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PENETRATION DEPTH STUDIES IN Ni AND Zn DOPED YBa$_2$Cu$_3$O$_{7-\delta}$

DISSERTATION

Presented in Partial Fulfillment of the Requirements for the Degree Doctor of Philosophy in the Graduate School of The Ohio State University

By

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To everyone who supports the cause of Jesus Christ
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Chapter I

Introduction

The discovery of the cuprate class of superconductors in 1987 boosted the highest known transition temperature by a factor of 4 in less than a year.[1,2] Most of the previously known superconductors with the exception of the heavy fermion superconductors and some organic superconductors were well explained by assuming electron-phonon coupling. This mechanism was often believed to be incapable of producing $T_C$ values as high as seen in the cuprates.[3] The discovery of the "high-$T_C$" superconductors therefore led to the development of theories based on other electron coupling mechanisms that predict a $d$-wave symmetry to the order parameter, rather than the usual BCS isotropic $s$-wave result.[4] The magnetic field penetration depth [$\lambda(T)$] would be expected to have a quite different temperature dependence if the order parameter possessed $d$-wave rather than $s$-wave symmetry. In addition, the high $T_C$ values, coupled with the small coherence lengths, could lead to the existence of a large temperature regime near $T_C$ in which superconducting fluctuations are important.[5] Measurements of the penetration depth (i.e. the complex conductivity) near $T_C$ would also be a good probe of the type of fluctuations present in cuprate superconductors.

This thesis focuses on the temperature dependence of $\lambda(T)$ in YBa$_2$Cu$_3$O$_{7-\delta}$ thin films. The standard $s$-wave BCS theory of superconductivity predicts that $\lambda(T) - \lambda(0)$ has
a thermally activated behavior at low temperatures because of the existence of a real energy gap in the electronic density of states. $d$-wave theories predict $\lambda(T) - \lambda(0) \propto T$, a dependence that has been seen in crystals,[6] and some films.[7,8] Early results on films had shown different results than those obtained in YBa$_2$Cu$_3$O$_{7-\delta}$ crystals,[9-12] leading to the uneasy feeling that the results on films were untrustworthy. More recently, it has been shown that the differences can be explained in terms of the effects of small amounts of disorder in a $d$-wave superconductors,[9,13] leading to the conclusion that the films are slightly more disordered than crystals, but otherwise trustworthy.

This thesis extends the $\lambda(T)$ measurements to higher levels of disorder by intentionally doping small amounts of Ni and Zn (2-6%) for the Cu atoms. Superconductivity in the high-$T_c$ compounds is widely believed to occur in the CuO$_2$ planes. As Ni and Zn substitute preferentially on the plane Cu sites, they should have a larger effect on the superconducting properties than, for example, Co which goes into the Cu chain sites.

Also, the mean-field BCS theory predicts that $1/\lambda^2(T)$ should be linear in $(T_c - T)$ near $T_c$. Recent measurements on crystals have shown $1/\lambda^3(T)$ to be linear in $(T_c - T)$,[14] implying the existence of 3D-XY critical fluctuations near $T_c$, which might be expected to exist in YBa$_2$Cu$_3$O$_7$ given its high transition temperature and small coherence length.

In this thesis, I have prepared pure and Ni-doped YBa$_2$Cu$_3$O$_{7-\delta}$ films by codeposition and helped prepare Ni-doped films by laser ablation. I have developed an apparatus for measuring $\lambda(T)$ by measuring the change in the mutual inductance of two
coils on opposite sides of the films. This is the only such apparatus capable of measuring the absolute value of $\lambda(0)$ in films, rather than just the change in $\lambda(T)$, and the combination of accuracy and precision on each data point makes this the best apparatus for measuring $\lambda(T)$ in the world. I measure $\lambda(T)$ in pure, Ni and Zn-doped $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ films to determine the effect of disorder on $\lambda(0)$ and distinguish between $s$ and $d$-wave theories. $\lambda(0)$ increases rapidly in the doped films, and the small slope $d\lambda/dT$ at low temperatures is used in Chapter VII to eliminate phase fluctuations as an explanation for this slope. Finally, I show measurements of the complex conductivity near $T_c$ to determine the nature of the fluctuations near the transition.
Chapter II

The nature of the order parameter in YBa$_2$Cu$_3$O$_{7.8}$

The nature and symmetry of the order parameter in the high-T$_c$ superconductors has been a subject of much debate. All superconductors discovered prior to 1987 had been well described by the standard BCS theory, which postulates an attractive interaction between electrons at the Fermi energy. The interaction potential was assumed to be independent of energy and direction, which led to a self-consistency equation for the order parameter that was also independent of direction. This leads to an energy gap that is the same size in all directions in k-space and an order parameter whose phase is the same in all directions. No electronic states exist below $\Delta$, and the electronic density of states is changed from the usual constant $N(0)$ to $N(0)E/(E^2 - \Delta^2)^{1/2}$ above $\Delta$.[15]

