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The Momentum Resolved Density of States: Pair-forming and Pair-Breaking in the Cuprate Superconductors

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The Momentum Resolved Density of States: Pair-forming and Pair-Breaking in the Cuprate Superconductors

by

Theodore Reber

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has been approved for the Department of Physics

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Date ________________

The final copy of this thesis has been examined by the signatories, and we find that both the content and the form meet acceptable presentation standards of scholarly work in the above mentioned discipline.
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The Momentum Resolved Density of States: Pair-forming and Pair-Breaking in the Cuprate Superconductors

Thesis directed by Prof. Daniel Dessau

As a direct measure of the electronic interactions in a solid, knowledge of the electronic scattering rates is essential for understanding a material's behavior. Since angle resolved photoemission spectroscopy (ARPES) can probe an individual momentum state, it holds great promise for the most detailed and accurate measurements of the \( k \)-dependent electron scattering rates. Unfortunately, the scattering rates determined from ARPES are typically an order of magnitude greater than those obtained from other probes, (e.g. optical spectroscopy). Here we present a new type of spectrum, the ARPES tunneling spectrum (ATS), which resolves this discrepancy, as well as provides a qualitatively different understanding of the electronic interactions in the cuprates. We find that the electronic states at the Fermi surface are governed by not one but two energy scales, the gapping order parameter, \( \Delta \) and the pair-breaking scattering rate, \( \Gamma \). We find that much of the exotic phenomena found in the cuprates, such as Fermi arcs, pseudogaps and two gap models can trace their origin to the interplay between these two energy scales.
Dedication

For Mel
Acknowledgements

First and foremost I would like to thank my parents. Without their love and constant support, I wouldn’t be where I am today.

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Chapter 1

Introduction

In 1911 in a series of papers, Kammerlingh Onnes reported the complete lack of resistance in mercury at 4.19K\cite{1}. This finding was the first observation of superconductivity. While some theorists had predicted no resistance at absolute zero, zero resistance at finite temperature was unexpected. For this discovery Onnes received the 1913 Nobel prize in physics. As more materials were investigated, the highest critical temperature, $T_C$, (onset of superconductivity) increased with lead (7.65K)\cite{2} in 1913 and by 1941 niobium nitride (16K)\cite{3}.

Describing superconductivity adequately in theoretical models proved intractable for many years. The electro-magnetic properties were initially well described by the London-London equations and full macroscopic phenomenological model by Ginzburg and Landau\cite{4}. But a microscopic theory describing the individual behavior of electrons in a superconductor did not emerge until 1957 with the work of Bardeen, Cooper and Schrieffer\cite{5}.

Bardeen, Cooper and Schrieffer, proposed that since the Coloumb repulsion is near perfectly screened in a solid, the smallest attractive potential could couple two electrons into a pair, known as a Cooper pair. With an integer spin this pair is a boson, such that a collection can collapse into a single macroscopic quantum state. The energy to scatter an electron out of the macroscopic state is so large that simple impurities and thermal excitations are incapable of scattering an electron. Consequently, with no scattering, resistance is zero. Later Gorkov showed that BCS recovers the Ginzburg-Landau equations\cite{6}. For their work Bardeen, Cooper and Schrieffer received the Nobel prize in 1972.
The coupling boson responsible for Cooper pairs is a phonon. The phonon is the quantized limit of a lattice distortion. A common illustration of phonon mediated pairing is shown in figure 1.1. An electron scatters off an ion shifting the electron’s momentum from \( k \) to \( k' \) while the ion receives a kick of \( k' - k \). This distortion forms a net positive charge density that attracts a second electron. This second electron scatters from \(-k\) to \(-k'\) relaxing the distortion. However, while intuitive this picture has several flaws. First, this sort of interaction is a single scattering event, while a true Cooper pair interacts continually. Second the billiard ball picture is a fundamentally incorrect picture of the electrons in a solid, where electrons are better described as extended waves with well defined momentum, \( k \). A better picture is to imagine a circular Fermi surface shown in figure 1.1. An electron scatters off of a phonon from \( k \) to \( k' \). This phonon can relax in only two ways. Either the original electron scatters back to its starting position such that the phonon interaction is included in the self energy or the electron on the either side of the Fermi surface can scatter from \(-k\) to \(-k'\). With multiple phonon interactions, these two electrons dance around the Fermi surface always staying opposite of each other. This strong correlation results in the formation of a Cooper pair that is a boson with a net momentum of zero.

Slowly new materials were found with ever higher temperatures culminating in 1973 with a \( T_C \) of 23K for \( \text{Nb}_3\text{Ge} \) [7] (Fig. 1.2). Further theory placed an upper limit on the critical temperature of 30K. However, in 1986, Bednorz and Mueller reported that the cuprate \( \text{LaBaCuO} \) superconducts at 35K, breaking the BCS threshold [8] [9]. Quickly, more materials in the same cuprate family were found with \( \text{YBCO} \) (90K)[10] breaking the liquid nitrogen threshold (1987) followed by \( \text{BSCCO} \) at 110K [11], culminating in \( \text{Hg-BCCO} \) at 135 K[12].

While the cuprate family has numerous members, they all share some key characteristics:

1. Cuprates are layered compounds with copper oxide planes for which they have been named. Figure 1.3A shows the crystal structure for the three members of the BSCCO group: \( \text{Bi2223} \), \( \text{Bi2212} \) and \( \text{Bi2201} \). The difference between the three members is the number of CuO planes[13].
Figure 1.1: **Comparison of Cartoon Models for Phonon Coupling** A, A typical cartoon for the electron phonon interaction where an initial electron scatters of an ion displacing it from the equilibrium position. To relax an electron of equal an opposite momentum scatters off the ion, thus coupling the two electrons into a Cooper pair. B, A slightly more accurate picture where the electrons are well defined by their momentum on the Fermi surface. Scattering off of a phonon causes a distortion with the momentum $q$ that can only relax with the electron with equal and opposite momentum from the initial electron. The pair can thus circle the Fermi surface with a net momentum of zero.
Figure 1.2: A Short History of Superconductivity Since the discovery of superconductivity, materials with higher critical temperatures were slowly discovered with a average rate of increase of one degree every five years. Then in 1986, the highest measured $T_C$ shot up by 100K in just two years.
(2) The electronic structure is two dimensional. Along the $k_Z$ direction the dispersion is minimal compared to $k_X$ and $k_Y$. The two dimensionality greatly eases ARPES as the spectra are identical at all photon energies[14].

(3) The Fermi surface is a hole pocket centered at the Y-point and X-point as shown in Figure 1.3B. This Fermi surface requires a van Hove singularity at M-point the energy of which varies with doping level[13].

(4) The superconducting gap is d-wave (Figure 1.3 C) in sharp contrast to the conventional superconductors that have an s-wave gap[15, 16]. The adjacent lobes of the d-wave gap have opposite phase, with consequences for the rate of pair-breaking that will be discussed in detail in this thesis. Along the zone diagonals the gap magnitude is zero and is therefore named the node. All angles reported in this thesis are measured relative to the node about the Y-point.

(5) The phase diagrams (temperature vs doping) are very similar in the cuprates (Figure 1.3D). The parent compound (i.e. zero doping) is an anti-ferromagnet. The superconducting region is a dome centered at a significant doping level (.16). The normal state has an extremely unusual doping dependence starting with a traditional Fermi liquid in the heavily overdoped region. However the optimally doped region (maximum of the superconducting dome) is a strange metal with unconventional resistivity[17], that is often referred as a Marginal Fermi Liquid. Finally, the normal state of the underdoped cuprates is extremely odd and has been the subject of intensive study. Although no longer superconducting, this state is still gapped, the famous pseudogap [18]. Whether this pseudogap is a precursor to the superconducting state (i.e. composed of Cooper pairs that have not condensed into single quantum state) or that it is from a competing process like a charge density wave.

Despite these findings, the mechanism for superconductivity in the cuprates remains unknown.

The mystery of high $T_C$ superconductivity has spurred the development and improvement of many experimental techniques to probe different aspects of the cuprates. Inelastic neutron
Figure 1.3: **Cuprate Fundamentals**

A Crystalline structure for the BSCCO family

B Typical Fermi Surface of a cuprate.

C Gap structure of cuprates. Along the diagonals the gap value is zero and referred to as the node (orange). All angles reported in this thesis are measured from the node about the Y point (purple).

D Generic phase diagram for a cuprate.
scattering can directly investigate the collective modes of the systems, lattice or magnetic\cite{19, 20}. Scanning tunneling spectroscopy (STS) measures the electron population on a sub-atom scale providing key insights into impurities and charge densities\cite{21}. Finally, angle resolved photo-emission spectroscopy (ARPES) measures the electron population in momentum space, providing a direct probe of the band structure of the materials\cite{13}.

In Bardeen’s Nobel lecture he cites the discovery by London and others\cite{22} that the superconducting state has a characteristic energy scale representative of an energy gap forming at the Fermi Surface as essential to formulating the BCS theory\cite{23}. In this dissertation, I will show that cuprates possess a second energy scale $\Gamma$, that is imaginary in contrast to the real $\Delta$ and representative of the pair-breaking rate in the superconducting state. This hidden energy scale can help account for many unusual features of the cuprates, such as Fermi arcs, the filling gap, non-Fermi liquid behavior and the pseudogap region.

To reach these conclusions, we invented a new analysis technique for ARPES spectra. By integration along a particular momentum we extract an effective momentum resolved density of states. We can now analyze this curve with established techniques from the tunneling. As these spectra are a combination of ARPES data and tunneling analysis we have named them ARPES tunneling spectra or ATS. The ATS allows us to extract the gap magnitude with unprecedented accuracy and resolve the gap when other techniques (leading edge method or symmetrized EDCs) fail to resolve any gap. Even more importantly, we extract $\Gamma$ from the ATS, whereas traditional ARPES analysis can not separate the pair-breaking rate from other scattering processes.

I will start with a brief overview of the ARPES technique, accompanied with a more detailed description of the effects of detector resolution, detector nonlinearity and surface quality. Chapter 3 will review the theory of electrons in materials to ground the later findings in a theoretical framework. In chapter 4, I will describe a new phenomenological model to describe the doping dependence of the scattering rates in the normal state of the cuprates. Chapter 5 will contain the introduction of the ARPES Tunneling spectra (ATS) and it’s strength and weaknesses over more conventional analysis. Chapter 6 will explore the angular dependence around the Fermi surface and
it’s implications on Fermi Arcs. Chapter 7 will investigate the temperature dependence to explain the filling gap, the pseudogap phase and the determination of activation energy scale. Chapter 8 will discuss the evolution of the ATS with doping and it’s implications for the coupling mode in the cuprates. Chapter 9 briefly addresses the antinodal states particularly how our near-nodal results inform the observations of the nodal-antinodal dichotomy. Chapter 10 will summarize and paint the fundamentally different conception of the cuprates that has been advanced by the ATS.
Chapter 2

Principles of ARPES

2.1 Introduction

As the number of unexplained materials has exploded in recent decades the techniques of experimental condensed matter physics have also grown in number and sophistication. The arsenal of old and new probes now available include neutron inelastic scattering[19], Raman scattering [24], scanning tunneling microscopy[21], optical conductivity[25], De Haas van Alphen[26], and Angle Resolved Photo-emission Spectroscopy (ARPES)[27]. ARPES is the most direct probe of momentum states of electrons. However, due to a lack of resolution, analytical tools, and understanding, ARPES has been relegated to be mostly qualitative in its results. That need not be the case as I will show in this thesis, that ARPES can be quantitative, and provide an unprecedentedly clear view of the process of pairing in the high $T_C$ superconductors. The development and details of ARPES are well documented in multiple theses and reviews [13, 28, 29], so in this chapter I will limit my discussion to a brief overview followed by a more detailed discussion of my contribution proper treatment and analysis of ARPES data. In particular, I will show how numerous experimental artifacts manifest as a low energy step in the widths and should not be construed as a low energy mode.

2.2 Experimental Principles

ARPES is based on the photo-electric effect, for the explanation of which Einstein was awarded the Nobel prize in 1921[30]. In the photo-electric effect, light is shined on a material, and if the energy of the photon is greater than the work function of the material, then an electron
is emitted. The photoemission process can be described by the equation below:

\[ E_f - E_i = h\nu - \Phi \]  

(2.1)

Where \( E_f \) and \( E_i \) are the final and initial energies of the electron, \( \nu \) is the frequency of the light and \( \Phi \) is the work function of the material. That the energy of the emitted electrons scale with frequency of the light rather than intensity was one of the primary evidence that light is actually composed of photons.

The first adaption of the photoelectric effect, to studying solids was photoemission spectroscopy (PES)[13]. PES measures the number of emitted electrons as a function of their final state energy. Provided the work function, and photon energy are known it is simple to use equation 2.1 to extract how many electrons exists at each energy in the solid, i.e. the density of states. Figure 2.1 illustrates this process.

To truly understand a solid, knowing the energy distribution of electrons is not enough, one also needs to know their momentum states. During the photoemission process the momentum states are preserved or rather they are mapped onto angles. While the energy of the photon is larger than the electron’s, the momentum of the photon is negligible compared to the momentum of the electron. Consequently, the momentum state of the excited electrons is preserved during the excitation process. Though the momentum state is preserved the momentum of the electron is changed such that the excited electrons still obey the energy momentum relationship of free particles. If the momentum was not changed then in plane electrons would never be emitted. Instead the momentum state is transformed into an angular state. This process is well understood and described by the following equation:

\[
k_{\parallel} = (\frac{2m}{\hbar}E_{\text{kin}})^{1/2} \sin \theta = (\frac{2m}{\hbar}(E_i - h\nu - \Phi))^{1/2} \sin \theta \]

(2.2)

where \( k_{\parallel} \) is the momentum in the plane of the surface, \( m \) is the mass of the electron and \( \theta \) is the azimuthal angle[28]. Consequently, provided the photon’s energy and the work function are well known then the proper momentum state can be well determined from a given angular state.

Angle resolved photoemission spectroscopy works the same way as PES but resolves the indi-
Figure 2.1: **Principle of Photoemission Spectroscopy** During the photoemission process the density of states of electrons in the solid are preserved as the electrons are emitted to free space.
individual angular state and thus the momentum state of the electrons in the sample. By sampling all
angles the full momentum and energy dependence of the electrons in the sample can be determined.
Figure 2.2 shows a cartoon of the ARPES process as well as sample data.

2.3 Apparatus

An ARPES analyzer is composed of three parts: the lens, the hemisphere, and the detector
as detailed in Fig. 2.3[31]. The lens collects the electrons and maps the angular distribution onto a
position at the slit. The hemisphere disperses the electrons orthogonally to the slit proportional to
the energy of the electrons. Finally the detector amplifies the individual electrons into a detectable
signal and records the energy and angular distribution as an image.

ARPES was originally performed by placing a pinhole over the entrance to a photo-emission
spectrometer, such that only electrons leaving the sample at a particular angle would enter the
analyzer[32]. However with this approach only a single momentum state could be probed at time.
The innovation of the electron lens was a major advance for it allowed a multiplexing of data.
Rather than the previous single state, now 360 angular channels and thus 360 momenta are regularly
measured simultaneously. Furthermore, the electron lens collects a cone of electrons being emitted
from a sample, then through a series of carefully designed and controlled electrodes, the angular
dependence is mapped onto position-space at the slit as shown in figure 2.3B. This collection and
mapping allows for an enhanced signal without the typical lack of resolution for enlarging the
pinhole. The improvement due to the addition of an electron lens is analogous to the improvement
in a photography between a pinhole camera and a modern lens imaging system.

After passing through the slit, the electrons enter the hemisphere which is actually composed
of two concentric hemispheres which the electrons pass through as shown in figure 2.3C. A static
electric potential is applied between the spheres such that the electrons experience a force orthogonal
to their momentum. Consequently, the electrons curve around the inner sphere. Electrons that are
too energetic will not curve enough and impact the outer hemisphere (red contour). If they are not
ergetic enough the radius of curvature will be so great that the electrons hit the inner hemisphere
(purple contour). Only electrons in a narrow energy range can navigate all the way around the
Figure 2.2: **Principle of ARPES** In addition to the energy states, the momentum states are preserved during photoemission but mapped into angular space. The ARPES analyzer resolves this angular distribution allowing the complete mapping of energy and momentum of the electrons (i.e. the band structure) of a solid.
Figure 2.3: Components of ARPES Analyzer

A Schematic of the three components of an ARPES analyzer: The electron lens, the hemispherical energy analyzer and the electron detector.

