is stronger near \( q = (\pi, 0) \) and weaker along the diagonal from \( q = \Gamma \) to \( q = (\pi, \pi) \). There is also a significant drop in the DOS at the Fermi level, indicative of pseudogap type behavior. The temperature dependence of this feature show that as the temperature is lowered from high temperatures, the Fermi surface transitions from a full Fermi surface to one with Fermi arcs near \( q = (\pi/2, \pi/2) \). More work is needed on this topic to verify that pseudogap physics is indeed what is being observed but the preliminary calculations show promising results.
Appendix B

Multiband Susceptibility

Bare Susceptibility

In linear response theory, the linear response function describes how a system reacts to some input. The susceptibility is a linear response function which can be calculated in quantum field theory through the expectation value of two operators. Here, a general expression for the susceptibility due to spin and charge fluctuations in a multiband \((\pi, \pi)\) antiferromagnetic system will be derived. To start, the bare susceptibility will be defined.

The bare susceptibility is simply a sum of bubble diagrams with no vertex corrections. The bare susceptibility at scattering momenta \(q\) and \(q'\) and imaginary time \(\tau\) is given by,

\[
\chi_{nm}^{0,\lambda_1,\lambda_2,\lambda_3,\lambda_4}(q, q', \tau) = \frac{1}{N} \langle T_{\tau} S_{\lambda_1,\lambda_2,\lambda_3,\lambda_4}^n(q) S_{\lambda_3,\lambda_4,\lambda_2,\lambda_1}^m(q') \rangle, \tag{B.1}
\]

\[
S_{\lambda_1,\lambda_2,\lambda_3,\lambda_4}^n(\tau) = \frac{1}{2} \sum_k \sum_{\alpha,\beta} \sum_{\alpha',\beta'} c_{k+,\lambda_1,\alpha}(\tau) \sigma_{\alpha\beta}^n c_{k,\lambda_2,\beta}(\tau), \tag{B.2}
\]

where \(S^n\) is the spin operator, \(c_{k,\lambda,\sigma}(c_{k,\lambda,\sigma}^\dagger)\) is the annihilation(creation) operator for an electron of momentum \(k\), in band \(\lambda\), with spin \(\sigma\), and \(\sigma_{\alpha\beta}^n\) is the \(\alpha, \beta\) component of any linear combination of the \(2 \times 2\) Pauli spin matrices indexed by \(n\). \(T_{\tau}\) is the time ordering.
operator. Using the definition of $S^n$ the susceptibility becomes,

$$
\chi_{0,\lambda_1,\lambda_2,\lambda_3,\lambda_4}(q,q',\tau) = \frac{1}{4N} \sum_{k,k'} \sum_{\alpha,\beta,\alpha',\beta'} \sigma_{\alpha \beta} \sigma_{\alpha' \beta'} \left( \langle T_\tau c_{k+q,\lambda_1,\alpha}(\tau) c_{k',-q',\lambda_3,\alpha'}(\tau) c_{k',q,\lambda_2,\beta}(\tau) c_{k-q',\lambda_4,\beta'}(\tau) \rangle \right)
$$

To include $(\pi, \pi)$-AFM order we take the momentum space sums to be only over the reduced Brillouin zone (RBZ), requiring terms outside the RBZ to be written explicitly by adding $Q = (\pi, \pi)$ to their momenta. The susceptibility becomes

$$
\chi_{0,\lambda_1,\lambda_2,\lambda_3,\lambda_4}(q,q',\tau) = \frac{1}{4N} \sum_{k,k'} \sum_{\alpha,\beta,\alpha',\beta'} \sigma_{\alpha \beta} \sigma_{\alpha' \beta'} \left( \langle T_\tau c_{k+Q,\lambda_1,\alpha}(\tau) c_{k',-Q',\lambda_3,\alpha'}(\tau) c_{k',Q,\lambda_2,\beta}(\tau) c_{k-Q',\lambda_4,\beta'}(\tau) \rangle \right), \tag{B.3}
$$

$$
= \frac{1}{4N} \sum_{k,k'} \sum_{\alpha,\beta,\alpha',\beta'} \sigma_{\alpha \beta} \sigma_{\alpha' \beta'} \left[ \langle T_\tau c_{k+q,\lambda_1,\alpha}(\tau) c_{k',-q',\lambda_3,\alpha'}(\tau) c_{k',q,\lambda_2,\beta}(\tau) c_{k-q',\lambda_4,\beta'}(\tau) \rangle \right]
$$

