Chapter 9

Summary and Future Directions

My PhD work has focused on two main goals: the realization of a viable ARPES system based on a laser photon source, and the use of this system to study electronic interactions in solid state systems. The first portion of this dissertation was dedicated to the design, construction and calibration of the ARPES system itself, including discussion of the special techniques required to overcome the difficulties inherent to working at the very low photon energies of laser-based sources. Much of my PhD career was in fact dedicated to the task of developing these techniques, none of which had been previously discussed in the literature. Today, our laser ARPES system not only offers increased bulk-sensitivity and reduced operating cost compared with synchrotron-based sources, but actually represents the state-of-the-art in resolution and overall data quality. The greatly increased photon flux of the laser also represents a considerable advantage since time is of the essence when performing ARPES experiments.

The second half of this thesis was dedicated to the study of the high T_c superconductor Bi2212, which is perhaps the material most heavily studied with high resolution ARPES. Directly relevant to the viability of the laser ARPES technique itself is the fact that the overall characteristics of the near-Fermi spectra, although much sharper and clearer, were found to be in qualitative agreement with the vast body of higher photon energy studies of this material. This is an indication that the laser ARPES experiments are indeed in the sudden regime with respect to the near-Fermi physics. This finding also lends strong support to the vast body of previous ARPES studies of Bi2212, as we now see the same overall picture near the Fermi surface with a probe that is significantly more bulk sensitive.

The improved resolution of laser ARPES enabled us to accurately fit EDCs with Lorentzians and to directly observe nodal quasiparticle-like peaks, reopening the possibility of applying some of the techniques of Fermi liquid theory to the normal state of the high T_c cuprates. As a natural extension of this discovery, I fit the nodal data with the ARPES lineshapes predicted by Fermi liquid theory and marginal Fermi liquid theory, both of which showed improvements over the simple Lorentzians. Although at this stage one cannot say with certainty if either of these theories is appropriate for the nodal cuprate physics, this work represents progress in the data quality and analysis techniques that will be required to do so.

In the study of underdoped Bi2212, we observed the emergence of a high energy hump in the EDCs. I introduced the PDH lineshape to fit this hump and tracked its spectral properties. Like the optimally doped Bi2212, these samples displayed quasiparticle-like peaks in the raw data, although not necessarily in the normal state. The superconducting gap symmetry was measured with very high momentum resolution in order to determine the range of the Cooper pairing interaction. In agreement with previous studies, significant amounts of second order *d*-wave harmonic were needed to fit the momentum dependence. Although some have argued that this may be due to impurity scattering, our data indicates that it is more likely to be an effect intrinsic to the pairing interaction. Underdoped samples with lower levels of impurity scattering, if possible to grow, would be an ideal way to resolve this issue.

We observed a strong linear temperature dependence to the nodal Fermi velocity at all doping levels, which may have important implications for transport in the cuprates. Analysis of this effect, in combination with the temperature dependence of the nodal dispersion kink, could shed light on the interactions at work. I simulated the temperature dependent effects of electronic coupling to various collective modes. According to these simulations the spectrum α^2F of the collective mode(s) responsible for the observed effects must have two qualities: it must have a sharp peak to produce the nodal dispersion kink, and it must have a broad tail to give a linear temperature dependence to the Fermi velocity. whether these qualities arise from a single collective mode, or many in harmony, is still unknown.

The work I have presented in this thesis is merely the beginning of what promises to be a very productive experimental technique for the study of electronic structure in solids. In our lab alone, no less than 3 new laser systems are currently under development for use as ARPES light sources. This includes a very high energy resolution 6.29 eV CW-laser source based on intra-cavity second harmonic generation.[1] Coupled with our new flowing-helium cryostat, this laser is capable of performing ARPES with sub 5 meV resolution. Similarly, a 3.5 eV quasi-CW laser is being used to generate low bandwidth 7 eV light, through second harmonic generation in the rare non-linear crystal $KBe_2BO_3F_2$ (KBBF) using the prism coupling technique.[2] The additional eV of photon energy greatly reduces concerns about angular resolution in the presence of stray magnetic fields.

While these new sources will likely become the next generation of laser photon sources for direct photoemission, the Ti:Sapphire system will be given the task of performing time-resolved pump-probe ARPES.[3] This will be the sole

[1] This development is being lead by Qiang Wang.
[2] This work is being lead by Fraser Douglas.
[3] This work is now being lead by Nick Plumb and Ted Reber.

technique capable of measuring the dynamics of the unoccupied states in a **k**-resolved manner. A pump pulse, below the photoemission threshold, will promote electrons to states above the Fermi energy. Following this, a probe pulse will photo-emit the hot electrons, which can then be measured using the ARPES spectrometer. Because the delay between these femtosecond pulses can be varied, we will be able to directly measure the lifetimes of the electronic excitations. This would be the most direct way to measure ImΣ, which we could then compare to our standard ARPES lineshapes. These devolvements, and the results of this thesis, indicate a bright future for laser ARPES in the study of electronic interactions in solids.