B The electron lens maps the different angles of the electrons onto a spatial distribution.

C The hemispherical analyzer has electric potential such that the entering electrons experience a force orthogonal to their velocity curving their path. Electrons that are too energetic (red) will not curve enough and be absorbed on the outer hemisphere. Electrons that are not energetic enough (purple) impact the inner hemisphere. The electrons that do make it through the entire hemisphere are now spatially distributed from low (blue) to medium (green) to high (orange) energy but orthogonal the spatial momentum distribution created by the electron lens.

D To amplify individual electrons into a detectable signal, the electrons are first accelerated via a pair of fine copper meshes. Then they are amplified in number with a multi-channel plate before impacting a phosphor causing the phosphor to fluoresce. The resulting glow of the phosphor is recorded by a camera.
hemisphere to the detector. By changing the potential difference, electrons at any specific energy can be selected. At the end of the hemisphere, the electrons pass through a fine mesh at zero potential to prevent fringe fields that can distort the electron trajectories.

To turn a single electron into a detectable signal is not a simple process. After emerging from the hemisphere, the electrons are now resolved in energy parallel to the radius of the spheres and in angle (i.e. crystal momentum) parallel to the circumference of the spheres. A strong DC field (3000 V) accelerates the electrons into the multi-channel plate (MCP). The multi-channel plate is an array of miniaturized photo-multiplier tubes, that transform the single electrons into an avalanche of millions of electrons. These electrons emerge from the MCP and impact a phosphor coating on a window. The electrons excite the phosphor causing it to briefly glow which a camera images and records as a single data point. If enough data is collected then the full $E$ vs $k$ dispersion of electrons is revealed. Currently, the data can be acquired in two distinct modes: gray scale and pulse counting. In gray scale, the camera has a relatively long exposure (100-1000 ms) and the phosphor blips sum over time. However, the phosphor blips can vary in intensity by more than a factor of two, such that the summed brightness is only approximately related to the electron numbers. Pulse counting mode was developed to get around this problem, by taking very short exposures (< 100 ms) so that every phosphor blip was isolated. Then each blip is measured and counted as a single event no matter the brightness. However, if two blips happen to occur close enough to each other that they overlap then those two blips count as a single event. This undercounting can be prevented by reducing the exposure time or the electron flux, but with the exposure time set to a minimum, the flux still needs to be set so low that the data acquisition takes an order of magnitude more time then gray scale mode. Consequently, most data is taken in gray scale.

2.4 Photon Energy Selection

In this thesis, all the data presented was taken with low energy (6-10 eV) photons. Low energy photons have three distinct advantages when studying Bi2212 [33, 28, 29]. First, the scattering length of excited electrons in solids are strongly energy dependent and follow the “universal
curve” (2.4). Since low energy ARPES has a scattering length of several nanometers vs the several angstrom path of the more typical excitation of 20-50 eV, low energy ARPES is much more bulk sensitive and less likely to be contaminated by surface states. Second low energy ARPES spectrum are profoundly sharper allowing much better extraction of pertinent parameters. Finally, the bilayer nature of Bi2212 allows for coupling between the layers. This coupling can either be bonding or anti-bonding each with a slightly different chemical potential. This shift manifests in ARPES spectra as two adjacent bands significantly muddying the analysis. However, the relative intensity of these bands is strongly photon energy dependent. For 7 eV photons the spectra are completely dominated by the anti-bonding band. The simplification of the single band greatly eases the difficulty of analysis so nearly all data presented in this thesis are taken with 7 eV photons.

![Figure 2.4](image)

**Figure 2.4: Advantages of Low Energy ARPES**

A, the mean free path for excited electrons in a solid is an order of magnitude longer for 6-7 eV excitations than for the more typical synchrotron excitations at 20-50 eV. Consequently, Low Energy ARPES probes the bulk states much more efficiently. B, Low Energy ARPES has dramatically improved energy and momentum resolution allowing for unprecedented extraction of the fine details of the electronic structure.

### 2.5 Spectra and Analysis

A typical ARPES spectra is shown in 2.5, in this case a nodal spectra of optimally doped Bi2212 at 10K. There are two conventional approaches to analyzing such data, breaking the spectra into Energy Distribution curves (EDCs) and into Momentum Distribution Curves (MDCs)[34].
The needs of analysis and nature of the spectra heavily influence these complimentary techniques. EDCs are created by taking vertical slices through the spectrum which results of plots of intensity vs energy. EDC peaks are viewed as representative of the dispersion while EDC widths are representative of the scattering rate. In contrast, MDCs are horizontal slices taken at specific energies. But again the MDC peaks are supposed to be representative of the dispersion while the MDC widths are representative of the scattering rate. However, the EDC and MDC peak locations agree only approximately. Sometimes, that is due to forcing a search for a peak where there is none (e.g. below the band bottom for MDCs or well above \( k_F \) for EDCs), but there are also more intrinsic concerns. The self energy is strongly energy dependent while only weakly \( k \)-dependent so it makes sense that MDC widths that are a function of energy are more representative of the self energy than the EDC widths. Furthermore, the strongly energy dependent self energy makes the EDCs have an unusual line-shape (e.g. the famous peak-dip-hump structure) making the FWHM less representative. Finally the observable band always sits on top of a energy dependent background that can fundamentally shift the peak and width measurements of EDCs. However as the background is momentum-independent the MDCs are modified by only a constant offset that is easily accounted for.

While the self energy is most directly determined with MDC analysis, MDCs do not work in every situation. When the band becomes very flat either at the band bottom or when it is bent back due to the formation of the gap the MDC method fails (Fig. 2.6). This failure can be observed in both the dispersion and the width. By fitting the MDCs through the gap we determine a peak where there is no quasiparticle (e.g. inside the gap or below the band bottom) and fail to track the bent back portion of the band, which is most clear in the 12° example. Similarly the MDC width inside the gap is no longer representative of the scattering rate, because no quasiparticles exists with in the gap. Furthermore, the bent back band causes an increase in MDC width that does not correspond to an increase in the scattering rate. These two effects combine to form a characteristic triple peak feature in the widths. The underlying energy dependence of the MDC widths cause the first peak manifest as a flattening followed by a sharp drop (Fig. 2.6C). This sharp drop should not be misconstrued as a low energy bosonic mode. As even the 6° shows a similar deviation in
Figure 2.5: **Approaches to Analyzing an ARPES Spectrum** The ARPES spectrum can be analyzed in two ways. By taking horizontal cuts one extracts momentum distribution curves (MDCs), which for a linear bare band and a momentum independent self energy are perfect Lorentzians (Top Panel). Taking vertical cuts one extracts energy distribution curves (EDCs), whose line shape does not reduce to a simple form because of the strongly energy dependent self energy.
the widths, the node must be determined accurately before analysis of any low energy mode.

While the MDCs are representative of the scattering rate it is important to know that they are not the scattering rate. As Chapter 3 shows in detail, the scattering rate, $\Gamma_{MDC}$ is:

$$\Gamma_{MDC} = \text{Im}(\Sigma) = 2 \frac{\sigma_{MDC}}{v_{BB}}$$

(2.3)

where $\sigma_{MDC}$ is the MDC width and $v_{BB}$ is the bare velocity. While $v_{BB}$ is not expected to change with temperature it is expected to change with angle. To find the bare-band, we perform a Kramers-Kronig transformation of the energy dependence of the MDC widths to determine the real part of the self energy, which we subtract from the dispersion to find the bare-band. The extracted bare-bands for three different angles are shown in Fig. 2.6A. Again inside the gap the bare-band should not be trusted but we find that below 40 meV, the bare bands are linear enough to extract a bare velocity. Using these bare velocities and the measured MDC widths(Fig 2.6C). We determine the angle dependence of the scattering rates, $\Gamma_{MDC}$ (Fig. 2.6D). While the MDC widths show a strong angular dependence, the scattering rates show no such angular dependence.

2.6  Detector Non-linearity and Correction

As the technology of ARPES continues to advance and new discoveries are made, one must be sure to eliminate all experimental artifacts from the data. One well-known but commonly ignored effect is the detector nonlinearity. The nonlinearity of photo-electron detectors used by Scienta, which dominates the ARPES field, was first detected by Fadley et al. during a multi-atom resonant photo-emission spectroscopy (MARPES) experiment [35, 36, 37]. As angle resolved photo-emission spectroscopy (ARPES) is in a fundamentally different regime, such nonlinearity has to our knowledge been ignored. However, we find the effects of the nonlinearity are subtle but pernicious and must be compensated before any analysis beyond the most rudimentary can be trusted. Here, we present the first discussion of the effects of this nonlinearity as well as the first to extract and correct for this nonlinearity.

In this section, I’ll detail the effects of the nonlinearity on a simple spectrum and the method of extraction of the detector nonlinearity. In Fig. 2.7 we show the effect of a linear detector (blue),
Figure 2.6: **Effects on MDC when Spectrum is Gapped**

A Spectra for the node and two angles in the near nodal region with dispersion(red) and the Kramers-Kronig determined bare-band(black).

B Comparison of dispersion for the three locations on the Fermi surface.

C Comparison of MDC widths

D Comparison of Extracted $\Gamma_{MDC}$
Figure 2.7: **Effects of a Nonlinear Detector on Typical ARPES Spectrum**

A. Example detector nonlinearities showing both a smooth (green) and a discrete (red) from a linear response (blue)

B. Spectra before (top) and after (bottom) nonlinearity inclusion

C. Sample MDC widths showing that the nonlinearity is one of the few experimental artifacts that make spectra sharper rather than broader

D. As the nonlinearity is monotonic the peaks remain the peaks, so the dispersion is unaffected by the nonlinearity

E. The energy dependence of the MDC widths show the narrowing expected below $E_F$, but above $E_F$ the falling spectral intensity shifts the entire MDC into the low count linear regime causing the MDC widths to return to the intrinsic value. The resulting asymmetry in the widths should not be confused for a signal of electron-hole asymmetry.

F. The spectral weights for the linear and nonlinearity spectrum, showing that the asymmetric enhancement around the $E_F$ results in an apparent shifting of the Fermi energy.
and two nonlinear detectors: one with a discrete change in slope (red), which makes the effects more obvious and a smoothly varying one (green) which makes the effects less obvious but is closer to what we observe. In 2.7A we show the nonlinearity of the detector in measured counts vs true counts. To elucidate how the actual spectra are affected, we depict the two spectra, a linear one and a continuously nonlinear one, side by side in figure 2.7B. We assumed a linear bare band and Fermi-liquid self energy. In 2.7 we show the effects of the nonlinearity on a sample MDC. While the deviation from a Lorentzian is obvious in the discrete case, the smoothed one is still well described by a Lorentzian. Consequently, detecting nonlinearity from a lineshape is difficult. The peak of the Lorentzian does not shift when the nonlinearity is applied, so analysis based on peak locations (e.g. band mapping, dispersions, Re(Σ)) are robust against the nonlinearity (2.7D). However, the peak enhancement also raises the half max level, effectively narrowing the MDC width. However the widths are significantly sharpened by the nonlinearity (2.7E). However as the intensity above $E_F$ is rapidly suppressed by the Fermi edge, the distorted nonlinear widths quickly return to the linear values. This creates a noticeable asymmetry in the widths that is roughly centered at $E_F$, which could be incorrectly interpreted as electron-hole asymmetry. Finally, the spectral weight, determined by integrating the MDC’s shows a clear enhancement due to the nonlinearity (Fig. 2.7F). However, this enhancement can be hard to detect, but if the nonlinear spectral weight is normalized, the nonlinearity can manifest as a Fermi Energy that drifts to deeper energies as the sample is warmed. Such a drifting Fermi Edge could have deleterious effects on procedures like the symmetrized EDCs, which require accurate determination of the Fermi Energy.

Since the asymmetry is an effect of the Fermi edge, it is strongly temperature dependent. To illustrate this behavior, we show a temperature dependence of the widths for a simulation of Fermi Liquid (quadratic energy dependence, discussed in detail in Chapter 3) in figure 2.8A and the corresponding nonlinear ones in 2.8B. Note that the asymmetry is strongest in the coldest sample but the other effect of the nonlinearity is softening of the width minimum with decreasing temperature. This softening is unphysical in that the minimum of the scattering rate is pegged to $E_F$, which can be understood from considering the allowable phase space for decay channels which is minimized at the $E_F$. This softening is more easily observed than the asymmetry so it is a clear
sign of nonlinearity in spectra. The temperature dependence of the linear and nonlinear spectral weights show another symptom of nonlinearity (Fig. 2.8C and 2.8D). Namely the isosbestic point (point of constant spectral weight) for the linear term is centered at $E_F$ in energy and half filling in weight as expected for particle conservation. However in the nonlinear case case the isosbestic point is still at $E_F$ but its filling is less than half of the max value and particle conservation is clearly broken. If the detector is nonlinear one of the easiest observations is a temperature dependent $E_F$ and minimum width location. For the simulations already described we show the temperature dependence of both in Fig. 2.8E.

One of the simplest tests for the detector nonlinearity is the temperature dependence of an amorphous gold sample. Amorphous (non-crystalline) gold is an ideal reference when taking ARPES data. The non-reactive nature of gold makes it resistant to aging, and the amorphous nature averages over all the bands such that the spectra are uniform in k but still show the Fermi Edge at $E_F$. Consequently, gold is regularly used to correct for detector inhomogeneity, as well as empirically determine both the Fermi energy as well as the resolution of the instrument. Even this simplest of spectra manifest the shifting Fermi edges due to the nonlinearity, but after correction with the curve extracted from Bi2212 spectra the Fermi edges no longer show any sort of thermal drift as expected (Fig. 2.9). Furthermore, we show on real data the difference between nonlinear and linearized Bi2212 results (Fig. 2.10), showing many unusual features: drifting minimum widths, E-H asymmetric widths, low isosbestic, are all significantly less pronounced in the linearized data. The remnant oddities are likely due to an imperfect linearization rather than representative of true features.

Previous measurements of the nonlinearity were done by the very labor and time consuming process of changing the photon flux by tiny increments [35, 36]. While the most direct method, it is an impractical calibration within the limited time available at synchrotrons. Instead we developed a method that takes advantage of the large dynamic range inherent in a Bi2212 spectrum to determine the detector nonlinearity. We take two spectra ostensibly identical except for changing the photon flux by a factor of two. Next we plot the high count image versus the low count image. If the detector was perfectly linear this image should be a scatter plot with a clear linear dependence
Figure 2.8: **Effects on Nonlinearity on Temperature Dependence Studies**

A Example temperature dependence of MDC widths
B Temperature dependence of widths after addition of nonlinearity, showing formation of asymmetry and shifting minimum width
C Example of temperature dependence of spectral weight with isosbestic point centered at $E_F$ and half filling
D Temperature dependence of spectral weights after addition of nonlinearity showing the isosbestic point holds at $E_F$ though shifted away from half filling
E Temperature dependence of the width minimum and the Fermi edge location after addition of nonlinearity
Figure 2.9: **Linearizing Amorphous Gold** The shifting fermi edge with temperature from non-linearity is evident in amorphous gold and can be corrected with nonlinearity extraction.
Figure 2.10: **Effects of Linearizing Data**

- **A** Example spectrum before and after linearization
- **B** Effects of linearization on sample MDC
- **C** Effects of linearization on dispersion
- **D** Effects of linearization on MDC widths
- **E** Temperature dependence of raw MDC widths
- **F** Temperature dependence of linearized MDC widths
- **G** Effects of linearization on spectral weight
- **H** Temperature dependence of raw spectral weights with isosbestic point well below half filling
- **I** Linearized spectral weights
proportional to the change in photon flux. Saturation of the image would manifest as a flattening of the high count vs low count at high counts. However, in the low count region, the Scienta detectors show the opposite effect, a bending up away from the low count extrapolation of a linear dependence. This effect may seem minor but can significantly alter the spectra.