$$
+ \langle T_\tau c_{k+q,\lambda_1,\alpha}(\tau) c_{k',-q',\lambda_3,\alpha'}(\tau) c_{k',Q,\lambda_2,\beta}(\tau) c_{k+Q,\lambda_4,\beta'}(\tau) \rangle
$$

$$
+ \langle T_\tau c_{k+Q,\lambda_1,\alpha}(\tau) c_{k',Q,\lambda_2,\beta}(\tau) c_{k'-q',\lambda_3,\alpha'}(\tau) c_{k,Q,\lambda_4,\beta'}(\tau) \rangle
$$

$$
+ \langle T_\tau c_{k+Q,\lambda_1,\alpha}(\tau) c_{k+Q,\lambda_2,\beta}(\tau) c_{k'-q',\lambda_3,\alpha'}(\tau) c_{k'+Q,\lambda_4,\beta'}(\tau) \rangle, \tag{B.5}
$$

where the sum over the RBZ is denoted by $\sum_{k,k'}$. Using Wick’s theorem the above equation is expressed as creation annihilation pairs. Disconnected diagrams result from pairs with equal time components and are considered to be fluctuations in the vacuum.

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Because the solution will be normalized by the vacuum fluctuations, these disconnected diagrams will divide out in the final solution and can be discarded ahead of time. The resulting susceptibility is

$$
\chi_{0,0,n,n}^{nm}(q,q',\tau) = -\frac{1}{4N} \sum_k \sum_{a,b} \sigma_{a,b}^{n} \sigma_{a,b}^{m} \delta_{a',b'} \times \left[ T_+ c_{k,0,0} (\tau) c_{k+q,0,0}^\dagger (\tau) \right]
$$

(B.6)

The result of the above equation will be zero unless $k' = k + q'$. This is true for all terms because $c_k = c_{k+q}$ meaning that shifting a particle by a momentum value equal to the periodicity of the RBZ, Q, will leave the particle in an identical state. With this simplification the sum over $k'$ can be eliminated by inserting a Kroniker delta such that

$$
\chi_{0,0,n,n}^{nm}(q,q',\tau) = -\frac{1}{4N} \sum_k \sum_{a,b} \sigma_{a,b}^{n} \sigma_{a,b}^{m} \delta_{a',b'} \delta_{k+q'} \times \left[ T_+ c_{k,0,0} (\tau) c_{k+q,0,0}^\dagger (\tau) \right]
$$

(B.7)
To write the susceptibility in terms of Green’s functions, a unitary transformation is applied to the creation and annihilation operators such that

\[ c_{k,\lambda,\sigma}(\tau) = \sum_a v_{k,a}^{\lambda,a} \gamma_{k,a}(\tau) \quad \text{and} \quad c_{k+Q,\lambda,\sigma}(\tau) = \sum_a v_{k+Q,a}^{\lambda,a} \gamma_{k,a}(\tau) \]  

(B.8)

\[ c_{k,\lambda,\sigma}^\dagger(\tau) = \sum_a \left( v_{k,a}^{\lambda,a} \right)^* \gamma_{k,a}^\dagger(\tau) \quad \text{and} \quad c_{k+Q,\lambda,\sigma}^\dagger(\tau) = \sum_a \left( v_{k+Q,a}^{\lambda,a} \right)^* \gamma_{k,a}^\dagger(\tau) \]  

(B.9)

The summed over index contains all possible bands. The symbol \( a \) is a band index. This is because the above unitary transformation diagonalizes the Hamiltonian with now \( a \) being the good quantum number instead of \( \lambda \) and \( \sigma \).