Relaxing the assumption that the interaction potential is independent of energy and direction leads to the Eliashberg equations, which give an "energy gap" that depends on energy. The energy gap will take advantage of the potential to minimize the energy of the superconductor by becoming larger in some directions and smaller in others. This is the situation in lead, where the minimum gap may be as small as 10% of the maximum gap.[16,17] Figure 1 shows the angle-dependence of the order parameter for several different s-wave states. The top figure is an isotropic s-wave state, the middle is a slightly anisotropic state, the third is a state where the minimum gap is 10% of the
Fig. 1. Dependence of the order parameter on direction of $k$ for various anisotropic $s$-wave states.
maximum gap, and the fourth is a state where the gap goes to zero in certain directions. Figure 2 shows the angle-integrated densities of states that correspond to these order parameters.

If the interaction potential actually changes sign in different directions in k-space, the "gap" or order parameter can also change sign to take maximum advantage of the interaction potential. A sign change corresponds to a phase change of 180 degrees, so the phase of the order parameter depends on direction. The symmetry of the order parameter determines whether the superconducting state is classified as $p$-wave, $d$-wave, or even higher wave state. Figure 3 shows the angle dependence of the order parameter for the $d_{x^2-y^2}$ state that may exist in the high-$T_c$ superconductors and the density of states that corresponds to this order parameter.

The BCS theory assumed that the attractive interaction potential comes from the electron-phonon interaction. It seemed unlikely, though, that this coupling mechanism could produce the high $T_c$'s of the high temperature superconductors. A much more recent theory by Monthoux, Balatsky and Pines[4] suggests antiferromagnetic spin fluctuations as the origin of the attractive interaction in the high-$T_c$ superconductors. Their theory unambiguously predicts that the pairing state in the high-$T_c$ superconductors should be $d_{x^2-y^2}$. By this time, some evidence was already pointing to a $d$-wave state in the high $T_c$ superconductors, but now the search began for experimental tests to clearly distinguish between $s$ and $d$-wave superconductors.

There are several types of experiments that can distinguish between $s$-wave and $d$-wave states. The first type of experiments examine the low-temperature behavior of
Fig. 2. Electronic densities of states corresponding to the order parameters in Fig. 1.
Fig. 3. $d_{x^2-y^2}$ order parameter and density of states.
various transport properties. The second type of experiments are those that are directly sensitive to the directional dependence of the phase of the order parameter, and the third type examines the sensitivity of the superconducting state to disorder. Examples of these three types of experiments are given below.

The first type of experiments examine the low-temperature behavior of transport properties. Because the energy gap goes to zero in some directions in k-space, the density of states for $d$-wave pairing states has a power law behavior in energy for the lowest excitations, rather than the usual energy gap. Because of this, the low temperature dependence of properties such as the specific heat, penetration depth, ultrasonic attenuation, and NMR relaxation rate is a power law in temperature. Since a thermally activated temperature dependence is expected for isotropic $s$-wave superconductors, these measurements provide a method of distinguishing between $s$-wave and higher wave states. These power laws depend only on the energy dependence of the density of states, and have nothing to do with the phase of the order parameter. An $s$-wave state that is sufficiently anisotropic as to have a zero, or nearly zero, gap in some k-space directions, would also have a gapless, or nearly gapless, density of states, and would also produce the temperature dependences expected of higher partial-wave states.

NMR measurements provided some of the earliest evidence of this type that the High-$T_c$ superconductors might be $d$-wave. Imai et al.[18] and Martindale et al.[19] find that the relaxation rate below $T_c$ cannot be fit by the usual $s$-wave BCS dependence (i.e. a coherence peak just below $T_c$ and $1/T_1 \propto e^{-\Delta/T}$ at the lowest temperatures). Instead, they find no coherence peak and power law behavior (i.e. $1/T_1 \propto T^3$) at the lowest
temperatures. This suggests a line node in $\Delta(k)$ and a density of states that is linear in energy at low energies, all of which are expected from $d_2\gamma 2$ pairing. The Knight shift \cite{20,21} is also better explained in terms of a gap with line nodes.

Infrared reflectance measurements\cite{22-26} have also been used to probe the density of states in high-$T_c$ superconductors. In conventional $s$-wave superconductors, there should be no absorption of light until the frequency $\omega$ is greater than the gap frequency $2\Delta/\hbar$. As there is no gap in the density of states, $d$-wave superconductors would allow absorption at any frequency. The situation is complicated, however, because the high-$T_c$ superconductors are in the clean limit, and the amount of absorption expected at $2\Delta/\hbar$ is small and difficult to distinguish from no absorption at all. The addition of impurities to increase the scattering rate would make the gap visible if it were an $s$-wave superconductor.\cite{27} No gap has been seen, either in pure,\cite{22} Ni-doped\cite{23} or Zn-doped,\cite{24} or ion irradiated YBa$_2$Cu$_3$O$_{7.4}$,\cite{25} or irradiated Bi$_2$Sr$_2$CaCu$_2$O$_{8.}$\cite{26} A discussion of the effect of dopants on $d$-wave superconductors is left until later in the chapter.

