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Disorder and interactions in high-temperature superconductors

Sulangi, M.A.

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INTRODUCTION

Ever since their discovery over thirty years ago [18], the cuprate high-T_c superconductors have provided a unceasing torrent of mysteries that have bedeviled generations of physicists. The initial astonishment at the very high transition temperatures found for these materials—several times larger than the largest T_c hitherto known—was soon augmented by bewilderment about the fundamental nature of these materials. The very presence of superconductivity in these materials was itself surprising: the ground state of the parent compounds of these materials is known to be an antiferromagnetic Mott insulator, which disappears fairly quickly upon hole-doping and from which superconductivity emerges. The close proximity of the superconducting and magnetically-ordered phases to each other was a clear hint that strong correlations among the electrons in these materials play a central role, as conventional wisdom suggests that magnetism and superconductivity arise from different electron-electron interactions and are therefore unlikely to be found near each other in materials. The necessity of a strongly-interacting description of these materials became clearer upon further exploration of their phase diagram. At temperatures close to zero, with increasing doping one encounters the antiferromagnetic Mott insulator, a d-wave superconductor, and, eventually, a normal phase which appears to be well-described by Fermi-liquid theory. On the underdoped side there are various charge- and spin-ordered phases which appear to coexist—or compete—with the *d*-wave superconductivity.

When temperature is raised, a veritable Pandora's box of problems is unleashed. It is in fact at this finite-temperature regime where much of the most persistently confusing aspects of the cuprates are found. At low dopings, antiferromagnetic order persists at finite T. The *d*-wave superconducting state at low doping transitions, at T_c, into a mysterious phase called the "pseudogap," which, unlike a normal metal, features an extremely pronounced depletion of electronic excitations near the Fermi energy. The superconducting state reaches its highest T_c at a special value of the doping—"optimal doping"—and, at temperatures higher than T_c near optimal doping, the superconductor gives way to an extremely unusual metal whose anomalous properties defy any description in terms of Fermi-liquid theory. This "strange metal" is seen to have a linear-in-T resistivity, unlike that of a conventional metal, for which the resistivity scales with a higher power. The pseudogap in turn is found to cross over to the strange metal even at very high temperatures, with the crossover scale set by a doping-dependent temperature T^*. The strange metal occupies a fan-shaped segment of the doping-temperature phase diagram; on the overdoped side of this fan a return to more conventional Fermi-liquid-like behavior occurs.

A consistent and unified description of *all* of these phases of the cuprates is at the moment not known. Even the superconductor, perhaps the most well-understood phase of the cuprates, is still unusual. For one, it is more stable against disorder than Bardeen-Cooper-Schrieffer theory suggests. The *d*-wave symmetry of the order parameter means that Anderson's theorem [12] does not apply to this situation, and quenched disorder should rapidly kill the superconductivity. However, the inhomogeneities present in the cuprate materials happen to coexist with a stable d-wave superconducting state, thus suggesting that some decidedly beyond-BCS mechanism ensures its survival in this disordered setting. Another instance in which the cuprate superconductors deviate from the BCS expectation is in how the superfluid density ρ_s—which quantifies the stiffness of the superconducting condensate against twists in the phase of the order parameter—behaves. The underdoped cuprates have a very small superfluid density which is found to scale with T_c [166]. This suggests that fluctuations of the phase of the superconducting order parameter acquire an outsize importance in the underdoped cuprates, even inside the superconducting state, with the possibility that a "preformed-pairing" picture—*i.e.*, pairs form below T^*, but acquire phase coherence only upon reaching T_c—may explain the pseudogap phase above T_c [44]. This is in stark contrast to the BCS picture, where the characteristic temperature scale of the phase fluctuations is much larger than T_c, leading to the relative unimportance of these fluctutations within the superconducting state. Finally, angle-resolved photoemission spectroscopy (ARPES) experiments suggest that the gap does not close at T_c, as BCS theory predicts; instead, it *fills* [140]. T_c appears to be set by the temperature at which the gap and the quasiparticle scattering rate energy scales cross over into each other, and the temperature at which the gap ultimately closes is higher than T_c. Nevertheless, despite these non-BCS-like features, the d-wave superconducting state is known to host coherent excitations which, as seen in ARPES [82, 176] and scanning tunneling spectroscopy (STS) experiments [70, 112], behave exactly as d-wave Bogoliubov quasiparticles do.