Applying the unitary transformation to the susceptibility gives

\[
\chi_{nm}^{\lambda_0,\lambda_1,\lambda_2,\lambda_3}(q, q', \tau) = -\frac{1}{4N} \sum_k \sum_{\alpha,\beta,\alpha',\beta'} \sigma_{\alpha,\beta}^n \sigma_{\alpha',\beta'}^m \times \left[ \sum_a \left( v_{k,a}^{\lambda_2,a} \right)^* \left( v_{k,a'}^{\lambda_3,a'} \right) \right. \\
\left. \times \left( T_{\tau} \gamma_{k,a}(\tau) \gamma_{k,a}^\dagger(\tau) \right) \sum_b \left( v_{k+Q,a}^{\lambda_1,b} \right)^* \left( v_{k,a}^{\lambda_4,b} \right) \left( T_{\tau} \gamma_{k+Q,b}^\dagger(\tau) \gamma_{k+Q,b}(\tau) \right) \right] \\
+ \sum_a \left( v_{k+Q,a}^{\lambda_2,a} \right)^* \left( v_{k,a}^{\lambda_3,a} \right) \left( T_{\tau} \gamma_{k+Q,a}(\tau) \gamma_{k+Q,a}^\dagger(\tau) \right) \sum_b \left( v_{k+Q,a}^{\lambda_1,b} \right)^* \left( v_{k+Q,a}^{\lambda_4,b} \right) \left( T_{\tau} \gamma_{k+Q,b}^\dagger(\tau) \gamma_{k+Q,b}(\tau) \right) \\
+ \sum_a \left( v_{k+Q,a}^{\lambda_2,a} \right)^* \left( v_{k,a}^{\lambda_3,a} \right) \left( T_{\tau} \gamma_{k+Q,a}(\tau) \gamma_{k+Q,a}^\dagger(\tau) \right) \sum_b \left( v_{k+Q,a}^{\lambda_1,b} \right)^* \left( v_{k+Q,a}^{\lambda_4,b} \right) \left( T_{\tau} \gamma_{k+Q,b}^\dagger(\tau) \gamma_{k+Q,b}(\tau) \right) \\
+ \sum_a \left( v_{k+Q,a}^{\lambda_2,a} \right)^* \left( v_{k,a}^{\lambda_3,a} \right) \left( T_{\tau} \gamma_{k+Q,a}(\tau) \gamma_{k+Q,a}^\dagger(\tau) \right) \sum_b \left( v_{k+Q,a}^{\lambda_1,b} \right)^* \left( v_{k+Q,a}^{\lambda_4,b} \right) \left( T_{\tau} \gamma_{k+Q,b}^\dagger(\tau) \gamma_{k+Q,b}(\tau) \right) \right].
\]  

(B.10)

The term \( \left( T_{\tau} \gamma_{k+q',b}^\dagger(\tau) \gamma_{k+q,b}(\tau) \right) \) will be zero unless \( q' = q \) or \( q' = q + Q \) because \( \gamma_{k,a} = \gamma_{k+Q,a} \).

The diagonalized Green’s function in quasiparticle space is defined as,

\[
\left( T_{\tau} \gamma_{k,a}(\tau) \gamma_{k,a}^\dagger(\tau) \right) \equiv g_{a}(k, \tau) \quad \text{and} \quad \left( T_{\tau} \gamma_{k,a}^\dagger(\tau) \gamma_{k,a}(\tau) \right) \equiv g_{a}(k, -\tau),
\]  

(B.11)
with Fourier transforms

\[
g_a(k, \tau) = \sum_{p_\eta} g_a(k, i p_\eta) e^{-i p_\eta \tau},
\]

\[
g_a(k, i p_\eta) = k_B T \int_0^\tau d \tau g_a(k, \tau) e^{i p_\eta \tau}.
\] (B.12)

The Fourier transformed susceptibility becomes the following with application of the convolution theorem,

\[
\chi_{nm}^{\lambda_0, \lambda_1, \lambda_2, \lambda_3, \lambda_4} (q, q', i \omega_\mu) = -\frac{1}{4 N} \sum_{k} \sum'_{\alpha, \beta, \alpha', \beta'} \sigma^m_{\alpha' \beta'} \sigma^n_{\alpha \beta} \times
\]

\[
\sum_{a, b} \left[ k_B T \int_{p_\eta} g_a(k, i p_\eta) g_b(k + q, i \omega_\mu + i p_\eta) \right],
\] (B.13)

which can be simplified with the known frequency sum,

\[
k_B T \sum_{p_\eta} g_a(k, i p_\eta) g_b(k + q, i \omega_\mu + i p_\eta) = - \frac{\left[ f(E^a_k) - f(E^b_{k+q}) \right]}{i \omega_\mu + E^a_k - E^b_{k+q}} = \chi^{ab}_{00} (k, q, i \omega_\mu).
\] (B.14)
The notation becomes compact by defining the coherence factor,