Angle-resolved photoemission spectroscopy can actually probe the directional dependence of the density of states. Shen et al. find an energy gap that is strongly anisotropic, suggestive of $d$-wave superconductivity.\cite{28} Again, this measurement is only sensitive to the magnitude of the order parameter and an anisotropic $s$-wave state would produce the same result.

Measurements of the magnetic field penetration depth also fall into this category of experiments. Initial measurements of $\lambda(T)$ were interpreted as consistent with $s$-wave
superconductivity,[29] but the low-temperature behavior was not closely examined. More recent measurements that paid closer attention to the low-temperature behavior showed inconsistencies between film data and crystal data. Film data generally showed \( \lambda(T) - \lambda(0) \propto T^2,[9-11] \) while the best crystal data showed \( \lambda(T) - \lambda(0) \propto T.[6] \) The crystal data suggest a line of nodes in \( \Delta(k) \), and both results are inconsistent with \( s \)-wave superconductivity and suggest a gapless density of states. The disagreement between crystal and film data will be discussed in greater detail in chapter 3.

All of these measurements are sensitive only to the energy dependence of the density of states. A strongly anisotropic \( s \)-wave superconductor could have a density of states similar to that of a \( d \)-wave superconductor, and none of these experiments would distinguish between these two states. Very recently, experiments have been performed that are sensitive to the phase of the order parameter in different directions. These experiments are superior in the sense that they can distinguish between a \( d \)-wave state and a strongly anisotropic \( s \)-wave state.

In the pioneering experiment, Wollman et al.[30] made a SQUID with Josephson junctions between a lead film and the two perpendicular faces of an untwinned \( \text{YBa}_2\text{Cu}_3\text{O}_{7.8} \) crystal. In an \( s \)-wave superconductor, the Josephson coupling is independent of direction, and the SQUID should exclude flux from its interior, or allow currents to flow so that the flux inside the junction is an integral number of flux quanta. The critical current of the SQUID would be a maximum at zero magnetic field. A \( d \)-wave superconductor would have different coupling in different directions. The ground state of the SQUID therefore has a half-integral number of flux quanta inside the junction. The
critical current would be a minimum at zero field, and rise as the field is slowly increased. Wollman et al. find that the SQUID behaves as expected for a \( d \)-wave superconductor. These experiments have been extended by others to look for the half-integral flux,[31-32] with the consistent conclusion that the phase of the order parameter depends on direction, and is therefore inconsistent with even anisotropic \( s \)-wave states.

A final class of experiments that can distinguish between \( d \)-wave and even strongly anisotropic \( s \)-wave states probes the effect of scattering on the superconducting density of states. An \( s \)-wave state is not sensitive to the addition of nonmagnetic impurities until the mean free path is on the order of the coherence length, \( \xi \approx 15 \text{ Å} \) in \( \text{YBa}_2\text{Cu}_3\text{O}_{7-\delta} \).[27] The undoped high-\( T_c \) superconductors are therefore in the clean limit. In isotropic \( s \)-wave superconductors, the primary effect of impurities is to change the matrix element for transitions from one state to another. In anisotropic \( s \)-wave superconductors, impurity scattering causes the gap to become more isotropic as information about the gap in one direction is carried into another direction with the scattered electrons.[33] \( s \)-wave states that are sufficiently anisotropic to be gapless would develop an actual gap in the density of states as impurities are added, and the transport properties would regain the BCS activated low-temperature behavior.

All impurities are pair-breaking, however, in \( d \)-wave superconductors, which leads to drastic changes in the density of states in such materials.[13,34] Even at small impurity concentrations the density of states becomes finite at \( E = 0 \), and the peak at \( E = \Delta \) is drastically reduced. This causes properties like the specific heat and infrared
reflectance to approach their normal state values when the scattering rates are equivalent to only a few percent of dopant substituted for Cu in YBa$_2$Cu$_3$O$_{7.4}$.

Measurements of the specific heat in Fe-doped YBa$_2$Cu$_3$O$_{7.4}$[35] find that the specific heat jump near $T_c$ decreases by about a factor of 3 at 2% Fe substituted for Cu, and is undetectable at 4% Fe. 1% Zn decreases the specific heat jump by factor of 2, and 5% Zn moves it below detectability.[36] Cr,[36,37] Li,[38] Ni,[39] and Al[39] substitutions all have similar effects. Direct comparison with the low-temperature specific heat data is complicated by the fact that $T_c$ is nearer the Debye temperature in YBa$_2$Cu$_3$O$_{7.4}$ than in lower $T_c$ superconductors. Therefore, a large phonon background contribution has to be identified and subtracted off to determine the effect of dopants on the electron density of states. Data of Loram et al.[40] on bulk sintered samples of Zn-doped YBa$_2$Cu$_3$O$_{7.4}$ indicate a gapless density of states. The superconducting density of states at the Fermi energy, $N_s(0)$, in 3% Zn-doped YBa$_2$Cu$_3$O$_{7.4}$ appears to be about 60% of its normal state value.