The contrast between the comparatively well-understood superconducting state and the proximate strange metal near optimal doping cannot be any more stark. Because the strange metal is not a Fermi liquid, there is no sense in which the normal-superconducting transition is describable by anything resembling BCS theory. However, it is possible to understand many features of this anomalous state via the marginal Fermi liquid theory proposed by Varma *et al.* [173]. In this phenomenological model, the plain-vanilla Fermi liquid is augmented by a momentum-independent but frequency-dependent self-energy whose imaginary part depends as T when the temperature is greater than the frequency. This has rather drastic consequences: long-lived quasiparticles, the backbone of Fermi-liquid theory, cease to exist, as the quasiparticle weight $Z \to 0$ at the Fermi surface as $T \to 0$. While the microscopic origin of this behavior is not known, on an effective-field-theory level this succeeds in reproducing the strange transport anomalies present in the cuprates. In addition, ARPES experiments find that the strange metal features a very incoherent spectrum whose behavior could be reasonably fit into the marginal-Fermi-liquid description [2]. However, it remains an open problem how coherent quasiparticles in the superconducting state form upon moving from the strange metal, where the excitations are incoherent, and how the pseudogap transitions into both the strange metal and the superconductor.

The landscape of the phases of the cuprates, as outlined above, is rich, complex, and, four decades on, still incredibly confusing. Despite this rather daunting state of affairs, this thesis will try to describe portions of the phase diagram of these materials. Given the immense difficulty of constructing a global theory of the cuprates, a more barebones and phenomenologically-minded approach can help illuminate which bits of physics are important and which are only of secondary importance. Much insight can be derived by considering fairly simple but well-understood models of these phases which can nevertheless be augmented by bells and whistles that account for deviations from the models we started out with. The marginal Fermi liquid theory of the strange metal is perhaps the paradigmatic example of this approach: the Fermi-liquid starting point of the model is weakly coupled, but the addition of the self-energy incorporates the nontrivial effects of interactions, leading to the destruction of long-lived quasiparticles in the theory. Also, the evidence from spectroscopy suggesting that coherent quasiparticles are present in the superconductor allows considerable leeway in treating the superconducting state as a mean-field BCS superconductor with d-wave symmetry.

It is in this spirit that this thesis will examine both the superconducting and the normal state of the cuprates. A dominant theme underlying the work presented here is the nontrivial effect of disorder on various electronic properties of these materials. Disorder plays two distinct, almost antithetical roles in the cuprates, but it is often taken for granted how interrelated these two are. First, in the limit of weak disorder, it acts as a probe of the underlying electronic structure of these materials. In a normal metal, the presence of an impurity induces Friedel oscillations—which are simply modulations in the local density of states—whose spatial structure reveals details about the Fermi surface [31, 159, 71, 137]. The situation in the cuprates is no different. These Friedel oscillations have been observed in the superconducting state of the cuprates using STS, leading to the phenomenon dubbed "quasiparticle scattering interference" (QPI) [70, 112, 61, 90, 50]. The modulations found in the real-space differential conductance maps from STS can be Fourier-transformed, revealing a rich set of dispersive peaks in the power spectrum whose behavior can be used to reconstruct the band-structure details of the cuprates [182, 25]. Importantly, the quantum-mechanical-wave interference underlying this phenomenon illustrates how the Bogoliubov quasiparticles are well-defined and coherent excitations [189]. Second, under some circumstances it generates low-energy electronic states in the superconducting state, in the process irrevocably altering the electronic spectrum of the clean case [68, 100]. The presence of a finite density of states at the Fermi energy deep in the superconducting phase has long been known throughout the cuprate family from specific heat experiments, and standard lore has it that these are generated by disorder [115, 116, 144]. In addition, doping these materials by zinc—a strong local scatterer—leads to impurity resonances being generated near the Fermi energy, seen vividly also by STS [129, 16, 17]. .

Given that we know with definiteness that the cuprates are macroscopically disordered materials, it becomes imperative to consider disorder both as a probe of electronic structure *and* as an orgin of low-energy excitations seen in specific heat. The situation is complicated even further by subtleties present in the very nature of disorder in the cuprates. In particular, the copper-oxide planes—which host the physics of most relevance to experiments—are clean. Aside from rare defects which are thought to be Cu vacancies, no strong impurities are seen within the copper-oxide planes using STS, due to the strong copper-oxygen bonding present. It is possible to induce these strong impurities via chemical substitution of zinc or nickel atoms, but these would necessarily result in resonances near the Fermi energy which are not seen in cuprates without Zn or Ni dopants. What appears to be the case instead is that dopants located in the insulating layers adjacent to the copper-oxide planes are the source of disorder—but unlike the aforementioned Zn or Ni dopants, which act as pointlike scatterers, these would generate a smoother and longer-ranged disorder potential which affects the electrons in the copper-oxide planes [2, 126, 124, 125, 161]. However, unlike pointlike forms of disorder, these smoother disorder potentials are far less amenable to analytical treatment, and are accessible only with large-scale numerical methods.