\[ C^{\alpha,\beta,\alpha',\beta',a,b}_{\lambda_1,\lambda_2,\lambda_3,\lambda_4}(k, q, q') \equiv v_{\lambda_1,\lambda_4}^{\alpha,b} \left( v_{k,\alpha'}^{\alpha',a} \right)^* v_{k+q',\beta'}^{b,\lambda_1} \left( v_{k+q+a}^{b,\lambda_1} \right)^* + v_{\lambda_2,\beta}^{\alpha,a} \left( v_{k+Q,\alpha'}^{\alpha',a} \right)^* v_{k+q',\beta}^{b,\lambda_1} \left( v_{k+q+a}^{b,\lambda_1} \right)^* + v_{k+Q,\beta}^{\alpha,a} \left( v_{k+Q,\alpha'}^{\alpha',a} \right)^* v_{k+q',\beta'}^{b,\lambda_1} \left( v_{k+q+a}^{b,\lambda_1} \right)^* + v_{\lambda_2,\beta}^{\alpha,a} \left( v_{k+Q,\alpha'}^{\alpha',a} \right)^* v_{k+q'+Q,\beta'}^{b,\lambda_1} \left( v_{k+q+Q,a}^{b,\lambda_1} \right)^*. \] (B.15)

The resulting susceptibility is

\[ \chi^{nm}_{0,\lambda_1,\lambda_2,\lambda_3,\lambda_4}(q, q', i\omega_\mu) = \]

\[ -\frac{1}{4N} \sum_k \sum_{\alpha,\beta,\alpha',\beta'} \sum_{a,b} C^{\alpha,\beta,\alpha',\beta',a,b}_{\lambda_1,\lambda_2,\lambda_3,\lambda_4}(k, q, q') \chi^{ab}_{00}(k, q, i\omega_\mu). \] (B.16)

The susceptibility is a function of the final momenta of the scattering process \( q \) and \( q' \). Because \( (\pi, \pi) \)-AFM order has been explicitly included, there are two distinct types of scattering. The first is normal scattering where electrons stay within their initial BZ. The second is Umklapp scattering where electrons scatter to a different BZ. Normal scattering is defined by \( q' = q \) and Umklapp scattering is defined by \( q' = q + Q \). Representing the susceptibility as a matrix,

\[ \hat{\chi}^{nm}_{0}(q, i\omega_\mu) \equiv \begin{bmatrix} \chi^{nm}_{0}(q, q, i\omega_\mu) & \chi^{nm}_{0}(q, q + Q, i\omega_\mu) \\ \chi^{nm}_{0}(q + Q, q, i\omega_\mu) & \chi^{nm}_{0}(q + Q, q + Q, i\omega_\mu) \end{bmatrix}, \] (B.17)

captures both normal and Umklapp components, where band indices \( \lambda_i \) have been dropped for readability.
Multi-orbital Interactions

Understanding the electron-electron interactions is important for deriving the RPA susceptibility. Here, the ways electrons can interact will be described by building the interaction term of the real space Hamiltonian for an interacting multi-orbital system.

First the standard intra-orbital Coulomb interaction, also known as the Hubbard interaction, has the form,

$$\frac{U}{2} \sum_{i\mu\sigma} n_{i\mu\sigma} n_{i\mu\bar{\sigma}}, \quad (B.18)$$

where $i, \mu,$ and $\sigma$ are the site, orbital, and spin indices respectively. Next, electrons on different orbitals of the same ion will repel each other, incurring another interaction energy called intra-orbital Hubbard repulsion given by,

$$\frac{U'}{2} \sum_{i\mu\sigma} \left( n_{i\mu\sigma} n_{i\nu\sigma} + n_{i\mu\sigma} n_{i\nu\bar{\sigma}} \right). \quad (B.19)$$

Since electrons are indistinguishable, electrons of the same or different spin can interchange between orbital positions. This is the exchange interaction,

$$\frac{J}{2} \sum_{i\mu\sigma} \left( c_{i\mu\sigma}^\dagger c_{i\mu\sigma}^\dagger c_{i\mu\bar{\sigma}} c_{i\mu\bar{\sigma}} + c_{i\mu\sigma}^\dagger c_{i\mu\sigma} c_{i\mu\bar{\sigma}} c_{i\mu\bar{\sigma}} \right). \quad (B.20)$$