Sumner et al.[23] find that the infrared reflectance of 2% and 4% Ni-doped YBa$_2$Cu$_3$O$_{7.4}$ films in the superconducting state can be fit by assuming no change at all in the density of states as the film becomes superconducting. Similar results are obtained by Kim et al. on Zn-doped films,[24] Mandrus et al. on ion irradiated YBa$_2$Cu$_3$O$_{7.4}$,[25] and Basov et al. on irradiated Bi$_2$Sr$_2$CaCu$_2$O$_8$.[26] None of these workers find the perfect reflectance below $2\Delta/h$ and sudden drop at the gap frequency that would be expected from an isotropic (or anisotropic) $s$-wave superconductor in the dirty limit.
Penetration depth measurements also probe the density of states. Measurements of $\lambda(T)$ are simpler to interpret than specific heat or infrared reflectance measurements because it is a purely superconducting property with no normal state analog. Specific heat measurements are complicated by the necessity of subtracting the phonon and other background contributions. Infrared reflectance measurements are complicated by the existence of an "mid-infrared band" that may change with dopant concentration, and by substrate effects when thin films are used. Therefore, we turn to a more detailed analysis of penetration depth experiments in the next chapter.

So far, the available evidence suggests an non-BCS electronic density of states with no gap, a feature of both $d$-wave and some anisotropic $s$-wave states. The best experiments for distinguishing these states are those that are sensitive to either the phase of the order parameter, or those that can measure the change in the density of states with disorder. In chapter VI, this thesis examines the effect of disorder induced by Ni and Zn doping in the CuO$_2$ planes to distinguish between these possible states.
Chapter III

Previous penetration depth measurements in YBa$_2$Cu$_3$O$_{7-\delta}$

Measurements of the temperature dependence of the magnetic field penetration depth $\lambda(T)$ provide some of the best evidence for unconventional pairing in YBa$_2$Cu$_3$O$_{7-\delta}$. $s$-wave pairing would give thermally activated behavior with $\lambda(T) - \lambda(0) \propto e^{-\Delta H T}$. $d$-wave pairing with line nodes (e.g. a $d_{x^2-y^2}$ state) should give a linear temperature dependence (i.e. $\lambda(T) - \lambda(0) \propto T$). While other measurements like infrared reflectance, Raman spectroscopy, specific heat, etc. give evidence for excitations below the gap, it is possible that these excitations are part of the nonsuperconducting background, either phonons, bound electrons, or some other type of quasi-particle. The penetration depth, on the other hand, is purely an effect of the superconducting electrons. No nonsuperconducting background excitations could be responsible for non-activated behavior in $\lambda(T)$.

Initially, the interpretation of the measurements of $\lambda(T)$ was confusing. Much data was said to be consistent with $s$-wave superconductivity because it could be fit to the Gorter-Casimir two-fluid form $\lambda^2(T) = \lambda^2(0) [1-(T/T_c)^4]$. However, this function is only an approximation to the actual $s$-wave dependence, and is a better fit to strong-coupling $s$-wave than to weak-coupling $s$-wave. It was not appreciated until later that because this function has a $T^4$ dependence at low temperatures rather than a thermally activated dependence it doesn’t distinguish well between $s$-wave and more exotic pairing
states. In fact, if the actual \( \lambda(T) \) in \( YBa_2Cu_3O_{7.4} \) followed the Gorter-Casimir form at low temperatures, it would show that there was, in fact, no gap in \( YBa_2Cu_3O_{7.4} \). And \( d \)-wave penetration depth functions can be constructed that follow the Gorter-Casimir form at higher temperatures.

Much of the earliest data was shown by Annett et al. to be consistent with \( \lambda(T) - \lambda(0) \propto T^2 \) at the lowest temperatures.\[^{[29,45]}\] The techniques included dc magnetization in single crystals,\[^{[41]}\] \( \mu \)SR in single crystals,\[^{[42]}\] and mutual inductance measurements on thin films.\[^{[43,45]}\] A few early workers\[^{[46,47]}\] reported \( \Delta \lambda \propto T \). Although none of the measurements showed activated behavior, the inconsistent results made interpretation difficult.