While representative of the nonlinearity, the high counts vs low counts curve is not an actual nonlinearity curve. However it does contain all the information necessary to extract the nonlinearity correction. The algorithm to extract the nonlinearity is composed of two steps which allow us to first iteratively reach the linear low count regime and then extrapolate back to the underlying true counts. We start with a given pixel on the high count image, then we transfer to the same pixel in the low count image, for a change in the true counts of a ratio of photon fluxes. Then we return to the high count image but change pixels and find a new pixel whose counts match the low count pixels magnitude. This process is iterated until we enter the linear regime. In the linear regime the measured counts are the true counts, and we know the number of iterations and thus the number of flux ratios we traverse, so it is simple extrapolation back up to find the underlying true counts for the original high count value. We repeat the process for every high count value and we can build up the entire nonlinearity curve. The nonlinearity extraction algorithm is shown in the next few lines.

\[
\text{Iter} = \frac{HC(x_1) \cdot HC(x_2) \cdot HC(x_3) \ldots HC(x_i)}{LC(x_1) \cdot LC(x_2) \cdot LC(x_3) \ldots LC(x_i)} \quad (2.4)
\]

Which if we express in terms of the nonlinearity function acting on the original True count rate at the \(x_1\).

\[
\text{Iter} = \frac{NL(TC) \cdot NL(TC/R_F) \cdot NL(TC/R_F^2) \ldots NL(TC/R_F^{i-1})}{NL(TC/R_F) \cdot NL(TC/R_F^2) \cdot NL(TC/R_F^3) \ldots NL(TC/R_F^i)} \quad (2.5)
\]

This can be heavily simplified to:

\[
\text{Iter} = \frac{NL(TC)}{NL(TC/R_F^i)} \quad (2.6)
\]

Since we stop the iteration in the linear regime

\[
NL(TC/R_F^i) = TC/R_F^i \quad (2.7)
\]
Figure 2.11: Extracting the Nonlinearity

A High count image and low count image

B High Count vs Low Count scatter plot (red), high order polynomial fit (green) and low count linear extrapolation (blue) and orange arrows tracing the nonlinear extraction method’s iterations.

C Extracted nonlinear curve (red) and low count linear extrapolation (blue)
This can be heavily simplified to:

\[
\text{Iter} = \frac{NL(TC)}{TC/R_F}
\]  

(2.8)

Since we know the values of \(\text{Iter}, NL(TC), R_F\) and \(i\), it is simple to extract \(TC\). Repeating this procedure for each point on the High count vs Low count fit, we can extract the full nonlinear curve.

### 2.7 Effects of Resolution on the ARPES spectrum

In addition to detector nonlinearity, other experimental artifacts can cloud the picture of ARPES. Experimental resolution, both energy and angular, can distort the spectrum. Our findings confirm the results of Kaminski and Fretwell [38]

The angular resolution is affected by many components including the inherent resolution of the analyzer, the beam spot size on the sample and any warping of the sample surface to name a few. The angular resolution can be calculated and estimated but not accurately measured. Luckily, the effect of angular resolution is to convolve a Gaussian from the resolution with the intrinsic Lorentzian. The resulting MDCs are broadening into a Voigt function, which can range from a near Lorentzian to a Gaussian depending which component (intrinsic or resolution) is dominant. In the cuprates, with current analyzers and low energy photons < 10 eV, momentum resolution is likely a minor contribution to the over all width as evidenced by the decidedly Lorentzian line-shape. Ultimately, angular resolution leaves the MDC peaks and the spectral weight unchanged while broadening the MDC widths a varying amount dependent on the ratio of the MDC width to the resolution.

The effects of energy resolution are not so easily dealt with. In particular, the rapid change in spectral weight of the Fermi edge can result in confusing and counterintuitive results for both the MDC widths and the dispersion. For example take a simulated Fermi liquid like spectra in figure 2.13. Energy resolution will shift a portion of the weight from the deeper energies up and smaller energies down. Consequently, the dispersion maps energy broadening on to the MDC widths. For a given MDC well below the Fermi energy, the contribution from above and below are equivalent. Thus, the width is broadened but the peak remains in the same location. However as the Fermi
Figure 2.12: **Effects of Momentum Resolution**

A Simulated spectra showing the effects of momentum resolution for none, typical and 2×

B Dispersions showing momentum resolution has no effect.

C MDC widths showing the expected increase in widths with the addition of momentum resolution

D Spectral weights showing momentum resolution has no effect
edge is approached the positive contribution is rapidly suppressed by the dying spectral weight. Therefore, only the deep energy contribution is significant so the MDC is broadened asymmetrically, shifting the peak and receiving only half of the broadening. Above $E_F$ the weight dies exponentially such that shifted weight from deeper energies dominates the original weight. Consequently, the widths return to the proper value but offset in binding energy and the dispersion is now offset from the underlying dispersion. All of these results are summarized in Fig. 2.13, with a diagram explaining the asymmetric broadening in Fig. 2.14. As a rule of thumb when the energy resolution is greater than the Fermi width, the asymmetric contributions at $E_F$ manifest as a distinct dip in the widths. Again, this change should not be confused with a low energy bosonic mode.

2.8 Surface concerns

In an ARPES experiment, the electrons are forcibly ejected from the bulk to the vacuum. The boundary between the bulk and vacuum must be nearly perfectly clean and flat to avoid contaminating the spectrum. Bi2212 is a layered compound, with paired bismuth oxide layers that are only weakly bound together by van-der-Waals forces, they have a natural cleavage plane much like mica. A freshly cleaved surface is perfectly clean by definition, and the cleanliness is maintained by only cleaving in an ultra-high vacuum environment such that nothing exists that could adsorb on to the surface. Methods to attain and maintain a clean surface are well documented and will not be discussed in detail here.

However the act of cleaving is a violent act, literally ripping a sample in two. Consequently, samples can be damaged, warped, bent or filled with flakes. Non-parallel surfaces clutter the ARPES spectra as we sample two or more different k-states simultaneously. To limit these damages, it is customary to cleave the sample when it is cold ($< 90K$). The idea being that the cold makes the sample harder and more brittle so the act of cleaving does not bend the sample. However, we find the act of cleaving cold damages the surface in a subtle but uniform manner, which can lead to misleading spectra.

To study the surface quality we use a type of imaging that to our knowledge has not been used in conjunction with ARPES, Nomarski microscopy (Fig. 2.15)[39]. Nomarski microscopy is
Figure 2.13: Effects of Energy Resolution

A Simulated spectra showing the effects of energy resolution for none, typical and 2× typical.

B Dispersions showing energy resolution creates a strong kink with higher velocity near $E_F$.

C MDC widths showing the expected increase in widths with the addition of energy resolution as well as an unusual dip in the widths near $E_F$ that should not be construed as an intrinsic effect.

D Spectral weights showing energy resolution has the expected effect of broadening the Fermi edge.
Figure 2.14: **Source of Energy Resolution Effects**

A Contributions to energy resolution broadened MDC (black) from deeper energies (red), intrinsic (green) and higher energies (purple) for an MDC well below $E_F$.  

B At $E_F$ the deep energy contribution swamps the higher energy contribution shifting the peak in momentum. As very little weight is contributed from the high energies due to the falling spectral weight of the Fermi edge, the MDC does not experience as much broadening near $E_F$ causing the widths to noticeably sharpen though still broader than the intrinsic width.  

C Above $E_F$ the spectral weight falls exponentially such that the MDC is dominated by the deep energy contribution resulting in a constant offset in dispersion.
a white light differential interference contrast microscopy. The illuminating light is split into two polarizations and translated relative to each other via Nomarski prism. Then the light hits the sample, where it is reflected back along the same path. On the return trip the Nomarski prism recombines the two polarizations such the light can interfere. If the path length difference between the two polarizations is a half of a wavelength, that color of light is enhanced, but if the path length difference between the two polarizations is a quarter of a wavelength, that color of light is minimized. Consequently, different colors correspond to surfaces at slightly different angles.

Nomarski images are shown in 2.16. Many surface defects are visible. In general the sample is composed of many regular steps due different cleavage planes. Warping of the sample surface is visible through the gradual change in intensity, particularly noticeable near some sample edges. Both of these defects are expected. However, the regular parallel lines seen cutting through the different layers were not. We believe these lines represent and an accordion-like folding of the sample surface. Such an folding will have particularly deleterious effects on an ARPES spectrum, when the angle emission from both surfaces will be measured simultaneously.

To determine the source of this folding we compare the orientation of the lines to the crystal orientation determined from x-ray diffraction 2.17. We see that the lines are always parallel to the Γ-X direction. The exactness of these lines suggest that they could even be used as a method to align the crystal. Their orientation along the Γ-X direction suggests their source: superstructure.

In the cuprates, the crystal lattice period for the BiO layer is slightly different than the CuO layer. This strain is resolved by a regular but slight warping of the CuO plane as shown in Figure 2.18. This warping is periodic not random with a periodicity of about 5 lattice constants along the Γ-Y direction. The periodic dependence in real space manifests as a replication of the main band structure, but translated 1/5 of the full Γ-Y periodicity as shown in figure 2.18. The replicated bands, known as superstructure bands, are weaker in intensity but can significantly complicate analysis. The strong overlap of bands along the Γ-X quadrant prohibits clean analysis, so most data is actually taken along Γ-Y. Other methods such as doping the material with Pb disrupts the lattice constant mismatch greatly increasing the periodicity and and decreasing the magnitude. The superstructure does not just complicate analysis but may provide a fundamental instability in
Figure 2.15: **Diagram of Normarski Microscopy** The Nomarski microscope works by splitting the illuminating light into two polarizations, displacing the polarizations spatially from each other with a Nomarski prism, reflecting off the sample and recombining to create an white light interference pattern. Small differences in the total path length manifest as different colors.
Figure 2.16: Cleaved Bi2212 Under Nomarski Illumination Four cleaved samples showing the large defects as well as regular straight lines suggesting some fundamental weakness of the samples flatness.
Figure 2.17: **Cleaved Bi2212 with Crystal Alignment** Sample alignment shows the Nomarski lines always run in the Γ-X direction.
the structure. During the act of cleaving, the sample undergoes intense stress, but when cold it is also in an unstable state due to the contraction of the copper supporting post. The layers relieve this tension by folding like an accordion.

![Figure 2.18: Superstructure in Real and Momentum Space](image)

The mismatch in lattice constants of the various planes in Bi2212 cause strain that is relieved by a subtle warping of the CuO plane along Γ-Y. This periodicity cause a replication of the Fermi surface (right).

While we estimate the angle between folds is only 1-3 degrees, the effect on the spectra is pernicious and devious. Rather than just smearing the band as typical continuous warping will do, this accordion folding causes the spectrum to be the sum of two distinct angles. When fit with a Lorentzian, the width is broad and the peaks track the space between the two bands. However if the desired band is along the node then the secondary band will be gapped. To illustrate this effect, we show simulated spectrum for the node (Fig. 2.19A) and slightly offnode (Fig. 2.19B) and their sum in (Fig. 2.19C). Because of the gap the secondary band’s weight near \( E_F \) will plummet. So much so that within the secondary’s gap the primary band emerges unadulterated. Consequently the MDC widths show a sharp low energy kink (Fig. 2.19D), and the dispersion has corresponding kink (blue in Fig. 2.19C). The combination of a step in the width and the kink in the dispersion can lead to the mistaken assumption that a low energy bosonic mode does exist, when in reality it is consequence of surface flatness.

While surface flatness is a significant concern, it is not the only concern. When cleaving warm, the sample surface is directly exposed to the vacuum at room-temperature. In these conditions,
Figure 2.19: Effects of Imaging Two Bands Simultaneously If we measure across the Nomarski line, we will simultaneously measure the node and the slightly gapped state. The resulting spectrum has an unusual low energy kink in the widths. But the corresponding kink in the dispersion is opposite of that observed by Plumb et al. [40]
weakly bound oxygen dopant atoms can diffuse into the vacuum, altering the doping concentration. Consequently, we recommend quenching the sample immediately after the warm cleave to secure the dopant atoms. However, if precisely maintaining the dopant concentration is more important than surface quality, a cold cleave is recommended.

2.9 Removing the Incoherent Background

A common concern in fitting EDCs is the inclusion of the background. Typically inelastic scattering is presumed to be the dominant contribution. This type of background can be well described by the Shirley approach [41, 28], which argues that the background grows monotonically with energy and is proportional to the total number of unscattered states at lower energies. Employing a recursive method the most likely background and intrinsic line shape can be determined for a given EDC.

We find that another background is present in ARPES spectra. This background is energy dependent but isotropic in momentum. As the electrons composing this background lack a energy momentum relationship we call it the incoherent background. We think this background has two likely sources. First, impurities in the materials could trap electrons in spatially localized states. As these electrons are well defined in real space, they are extremely poorly defined in momentum space. So poorly defined they are spread over the full Brillouin zone creating the momentum independent background. Alternatively, this background may be extrinsic and specific to the ARPES process. Even a very good surface may have small defects or molecules adsorbed on to it[42]. As the electrons are emitted from the surface, a small fraction scatter off these impurities destroying the coherence resulting in an incoherent background.

To remove the incoherent background we slice the image into MDC’s and fit with a Lorentzian with a constant offset. This offset is the momentum independent background. We then subtract that contribution from the image leaving a spectrum composed of only the coherent band. Figure 2.20, shows the effect of removing the incoherent background. For this nodal case, this background appears to be minimal as the spectrum does not visually change when it is removed (Fig. 2.20C) and at it’s strongest it is only 10% of the peak height (Fig. 2.20A). However as the background is
Figure 2.20: **Removing the Incoherent Background** a, Sample MDC (red) 100 meV from $E_F$, its fit (teal), background weight (blue), and band weight (Green). The MDC at $E_F$ (dashed) is shown for comparison. b, Energy dependence of spectral weights for a nodal spectrum at 50K c, Comparison of the original nodal spectrum to one with the background removed.
extended over the full window it’s contribution to the integrated weight of the spectra rivals the weight of the band (Fig. 2.20B). Consequently, any study of the spectral weight of the band requires the proper removal of incoherent weight.
Chapter 3

Formalism and Theory

3.1 Introduction

Placing a chapter on theory this early in an experimental thesis can be daunting to many readers. However, I’ve found this formalism to be helpful to understanding the major results presented here. Such methodology is discussed in much more detail in numerous text books [43, 44, 45, 46]. These texts can be overwhelming so I have attempted to reduce the complexity such that it is approachable to beginning experimental graduate students. In this chapter I will detail the principle as well as the multiple formalisms of the electron Green’s function. I will then explore the spectral function which ARPES directly measures, followed by the electron self energy, including Kramers-Kronig self consistency. I will conclude with a short description of how to incorporate BCS theory into the Green’s function framework.

3.2 Green’s Function

Most physicists are first introduced to Green’s functions in graduate electricity and magnetism, where Green’s functions are used to solve the intermediate electric field for a given volume based on the geometry of the boundaries and the location of any isolated charges. While not true in the strictest mathematical definitions of Green’s function, many-body theory has appropriated the terminology with the caveat that rather than spatial boundary conditions, particles experience temporal boundary conditions (i.e. initial and final states). For a non-interacting free particle the solutions are almost trivial and the Green’s function is:
\[ G(\omega, k) = \frac{1}{\omega - \frac{p^2}{2m} - i\delta} \]  

(3.1)

where \( p \) is the free space momentum of the particle, \( m \) is the mass of the particle and \( \delta \) is an infinitesimal quantity to avoid singularities.

Placing the electron inside of a solid modifies the Green’s function by adding in the band structure, \( \epsilon_k \), due to the symmetries and periodicities of the lattice.

\[ G(\omega, k) = \frac{1}{\omega - \epsilon_k - i\delta} \]  

(3.2)

Note that without the infinitesimal \( \delta \), the Green’s function would composed of singularities where \( \omega = \epsilon_k \). These singularities are mathematical poles that trace the band dispersion. These poles can be broadened and renormalized, but their presence defines the energy and momentum of quasiparticles. Consequently, confirming the location of the poles is essential in any analysis of quasiparticles.

While a nice framework, the noninteracting Green’s function clearly can not describe the most interesting physics which occurs when electrons interact. We resort to a perturbation terminology where the interactions modify or “dress” the noninteracting term. This modification is expressed:

\[ G(\omega, k) = \frac{1}{\bar{\omega} - \epsilon_k - i\delta} \]  

(3.3)

where

\[ \bar{\omega} = \omega - \Sigma' - i\Sigma'' \]  

(3.4)

where \( \Sigma' \) and \( \Sigma'' \) represent the real and imaginary parts of the self-energy. While the addition of the self-energy may seem like a minor modification, the potential physics contained is immense and complicated. Consequently much effort has been expended to accurately determine the electron self energy.