Finally, two electrons of opposite spin on a given orbital can simultaneously jump to another orbital. This is called pair hopping and is given by,

$$\frac{J'}{2} \sum_{i\mu\sigma} c_{i\mu\sigma}^\dagger c_{i\mu\sigma} c_{i\nu\bar{\sigma}} c_{i\nu\sigma}. \quad (B.21)$$
These terms can be combined into an interaction Hamiltonian of the form,

$$H_{int} = \frac{U}{2} \sum_{i\mu\sigma} n_{i\mu\sigma} n_{i\mu\bar{\sigma}}$$

$$+ \sum_{i\mu\sigma \atop \mu \neq \nu} \left\{ \frac{U'}{2} n_{i\mu\sigma} n_{i\nu\bar{\sigma}} - \frac{J}{2} c_{i\mu\sigma} c_{i\mu\bar{\sigma}} c_{i\nu\sigma} c_{i\nu\bar{\sigma}} + \frac{U'}{2} n_{i\mu\sigma} n_{i\nu\sigma} + \frac{J'}{2} c_{i\mu\sigma} c_{i\mu\bar{\sigma}} c_{i\nu\sigma} c_{i\nu\bar{\sigma}} \right\}.$$  

(B.22)

The interaction energies can be grouped into an object with both spin (superscript) and orbital (subscript) indices called $g^{\alpha\beta\gamma\delta}_{abcd}$. With this notation, $H_{int}$ can be generalized. It can also be written in momentum space as,

$$H_{int} = \frac{1}{2} \sum_{k_1k_2k_3k_4} \sum_{\mu\nu\mu'\nu'} \sum_{\sigma\bar{\sigma}} g^{\sigma\bar{\sigma}\sigma\bar{\sigma}}_{\mu'\nu'\mu\nu} c_{k_1\mu'\sigma} c_{k_2\mu\sigma} c_{k_3\nu'\bar{\sigma}} c_{k_4\nu\bar{\sigma}}.$$  

(B.23)

By conserving momentum the number of sums in momentum space can be reduced and the resulting interaction is given by,

$$H_{int} = \frac{1}{2} \sum_{kk'q \mu
u\mu'\nu'} \sum_{\sigma\bar{\sigma}} g^{\sigma\bar{\sigma}\sigma\bar{\sigma}}_{\mu'\nu'\mu\nu} c_{k+q\mu'\sigma} c_{k\mu\sigma} c_{k'\nu'\bar{\sigma}} c_{k'\nu\bar{\sigma}}.$$  

(B.24)
The diagrams for the interactions contained in the above equation are as follows,

\[
\begin{align*}
\text{k, } a, \uparrow & \quad \text{k + q, } a, \uparrow \\
\text{k', a, } & \downarrow \quad \text{k' - q, } a, \downarrow \\
\text{k, } b, \uparrow & \quad \text{k + q, } a, \uparrow \\
\text{k', a, } & \downarrow \quad \text{k' - q, } b, \downarrow \\
\text{k, } a, \uparrow & \quad \text{k + q, } a, \uparrow \\
\text{k', a, } & \downarrow \quad \text{k' - q, } a, \downarrow \\
\end{align*}
\]

\[g_{aaaa}^{\uparrow\uparrow\downarrow\downarrow} = U\]
\[g_{abba}^{\uparrow\uparrow\downarrow\downarrow} = J\]
\[g_{aabb}^{\uparrow\uparrow\downarrow\downarrow} = U' - J\]

The spin indices on the g’s can be flipped resulting in equivalent interaction energies.

Equation B.24 can also be written in the spin flip channel where three commutations are performed in order to flip the order of the two \(c^\dagger\) terms. The operation incurs a negative sign which can be absorbed into \(g_{abcd}^{\alpha\beta\sigma\bar{\sigma}}\) such that the interaction becomes,

\[
H_{\text{int}} = \frac{1}{2} \sum_{kk'q_{\mu\mu'}\nu'\sigma\bar{\sigma}} \sum g_{\mu'\mu\nu'\nu}^{\sigma\sigma\bar{\sigma}\bar{\sigma}} c_{k+q_{\mu\mu'}\nu'} c_{k'q_{\mu'\nu}'} c_{-q_{\mu'\nu}} c_{\nu}\bar{\sigma}.
\]
Note that, after the commutations, the momentum indices are redefined such that they are in the same order as in equation B.24. The resulting diagrams are,

\[ g_{aa}^{\uparrow\uparrow\downarrow\downarrow} = -U \]
\[ g_{ba}^{\uparrow\uparrow\downarrow\downarrow} = -J \]
\[ g_{ba}^{\uparrow\uparrow\downarrow\uparrow} = -U' + J \]

(B.27)