The situation became clearer with the unambiguous measurement of \( \lambda(T) - \lambda(0) \propto T \) from 2 - 40 K in \( YBa_2Cu_3O_{7.4} \) crystals by Hardy et al.\[^{[6]}\] It was also noted that even small amounts of impurities would distort a \( d \)-wave density of states and produce \( \Delta \lambda \propto T^2 \).\[^{[13]}\] It seems reasonable that films and the earlier crystals had residual amounts of disorder that destroyed the expected linear dependence.

Hirschfeld and Goldenfeld explored theoretically the effect of small amounts of disorder on the density of states and penetration depth of \( d \)-wave superconductors.\[^{[13]}\] They found that all scattering is pairbreaking, leading to sharp reductions in \( T_c \) and causing the density of states to be finite at \( E = 0 \). This causes a rapid decrease in the density of superconducting electrons \( n_s \), or a correspondingly rapid increase in the zero-temperature penetration depth \( \lambda(0) \). It also causes a crossover in \( \lambda(T) \) from linear in \( T \) at high temperatures to \( T^2 \) at low temperatures with a crossover temperature \( T^* \) that
depends only on the scattering rate. The increase in $\lambda(0)$ and the crossover temperature $T^*$ are functions of a disorder parameter, which depends on the scattering rate and order parameter.

With this in mind, Lee et al. measured the penetration depth in several pure YBa$_2$Cu$_3$O$_{7.4}$ films. They discovered that $\lambda^2(0)/\lambda^2(T)$ between 25K and 90K agreed quite well with the temperature dependence obtained by Hardy et al., given a slightly larger value of $\lambda(0)$ in the films. Specifically, they found a linear region in $1/\lambda^2(T)$ above 25K, with a crossover to $T^2$ behavior below $T^* = 25$ K. The increase in $\lambda(0)$ and curvature of $\lambda^2(0)/\lambda^2(T)$ are what would be expected for an order parameter $\Delta_0/kT_c$ between 3 and 3.5, and a scattering rate of $\Delta_0/\hbar$, about the scattering rate expected for .5% Ni or Zn doping. The odd data on films were now capable of being explained quantitatively within the framework of $d$-wave superconductivity. Figure 4 shows the density of states calculated by Borkowski and Hirschfeld at this level of disorder for comparison to the pure $d$-wave density of states.

Bonn et al. then measured the effect of small amounts of Zn (0.15%-0.31%) and Ni (0.15%-0.7%) doping on the temperature dependence of $\lambda(T)$ at temperatures below 20K. They find that Zn doping also produces a crossover in $\lambda(T)$ from linear to $T^2$ below $T^*$. They find $T^*$ for the pure, .15% Zn and .31% Zn sample were 3, 10, and 28K respectively. These crossover temperatures are in reasonable agreement with the predictions of Hirschfeld and Goldenfeld given the impurity scattering rate deduced from microwave measurements of the conductivity and an order parameter $\Delta_0/kT_c \approx 3.0$. Bonn et al. were unable to measure $\lambda(0)$ and reported only $\Delta \lambda$, so no comparison of the
Fig. 4. Density of states from Ref [48] for a $d$-wave superconductor with a small amount of disorder. The dotted line is the pure $d$-wave density of states.
increase in $\lambda(0)$ between theory and experiment is possible.

Ni doping did not produce such dramatic results, however. Even the substitution of .75\% Ni was incapable of producing a $T^2$ dependence in $\lambda(T)$ at the lowest temperatures, despite the fact that .75\% Ni suppresses $T_c$ nearly as much as .31\% Zn. They suggest the possibility that Ni impurities might be Born scatterers rather than unitary scatterers, which would predict much lower crossover temperatures. Again, it is not possible to see if the actual value of $\lambda(0)$ in these crystals is consistent with this interpretation.

In an attempt to explain the linear term in $\lambda(T)$, Roddick and Stroud examined the effect of fluctuations in the phase of the order parameter. They find that phase fluctuations in an $s$-wave superconductor would produce a linear term in $\lambda(T)$ in both the classical limit and after including quantum effects of charging and dissipation. However, the slopes, $d\lambda/dT$, that they predict are a factor of ten smaller than the slope observed by Hardy et al. when $\lambda(0) = 1400\text{Å}$. The predicted slopes are largest when the c-axis anisotropy is large. Increasing the anisotropy won't help, however, because $\text{YBa}_2\text{Cu}_3\text{O}_{7.8}$ is essentially 2 dimensional as far as this calculation is concerned. Slopes near 4 Å/K are not expected until $\lambda(0)$ is about 2600Å, which is significantly larger than any experimental results. It appears unlikely that fluctuations are responsible for the linear $T$ dependence in $\text{YBa}_2\text{Cu}_3\text{O}_{7.8}$.