### 3.3 Spectral Function

While ARPES directly probes the electrons in a solid, it does not measure the electron Green’s function. Rather it measures the spectral function which is defined as
\[ A(\omega, k) = -\frac{1}{\pi} ImG(\omega, k) \] (3.5)

Consequently,
\[ A(\omega, k) = -\frac{1}{\pi} \frac{\Sigma''(\omega, k)}{(\omega - \Sigma'(\omega, k) - \epsilon_k)^2 + \Sigma''(\omega, k)^2} \] (3.6)

In the cuprates, the self energy varies weakly with angle such that for any given spectrum it is effectively k-independent.

There are two competing and complimentary approaches to analyzing ARPES data. One can break the energy vs. momentum spectra and break it down into curves at each momentum called Energy distribution curves or EDCs. Alternatively, we select curves at each individual energy called momentum distribution curves or MDCs. Historically, the first ARPES data was taken with a pinhole to select a single momentum state before scanning a spectrum. The peak positions would map the band while the widths would roughly follow the scattering rate. However, a close investigation of (3.6) reveals that the peak positions map the dressed band only well away from the Fermi energy and that the peak width is poorly defined as the line shape is significantly complicated with the addition of the self energy.

However, in the cuprates, the self energy varies weakly with angle such that for any given spectra we can assume it is effectively momentum-independent. Consequently
\[ A(\omega, k) = -\frac{1}{\pi} \frac{\Sigma''(\omega)}{(\omega - \Sigma'(\omega) - \epsilon_k)^2 + \Sigma''(\omega)^2} \] (3.7)

which, if we assume a linear bare band, \( \epsilon_k = v_{BB}(k - k_F) \), can be rewritten as:
\[ A(\omega, k) = -\frac{1}{\pi} \frac{\Sigma''(\omega)/v_{BB}^2}{(k - (k_F - \omega + \Sigma'(\omega))/v_{BB})^2 + (\Sigma''(\omega)/v_{BB})^2} \] (3.8)

The spectral function at a particular energy is thus a perfect Lorentzian in momentum with the peak positions matching the dressed band and the widths proportional to the imaginary part of the self energy. The ease at extracting the self energy has made the MDC method the preferred technique to extract the self energy.
3.4 Components of and Models for the Self Energy

The self energy has two components: the real and imaginary parts. The real part is a measure of the dispersion of the electrons (\( E \) vs \( k \) just as the dispersion of light in material is \( \omega \) vs \( \lambda \)). The imaginary part represents the rate of scattering corresponding to the absorption of light in a material. Just as the dispersion and absorption of a dielectric are connected through the Kramers-Kronig relations, the dispersion and the scattering rate of electrons are connected. Consequently, these components are deeply and profoundly linked such that each contains the same information. This duplication of information is a built in method to confirm on any finding. However the dispersion is complicated by the unknown bare band making the extraction of relevant values much more difficult. The imaginary part of the self energy is a much more direct measure of the electron’s self energy, which is where I will focus most of my energy.

The self energy is widely considered electron hole symmetric with the real part point symmetric about \((E_F, k_F)\) with the imaginary part plane symmetric about \(E_F\). This symmetry seems logical from the the following common argument. For an electron to transition from one state to another, it is actually swapping states with a hole. Consequently all electron transitions are mirrored by a hole transition. However the exact proof is more subtle. The electron lifetime is not symmetric about \(E_F\), but is the mirror image of the hole’s lifetime about \(E_F\). But, any experiment ejecting an electron involves the injection of a hole. Consequently, the hole’s lifetime and the resident electron’s are added such that we sample a summed electron-hole lifetime which is symmetric[44].

The imaginary part is composed of two distinct contributions: fundamental electron-electron interactions that extend over all energies and bosonic excitation modes that are localized to specific energies[47]. Lev Landau provided the first theoretical description of interacting fermions, now known as a Fermi liquid (FL) [48]. From phase space considerations for a 3D metal with a roughly spherical Fermi surface, the imaginary part of the self energy is quadratic in energy and temperature.

\[
\Sigma(\omega, T) = \alpha(\omega) + i\beta(\omega^2 + (\pi k_B T)^2) \tag{3.9}
\]

While this picture is believed to reasonably describe strongly overdoped cuprates, it fails
for all other dopings. To explain the decidedly linear temperature dependence, Varma proposed a phenomenological model called the marginal Fermi liquid (MFL):

$$\Sigma(\omega, T) = \omega \ln\left(\frac{\omega_C}{\text{Max}(|\omega|, \pi k_B T)}\right) + i(\text{Max}(|\omega|, \pi k_B T))$$  \hspace{1cm} (3.10)

However, satisfactory description of the underdoped case remains elusive.

In addition to the gradual evolution with energy of the electron-electron scattering, localized bosonic modes (e.g. the famous 70 meV kink)[49] manifest as step in the imaginary part and sharp bends in the dispersion, often called kinks. The two main sources for bosonic modes are the magnetic interactions via the electrons’ spins and phonons where the electrons scatter off the lattice. To determine the nature of an observed mode the most conclusive test is the isotope effect. By swapping all the atoms in a crystal structure for a different isotope the electron configurations remain the same but the mass of the lattice does not. Consequently, the energy of the phonon modes is altered. The observation of an isotope effect in the conventional superconductors was essential to determining that the pairing was mediated by phonons[23]. For the a given bosonic mode the corresponding self energy can be calculated as shown by Shulga et al. [50] and Vonsovsky, Izyumov, and Kurmaev[51].

### 3.5 Incorporating BCS

One might assume that the pairing interaction in superconductivity would best be described by inclusion in the self energy. However, the formation of a Cooper pair breaks an unstated assumption of the use of many-bodied Green’s functions: particle conservation. The Green’s function methodology assumes that an electron in equals an electron out. However, when a Cooper pair forms, it is now a separate entity. Specifically, the eigenstate of a Cooper pair is not an electron but the mixed state of an electron and a hole, called a Bogoliubov quasiparticle [52].

Now is a good time to discuss electrons vs holes. When solving for the band structure, when one assumes that electrons are the carriers then the band structure is $\epsilon_k$ but if one assumes holes then it is $-\epsilon_k$. This second and normally ignored solution is the hole band. $E_F$ is often considered the boundary between the electrons and the holes, which it is. However it is not the mixing of
holes and electrons in the electron band that forms a Bogoliubov quasiparticle but the mixing of electron and hole bands.

The easiest way to write the complete Green’s function is the Nambu formalism [53] where the electron and hole bands are orthogonal states of the system such that

\[ |e\rangle = \begin{pmatrix} 1 \\ 0 \end{pmatrix} \quad \text{and} \quad |h\rangle = \begin{pmatrix} 0 \\ 1 \end{pmatrix} \] (3.11)

And the Green’s function is now:

\[ G^{-1} = \begin{pmatrix} \omega - \epsilon_k & \Delta \\ \Delta^* & \omega + \epsilon_k \end{pmatrix} \] (3.12)

It is now evident that the addition of the superconducting order parameter mixes the electron and hole states.

Though common, the matrix notation for the superconducting Green’s function is not the exclusive representation. Sometimes it is written equivalently with the Pauli matrices:

\[ G(\omega, k) = \frac{\omega \tau_0 + \epsilon_k \tau_3 + \Delta \tau_1}{\omega^2 + \epsilon_k^2 + \Delta^2} \] (3.13)

Neither of these representations were used by Gorkov when he first introduced the concept[6, 54]. Instead he introduced the normal Green’s function

\[ G(\omega, k) = \frac{1}{\omega + \epsilon_k} \] (3.14)

and the anomalous Green’s function to represent the transition from electron to hole states:

\[ F(\omega, k) = \frac{1}{\Delta} \] (3.15)

### 3.6 Simulating the ARPES Spectrum

Using the electron Green’s function from the Nambu formalism, it is simple to try to recreate our observed ARPES spectra. Including the Fermi edge, energy dependent self energy and experimental resolution, the ARPES spectra can be written:
\[ I_{\text{ARPES}}(\omega, k) = (F(\omega) \times \text{Im}(G(\omega, k))) \otimes R(\omega, k) \] (3.16)

Where \( F(\omega) \) is the Fermi function \( R(\omega, k) \) is the experimental resolution [13].

For these simulations, we used a reasonable interpretation of \( \text{Im}(\Sigma) \) linear and symmetric about \( E_F \) with a step at 70 meV. We took the Kramers-Kronig transformation of \( \text{Im} \Sigma \) to extract the \( \text{Re} \Sigma \). We assumed a linear bare band with \( v_{BB} = 2.9 \text{ eV } \AA \) and \( k_F = .42 \text{ } \AA^{-1} \). We assumed \( T=50K \) and the energy resolution was 4 meV. For the nodal (\( \Delta = 0 \)) case this simulation does an excellent job of recreating the observed spectrum Fig. 3.1. To simulate the off-nodal spectra we should only have to add the presence of a small but finite \( \Delta \). However the addition of a finite \( \Delta \) has no observable effect on the on the simulation, even though the gap is clearly resolvable in the data.

We find that the minimum \( \text{Im}(\Sigma) \), is just too large to get as significant of a depreciation in weight at the Fermi energy for the off-nodal spectrum. We can achieve the expected reduction in spectral weight by drastically reducing the minimum \( \text{Im} \Sigma \) by an order of magnitude. However the band’s width is dramatically too narrow. So we compensate by broadening the spectrum as if it was a final state effect by convolving a Lorentzian in momentum with the spectrum. This contradiction spurred the creation and development of the ATS as will be discussed in chapter 5. This simulation can easily be expanded into a three dimensional simulation by replacing the linear bare-band with a band structure calculated from a tight binding model and the momentum dependent \( \Delta \) to account for the d-wave gap.
Figure 3.1: **Simulations from Green’s Function** While we can accurately simulate the nodal spectra with the $\Gamma$ extracted from the MDC, we cannot recreate the observed gapping for the off-nodal spectra. Instead we must lower $\Gamma$ by an order of magnitude and then broaden it in momentum by convolving a Lorentzian in $k$. 
Chapter 4

Phenomenological Model For Scattering in the Normal State

4.1 Introduction

While the superconducting state is the ultimate driver for research in the cuprates, the normal state’s unusual behavior has gathered particular interest. Fig. 4.2 shows a compilation of the results of transport studies on the cuprates [55]. While we commonly divide the normal state into four sections: anti-ferromagnetism, pseudogap, strange metal and Fermi liquid only the antiferromagnetism transition is definitely observed as a phase transition. Furthermore, while it is commonly stated that overdoped cuprates are Fermi liquids, and optimal doped are strange metals with a linear temperature dependence or marginal Fermi liquids, the story is much more complex. Nigel Hussey points out that a mixed linear/quadratic region exists between optimal and overdoped and an even odder s-curved region in the underdoped.

Some ARPES experiments have attempted to study the electron-electron self energy. Koitzsch et al. studied the doping and energy dependence of the electron-electron self energy in the superconducting state [56] They argue for a sharp transition from marginal Fermi Liquid to Fermi Liquid at optimal doping, counter to the transport studies summarized by Hussey [55]. Evtushinsky et al. studied the doping and temperature dependence, and found no doping dependence of the low temperature limit and no significant features in the temperature dependence, also counter to the transport studies [57]. As both of these studies disagree with transport experiments, further investigation is warranted to bring ARPES and transport into agreement.

With excellent resolution of low energy ARPES, we can now resolve the fine evolution of the electron-electron scattering rate with energy and temperature. With this improved data we can, for the first time, fit the energy and temperature dependence simultaneously. In order to fit this high
Figure 4.1: **General Phase Diagram of Transport in the Cuprates** Transport phase diagram from Hussey [55] showing the resistivity ranges from Fermi liquid ($T^2$) to marginal Fermi liquid ($T$) to a strange “S”-shaped behavior in the underdoped case.
Figure 4.2: Prior Studies of the Energy, Temperature and Doping Dependence of the Electron-electron Scattering Rate

A Energy and doping dependence study by Koitzsch et al. at $T = 30K$ which concluded optimal doping marks a sharp transition from marginal Fermi Liquid to Fermi Liquid behavior.\textsuperscript{[56]}  

B Temperature and doping dependence study by Evtushinsky et al. which concluded no significant doping dependence to $\Gamma(T)$. \textsuperscript{[57]}
quality data we need a model for the electron-electron scattering. In response, we developed a new phenomenological model for the electron-electron self energy, the Variable Fermi Liquid (VFL). In this chapter, we will show the VFL appears to completely describes the energy, temperature and doping dependence of the imaginary part of the electron-electron contribution to the self energy.

4.2 Variable Fermi Liquid

The Variable Fermi Liquid can be succinctly written as:

$$\Sigma''_{VFL}(\omega, T, x) = A(\omega^2 + (\beta k_B T)^2)^{\alpha x} + \Gamma_0$$

(4.1)

where $A$ is the amplitude. $\beta$ governs the comparative strength of the energy and temperature dependence. For Fermi liquids, $\beta = \pi$. $\alpha$ is a prefactor on the doping fraction, $x$. To account for the possibility of impurity scattering, we include a $\Gamma_0$.

The excellent quality of low energy ARPES data allows us to resolve the subtle evolution of the scattering rate well into the normal state. Fig. 4.3 shows how well the this form fits the extracted self energies from three dopings. The quality of the fits suggest that the VFL is a good description of the normal state, energy, temperature and doping dependence of the self energy of Bi2212. We have chosen to study the scattering rates exclusively at the node for three reasons. First, as we have shown in Fig. 2.6, the self energy is weakly angle dependent, so the nodal states are likely universal. Second, by studying the node we avoid any possible complications from the presence of a pseudogap in the normal state, which would corrupt the widths as previously described in chapter 2. Third, the Fermi velocity is largest at the node and thus these states should dominate the transport studies.

Note that $\alpha$ is very close to $\pi$ in value. In fact we can achieve reasonable fits assuming $\alpha = \pi$ (Fig. 4.4). Consequently the full energy, temperature and doping dependence can be fit with just three free parameters: $A$, $\beta$ and $\Gamma_0$. The amplitude, $A$, includes many effects, it’s doping dependence is not immediately telling of underlying physics, but further studies may find some. The constant offset, $\Gamma_0$, is likely due to impurity scattering which can vary greatly from sample to sample. We find $\beta$ varies but is always within 20% of $\pi$, reasonably similar to the original Fermi
Figure 4.3: **Fitting the MDC widths with the Variational Fermi Liquid** Fitting spanning the doping diagram showing the accuracy of the VFL.
liquid. Further studies can determine whether the observed doping dependence is real or just a statistical fluke.

4.3 Doping Dependence of VFL

One of the compelling aspect of the VFL is that the energy, temperature and doping dependence can be well described with just the three free variables, $A$, $\beta$ and $\Gamma_0$. While not unconventional the overdoped region is worth discussing for it follows the conventional understanding of interacting fermions, the Fermi liquid. The overdoped case is the simplest and most straightforward. Assuming an $\alpha$ of $\pi$ then for a doping level .32 the self energy of the VFL is:

$$\Sigma_{VFL}(\omega,T,.32) = \omega^2 + (\pi k_BT)^2 + C$$  \hspace{1cm} (4.2)

And the normal Fermi liquid is recovered. However for only lightly overdoped cases (e.g. the data in panel 4.3A) the data is only Fermi liquid-like and goes as $T^{1.25}$.

The self energy of the optimally doped case is one of the most studied and contentious areas of research. Widely considered a strange metal it is marked by a linear temperature dependence. However, we observe a quadratic energy dependence near $E_F$. These different orders for $\omega$ and $T$ are surprising for an excitation in energy should be indistinguishable from an excitation in temperature.