It cannot be emphasized enough that the aspects of disorder considered in this thesis remain central to some staggeringly persistent mysteries about the cuprates. One example of this is "QPI extinction" [90, 50]. STS experiments on underdoped cuprates in the superconducting state, across a fairly wide doping range, show the usual signatures of QPI—modulations in the differential conductance maps, prominent peaks in the power spectrum—right until the bias voltage is such that the tips of the contours of constant energy cross the antiferromagnetic zone boundary—*i.e.*, the portion of the Brillouin zone enclosed by the four lines connecting $(0, \pm\pi)$ and $(\pm\pi, 0)$. Beyond that voltage, most of the dispersing peaks seen in the power spectrum vanish, and what do remain are seen not to disperse as the voltage is further changed. Why this happens is not presently known, and numerous explanations using a mean-field free-fermion description—*e.g.*, unusual impurities [176], coexisting spin-density wave order [11]—appear unsatisfactory, or are simply infeasible, due to the lack of supporting experimental evidence for them. If one takes the results at face value, this phenomenon is a remarkably lucid demonstration of the breakdown of the quasiparticle description as one nears the antinodes—the famous "nodal-antinodal dichotomy" in action—but to bolster that interpretation, it first has to be understood what precisely is happening at the point where these peaks are extinguished, and here STS finds itself in disagreement with results from ARPES, for reasons that are not completely understood. Many aspects of this problem remain unclear even to this day.

Another example of this is a recent set of specific heat measurements on underdoped cuprates in the presence of magnetic fields [144]. The cuprate samples used in these experiments are some of the cleanest known—far more orderly than most other members of the cuprate family. Despite this, a residual linear-in-temperature term in the specific heat at zero field is seen in the data, which is indicative of a nonzero density of states at the Fermi energy. It is not trivial to attribute this simply to disorder, as the coefficient of this T-linear term—which is proportional to the density of states at $E = 0$—is larger than that seen in ostensibly dirtier cuprates. It has almost been taken for granted since the early years of the cuprates that disorder generates these low-lying excitations detected in specific heat. So, what is happening here? There is as yet no definitive answer, as the cuprates in question are not especially amenable to surface probes such as ARPES and STS, and thus one cannot examine the copper-oxide planes directly to see what the underlying source of the low-energy electronic excitations is. (NMR is a local probe which could in principle measure various local quantities directly, but one has to note the fact that a magnetic field is present in these experiments, and thus the decidedly nontrivial effects of this field on the superconducting state have to be carefully taken into account.) One idea that has been proposed is intra-unit-cell loop-current order which coexists with the d-wave superconducting state [21, 4, 88, 181], but this idea appears to run into difficulties when one tries to constrain the parameters of the model from the observed data. Disorder due to dopants within the buffer layers may be another way out—and as a matter of fact this is discussed in great detail in this thesis—as the dopants present in these cuprates could remain away from the copper-oxide planes while still disordering the electrons within the plane via a random screened Coulomb potential [162]. However, as we have only indirect means of probing disorder and any putative coexisting order, it is safe to say that there is still no firm resolution to this problem.

One admittedly heuristic takeaway from these two examples is that whenever disorder is involved in the cuprates, there is more than meets the eye. In the first case, QPI is Friedel oscillations, but it also is much more than that. For one, the system is macroscopically disordered: there is no single isolated impurity, but rather the disorder—wherever it may come from—is of a distributed nature. Second, the tunneling process between the STM tip and the copper-oxide plane is highly nontrivial [109, 93]. Third, despite these two considerations, the peaks seen in experiment are somehow well-defined—almost miraculously so. In the second case, specific heat experiments provide a very beautiful probe of *all* low-energy excitations, but are blind to the precise mechanism through which these excitations are generated. In addition, in the case of the cuprates, what constitues "disorder" can be ambiguous, thanks to the complex layered structure of these materials. The dopants reside off the plane, but for some cuprates the question of *where* exactly these reside is difficult to resolve. There are also non-stoichiometric alterations to the structure of the compounds, unrelated to hole doping, which induce further off-plane disorder [42]. All of these complicating factors mean that disorder is not a simple matter at all. There is only so much one can understand about its impact on the electronic properties of the cuprates without taking into accout all these tricky caveats. One thus sees the need to thoroughly revisit disorder and to see the extent to which its effects manifest themselves on experimentally-measurable quantities.