On the other hand, a $d^2\gamma$ order parameter leads to the same Ginzburg-Landau equations that an $s$-wave order parameter would, so these phase fluctuations should be present, regardless of whether $\text{YBa}_2\text{Cu}_3\text{O}_{7.8}$ is $s$-wave or $d$-wave. It is possible that most
of the linear term in $\lambda(T)$ seen by Hardy et al. is due to a $d$-wave density of states, while 10% or so is due to phase fluctuations. These fluctuations should also show up in YBa$_2$Cu$_3$O$_{7.4}$ films as a small linear term in addition to the $T^2$ term seen by Lee et al.[9] and others.[10-12] This linear term has not as yet been seen.

There is also evidence that fluctuations are responsible for the curvature of $1/\lambda^2(T)$ near $T_C$. The BCS theory is a mean-field theory which leads to $1/\lambda^2(T)$ linear in $(T - T_C)$ near the transition. The first order correction of Gaussian fluctuations describes low-$T_C$ superconductors well. The combination of high transition temperature and small coherence length in YBa$_2$Cu$_3$O$_{7.4}$ means that 3D-XY fluctuations may be responsible for the temperature dependence of $\lambda$ within a few degrees of $T_C$.[5] Specific heat measurements on YBa$_2$Cu$_3$O$_{7.4}$ crystals can be explained by assuming 3D-XY fluctuations near $T_C$.[51]

3D-XY fluctuations would alter the behavior of $\lambda$ near the transition, making $1/\lambda^3(T)$ linear near $T_C$. Kamal et al. have measured $\lambda(T)$ near the transition and find $1/\lambda^3$ is linear within 20 degrees of $T_C$.[14] Agreement over a region this large is likely accidental. The fluctuation region is only expected to be a few degrees wide, and even the mean-field BCS result would look linear in $1/\lambda^3$ if one looked far enough away from $T_C$. The 3D-XY behavior persists when small concentrations of Ni or Zn impurities are added, as expected. This result has been confirmed by microwave measurements on thin films by Anlage et al.[52] Lin et al.[53] however, see mean-field behavior and suggest that incomplete analysis would cause the wrong conclusions to be drawn. This will be expanded upon in later chapters.
There is much than can be learned from a detailed study of $\lambda(T)$ in all temperature ranges. The change in $\lambda(0)$ with doping at higher concentrations of dopant will yield information about the density of states, and the symmetry of the order parameter. The exact low-temperature and high-temperature dependence of $\lambda(T)$ yield information about the type of fluctuations present in these materials. The situation is still incomplete because a good method for measuring both the absolute value of $\lambda(0)$ and the temperature dependence in all ranges with both accuracy and precision did not exist until recently. This thesis presents measurements of the change in the absolute value of $\lambda(0)$ with doping with an accuracy of better than 10%, and presents measurements of $\lambda(T)$ with a precision of better than 3 Å on each data point.
Chapter IV

Sample preparation and characterization

Measuring the change in $\lambda(T)$ with dopant concentration, or oxygen concentration, can provide much useful information about the nature of superconductivity and superconducting fluctuations in YBa$_2$Cu$_3$O$_{7.8}$. A signature of $d$-wave superconductivity is its high sensitivity to elastic scattering from impurities. $d$-wave theory would predict a drastic increase in $\lambda(0)$ at dopant concentrations of a few percent, as well as a similarly drastic reduction in $T_c$.

It is already well known that the transition temperature of YBa$_2$Cu$_3$O$_{7.8}$ decreases sharply with dopant concentration.[54-56] The effect is more pronounced when atoms substitute for Cu than when they substitute for Y. It is most prominent when Zn and Ni substitute for Cu. It is believed that Ni and Zn primarily replace the Cu plane atoms. Co and Al substitutions have a less drastic effect on $T_c$ and primarily replace the chain Cu atoms. This, combined with the fact that the vast majority of High-$T_c$ compounds have some form of CuO$_2$ planes, makes it likely that the superconductivity in YBa$_2$Cu$_3$O$_{7.8}$ is primarily in the CuO$_2$ planes. Therefore, it makes sense to study the effects of Ni and Zn substitution on $\lambda(T)$, as these should be most drastic.

The samples used for this study were thin films of YBa$_2$(Cu$_{1-x}$M$_x$)$_3$O$_{7.4}$ where M = Ni or Zn. The dopant concentrations ranged from nominally pure ($x = 0$) to $x = .06,$
and the oxygen concentration was also varied in several doped films. The films were made by several techniques, some by codeposition of $Y$, $BaF_2$, $Cu$, and dopant, and some by laser ablation from a stoichiometric target. The results of the penetration depth study were independent of the method used to produce the films.