Figure 4.4: Fitting the MDC widths with the VFL with $\alpha = \pi$
Figure 4.5: **Features of the VFL Model** The VFL ranges from quadratic in the overdoped limit to a hyperbola (quadratic around $E_F$ but linear at deeper energies) at optimal doping to the strange ‘S’-shaped behavior in the underdoped case.
Phil Anderson has focussed considerable effort addressing this contradiction proposing the Hidden Fermi Liquid [58]. However the most accepted explanation is the phenomenological model proposed by Varma [17], the Marginal Fermi Liquid which was originally written as:

\[ \Sigma_{MFL}(\omega, T) = \omega \ln\left(\frac{\omega}{\text{Max}(|\omega|, \pi k_B T)}\right) + i(\text{Max}(|\omega|, \pi k_B T)) \]  (4.3)

He has mentioned as an aside that the imaginary part of the marginal Fermi liquid can be written as a hyperbola [59]

\[ \Sigma''_{MFL}(\omega, T) = \left(\omega^2 + (\pi k_B T)^2\right)^{1/2} \]  (4.4)

which is the exact form the VFL simplifies to for optimally doped samples

\[ \Sigma''_{VFL}(\omega, T, .16) = \left(\omega^2 + (\beta k_B T)^2\right)^{1/2} \]  (4.5)

provided \( \beta = \pi \). The hyperbola also appears to be able to account for the contradiction that has so troubled Anderson. The resistivity measurements are determined by the temperature dependence at \( E_F \).

\[ \Sigma''_{VFL}(0, T, .16) = \beta k_B T \]  (4.6)

which is linear. However if we expand about \( E_F \) the VFL recovers a parabola in \( \omega \) as shown in Fig. 4.5.

\[ \Sigma''_{VFL}(\omega, T, .16) = (0^2 + (\beta k_B T)^2)^{1/2} + \omega - \frac{2 \times 0}{(0^2 + (\beta k_B T)^2)^{1/2} + 1 - \frac{1}{(0^2 + (\beta k_B T)^2)^{3/2}}} + \frac{0^2}{(0^2 + (\beta k_B T)^2)^{3/2}} \]  (4.7)

\[ \Sigma''_{VFL}(\omega, T, .16) = \frac{\omega^2}{2\beta k_B T} + \beta k_B T \]  (4.8)

The reverse is true at deep energies. So in summary, near \( E_F \) the hyperbolic form leads to a quadratic energy dependence and a linear temperature dependence but at deep energies, it has a linear energy dependence and a quadratic temperature dependence, which is why the scattering rates at deep energies converge as we see in the data (Fig. 4.3.

As we go to the underdoped region the self energy takes on a truly unique S-curve like shape. This S-curve should not be confused with a bosonic mode for it is a natural out growth of the
VFL. This S-curve is not unique to the ARPES results as Hussey also reports an S-curve resistivity for the underdoped samples in fig 4.2. While it is a natural aspect of the VFL it is not possible with a linear combination of FL and MFL, lending support to the possibility that the VFL is the more complete model. Ultimately, the VFL is just a phenomenological model without a theoretical underpinning yet. However, its accuracy at describing the full energy, temperature and doping dependence of the normal state of Bi2212 suggests that it may be helpful in understanding the electron-electron interactions in the cuprates.
Chapter 5

The ARPES Tunneling Spectra

5.1 Introduction

As the most direct measure of the momentum state of electrons, ARPES has long been heralded as the definitive measure of an electron scattering rate. However several flies are in that ointment. First, the scattering rates extracted from even the sharpest of the MDC widths are an order of magnitude larger than those determined from optical conductivity and other experiments. Second, ARPES MDC widths were wildly inconsistent from sample to sample, varying by as much as a factor of three. This wide variation in widths is also strikingly suspicious since presumably related properties like resistivity and $T_C$ do not show a similar variation.

The disagreement between ARPES and optical conductivity studies could be due to experimental limitations of ARPES which continual experimental improvements would resolve. Alternatively and more likely ARPES is sensitive to scattering processes that optical conductivity is not. If these processes are from a spatial disorder, then it would manifest solely as a broadening in momentum. Consequently, if we integrate over momentum we can extract a spectral weight that is independent of the additional broadening. Properly normalized, these integrated spectra are analogous to those of tunneling spectra of s-wave superconductors. However, in contrast to conventional tunneling, these new spectra that we call ARPES tunneling spectra (ATS), maintain the momentum selectivity that makes ARPES uniquely powerful.

5.2 Extracting the ATS

In most solids, the electron states are defined by the momentum states, however thermodynamical properties are actually governed by the density of states, i.e. the total number of electrons
at a given energy. The density of states has been directly probed by tunneling and the local density of states by STM. However, in the cuprates, the strong d-wave nature of the gap complicates the analysis for both of these techniques. The ATS, though maintains the angle dependence of ARPES while still extracting a density of states, effectively a momentum resolved density of states. The creation of the ATS is a three step process. First the incoherent background is removed. Second the band is integrated over to extract a spectral weight. Finally the spectral weight is normalized to remove the Fermi Edge.

The removal of the incoherent background as previously described isolates the coherent band, i.e. the electron’s that are well defined in momentum space. By integrating along momentum perpendicular to the Fermi surface, we extract a coherent spectral weight, equivalent to an angle resolved density of electron filled states. For the node (Fig. 5.1a), this spectral weight is dominated by the Fermi edge. The slight slope at deeper energies is due to the band dispersion, because like all density of states the integrated weight is proportional to $\frac{1}{dE/dk}$. Away from the node (6°) (Fig. 5.1b) the electron density of states now shows more structure due to the presence of a finite gap and thermally populated Bogoliubov excitations above and below $E_F$.

Figure 5.1: Creating an ATS a Nodal spectrum and the corresponding spectral weight. b Near-nodal spectrum and corresponding spectral weight with the thermally populated states above the gap clearly evident. c Near nodal spectrum normalized to the nodal spectral weight. The ARPES tunneling spectrum (ATS) is shown in pink.

To compensate for the Fermi edge, the nodal spectrum is the ideal reference as it is taken
under identical conditions including temperature, experimental resolution and surface aging. After normalization, the resultant curve is the ATS (purple Fig. 5.1C).

By normalizing to the node, we isolate the differences between the node and off-node spectra, specifically the presence of a gap. However this normalization also damps the subtle structure in the density of states due to the changes in the dispersion. This damping is not ideal as such structure was the conclusive result that proved BCS theory in the traditional superconductors. However, as we’ve already shown the detector nonlinearity and resolution make the determination of $E_F$ and the appropriate Fermi width difficult. Inaccuracy in determination of either would significantly alter the resulting ATS. Consequently, we use the nodal normalization method. Additionally, as our attention is focused on the states within the gap, this damping of the deep energy structure, while unfortunate, does not impact our findings.

Though the ATS may bare a resemblance to symmetrized EDCs that are commonly plotted in ARPES, the two curves are fundamentally distinct as the EDCs do not include the integration and the momentum broadening is mapped into an EDC width via the dispersion. In contrast, as a measure of the density of states, the ATS can be analyzed with well established methods first developed for tunneling experiments. Furthermore, the ATS are not symmetrized around $E_F$ - rather the ATS is a real measure of the spectral function above $E_F$ due to thermal occupation. Unlike the symmetrized EDC method, electron-hole symmetry need not and has not been assumed.

### 5.3 Fitting the ATS

The ATS is equivalent to a typical Giaever tunneling curve[60, 61], except localized to a particular portion of the Fermi surface, a significant advantage over conventional tunneling. For each ATS, the gap strength is single valued, which allows us to use the formula first proposed by Dynes to explain tunneling from s-wave superconductors:

$$I_{ATS} (\omega) = \rho_{Dynes} = \text{Re} \frac{\omega + i\Gamma}{\sqrt{(\omega + i\Gamma)^2 - \Delta^2}}$$

(5.1)

where $\omega$ is the energy relative to $E_F$, $\Gamma$ is the imaginary part of the energy corresponding to a finite lifetime and $\Delta$ is the superconducting gap[62]. As mentioned previously, each ATS is specific
to a location on the Fermi surface. For six degrees off the node, the fit returns a gap size of $6.0 \pm 0.1$ meV and minimum scattering rate, $\Gamma_{ATS}$, of $2.6 \pm 0.1$ meV (Fig 5.2). This value corresponds to a scattering length of 1200 Å and a lifetime of 40 cm$^{-1}$, which for the first time brings ARPES into agreement with results from other experiments such as optical conductivity[63], STM[64, 65], microwave conductivity [66] and tunneling[67].

Figure 5.2: **Fitting an ATS** The Dynes’s formula fits the ATS taken with a wide variety of sample conditions. The quality of fits despite the change in shape of the ATS further support our contention that the Dynes formula is an accurate description of the underlying physics of the ATS.

Qualitatively, this magnitude in the scattering rate should be expected, because a moderately deep depletion of weight in the gapped region requires a $\Gamma$ of the order of or smaller than $\Delta$. The Dynes formula accurately fits the ATS over a wide range of angles and temperatures. We show the quality of fits for an optimally doped sample at 10K and 90K at 5$^\circ$ and 10$^\circ$ from the node in figure 5.2. We find that the inclusion of an energy dependent term in $\Gamma$ or $\Delta$ is unnecessary.

Though energy dependence need not be included, the small but finite energy resolution does. Though small in Low Energy ARPES, the energy resolution is present and for the most accurate fits we need to include it. To properly include the energy resolution we can not simply convolve the Dynes formula with a Gaussian representing the energy resolution. That method fails to consider the effect of the rapidly decreasing weight from the Fermi edge. Instead we must consider when the experimental resolution is added, which occurs before the normalization. Consequently, we use the formula:
\[
F(\omega) = \frac{(F(\omega) \times \rho_{\text{Dynes}}(\omega)) \otimes R_{\omega}}{(F(\omega) \otimes R_{\omega})}
\] (5.2)

Where \(F(\omega)\) is the Fermi function, \(\rho_{\text{Dynes}}(\omega)\) is the Dynes formula, \(R\) is the experimental resolution. The main effect of including the resolution is the deviation from the expected "U" shape inside the gap to the "V" shape of the observed ATS.

We further justify the use of Dynes formula with a simulation. We simulate a simplified spectrum from the Green’s function, extract an ATS from the simulation and fit with the Dynes formula. Assuming a constant \(\Gamma\) as the imaginary part of the self energy, the real part of the self energy is zero. Consequently the Green’s function can be written as:

\[
G(\omega, k) = \frac{\omega + i\Gamma + \epsilon_k}{(\omega + i\Gamma)^2 + \epsilon_k^2 + \Delta^2}
\] (5.3)

For \(\Gamma = 3\) meV and \(\Delta = 10\) meV, the resultant spectrum is shown in figure 5.3. With no background and no Fermi edge a simple integration extracts the ATS. Fitting with the Dynes formula reveals that not only is the fit nearly perfect the original values of \(\Delta\) and \(\Gamma\) are both recovered.

A reasonable question to ask is why the lifetime term is only added to \(\omega\) and not \(\Delta\)[68].

\[
I_{\text{ATS}}(\omega) = \rho_{\text{Mitrovic}} = \text{Re} \frac{\omega}{\sqrt{\omega^2 - (\Delta + i\Gamma\Delta)^2}}
\] (5.4)

However, if we add the lifetime term to \(\Delta\) instead of \(\omega\) then at \(E_F\) the spectral weight would always be zero. As this is a clear contradiction to what we observe, we are confident the lifetime should only be added to \(\omega\). Finally Mikhailovsky showed that the Dynes formula can be extracted from first principles for a pair-breaking scattering[69].

5.4 Two Types of Scattering: Normal and Pair-breaking

The standard ARPES method for determining the scattering rate, MDC widths[34], give static values \(\Gamma_{\text{MDC}}(\omega = 0, T = 0)\) nearly an order of magnitude larger than the associated values for ATS. Resolving this disagreement will prove the power and value of the ATS method. While the ATS method returns almost identical scattering rates from many samples at the same doping, the MDC method varies by more than a factor of 2 for the low temperature values (fig. 5.4). These
Figure 5.3: **Accuracy of Dynes Formula** Simulations of a simplified Green’s function form can be fit accurately with the Dynes’s formula.
results indicate that the physical processes that are responsible for the $\Gamma_{MDC}$ are different from those that are responsible for the $\Gamma_{ATS}$. In particular, the wide variation in MDC widths without a similar variation in $T_C$ suggests the MDC is subject to scattering processes that do not affect the pairing interaction. But the consistency of ATS curves implies that $\Gamma_{ATS}$ represents intrinsic and universal scattering processes.

![Graph](image1.png)

**Figure 5.4: ATS vs MDC**

A The ATS for three different optimally doped samples taken under identical conditions agree quite well. B The MDC widths for the nodal spectra for the same three samples showing a variation of over a factor of 2. Furthermore, the extracted values of the $\Gamma_{MDC}$ are as much as an order of magnitude larger than $\Gamma_{ATS}$

In Dynes original study of Pb$_{0.9}$Bi$_{0.1}$ [62], the presence of a finite $\Gamma$ manifest mainly by broadening the coherence peak. However when $\Gamma$ is comparable to $\Delta$, the coherence peak is more than just broadened in fact a significant fraction of weight is shifted from the peak to the center of the gap. We find it instructive to illustrate the differences between a changing $\Gamma$ and $\Delta$, in Fig 5.5. A changing $\Delta$ has two main effects, the expected shifting of the coherence peaks and a changing weight at the center of the gap. In contrast, $\Gamma$ mostly just shifts weight from the coherence peak into the gap. Consequently, either a decreasing $\Delta$ or an increasing $\Gamma$ can add weight to the center of the gap. As the sole function of $\Gamma$ is shifting of weight from the coherence peaks into the gap we argue it is representative of a competing pair breaking process. Typical competing processes compete by forming a gap that removes available electrons from the Fermi surface, this competing
process instead acts by breaking already formed Cooper pairs restoring spectral weight to the Fermi energy.

Not all scattering processes are pair-breaking processes. The distinction between pair-breaking and non-pair-breaking is best illustrated by impurity scattering in the BCS superconductors \[70\]. Dirty superconductors (i.e. superconductors with significant impurity concentration) are split into two types based on whether the impurities are magnetic or not. For magnetic impurities, even a small concentration quickly disrupts the superconductivity. However, conventional s-wave superconductivity is robust against non-magnetic impurities. Anderson explained this dichotomy by pointing out that magnetic impurities break the time reversal symmetry by flipping the spin of one of the electrons. This interaction breaks the pair. In contrast, the non-magnetic impurities can not break the time-reversal symmetry leaving the pair intact. Anderson described how to approach the non-magnetic impurity scattering. One first must diagonalize the impurity scattering interaction and only then introduce the pairing interaction between the new eigenstates.

In the cuprates, the d-wave gap provides a second channel besides the spin-flip of a magnetic impurity to break a pair. Adjacent lobes of the d-wave gap are out of phase such that scattering
from one lobe to one of the orthogonal ones breaks the time-reversal symmetry and consequently the pair. However scattering within a lobe or to the opposite lobe does not break pairs. This difference is why the MDC scattering rates are so much larger than those from the ATS. Figure 5.6B schematically illustrates both a pair-breaking and non-pair-breaking scattering event.

These forward (small q) scattering events that constitute the majority of $\Gamma_{MDC}$ are likely a result of dopant atoms residing between the BiO$_2$ planes well away from the CuO planes (as is usually the case)[71]. In this case the dopant potentials will be well screened from the CuO planes, giving a shallow but spatially extended potential disturbance to the conduction electrons. This weak disturbance can on average only scatter the electrons by a small momentum shift forward, i.e. this scattering will be predominantly non-pair-breaking. We believe this process to contribute strongly to the MDC and EDC linewidths, but not to the much sharper ATS spectra (they will only contribute to the ATS spectra taken very near the node, possibly explaining the slightly enhanced $\Gamma_{ATS}$ for those states as will be shown in (fig 7.1)). In contrast, defects in the copper oxide plane (e.g. a Cu vacancy) are unitary scatterers allowing for a large momentum transfer during scattering. STM analysis estimates the Cu vacancy density as .2% with a diameter of 3 lattice constants, which for a lattice length 3.8 Å corresponds to a scattering length of 600 Å. Assuming the Cu vacancy acts is a unitary scatterer the final state is equally likely anywhere in the Brillouin zone, but only those events that flip the phase of the order parameter are pair-breaking exactly half of all scattering events are pair breaking resulting in a pair-breaking scattering length of 1200 Å. To convert from scattering length to scattering rate we use the bare velocity as previously discussed. We estimate the pair breaking rate due to the Cu vacancies as 2.5 meV consistent with our measured $\Gamma_{ATS}$.

