

## Chapter 1

### Introduction

The physical properties of solids are governed by electrons. Specifically, it is the electrons that are least bound to the ion lattice, those near the Fermi energy, which are able to move most easily about the solid carrying heat and current. In almost all materials, the electronic properties fall into one of three categories: metallic, insulating, and semiconducting, all of which are well described by band theory. Amazingly, even though there on the order of  $10^{24}$  electrons involved, the properties of most solids do not depend strongly on the interactions between these electrons. Band theory, which only deals with electron-electron interactions in an average way, captures the behavior of almost all materials with a high degree of accuracy. However, in a few states of matter, such as superconductors, the electron-electron interactions are of the utmost importance, and the full many-body problem must be taken into account in order to understand even the qualitative behavior of the material. These correlated electron systems, the most mysterious of which may be the high  $T_c$  superconductors, are at the forefront of modern condensed matter physics research.

The most common way to experimentally probe the electronic properties of solids is to observe how the electrons respond to being shaken, or driven, at different frequencies, which is accomplished by current transport measurements in the low frequency range (up to  $\sim 100$  GHz), and by optics for higher frequencies.

The drawback is that in order to extract information about the electron-electron interactions from these techniques, specific physical models of electron transport must be assumed. They are in a sense one step removed from the electrons they are trying to study. Angle-resolved photoemission spectroscopy (ARPES), on the other hand, is a technique which measures the electrons directly, and therefore requires fewer model assumptions in order to extract information about the electronic interactions. In an ARPES experiment, photons with sufficient energy to overcome the work function are used to eject electrons from the sample. Since the electron momentum is conserved in this process, the angular distribution of photoelectrons from a single domain crystal is representative of the initial momentum distribution of electrons in the solid. Measuring this photoelectron distribution is equivalent to imaging the electrons of the solid in momentum space. Improvements in electron spectrometer technology over the past decade have made it possible to not only measure the band structure and Fermi surface topology, but to observe fine structure in the electron self-energy, both the real and imaginary parts of which can be directly measured with ARPES. Perhaps the biggest drawback to the ARPES technique is surface sensitivity; the escape depth for electrons in typical ARPES experiments is only a few nm, and so the measured electronic structure may not be representative of the bulk crystal properties.

This thesis represents major advances in both angle-resolved photoemission and the physics of high  $T_c$  superconductivity. We have developed the first system to perform ARPES using a laser-based photon source, which offers many advantages over the synchrotron radiation sources usually required. In addition to being orders of magnitude smaller and less expensive to operate, the laser source offers far superior photon flux with reduced bandwidth. The relatively low photon energy of the laser also improves momentum resolution and bulk-sensitivity both by an order of magnitude. These improvements have enabled us to obtain

the clearest images yet of the electronic structure of the high  $T_c$  superconductor  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  (Bi2212), providing the most direct evidence yet for quasiparticles in the normal state.

*Chapter 2* is a general overview of the photoemission technique, with special attention paid to the issues important to this work. In particular, the low photon energy (6 eV) regime of the laser ARPES experiments has not yet been studied, and so it pushes the limits of our theoretical understanding of the photoemission process. The sudden approximation, in which one assumes that the photoelectrons are measured before the electron system in the sample has had time to relax, is expected to break down in this range. It is of critical importance to understand the nature of this possible breakdown.

*Chapter 3* is dedicated to the development of the laser-based ARPES system, including detailed discussions of the photon source, the analysis chamber, and the electron spectrometer. Special techniques and calibrations necessary for working at low energy are also discussed.

*Chapter 4* is an overview of superconductivity with discussions on both conventional BCS and high  $T_c$  superconductors. The section on BCS superconductivity is primarily a theoretical review, including some simple calculations using many-body quantum mechanics. It is not absolutely crucial to this thesis, but is relevant as there is general optimism that many of the methods of BCS will be useful in formulating a microscopic theory of superconductivity in the high  $T_c$  cuprates. The section on high  $T_c$  is an overview of the general phenomenology, with more emphasis on past experimental results.

In *Chapter 5* I present a brief study of sample surface ageing using a combination of laser ARPES and x-ray photoemission spectroscopy (XPS). Very high energy photons (1487 eV) are used for XPS, making it extremely surface sensitive in contrast to the bulk-sensitive 6 eV laser photoemission. This contrast allowed

us to study the effects of adsorbed gasses on the sample surface in a novel way, by comparing spectra from these two drastically different energy ranges.

*Chapter 6* is a laser ARPES study of optimally doped Bi2212, which represents perhaps our biggest contribution to the science of high  $T_c$  superconductivity. Contained in this chapter are the most detailed discussion of the specific techniques used to study Bi2212 with laser ARPES, including sample preparation and low energy  $\mathbf{k}$  conversion, as well as the most detailed description of the data analysis and fitting techniques which will be used throughout this thesis. I will compare laser ARPES results to those from higher energy synchrotron experiments, and discuss the possible breakdown of the sudden approximation. Kramers-Kronig consistency of the self-energy extracted from laser ARPES data is shown, and the measured scattering rates are compared to those measured with optics. We observe the first spectral peaks that are sharper than their binding energy, opening up the possibility of quasiparticles in the normal state. Finally, I use the spectral functions predicted by Fermi liquid theory and marginal Fermi liquid theory to fit the laser ARPES spectra.

In *Chapter 7* I will present laser ARPES studies of underdoped Bi2212 which are focussed on the superconducting state. The lineshapes are compared to those from optimally doped Bi2212 and a new peak-dip-hump fitting procedure is introduced to fit the underdoped data. Also presented is the highest resolution study of the superconducting gap symmetry, which is found to include higher order  $d$ -wave harmonics.

*Chapter 8* discusses the discovery of a temperature dependent nodal Fermi velocity in Bi2212. This is analyzed in the context of the coupling of electrons to collective modes, which although never explicitly discussed, has been previously observed with ARPES and optics. I will also present simulations of the temperature dependence of the self energy arising from electronic coupling to phonons

and spin fluctuations.

*Chapter 9* summarizes the scientific impacts of this thesis. I will discuss the future of laser ARPES, which shows great promise as a tool for the study of electronic interactions in solids.