The majority of the Ni doped films in this study were fabricated by codeposition of $Y$, $BaF_2$, $Cu$, and dopant onto a room temperature (100) $SrTiO_3$ substrate in a background oxygen pressure of $3\times10^{-6}$ torr.[56] The $Y$ was evaporated using an electron beam source, and the $BaF_2$, $Cu$ and dopant were evaporated from resistively heated boats. The evaporation rates were controlled by independent feedback controllers that made sure the proper ratio of atoms hit the substrate at all times. The rates were adjusted until measurements of the concentration by Rutherford backscattering gave 3 atoms of $Cu$ and 2 atoms of $Ba$ for every atom of $Y$ on a Si substrate deposited on simultaneously. The films were annealed after deposition in a tube furnace in flowing oxygen. The tube furnace dwells at 900 °C for 1 hour, where most of the crystal structure is formed. The oxygen is bubbled through distilled water before entering the tube furnace at this point. It is then cooled to 450 °C at 5°/min in dry oxygen. The film dwells at 450 °C for 1 hour, at which time the oxygen enters the sample. The pure samples were annealed at 850 °C instead of 900 °C, but it was found that high-quality doped films could not be made with a lower temperature anneal. The codeposited films were c-axis oriented, typically 1000-3000Å thick and circular with a 12mm diameter. The reproducibility of this geometry is important in determining the absolute value of $\lambda(0)$ with any accuracy.
One 4% Ni film was the subject of more detailed measurements. This film was made by laser ablation in the Electrical Engineering department at Ohio State from a stoichiometric Ni-doped YBa2Cu3O7-δ ceramic target. The target was prepared by a conventional coprecipitation technique, and the transition temperature and width were measured before the films were made. The transition temperature of the laser ablated films were typically a few degrees lower than the \( T_c \) of the target, or the \( T_c \) of codeposited films with the same nominal Ni concentration.

Figure 5 shows the inductive transitions of the pure and Ni doped films used in the study. The transitions were measured by measuring the change in the mutual inductance of two coils on opposite sides of the film. The same apparatus is used to determine the penetration depth, but the data is shown here only to show the width of the transition. The \( T_c \)'s fall with dopant concentration and are well within the range reported for bulk samples. The inductive pick-up decreases by a factor of 100-1000 within 2 degrees of \( T_c \), demonstrating that the films are homogenous. \( T_c \) is reduced by about 5K per atomic percent Ni, and an inhomogeneity of \( \pm 0.2\% \) would produce a 2K transition width, so the films appear homogenous to at least .2% dopant concentration.

The films were also characterized in several more stringent ways. X-ray diffraction measurements on the films indicate that codeposited films with up to 11% Ni are as good as pure films, with no evidence of second phases or misoriented grains. Raman signals from misoriented grains should be larger than that from c-axis grains, and Raman spectroscopy is also extremely sensitive to the existence of certain second phases such as BaCuO2. Despite this sensitivity, no misoriented grains or second phases was
Fig. 5. Change in mutual inductance of two coils on opposite sides of the Ni-doped YBa$_2$(Cu$_{1-x}$Ni$_x$)$_3$O$_{7-\delta}$ films.
seen in Raman spectra. Therefore, the vast majority of our film volume consists of c-axis oriented grains of Ni-doped YBa$_2$Cu$_3$O$_{7.4}$.

The resistivity of several codeposited films not used for this study were measured by a standard four-terminal resistance measurement. The films were patterned lithographically, and therefore could not be used for the penetration depth measurement. Figure 6 shows $\rho(T)$ for a selected set of Ni-doped films. The infrared reflectance was also measured on these films. It was hoped that this amount of scattering would move YBa$_2$Cu$_3$O$_{7.4}$ into the dirty limit, and a gap feature would become visible. It turned out that the reflectance was essentially indistinguishable from the sample reflectance in the normal state. The scattering rate was obtained from a Drude fit at low frequencies and agreed well with the resistivity measurements, implying that grain boundaries were only a small part of the total resistance of the films. The infrared scattering rates were used later for a quantitative comparison to the predictions of $d$-wave theories.

The Zn-doped films were made by laser ablation at Los Alamos National Labs, again using a stoichiometric target produced by a conventional coprecipitation technique. We were unable to make high-quality Zn-doped films by codeposition using either Zn or ZnF$_2$ as the evaporating material. The change in mutual inductance of the two coils on opposite sides of the Zn-doped films are shown in figure 7. The signal drops by a factor of 100-1000 within 2 degrees of $T_c$. Zn doping decreases $T_c$ by 12K per percent Zn. Therefore, these films are homogenous to $\pm .1\%$ Zn. The resistivities of patterned YBa$_2$(Cu$_{1-x}$Zn$_x$)$_3$ films are shown in figure 8. Again, the scattering rate obtained from the resistivities agrees well with the scattering rate obtained from infrared reflectance.
Fig. 6. Resistivities of a series of Ni-doped \( \text{YBa}_2(\text{Cu}_{1-x}\text{Ni}_x)_3\text{O}_{7-d} \) films.
Fig. 7. Change in mutual inductance of two coils on opposite sides of the Zn-doped YBa$_2$(Cu$_{1-x}$Zn$_x$)$_3$O$_{7.8}$ films.
Fig. 8. Resistivities of 2% and 4% Zn-doped YBa$_2$(Cu$_{1.4}$Zn$_x$)$_3$O$_{7.5}$ films.
measurements on the same films, implying that grain boundary resistance is a minimal contribution to the total resistance of the film. Both the Ni and Zn-doped films are of sufficiently high quality for a study of the dependence of $\lambda(T)$ on doping.