These considerations motivate Chapters 3 and 4 of this thesis, which reexamine respectively the imprint of disorder on the local density of states (as seen by STS) and the quasiparticle density of states at the Fermi energy (as seen by specific heat experiments) in the d-wave superconducting state. For the first case, the vast majority of prior theoretical work on QPI has centered on the case of a single isolated pointlike impurity [182, 25, 125, 176, 93]. The extent to which the experimental evidence for QPI can be reproduced using distributed models of disorder is in general not clear. For the second case, the prevailing explanation for the finite DOS at the Fermi energy in the cuprates is the so-called "dirty *d*-wave" theory, which implicitly assumes both a dirty copper-oxide plane and a local, pointlike model of disorder [54, 68, 100, 39, 14, 13], and as such fails to describe the realistic situation in which the copper-oxide planes are clean and a smooth disorder potential sourced by off-plane dopants is present.

Chapter 2 contains a review of the various phenomena seen in ARPES and STS, in addition to a brief overview of the basics of these two methods. We consider three phases of the cuprates individually—the superconductor, the pseudogap, and the strange metal—and exhaustively describe much of their known phenomenology. We will also discuss in detail some of the theories that have been put forth to explain these various phenomena, and point out regimes where these proposed theories fail.

In Chapter 3 we revisit quasiparticle interference in the cuprates and try to see if the strikingly sharp peaks seen in the experimental power spectra can be reproduced by an exhaustive array of models of distributed disorder—examples include an ensemble of weak pointlike scatterers, random chemical potential disorder, and smooth disorder—and the incorporation of a model of the STM tunneling process. What is found is that weak pointlike disorder and random chemical potential disorder best reproduce both the real-space and the Fourier-transformed spectra seen in experiment; that smooth disorder fails to fully reproduce the experimental power spectrum, as large-momentum scattering is suppressed in that particular case; and that the peaks in experiment are *sharper* than any of our simulations see, in a surprising reversal of what we usually expect.

Chapter 4 is devoted to a very thorough examination of various kinds of disorder and their impact on the quasiparticle density of states near the Fermi energy as the amount of disorder is increased.

While strong pointlike scatterers and random chemical-potential disorder do lead to a finite DOS, agreement with experiment is achieved only when the amount of disorder is unphysically large. Meanwhile, disorder due to off-plane dopants is found to lead to a realistic value of the DOS at the Fermi energy while leaving much of the *d*-wave state in intermediate and higher energies largely unaffected, and in addition sharp resonances *at* the Fermi energy are seen when the concentration of these smooth scatterers is very large. The localization length is also studied for various models of disorder, finding that for all three models of disorder the quasiparticles at the Fermi energy are localized, and that the dependence of the localization length on energy depends sensitively on the type of disorder present.

Chapter 5 spotlights QPI once more, this time focusing on the effects of self-energies on the LDOS power spectrum, with the advantage of knowing, from Chapter 2, that the single weak pointlike scatterer does lead to a phenomenologically accurate power spectrum. After discussing the effect of self-energies on the spectral function and the DOS, we proceed to analyze cases of interest to the cuprates. In the superconducting case, we study the "gap-closing/filling" phenomenology seen in ARPES experiments and attempt to analyze the extent to which STS measurements can also see this phenomenon, and contrast these with the BCS case, wherein the gap closes but does not simultaneously fill. The peaks seen in the superconducting power spectra are found to be rather sensitive to the amount of broadening present, with the peaks smearing and becoming incoherent at large self-energies. We also study the normal-state LDOS power spectra, assuming that the strange metal is well-described by a marginal Fermi liquid, and find that a key signature of this state is the presence of broad caustics in the power spectrum describing scattering wavevectors between points along the Fermi surface. The main difference between the marginal

Fermi liquid and the ordinary Fermi liquid is found to be simply in the amount of broadening present in these caustics: the LDOS power spectra of a marginal Fermi liquid has much more broadening than that of the ordinary Fermi liquid.

Finally, a summary of our results is shown in Chapter 6, along with a lengthy discussion of potential future directions, both experimental and theoretical, in the study of the cuprates.