Packaging the interactions into \( g_{ab}^{\alpha\beta\gamma\delta} \) makes it easy to form the spin dependent basis, which has been used in this section, in a spin independent basis. When performing RPA calculations this is a necessary step because one must know how the interactions couple to the bare susceptibility. Looking at equation B.3 shows that the bare susceptibility is, in fact, independent of spin. This is done by a unitary transformation using the Pauli spin matrices leaving the susceptibility dependent only on \( n \) and \( m \) which represent the 0, \( x \), \( y \), \( z \) and ± Pauli spin matrices. This transformation will be carried out, in detail, in the next section and applied to the RPA formalism in following sections.
Interactions in the spin channel

It is convenient to write the interactions in a spin independent basis or spin channels. This is done by taking the spin dependent basis, \( g_{\alpha\beta\alpha'\beta'} \), where the Greek letter subscripts indicate either spin up or down, and summing out the spin indicies resulting in the interaction in the spin independent basis, \( v^{nm} \). The transformation has the form

\[
g_{\alpha\beta\alpha'\beta'} = \sigma_n^{\alpha\beta} v^{nm} \sigma_m^{\alpha'\beta'}, \tag{B.28}
\]

where the sum over the \( n \) and \( m \) index is implied. For notational convenience, the orbital indicies have been dropped but must still be considered to exist. This method is outlined in two papers by Aryasetiawan and Biermann [123, 124]. The advantage of this transformation is that when constructing Feynman diagrams one no longer needs to be concerned with conserving spin at a vertex. This is because the spin independent nature of \( v^{nm} \) automatically conserves spin and results in susceptibilities for types of spin order which are uncoupled. These will be called the spin channels and are considered a good basis for these calculations. It should be noted that if one were to introduce spin orbit coupling the spin channels would no longer be uncoupled and a new basis would need to be constructed.
where the subscripts 0 and 1 on the $g$'s indicates either spin up or spin down. The results can be summarized in terms of the multi-orbital interaction energies given in table B.1.

This simple transformation has resulted in spin independent interactions which can couple to the bare multiband susceptibility. The following section will show how these interactions can be used to calculate the RPA susceptibilities for a multiband system.
Table B.1: Orbital components of the interaction in the spin channel. \((a \neq b)\)

<table>
<thead>
<tr>
<th>Component</th>
<th>(aaa)</th>
<th>(aabb)</th>
<th>(abba)</th>
<th>(abab)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(v^{00})</td>
<td>(\frac{U}{2})</td>
<td>(U' - \frac{J}{2})</td>
<td>(J - \frac{U'}{2})</td>
<td>(\frac{J'}{2})</td>
</tr>
<tr>
<td>(v^{zz})</td>
<td>(-\frac{U}{2})</td>
<td>(-\frac{J}{2})</td>
<td>(-\frac{U'}{2})</td>
<td>(-\frac{J'}{2})</td>
</tr>
<tr>
<td>(v^{+-} = v^{-+})</td>
<td>(-\frac{U}{4})</td>
<td>(-\frac{J}{4})</td>
<td>(-\frac{U'}{4})</td>
<td>(-\frac{J'}{4})</td>
</tr>
</tbody>
</table>

Random Phase Approximation Susceptibility

The RPA assumes the electrons only interact with the average of the total electric potential. Therefore, in momentum space, only the wave vector \(k\) contributes to the electric potential. The RPA corrected susceptibility is a sum of all of the connected bubble diagrams. Mathematically, the diagrams can be written as,

\[
\hat{\chi}_{nm}^\text{RPA} = \hat{\chi}_{nm}^0 + \sum_{lh} \hat{\chi}_{nl}^0 v_{lh} \hat{\chi}_{hm}^\text{RPA}.
\]  

(B.31)

This equation can be rearranged to remove the RPA susceptibility from the left hand side resulting in,

\[
\hat{\chi}_{nm}^0 = \sum_{lh} \left( \delta_{nh} - \hat{\chi}_{nl}^0 v_{lh} \right) \hat{\chi}_{hm}^\text{RPA}.
\]  

(B.32)

Introducing the quantity \(\hat{\Lambda}_{0}^{nh} = \sum_{l} \hat{\chi}_{0}^{nl} v^{lh}\) which describes inter and intra orbital interactions simplifies notation and makes equation B.32 become,

\[
\hat{\chi}_{0}^{nm} = \sum_{h} \left( \delta_{nh} - \hat{\Lambda}_{0}^{nh} \right) \hat{\chi}_{hm}^\text{RPA}.
\]  

(B.33)