This distinction between forward/non-pair-breaking and unitary/pair-breaking scattering also explains why optical conductivity measurements of the scattering rate agree with the $\Gamma_{ATS}$ but not with the $\Gamma_{MDC}$. Optical conductivity bases their calculation of the scattering rate on the Drude model[25]. Where the response of the electrons in an applied electric field can be written in terms of their momentum as:

$$\langle p(t_0 + dt) \rangle = (1 - \frac{dt}{\tau_U} - \frac{dt}{\tau_F}) (\langle p(t_0) \rangle + qEdt) + \frac{dt}{\tau_U} \langle p_U \rangle + \frac{dt}{\tau_F} \langle p_F \rangle \quad (5.5)$$
where \( \langle p \rangle \) is the average momentum, \( 1/\tau_U \) is the rate of unitary scattering, \( 1/\tau_F \) is the rate of forward scattering, \( \langle p_U \rangle \) is the average momentum after unitary scattering, and \( \langle p_F \rangle \) is the average momentum after forward scattering. However

\[
\langle p_U \rangle = 0; \quad \langle p_F \rangle = \langle p(t_0) \rangle
\]

Such that,

\[
\langle p(t_0 + dt) \rangle = (1 - \frac{dt}{\tau_U} - \frac{dt}{\tau_F})\langle p(t_0) \rangle + qE dt + \frac{dt}{\tau_U} \langle p(t_0) \rangle
\]

Simplifying and assuming terms that are second order in \( dt \) are negligible

\[
\langle p(t_0 + dt) \rangle = (1 - \frac{dt}{\tau_U})\langle p(t_0) \rangle + qE dt
\]

Which can be rewritten as:

\[
\frac{dp}{dt} = qE - \frac{\langle p(t_0) \rangle}{\tau_U}
\]

Note that the only lifetime is the unitary lifetime. Consequently, optical conductivity experiments are primarily sensitive to unitary scattering and are only weakly sensitive to forward scattering processes. This differential sensitivity could explain why optical conductivity agrees with the \( \Gamma_{ATS} \) rather than \( \Gamma_{MDC} \).

Alternatively, a variation of dopant density has been argued to manifest as a distribution of discrete doping domains within a single sample complete with matching distribution in gaps and Fermi volumes[72, 73]. As the doping domain size is on the nanometer scale, ARPES will always sample many of these domains simultaneously. Consequently, the band ARPES resolves will actually be the sum of many different narrows bands offset in energy due to the varying chemical potential. To illustrate this effect, we calculated Fermi surfaces from a tight binding model for three different doping levels (Fig. 5.6A). The inset shows how the multiple Fermi surfaces would sum to create a larger than expected MDC width. However as the superconducting gap is fixed at \( E_F \) in each domain, the doping inhomogeneity would not smear the gaps, leaving the ATS comparatively unaffected. Near the antinode the Fermi surfaces grow further apart, which could account for previous observations of a growing MDC width with angle [34]. Ultimately, both the forward scattering and the doping inhomogeneity explanations are rooted in the out of plane dopant distribution and are likely just two different views of the same underlying physics.
Figure 5.6: Sources of MDC Broadening Diagram of the Fermi surface detailing the difference between non-pair-breaking forward scattering (red) and pair-breaking unitary scattering (blue) B Illustration of how doping inhomogeneity leads to MDC broadening from sampling of multiple Fermi surfaces at different doping levels
Chapter 6

Pre-pairing, the Pseudogap and the “Filling” Superconducting Gap:
Temperature Dependence of the ATS

6.1 Introduction

The nature of the superconductive gap in the cuprates, the temperature at which it disappears, and how it disappears are critical questions that have resisted explanation due to a lack of unambiguous spectroscopic data. At the antinode, where the gaps are largest and most studies are focused, the ambiguity is unavoidable, because additional ordering potentially contaminates the region such that antinodal gaps may not be from the superconducting pairs. The near nodal region does not have these complications but has smaller gaps requiring more precision - a problem we overcome with ATS. We show that the near-nodal gap magnitude remains nearly constant through $T_C$ and only closes at a higher temperature, $T_{Close}$. This finding indicates pre-formed pairs in the pseudogap state, consistent with diamagnetism experiments which find pairs form at the temperature, $T_{Onset}$. We also show that the energy gain for superconductive gapping primarily disappears due to the increasing scattering rate ($\Gamma$) filling in the gap, rather than the gap closing via a shrinking $\Delta$.

6.2 Temperature Dependence of the ATS

To understand the normal state behavior of the ATS, we find it illustrative to show the temperature evolution through $T_C$ for two near-nodal cuts (12° and 18° away from the node) of a lightly underdoped ($T_C=85K$) Bi2212 sample (Fig 6.1). The raw data of panel A shows the clear formation of the superconducting gaps as temperature is lowered, as well as the start of the
“bending back” Bogoliubov-type dispersion, which is most apparent in the coldest $18^\circ$ spectra. We uncover a quantitative description of the temperature evolution through the use of the ATS spectra, which are shown in panels B and C for the two cuts. In both panels we see the clear evolution with temperature with the predominant effect being that the gap “fills in” as the sample temperature is raised - an effect which is clearly different than the “closing” of the gap that is observed traditional BCS superconductors. We also note that spectra taken above $T_C$ (i.e. the yellow 90K ATS and the red 100K ATS) still clearly show the effect of the gapping, even though the sample is no longer superconducting. These effects can be quantified by fitting the spectra with equation 5.1, the results of which are shown in figure 6.1D. The extracted $\Delta$’s for each angle decrease only slightly from low to high temperature, including above the superconducting $T_C$ where they become termed a pseudogap. These states have an extrapolated gap closing of 125K or above for this sample - a temperature consistent with the onset temperature $T_{Onset}$ measured from Nernst[74] and diamagnetism experiments[75], and also consistent with experiments of pre-pairing measured at the antinode[76].

6.3 Pseudogap as a Preparing Gap

As the prelude to superconductivity, the pseudogap state is widely believed to be one of the keys to understanding superconductivity in the cuprates[77, 78]. Two interpretations of the pseudogap have been advanced: a) the pseudogap is a precursor state to the superconductivity, with Cooper pair formation at $T^*$ and condensation of these pairs at the lower temperature $T_C$[79], b) the pseudogap is a separate, competing phenomenon from the superconducting state such as the formation of a density wave[80, 81]. One of the main keys to resolving this debate is to study the near-nodal regime of the Fermi surface, where known density waves are not active. Previous studies of the near-nodal states were incapable of separating the gap or pairing energy scales from the scattering rate or pair-breaking scale - one of the issues that we solve here.

That the superconducting gaps and pseudogaps merge so seamlessly (Fig. 6.1D) with each other as a function of temperature, even for the states very near the node, indicates quite conclusively that the pseudogap state is simply a continuation of the superconducting state above $T_C$, i.e.
Figure 6.1: **Temperature Dependence of the ATS**

A Temperature dependence of spectra taken at $\theta = +12^\circ$ and $\theta = +18^\circ$.  
B Corresponding ATSs at $12^\circ$.  
C Corresponding ATSs at $18^\circ$.  
D Interplay of $\Gamma$ and $\Delta$ with temperature as well as fitting $\Delta$ to a d-wave BCS (orange) and a guide for the eye (green) for the temperature evolution of $\Gamma$.
it is a precursor pairing state. Here we point out that while similar-sized pseudogaps and superconducting gaps have been previously observed for antinodal states, those results are complicated by competing orders such as various types of density waves or checkerboard order[80, 81] which are believed to affect the antinodal but not near-nodal states.

![Figure 6.2: Comparison of ATS(T) and Symmetrized EDC(T) ATS and symmetrized EDCs for a UD 85K Bi2212 sample at 12° from the node. While the ATS clearly resolves a gap at 90K (gold), the symmetrized EDC method would conclude that the gap closed between 80K (green) and 90K (gold) or approximately at $T_C$.

Our result is inconsistent with a recent ARPES result that stated that the nodal gap closes at $T_C$ - a key piece of evidence for the “nodal-antinodal dichotomy” of the behavior of the superconducting state of the cuprates. We believe that the main difference between our result and that of ref [82] is that near $T_C$ the scattering rate $\Gamma_{ATS}$ becomes comparable to $\Delta$ (figure 6.1). This regime is precisely where the old qualitative methods artificially indicate a zero gap, whereas in fact the gap remains finite. Figure 6.2 specifically show that if we analyze the present data using the symmetrized EDC method we also would interpret the near-nodal gap to close near $T_C$ (gold), while the ATS clearly show the existence of gap above $T_C$.

Almost all previous ARPES studies of the pseudogap in cuprates have focused on the antinodal states, as the larger gap there makes its detection easier, particularly in the presence of the
sizeable pair-breaking scattering rates. A few recent works have indicated that there may be an
order parameter besides superconductivity acting at the antinode, with the gapping effect of this
order parameter also termed a pseudogap. This possibility is different from but not inconsistent
with the present analysis, i.e. these two different types of pseudogaps could both exist. In fact the
measured $T_{Close}$ from fitting to a d-wave BCS gap is 119K, consistent with the $T_{Onset}$ (125K) found
from measurements of the Nernst effect, rather than the $T^*$ (175K) determined from the anti-nodal
gap opening.

6.4 Functional Form of Temperature dependence of $\Gamma_{ATS}$

The temperature dependence of the scattering rate has several different predicted forms
depending on the dominant source of scattering. If electron-electron scattering dominates then it
is expected to be cubic[83, 84], which has been reported to be experimentally confirmed [85]. The
reduced phases space of the d-wave gap limits the available states of Fermi liquid adding another
order of T to the quadratic Fermi liquid. If low energy phonons are to blame, then it should go as
$T^4$[83]. Inelastic quasiparticle scattering is predicted to follow a $T^{9/2}$ or $T^{7/2}$[86]. As we show in
fig 6.3, $T^3$ is not a horrible fit which possibly accounts for previous results. However, if we fit with
a simple power law, we find the best fit is between 5-6, for which there is no reasonable theory.

Alternatively, the pair-breaking may be a virtually coupling of the electrons to a higher
energy mode. In the limit of a delta function, this mode results in a scattering rate of the Arrhenius
form[87, 88]:

$$\Gamma(T) = \Gamma_0 + Ae^{-\frac{E_A}{k_BT}}$$  (6.1)

In fact this form of the Arrhenius is actually a simplified form of the full equation:

$$\Gamma(T) = A \frac{1}{e^{-\frac{E_A}{k_BT}} + 1}(1 - \frac{1}{e^{-\frac{E_A}{k_BT}} + 1}) + \Gamma_0$$  (6.2)

The Arrhenius may appear to be a simple exponential but it is a much more complicated
shape as we show in figure 6.4. The Arrhenius is zero initially with a sharp turn on around a
tenth of $E_A$. Note that simple and full form agree up to a quarter of $E_A$. As a rule of thumb the
Figure 6.3: **Functional Form of** $\Gamma(T)$ **Fitting** $\Gamma(T)$ **with a selection of possible forms predicted from theoretical work on a variety of scattering processes showing that the Arrhenius form caused by exciting a deep energy mode (dark blue) is the best fit.**
simplified form is a reasonable approximation up to temperature, \( T_{\text{Limit}} = 6 \frac{K}{\text{meV}} \times E_A \) where \( E_A \) is in meV. Above a quarter of \( E_A \) the simplified and full forms deviate but both saturate as the mode becomes fully activated. As the fits to the Arrhenius are excellent, we believe the temperature dependence of \( \Gamma \) indicate an activated mode is the dominant pair-breaking channel. Studying the effects of doping on \( \Gamma(T) \) will provide valuable insight into the exact nature of the pair-breaking mode and will be discussed in detail in Chapter 8.

Figure 6.4: **Behavior of the Arrhenius Form** The Arrhenius form is most often used as the simplified model but the full Arrhenius shows a stronger saturation at a lower level. For the activation energies found in all samples studied are large enough that the approximation of the simplified form is valid.

6.5 **Comparison of Cuprates to Conventional Superconductors**

We find it illustrative to directly compare these results for the cuprates to Dynes’s original study of \( \text{Pb}_{0.9}\text{Bi}_{0.1} \). Figure 6.5 shows a direct comparison of the \( \Delta \) and \( \Gamma \) for an underoped (85K) cuprate and the conventional superconductor, \( \text{Pb}_{0.9}\text{Bi}_{0.1} \). We normalized both \( \Delta(T) \) and \( \Gamma(T) \) to \( \Delta_{\text{Max}} \) and \( T \) to \( T_C \). The conventional \( \Delta \) closes at \( T_C \) as expected, while we observe a finite \( \Delta \) in the normal state as has been previously discussed.

The most dramatic change between the conventional and the cuprate is in \( \Gamma \). First, the low
Figure 6.5: Comparison of Cuprates and Conventional Superconductors. Comparison of the temperature dependence of $\Gamma$ and $\Delta$ for underdoped (85K) Bi2212 and a the strongly coupled conventional superconductor, Pb$_{0.9}$Bi$_{0.1}$. The temperature scales have been normalized to $T_C$ and the energy scales $\Delta_{Max}$, where $\Delta_{Max}$ is the anti-nodal gap magnitude for the Bi2212 and $\Delta(T=0)$ for Pb$_{0.9}$Bi$_{0.1}$. 

- $\Delta$ (UD 85K)
  - $T_{Close} = 1.42 T_C$

- $\Delta$ (Pb$_{0.9}$Bi$_{0.1}$)
  - $T_{Close} = 0.98 T_C$

- $\Gamma$ (UD 85K)
  - $\Gamma_0 = 0.08 \Delta_{Max}$
  - $E_A = 6.11 T_C$
  - $A = 56.4 \Delta_{Max}$

- $\Gamma$ (Pb$_{0.9}$Bi$_{0.1}$)
  - $\Gamma_0 = 0.002 \Delta_{Max}$
  - $E_A = 2.11 T_C$
  - $A = 0.333 \Delta_{Max}$
temperature limit is significantly higher in the cuprates 8% of $\Delta_{Max}$ while the conventional are just a few tenths of a percent. Furthermore, the activation energy for the cuprates is a triple that of the conventional superconductors and the amplitude representative of the coupling to said mode is a $200 \times$ that of the conventional superconductor. This dramatically different behavior suggests that the cuprates are in a completely different regime of strong coupling than the conventional strong coupled materials like the Pb$_{0.9}$Bi$_{0.1}$.

### 6.6 Filling vs. Closing Gaps

The results shown here indicate a qualitatively new type of superconductive pairing - not just the pre-pairing effect but also the way the superconducting gap disappears as the sample temperature is raised. In a conventional BCS superconductor the superconducting gap energy scale is reduced to zero as the temperature is raised to $T_C$, while the quasiparticle scattering rates in general remain very small, consistent with the weak electronic interactions. In the cuprates the gap energy scale does not close at $T_C$, but rather $\Gamma_{ATS}$ increases rapidly, filling in the gap such that it is mostly filled in at $T > T_C$. The filling in of the gap due to the increasing $\Gamma$ removes the energy gain for creating Cooper pairs, as opposed to the BCS mechanism where the gap magnitude, $\Delta$ itself closes. The evolution of the superconducting energy gain for gapping is clearly different from the conventional BCS one and appears to have limited precedent in other materials.

Though limited, this filling of the gap is not unheard of. For conventional superconductors doped with magnetic impurities, superconductivity can still occur even with finite weight in the center of the gap. This ‘gapless’ superconductivity is well understood [89]. However the evolution of the density of states due to increasing magnetic impurities (Fig. 6.7) is distinct from what observe. While the cuprates gap fills with increasing temperature, the gapless superconductor first closes before beginning to fill. However, scanning tunneling spectroscopy of vortex cores in both MgB$_2$[90] and LuNi$_2$B$_2$[91] both show a distinct filling of the gap as the center of the vortex 6.8. In most type II superconductors, the STS of the center of the vortex core has a pronounced zero bias conductance peak (ZBCP), which is attributed to bound states with in the vortex [21]. To explain the lack of a ZBCP both Eskildsen et al. an De Wilde et al. argue that the filled gap corresponds to
Figure 6.6: **Filling vs. Closing Gap**

A. Temperature dependence of the ATS for optimally doped Bi2212

B. Recreation of the temperature dependence by changing $\Gamma$ and holding $\Delta$ constant, showing the full temperature dependence can be explained by an increasing $\Gamma$

C. Recreation of the results originally reported by Dynes for Pb$_{0.9}$Bi$_{0.1}$, showing that the dominant behavior is still a closing of the gap for the conventional superconductors even when strongly coupled.
a region where the scattering length is shorter than the coherence length for a Cooper pair. In this scenario, the high magnetic field at the center of the vortex breaks any pairs but fails to localize the electrons. This view agrees with our picture of a filling gap due to a pair-breaking process represented by $\Gamma$.