One now wishes to invert the equation to be in terms of \(\hat{\chi}_{RPA}\). Equation B.33 has the form \(\hat{A} = \hat{B}\hat{C}\) which can be rearranged as \(\hat{B}^{-1}\hat{A} = \hat{C}\). Writing these equations explicitly in terms of their indices give \(\hat{A}_{ik} = \sum_{j} \hat{B}_{ij}\hat{C}_{jk}\) and \(\sum_{i} \hat{B}_{ji}^{-1} \hat{A}_{ik} = \hat{C}_{jk}\). Therefore
equation B.33 becomes

\[ \hat{\chi}_{RPA}^{hm} = \sum_n \left( \delta_{hn} - \hat{A}_n^{hn} \right)^{-1} \hat{\chi}_{0}^{nm} \]  

(B.34)

Finally, for readability, the indices can be relabeled by switching \( n \) and \( h \).

\[ \hat{\chi}_{RPA}^{nm} = \sum_h \left( \delta_{nh} - \hat{A}_h^{nh} \right)^{-1} \hat{\chi}_{0}^{hm} \]  

(B.35)

and the problem is reduced to determining \( \hat{A}_0 \) for each susceptibility.

\[ \hat{A}_0 = \begin{bmatrix}
\hat{\chi}_{0}^{00} v^{00} & \hat{\chi}_{0}^{0z} v^{zz} & \hat{\chi}_{0}^{0-} v^{-+} & \hat{\chi}_{0}^{0+} v^{+-} \\
\hat{\chi}_{0}^{z0} v^{00} & \hat{\chi}_{0}^{zz} v^{zz} & \hat{\chi}_{0}^{z-} v^{+-} & \hat{\chi}_{0}^{z+} v^{+-} \\
\hat{\chi}_{0}^{-0} v^{00} & \hat{\chi}_{0}^{-z} v^{zz} & \hat{\chi}_{0}^{-+} v^{+-} & \hat{\chi}_{0}^{++} v^{+-} \\
\hat{\chi}_{0}^{+0} v^{00} & \hat{\chi}_{0}^{+z} v^{zz} & \hat{\chi}_{0}^{+-} v^{+-} & \hat{\chi}_{0}^{--} v^{+-}
\end{bmatrix} \]  

(B.36)

At this point, everything needed to computationally calculate multiband susceptibilities has been derived. It is important to emphasize a few aspects of this derivation which should be kept in mind when considering the systems to apply this formalism to. The momentum vector \( Q \) of the ordered system can be treated as an index just like the orbital indices because it does not couple to the interaction. This is why \( Q \) and the orbital indices have been left out, for convenience, of the RPA derivation. The orbital indices become important in the RPA when spin orbit coupling is present. In a spin orbit coupled system the spin, \( S \), and orbital angular momentum, \( L \), combine such that the total angular momentum, \( J \), is now the good quantum number for the system. The transformation of the interactions to the spin independent channel in section B.3 will no longer use the Pauli spin matrices but the equivalent matrices associated with \( J \). This should be a straightforward extension to the formalism laid out here.
The remaining section will focus on using symmetries of the system to reduce the number of interaction parameters necessary to describe the system.

**Constraining Interactions**

In this section, physical properties of a condensed matter system will be used to reduce the number of free interaction parameters from four to two. Many of the arguments made in this section were given by A.M. Oleś in reference [125].

The field operator is given by

\[
\psi_{\sigma}(x) = \sum_{i\mu} \psi_{i\mu}(x) c_{i\mu\sigma},
\]  

(B.37)

where \(x\) is the real space coordinate, \(\psi_{i\mu}(x)\) is the electron wave function, and \(c_{i\mu\sigma}\) is the electron annihilation operator. The interaction can be written in terms of the field operator as

\[
H_{\text{int}} = \frac{1}{2} \sum_{\sigma\bar{\sigma}} \int d^3 x \int d^3 \bar{x} \psi_{\sigma}^\dagger(x) \psi_{\bar{\sigma}}^\dagger(\bar{x}) V(x - \bar{x}) \psi_{\sigma}(\bar{x}) \psi_{\bar{\sigma}}(x),
\]  

(B.38)

Where \(V(x)\) is the spatially dependent potential of the system. By comparing equation B.22 to equation B.38 one can write down the equations for \(U, U', J, \) and \(\bar{J} \)' in terms of the wave functions as

\[
U = \int d^3 x \int d^3 \bar{x} |\psi_{i\mu}(x)|^2 V(x - \bar{x}) |\psi_{i\mu}(\bar{x})|^2,
\]  