Finally, this filling due to an increasing $\Gamma$ also suggests that $\Gamma$ represents a competing process with superconductivity. Traditional competing processes (e.g. charge density waves or spin density waves) inhibit superconductivity by removing states from $E_F$ and thus reducing the total number of possible candidate electrons for Cooper pairs. However, the pair-breaking process represented by $\Gamma$, instead shifts weight from $\Delta$ back to $E_F$, a fundamentally different method of competition. Elucidating the implications of this difference as well as the pre-pairing effects may go far towards explaining the mechanism of high $T_C$ superconductivity.

Figure 6.7: **Density of States for Gapless Superconductor** With increasing magnetic impurities ($\alpha$), the gap first closes before filling in[89].
Figure 6.8: **Filling of the gap inside of a Vortex** Comparison of STS studies of the density of states inside a vortex of MgB$_2$[90] and LuNi$_2$B$_2$[91] to the temperature dependence of the ATS for Bi2212
Chapter 7

Fermi Arcs and Non-quasiparticle Nature: Angle Dependence of the ATS

7.1 Introduction

A Fermi arc is a disconnected segment of a Fermi surface[92, 93]. This simple description belies the fundamental inconsistency in the physics of Fermi arcs, namely that such segments violate the topological integrity of the band structure. Efforts to resolve this contradiction of experiment and theory have focused on connecting the ends of the Fermi arc back on itself to form a pocket[94, 95, 96, 97]. Here we present an alternative view that the Fermi arc is composed of real spectral weight, shifted into the center of the gap via a pair-breaking process. While real, this weight lacks the poles necessary to be a true quasiparticle at the Fermi Surface[98].

7.2 Angle Dependence of the ATS

Figure 7.1 shows the evolution of the ATS’s along the Fermi surface in the near-nodal regime. Panel A shows the evolution of the gap in the raw spectra around the node. Panel B shows the location for the spectral weights in Panel C and the corresponding ATS in panel D. The extracted values of $\Delta$ and $\Gamma_{ATS}$ from fitting the ATS are detailed in panel E. T By fitting to the Dynes formula, we find the gap is linear and symmetric about the node as expected for a d-wave gap (Fig 7.1E). In this near-nodal regime, ATS are essentially isotropic, contrasting with the behavior of MDC’s which is observed to increase going away from the node[99, 100]. The slight increase in $\Gamma$ near the node may be due to the a second pair-breaking channel opening. The momentum transfer during forward scattering is usually too small to flip the sign of the order parameter between initial and final states. However near the node even forward scattering can flip the sign of the order parameter breaking the pair. We find we can simulate the angle dependence of the ATS accurately.
assuming a constant $\Gamma$ and a d-wave $\Delta$ (Panel F).

Two other methods are conventionally used to find $\Delta$ from ARPES: leading edge and symmetrized EDCs. We compare those two methods to the ATS in Fig. 7.2. The leading edge method simply takes the EDC at $k_F$ and simply finds the location of the half maximum on the leading edge of the EDC (Fig. 7.2a). This method tends to underestimate $\Delta$ as well as return unphysical negative values near the node, which are often artificially set to zero. The symmetrized EDC compensates for the Fermi edge, by adding the reflection about $E_F$ of the EDC to itself, i.e.,

$$SymEDC(\omega) = EDC(\omega) + EDC(-\omega)$$

(Fig. 7.2b). The gap size is then defined as the location of the peak. Close to the node this method also fails to resolve the gap. Furthermore, symmetrized EDCs tends to overestimate the gap size, extrapolating to a finite value of $\Delta$ at the node. For the same sets of data, the ATS method clearly resolves a gap all the way to the node(Fig. 7.2c). Furthermore, we find the $\Delta_{ATS}$ are roughly the average of the “standard” techniques away from the node (Fig. 7.2d). The failure of the two ”standard” techniques near the node result in a small arc of gapless states. Though a Fermi arc in the superconducting state is not customarily discussed, this small arc fits the standard definition for a Fermi arc. By comparing Fig. 7.1E and 7.2d the threshold for the existence of an arc is when $\Gamma > \Delta$.

### 7.3 Fermi Arcs

To study the normal state Fermi arc, we move to an underdoped sample ($T_C=65K$) with finely gridded momentum map in the superconducting state ($T=10K$) and the normal state ($T=75K$) (Fig 7.3). The growth of the Fermi arc is evident in the Fermi surface maps(Fig 7.3a). The symmetrized EDCs show a small ($2.4^\circ$) Fermi arc at 10K but a much larger one at 75K, with a gap only definitively present at $12^\circ$ (Fig 7.3b). However the ATS paint a different picture, showing a smooth filling of the gap rather than a discrete change at $12^\circ$ (Fig 7.3c). Assuming a d-wave $\Delta$ and an isotropic $\Gamma$, we can fit the entire momentum dependence of the ATS at once (Fig 7.3d). Both the superconducting and normal states are well fitted with this form. While $\Delta_{Max}$ has shrink by 30%, $\Gamma$ has more than doubled, effectively completely filling in the gap for states close to the node. We argue these filled in states are the source of the Fermi arc.
Figure 7.1: **Angle Dependence of ATS for Optimally Doped Bi2212 at 50K**

A) Evolution of ARPES spectra with angle in the near nodal region

B) Region of the Brillouin zone covered by the positive angles in spectrum in A

C) Extracted spectral weight showing the clear formation and growth of that gap with angle

D) Corresponding ATS

E) Extracted values of $\Delta$ and $\Gamma$ showing $\Delta$ is well described by a d-wave symmetry but $\Gamma$ is isotropic

F) Simulation showing that a constant $\Gamma$ and a growing $\Delta$ can recreate the observe ATS temperature dependence
Figure 7.2: Comparing Methods for Determining $\Delta$ A EDCs at $k_F$ showing the leading edge method of gap determination would only determine a finite gap above 4.1° B Symmetrized EDCs from the same spectra showing the resolvable peaks only at 2.9° C The ATS from the same spectra showing a gap existing all the way to the node. D Extracted $\Delta$ values for the three methods. Not only does the ATS measure a finite $\Delta$ closer to the node than either the other methods it also extrapolates to value of zero at the node where as the leading edge extrapolates to a negative value and the symmetrized EDC extrapolates to a positive one.
Figure 7.3: **Source of the Fermi Arc**

A Fermi map of the near-nodal regime both in the superconducting (top) and pseudogapped (bottom) states, for underdoped (65K) Bi2212. **B** Symmetrized EDCs from which one would determine a finite range of gapless states at $E_F$ which grows with increasing temperature. **C)** Measured ATS spectra as a function of angle away from the node. **D)** Two parameter fits ($\Gamma_{ATS}$ and $\Delta_{Max}$) for each entire set of spectra, with parameters listed on the panels. $\Delta_{Max}$ is the max of the d-wave gap, occurring at the antinode, with the k-dependence of the gap forced to maintain the simple d-wave form. $\Gamma_{ATS}$ is held constant with momentum.
A more complete temperature dependence of the Fermi arc is shown in Fig. 7.4. The reported growth of the Fermi arc with temperature can be accurately simulated with a tight-binding band structure obtained from experiment[101, 29], a simple form of d-wave gap with max value at the antinode of 40 meV, and an electron scattering rate which is constant across the Fermi surface (figure 7.1) but which varies strongly with temperature (figure 6.1). The values of the electron scattering rate are directly obtained from the Dynes fits up to and including 90K. However at higher temperatures the gaps are nearly completely filled and fits become less reliable, so we have extrapolated those values. While the gap is seen to close slightly with increasing temperature (fig 7.3), it was held constant in Fig. 7.4 for simplicity.

The picture presented here has some qualitative similarities to a theoretical proposal by Chubukov et al.[102], though their idea did not gain much traction because there was minimal experimental evidence to back it up. Here, all parameters of the simulations for most temperatures are directly obtained from experiment with no adjustable parameters, and the excellent quantitative agreement with the independently measured arc data makes a very strong case for this interpretation of the Fermi arc.

The notion of discontinuous Fermi arcs put forward from the previous ARPES experiments[103, 77] is unphysical within the context of standard condensed matter theory. Therefore, significant effort has been expended to observe if and how these arcs close[95, 96, 94, 97, 104], or if they represent a new electronic state[105, 106, 107]. Much excitement thus came when small pockets were observed in quantum oscillation experiments, though a direct connection to something that would close the arcs has not been found. From ARPES there have been three recent experiments that presented evidence for arc closures, though these have been met with skepticism for reasons due to potential contamination from shadow bands, superstructure bands, or extended extrapolations. With the new understanding presented here, there is no physical reason why the arc would need to be closed because it is not a real locus of quasiparticle states. In this light, the difficulty in observing the “back” side of the arc is completely natural - that is, the arc is actually arc-like, as opposed to only being the front side of a small hole pocket.
Figure 7.4: **Temperature Dependence of the Fermi Arc** The unusual temperature-dependent arc length is seen in both the data and simulations. The main inputs to the simulations are an experimentally determined tight binding band structure, a simple d wave gap (both below and above $T_C$) with independently measured d-wave gap maximum 40 meV, and a temperature dependent scattering rate $\Gamma$ (shown) with units of meV. The majority of these $\Gamma$ values ($\sim 100$K) are also independently experimentally determined.
7.4 The lack of quasiparticle weight at $E_F$ and non-Fermi liquid physics.

Within the conventional analysis method of symmetrized EDCs, the Fermi arcs are considered loci of quasiparticle states that grow with increasing temperature. Instead, the above simulations and data argue that the arcs are a result of the interplay between $\Delta(k,T)$ and $\Gamma_{ATS}(k,T)$, which is dominated by $\Delta(k)$ and $\Gamma_{ATS}(T)$. Importantly, this result suggests the arcs are not composed of true quasiparticles because, besides the node, no quasiparticle poles exist at $E_F$ - rather the pole locations are the gap values, $\Delta(k)$. Rather than a true Fermi surface, the arcs are regions where real spectral weight has been scattered inside the d-wave gap, with this weight varying with Fermi surface angle and temperature. Though this picture is clearly different from the truncated locus of quasiparticles at $E_F$, it could be responsible for much of the exotic non-Fermi liquid behavior in the cuprates.

To put this into context, almost all of the machinery used in condensed matter physics to calculate transport, thermal properties, etc. is built around the presence of quasiparticle states or poles at the Fermi energy[43, 108, 44]. However, here we see that in the pseudogap state there are no quasiparticle poles at the Fermi energy but rather they are at the gap edges, which may be 30 or more meV away from $E_F$. Despite this lack of quasiparticle poles at $E_F$, there is spectral weight at the locations of the “remnant Fermi surface” which originates from the finite $\Gamma_{ATS}$, with this weight of an incoherent non-quasiparticle-like nature. Aside from the single quasiparticle state at the node, this non-quasiparticle weight comprises the only states available to contribute to the transport and thermodynamic properties. Therefore, these are strong candidates for the long-sought non-quasiparticle[98, 109] or non-Fermi liquid states that dominate the transport, heat capacity, etc. in the pseudogap state of the cuprates, though the way in which these non-quasiparticle states actually contribute to these other properties is of course not yet understood.
Chapter 8

Activation Energies and the Pairing Strength: Doping Dependence of the ATS

8.1 Introduction

In addition to the temperature and angle dependence the cuprates have one more dimension that can be probed with ARPES, doping. As previously discussed the phase diagram of the cuprates is profoundly interesting. Most notably the superconducting region is limited to a dome structure centered at finite doping. This doping dimension provides an excellent method to probe and study the superconducting state.

Previous studies of the doping dependence have focused most intently on the evolution of $\Delta$ with comparatively cursory investigations of $\Gamma$. Hufner compiled the results of many different experiments to create Fig. 8.1A [110]. He shows that many different measurements show two different energy scales one that follows the superconducting dome and is generally believed to be the near-nodal gap and a second energy scale that grows linearly and roughly follows the $T^*$ widely thought to be the antinodal gap that is distinct from the superconducting gap. Studies of the doping dependence of the pair-breaking rate are limited, but as I have shown optical conductivity are sensitive pair-breaking rate, so it is the best probe to compare with the ATS. Fig. ??B shows an example doping dependence of the scattering rate determined by optical conductivity[63]. Extracting conclusions from this data is difficult besides the general consensus that the scattering rates decrease with increase doping, likely from better screening from the increased carrier density. Our doping study of the ATS paints a more detailed and occasionally different picture than these results.
Figure 8.1: Prior Studies of the Doping Dependence of $\Delta$ and $\Gamma$

A. Compilation of energy scales charting both the near-nodal gap and the anti-nodal gap [110]

B. Energy, temperature and doping studies of scattering rate as determined by optical conductivity [63]
### 8.2 Doping Dependence of the ATS

In figure 8.2, we show a short summary of the doping dependence of the ATS. Column A contains temperature dependence of the ATS at $12^\circ$ from the node for different dopings covering most of the superconducting dome($0.08 < x < 0.22$). Note that at all dopings the primary effect of increasing temperature is the filling of the gap. Consequently, the competing process represented by $\Gamma$ is likely universal in the cuprates. In column B, we show the temperature dependence of $\Gamma$ at multiple angles for each doping. The agreement between angles at each doping shows that the finding of an isotropic $\Gamma$ for the near nodal region is universal across doping. The orange curve is the fit to the Arrhenius form for each doping. The quality of the fit at each doping again supports our conclusion that the Arrhenius form is an appropriate description of the temperature evolution of $\Gamma$. Finally, column C shows the extracted values for $\Delta$ for the full doping study. Note that at all dopings, $\Delta$ is finite at or above $T_C$. Consequently, we have evidence for pre-formed pairs over the full phase diagram. To quantify the behavior of $\Delta$, we fit the temperature dependence at all angles with a d-wave gap with a BCS temperature dependence (black). These fits are characterized by two parameters $\Delta_{Max}$ and $T_{Close}$.

### 8.3 Low Temperature Limit

I summarize the low temperature limit doping dependence of both $\Delta$ and $\Gamma$ in figure 8.3. We find that all dopings are well described by the d-wave $\Delta$. As most of our data is limited to the near nodal region we can not directly measure $\Delta_{Max}$; instead we find the increase of $\Delta(\Theta)$ to extrapolate to a d-wave $\Delta_{Max}$. We find that $\Delta_{Max}$ monotonically decreases with increased doping. However unlike previous studies of the antinodal gap, we find it is not linear but rather has a clear curvature that can be approximated as parabola centered at zero doping (Fig. 8.3). Furthermore, $\Delta_{Max}$ does not follow the superconducting dome suggesting that a competing process maybe suppressing the superconductivity in the normal state of the underdoped cuprates. This possibility will be discussed later in this chapter.

Likewise, the pair-breaking rate, $\Gamma_0$, decreases as doping increases. As already discussed, we attribute $\Gamma_0$ to in-plane impurities. At first glance this doping dependence may seem unexpected
Figure 8.2: **Doping Dependence of ATS**

A Temperature dependence of ATS curves at 12° from the node for dopings covering most of the superconducting dome.

B Temperature dependence of extracted $\Gamma$ for multiple angles and the fit to the Arrhenius (orange) showing the isotropic nature of $\Gamma$ across the phase diagram.

C Temperature dependence of $\Delta$ for the different dopings and the fit to a d-wave BCS (black). At all dopings the gap closes above $T_C$ at an different temperature $T_{Close}$.
Figure 8.3: Doping Dependence of $\Gamma$ and $\Delta$ at $T=0$ In the 0K limit both $\Delta$ and $\Gamma$ decrease with doping. Note that $\Delta_{Max}$ does not follow the superconducting dome.
as the in-plane impurity density should not change much with doping as dopant atoms are far from the CuO plane. However the doping does change the carrier density and thus the effective screening of the impurities. Consequently, the impurity potentials in the underdoped case are likely larger in size though not increased in number, resulting in a shorter scattering length and lifetime.

8.4 Differentiating the Temperature Scales

Three different temperature scales are reported to govern the behavior of the cuprates. The first and best defined and known is the critical temperature, $T_C$, at which superconductivity begins. The doping dependence of $T_C$ marks the well known superconducting dome. Studies of the antinodal gap found a second temperature scale, $T^*$, at which the pseudogap opens. Though generally poorly defined and hard to measure, $T^*$ is generally considered to grow linearly with decreased doping[110]. Finally, recent studies of the Nernst effect have found evidence for a third energy scale, $T_{\text{Onset}}$ peaked in the underdoped region generally located between $T_C$ and $T^*$. The Nernst experimental results can be explained if the Cooper pairs continue to exist above $T_C$ but phase fluctuations prevent condensation into a single macroscopic superconducting state. As only $T_C$ is well defined, I've sketched the approximate behavior of both $T^*$ and $T_{\text{Onset}}$ in Figure 8.4.

From the doping study of the ATS we also extract three potential temperature scales scales. In conventional superconductors, the ratio $\Delta/k_BT_C$ is generally around 1.7-2.5 and helps to govern whether it is a weakly coupled or strongly coupled. However to have the $\Delta_{\text{Max}}$ line up with the $T_C$, for the overdoped, i.e. most conventional, we had to assume an especially strong coupling ratio of 3.5. The resulting mean-field $T_C$ fails to follow either $T_C$ or $T^*$, though it roughly follows $T_{\text{Onset}}$, though we do not observe the decrease expected for the heavily underdoped samples. The second temperature scale extracted from the ATS is the $T_{\text{Close}}$ from the temperature dependence of $\Delta$ (Fig. 8.2. Again this temperature scale most closely matches the $T_{\text{Onset}}$, but now includes a reduction for the most heavily underdoped sample. That both temperature scales extracted from $\Delta$ match the Nernst results suggests that the near nodal gap is determined by the same pairing interaction found by Wang et al.[74] For the final temperature scale, note that in column C of Fig 8.2, $\Gamma(T_C) = \Delta(T_C)$ at $10^9$ for every doping. While the doping dependence of $\Gamma$, $\Delta$ and $T_C$ are
Figure 8.4: Doping Dependence of the Distinct Temperature Scales. We compare the three established temperature scales of the cuprates; $T_C$, $T^*$ and $T_{Onset}$ with three scales we extract from the ATS.
all different they conspire to agree for all dopings. The angle is unlikely to be critical rather it is likely the fraction of $\Delta_{\text{Max}}$ represented by that angle is most important. To demonstrate this we plot the temperature at which $\frac{\Delta(T)}{\Gamma(T)} = \pi$ which closely follows $T_C$. Consequently we believe the interplay of $\Delta$ and $\Gamma$ sets a threshold for superconductivity creating the superconducting dome. The most likely explanation for this picture is that below $T_{\text{Close}}$ Cooper pairs can form, but the pair-breaking process shortens their lifetime such that they fluctuate into and out of existence. This fluctuation prevents coherence between pairs and thus the formation of the macroscopic quantum state necessary for superconductivity.

8.5 Doping Dependence of Activated mode

Now we will focus on the doping dependence of $\Gamma(T)$, specifically extracting the activation energy of the pair-breaking mode as well as the coupling of the electrons to that mode. For this fitting we use a modified Arrhenius form derived on the Eliashburg theory for single narrow mode:

$$\Gamma(\omega) = \Gamma_0 + \lambda E_A e^{-\frac{E_A}{k_B T}}$$

(8.1)

where $\lambda$ is the coupling strength of the electrons to the mode at $E_A$ [111]. The results of this fitting are shown in figure 8.5.

First, we find $E_A$ is strongly doping dependent and roughly follows the superconducting dome. Furthermore, the coupling parameter, $\lambda$, also follows the superconducting dome or at a minimum is strongly peaked at optimal doping. Finally, note that lambda is simply massive in value ranging from 3 to 70. Typical strongly coupled superconductors (e.g. Pb) have a $\lambda$ of 1-3 while weakly coupled superconductors have a lambda $< 1$. Consequently, either the cuprates seem to be in a new class of extremely strongly coupled superconductors, or the Eliashburg theory fails for this case and a new approach is needed. In either case, that this mode's energy and coupling both peak at optimal doping is highly suggestive that it is critical to superconductivity.

Though we have focused on the pair-breaking processes represented by $\Gamma$, it can also provide invaluable insights into the pair-forming process. The electrons that form Cooper pairs are generally found at energies much lower than the energy of the coupling boson. This difference is evident in the classic measure of the phonons responsible for coupling in lead from tunneling where they are
Figure 8.5: **Doping Dependence of $\Gamma$ and the Activated Mode** The activated mode is defined by two parameters: the activation energy $E_A$ and the coupling parameter $\lambda$. Both roughly follow the superconducting dome and are peaked at optimal doping. $\Gamma_0$ is the constant offset due to impurity scattering.
at much deeper energies than the gap values[23]. Consequently, the coupling occurs via virtual phonons[5]. The fleeting nature of a virtual phonon also contributes to the rapid interactions between the component electrons of a Cooper pair. However when the sample is warmed enough that the phonons can be thermally excited they are now real, which suppresses the pair coupling. Consequently, the mode that breaks pairs when thermally activated could be the one responsible for the pairing to begin with.

The strong doping dependence of this mode is also suggestive of it origin. As neither the mass nor the binding strength between ions are affected by carrier density, phonon modes are not expected to be strongly doping dependent. However, inelastic neutron spectroscopy of the spin resonance mode find a strong doping dependence that follows the superconducting dome just as our $E_A$(Fig. 8.6) [112, 113]. This agreement strongly suggests that the coupling between electrons is through these virtual excitations of the spin resonance mode. However, when that mode is thermally activated to create real spin excitations, those excitations can break the pairs.

While the doping dependence of the activated mode follows the spin-resonance, the magnitude is consistently larger than that reported in the inelastic neutron spectroscopy experiments. This disagreement can be explained by the fact that the likely $q$ coupling the electrons is not the perfect($\pi, \pi$) studied in the neutron experiments. The spin-resonance mode has a distinctive hourglass shape as a function of $q$ [114, 101]. Consequently, the mode we are sensitive to is most likely the upper branch of the spin resonance mode.

### 8.6 The Paradox of $E_A$ and $\Delta_{Max}$

We have presented two measurements of the superconducting state with apparently contradictory conclusions. The doping dependence of $\Delta$ suggests a pairing interaction that weakens with increased doping. To account for the superconducting dome one would expect a competing process that is stronger in the underdoped region such that only in the superconducting dome does the pairing interaction win out. The zero temperature limit of $\Gamma$ actually follow this behavior, suggesting if screening due to carrier density was the dominant contribution to the pair-breaking rate, this picture of competing processes may make sense. Alternatively the dynamical nature
Figure 8.6: **Doping Dependence of Spin Resonance Mode from INS** Campagno et al showed the spin resonance mode follows the superconducting dome like our activated mode.
\( \Gamma \) is a direct probe of electron interactions. Here we find they couple to a bosonic mode whose energy dependence follows the superconducting dome, most likely a spin resonance. This result suggests the mode is responsible for the pairing. However, if this picture of coupling through the spin resonance mode is correct we would expect the gap magnitude to shrink in the underdoped region. Consequently, the ever increasing \( \Delta \) with decreasing doping must represent something other process than the coupling mode. Resolving this paradox, will ultimately provide an invaluable new understanding of the competing processes in the cuprates.
Chapter 9

A Brief look to the Anti-node

9.1 Introduction

The anti-nodal region has received considerable attention for it holds a wide variety of physical phenomena, but it almost all boils down to the question, “Is the anti-nodal gap caused by the same process as the near-nodal gap?” In this thesis, we have already presented evidence for the existence of a pairing gap above $T_C$ in the near nodal region, but that gap seems to close at $T_{Onset}$, where as the anti-nodal gap has been argued to close at $T^*[13]$. To settle the question of the anti-nodal gap we will first address the possibility of using the ATS at anti-nodal followed by a possible explanation of the observations of the nodal-antinodal dichotomy that has been informed by our results on the effects of the incoherent background and the doping dependence of $\Gamma$ and $\Delta$.

9.2 Limits of the ATS

The ATS are severely hampered at the anti-node for two reasons. First, without a proper removal of the background, the ATS can only be qualitative not quantitative. But the proper removal requires a single band for accurate MDC fitting. However, spectra taken with nearly all photon energies have weight in both, the bonding and the antibonding band. While 7 eV photons do isolate the anti-bonding band, the photon energy can not probe the anti-node(Fig. 9.1). Consequently, the full quantitative ATS can only study the near-nodal region of the Brillouin zone. Second, the spectral weight varies as the inverse of the the bare velocity. Near the anti-node the van-Hove singularity causes a rapid change in spectral weight causing a peak in the ATS that exists regardless of the presence of a gap. Furthermore, when the van-Hove singularity is close to
the Fermi surface, the coherence peak from the gap can merge with the peak from the band bottom, complicating the extraction of $\Delta$ and $\Gamma$. Further study and improvements to the method may allow for extension of a quantitative ATS over the full Brillouin zone, but at present the ATS cannot be applied to the anti-node. However, details learned from the near-nodal analysis can inform the traditional techniques of analysis at the antinode.

### 9.3 Nodal-Antinodal Dichotomy

Currently, the exact form of the gap function is extremely controversial. While the simple d-wave form has a long history and significant evidence\cite{15, 115, 13}, recent experiments have found evidence for a two gap model for the cuprates\cite{116}. In this picture, the gap near the node is the superconducting gap while the gap at the antinode is due to a competing process (e.g. a charge density wave). As the two gaps have fundamentally different origins, their magnitude, doping dependence and temperature dependence are expected to be different. Observations of such a nodal-antinodal dichotomy are foundation of the two gap model.

The simplest piece of evidence for the two gap model is from tracking the EDC peaks at $k_F$ along the Fermi surface\cite{116, 82}(Fig. 9.2). Near the antinode, the EDC peaks sharply deviate from the d-wave form. In fig 9.3A, we show the EDCs for an underdoped (67K) sample showing the same clear deviation near the anti-node. We propose this deviation may not be intrinsic but is consequence of the failure to consider the contributions of the background. As the antinode is approached the peak strength relative to the background continually drops until the background is comparable. The rapidly increasing background effectively shifts the EDC peak to deeper energies. We illustrate this effect in fig. 9.3B. Here we assumed a d-wave gap with decreasing coherent (i.e. peak) intensity relative to the background (dashed black line). We find we can accurately simulate the results with the d-wave + background model as shown by the green line in the inset of fig 9.3A.

The other significant evidence for the two gap model was the distinctly different doping dependence for the near nodal gap and the anti nodal gap\cite{116}. The anti-nodal gap was found to continually shrink with increasing doping while the near-nodal gap grew with increased doping.
Low energy photons cannot be used to study the antinode and are limited to only the near nodal region for Bi2212.

Figure 9.1: **Momentum Limit of 7 eV Photons** Low energy photons cannot be used to study the antinode and are limited to only the near nodal region for Bi2212.
Figure 9.2: **Two Gap Model Angular Dependence** Panel from [116] showing the deviation from a simple d-wave gap at the anti-node for a symmetrized EDC peaks of UD50K Bi2212.
Figure 9.3: One vs Two Gap: Angle Dependence of EDC peak Position Symmetrized EDCs and showing the nodal antinodal dichotomy with a sharp increase in peak location near the antinode (inset). We can simulate this behavior with a momentum independent background and a momentum dependent peak intensity (right) starting with a d-wave form. The accuracy of our simulation suggests the observations of two independent gaps may be an experimental artifact.
We believe this nodal anti-nodal dichotomy is a product of the same mistake that resulted in the Fermi Arc and the filled gap being confused for the closed gap, the failure to take the pair-breaking rate into account. With our results from the ATS for the doping dependence of the $\Delta$ and $\Gamma$, we can simulate our the EDC peak behavior along the Fermi surface. We show in fig 9.5, that for the $\Delta$ and $\Gamma$ found in fig 8.2, an artificial nodal-antinodal dichotomy would be observed in the EDC peaks.

![Figure 9.4: Doping Dependence of the Nodal-Antinodal Dichotomy from Tanaka et al.[116]](image)

A third argument for nodal-antinodal dichotomy is the claim that the near-nodal $\Delta$ closes at $T_C$, while the anti-nodal $\Delta$ closes at much high temperature $T^*$[82]. We have already shown (chapter 6) that the reported temperature dependence of the near nodal gap is flawed. Rather than closing at $T_C$, the gap actually closes at a higher temperature near $T_{Onset}$ as determined by the Nernst experiments. Unfortunately at this time we cannot address the behavior of the anti-node at $T^*$ with the ATS. However Kondo et al. [76] have found evidence for two energy scales, $T^*$ and $T_{pair}$ at the anti-node by looking at the spectral weight inside the gap at the anti-node from symmetrized EDCs. This spectral weight is sensitive to the interplay of $\Gamma$ and $\Delta$. With only the
Figure 9.5: Simulating the Nodal Antinodal Gap Dichotomy

Simulations of the expected symmetrized EDC angular and doping dependence based on our studies of the ATS showing the observed [116] behavior is a natural consequence of the doping dependence of $\Delta$ and $\Gamma$. 
spectral weight, it is impossible to tell whether the energy scale $T_{pair}$ is due to a sudden increase in \( \Delta \) or a sudden decrease in \( \Gamma \). However as the ATS method is improved and can be applied to the anti-node we will be able to address this last evidence for second gap at the anti-node and settle the question of one or two gaps.
Figure 9.6: **Evidence of the Third Temperature Scale, \( T_{\text{Pair}} \) from [76]** A Temperature dependence of symmetrized EDCs at the anti-node for Bi2212 B Same symmetrized EDCs but normalized to high temperature C Same EDCs as A to show temperature evolution of gap (bars) D Temperature evolution of weight in the middle of the gap showing a clear transition between \( T^* \) and \( T_C \), which Kondo et al. interpret as the onset of pairing.
Chapter 10

Conclusion: A New Picture of the Cuprates

In this dissertation I have presented several new additions to the processing of ARPES spectra (detector nonlinearity correction, resolution effects, background removal), as well as physical concerns on sample preparation and cleaving. Taking all of these techniques into account I presented a new phenomenological model for electron-electron scattering, the Variational Fermi Liquid. The VFL describes the energy, temperature and doping dependence of the normal state scattering rates in a simple form with just three free variables. Finally, I have introduced the ARPES tunneling spectra (ATS). The ATS is a powerful new technique that allows us to separate the pair-breaking processes from all other scattering processes as well as resolve the pairing interaction simply and more accurately than previous techniques.

The findings of the ATS can be summarized by the following eight points:

(1) $\Delta$ is d-wave

(2) $\Delta_{Max}$ shrinks with increased doping.

(3) $\Delta$ closes above $T_C$ at the temperature $T_{Close}$ well below $T^*$ but near $T_{Onset}$

(4) $\Gamma_{ATS}$ is finite at $T = 0$, likely from in-plane impurities like Cu vacancies

(5) $\Gamma_0$ decreases with increased doping, probably from better screening from the increased carrier density

(6) For all angles so far studied $\Gamma$, is isotropic in contrast to the d-wave $\Delta$. 
(7) $\Gamma(T)$ is best described with an Arrhenius form, which suggests the dynamical pair breaking is governed by a high energy mode rather than a low energy one or electron-electron scattering.

(8) The doping dependence of said mode and its coupling to the electrons follow the superconducting dome suggesting it is integral to the superconducting state, and likely the upper branch of the spin resonance.

(9) $T_C$ seems integrally linked to when $\frac{\Delta(T_C)}{\Gamma(T_C)} = \pi$ suggests a threshold condition for the coherence of the superconducting pairs.

(10) The interplay between $\Delta$ and $\Gamma$ is responsible for much of the cuprates’ exotic physics, like Fermi arcs, near-nodal pseudogaps, and non-quasiparticle behavior.

These findings paint the cuprates in a much simpler light. Gone are the mysteries of the Fermi arcs, excessively large ARPES scattering rates, and the pseudogap. In its place we have a model of electrons being torn between coupling via a virtual spin resonance interaction to form Cooper pairs and those pairs being broken apart by either unitary impurity scattering or shaking off of a real spin excitation. This tug-of-war is governed by the competing energy scales representing pair formation, $\Delta$, and pair-breaking, $\Gamma$. Future studies will push the ATS to the anti-node and further into the normal state, as well as deepen the dopings and species of samples studied. These studies will advance this new picture of the interplay of $\Gamma$ and $\Delta$ in the cuprates and other materials.
Bibliography