(B.39)

\[
U' = \int d^3 x \int d^3 \bar{x} |\psi_{i\mu}(x)|^2 V(x - \bar{x}) |\psi_{i\nu}(\bar{x})|^2,
\]  

(B.40)

\[
J = \int d^3 x \int d^3 \bar{x} \psi_{i\mu}^\dagger(x) \psi_{i\mu}(\bar{x}) V(x - \bar{x}) \psi_{i\nu}^\dagger(\bar{x}) \psi_{i\nu}(x),
\]  

(B.41)

\[
\bar{J}' = \int d^3 x \int d^3 \bar{x} \psi_{i\mu}^\dagger(x) \psi_{i\mu}(\bar{x}) V(x - \bar{x}) \psi_{i\nu}(\bar{x}) \psi_{i\nu}(x).
\]  

(B.42)
If the wave function is real, as is the case for d-orbitals, then $\psi_{i\mu}(\vec{x}) = \psi_{i\mu}^\dagger(\vec{x})$ and therefore $J = J'$.

In a system with orbital rotational symmetry one more constraint on the interaction parameters can be imposed. So far, it has been assumed that the onsite interaction $U$ is unique. However, if onsite intra-orbital interactions are considered in a system with orbital rotational symmetry, $U$ can be related to $U'$ and $J$. This is done by eliminating the condition that $\mu \neq \nu$ in equation B.22 resulting in

$$H_{\text{int}} = \sum_{i\mu\sigma} \left\{ \frac{U'}{2} n_{i\mu\sigma} n_{i\nu\bar{\sigma}} - \frac{J'}{2} c_{i\mu\sigma}^\dagger c_{i\mu\sigma} c_{i\nu\sigma} c_{i\nu\bar{\sigma}} + \frac{J'}{2} c_{i\mu\sigma}^\dagger c_{i\mu\sigma} c_{i\nu\bar{\sigma}} c_{i\nu\sigma} \right\} + \sum_{i\mu\sigma} \left( \frac{U' - J}{2} n_{i\mu\sigma} n_{i\nu\bar{\sigma}} - \frac{J'}{2} c_{i\mu\sigma}^\dagger c_{i\mu\sigma} c_{i\nu\sigma} c_{i\nu\bar{\sigma}} \right).$$  

(B.43)

The condition $\mu \neq \nu$ has been left on the $U' - J$ term because electrons of the same spin cannot occupy the same site. Now, the $\mu = \nu$ term can be pulled out explicitly. Doing this and using the identity $J' = J$ gives

$$H_{\text{int}} = \frac{U' + 2J}{2} \sum_{i\mu\sigma} n_{i\mu\sigma} n_{i\nu\bar{\sigma}} + \sum_{i\mu\sigma} \left\{ \frac{U'}{2} n_{i\mu\sigma} n_{i\nu\bar{\sigma}} - \frac{J'}{2} c_{i\mu\sigma}^\dagger c_{i\mu\sigma} c_{i\nu\sigma} c_{i\nu\bar{\sigma}} + \frac{U' - J}{2} n_{i\mu\sigma} n_{i\nu\bar{\sigma}} + \frac{J'}{2} c_{i\mu\sigma}^\dagger c_{i\mu\sigma} c_{i\nu\bar{\sigma}} c_{i\nu\sigma} \right\}.$$  

(B.44)

Here, the parameter $U$ can be written as $U = U' + 2J$ or, more conveniently, as $U' = U - 2J$. Given spin rotational invariance and orbital rotational symmetry, the number of free parameters have been reduce to just $U$ and $J$. It is important to note that the orbital rotational symmetry is broken in systems with crystal field splitting. In that case the degeneracy of the same orbitals on different sites is lifted and one can no longer write the on-site interaction energy in terms of the intra-orbital interaction energy.

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There is however, a potential contradiction in the two assumptions made in order to reduce these parameters. The wave functions of the d-orbitals are real only when they are not degenerate. Crystal field splitting is required in order to break the degeneracy of the d-orbitals in a lattice even though it was assumed that the wave functions are real and there is no crystal field splitting. Despite this contradiction, the reduction of parameters presented here can be used as a first approximation.

Conclusion

This appendix has given the details needed to calculate susceptibilities for a system with multiple bands. Currently, work is being done on extending the QPGW formalism, presented in appendix A, to multiband systems. The multiband susceptibility is of fundamental importance to the extension of the QPGW calculations. This will provide a method for studying momentum dependent self energies of systems in the intermediate coupling regime such as the pnictides and iridates.