It is also useful to study the effect of oxygen content on the behavior of the penetration depth. If YBa$_2$Cu$_3$O$_{7-\delta}$ is a $d$-wave superconductor, the penetration depth should increase as a result of the disorder introduced by Ni and Zn impurities even though the density of superconducting electrons is not changed much by the dopant atom. In contrast to this, oxygen depletion reduces the carrier concentration without introducing a substantial amount of disorder, which is an independent method of increasing $\lambda(0)$.[44,57] The low-temperature phase fluctuations predicted by Roddick and Stroud should be more pronounced when $\lambda(0)$ is large.[50] We can vary $\lambda(0)$ by varying the oxygen concentration in a single pure or doped film and study the evolution of the low-temperature linear term in $\lambda(T)$ with $\lambda(0)$. We remove oxygen by heating the film to 280 $^\circ$C in 1 atm. Ar for 10-30 minutes.

We conclude that these films of sufficiently high quality to justify a study of the evolution of $\lambda(T)$ with doping. The following chapters describe the apparatus for measuring $\lambda(T)$ in these films, currently the only method in the world capable of measuring both $\lambda(0)$ and $\Delta\lambda(T)$ to good accuracy, and examine $\lambda(T)$ to determine the effects of disorder and thermal fluctuations on $\lambda$. 
Chapter V
Experimental set-up and analysis

We measure the penetration depth by measuring the change in the mutual inductance of two coils on opposite sides of thin Ni and Zn-doped YBa$_2$Cu$_3$O$_{7-\delta}$ films. The method was invented by Hebard and Fiory,[58] and initially used to measure the penetration depth in more conventional superconductors. Later, Fiory et al. used the same method to measure the penetration depth in YBa$_2$Cu$_3$O$_{7-\delta}$.[43] Our present apparatus has several substantial improvements over the original method which will be detailed in this chapter. The most important is that, although the original authors claimed to only be able to determine changes in $\lambda^2(T)$, our present geometry is known well enough that the absolute value of $\lambda(0)$ can be obtained to $\pm 10\%$ accuracy, without the necessity of fitting to any functional form.[59]

The numerical modelling is straightforward. The vector potential $A(r)$ in the film due to the current density $J(r')$ in the film and the current in the drive coil is derived from the equation

$$A(r) = \mu_0/4\pi \int J(r')/|r-r'| \, dr' + A_{\text{drive}}(r).$$  \hspace{1cm} (1)
\( A_{\text{drive}}(r) \) is the contribution from the current in the drive coil, and it is chosen to be cylindrically symmetric. If the coil and film are cylindrically symmetric, the vector potential has zero divergence and is parallel to the film edge. It is therefore automatically in the London gauge, and no gauge correction is necessary to apply the London equation. Assuming \( \text{YBa}_2\text{Cu}_3\text{O}_{7-\delta} \) is in the local limit, this gives

\[
J(r) = -\frac{1}{(\mu_0 \lambda^2)} A(r). \tag{2}
\]

Combining eq. (1) and (2) leads to an integral equation for \( J(r) \),

\[
\mu_0 \lambda^2 J(r) + \mu_0/4\pi \int \frac{J(r')}{|r-r'|} \, dr' = A_{\text{drive}}(r). \tag{3}
\]

The integral over the film thickness can be easily approximated. If \( \lambda \) is large compared to the film thickness, \( d \), \( J \) is constant over the thickness and the equation becomes

\[
\mu_0 \lambda^2 J(\rho) + \mu_0/4\pi \int J(\rho')d/|\rho-\rho'| \, d\rho' = A_{\text{drive}}(\rho) \tag{4}
\]

as long as the film is thin enough that \( |\rho-\rho'| \approx |r-r'| \). If \( d \) is large compared to \( \lambda \), but still small compared to the other two dimensions, the current density falls off exponentially from its value \( J_0(\rho) \) at the surface and the integral becomes

\[
\mu_0 \lambda^2 J_0(\rho) + \mu_0/4\pi \int J_0(\rho')\lambda/|\rho-\rho'| \, d\rho' = A_{\text{drive}}(\rho). \tag{5}
\]
An expression that is approximately correct in all ranges can be obtained by replacing the thickness \( d \) in eq. (4) by an effective thickness \( d_{\text{eff}} = \lambda \tanh(d/\lambda) \).[60] The sheet current \( K(\rho) \) is equal to \( J_0(\rho)d_{\text{eff}} \) and the equation becomes two dimensional:

\[
\mu_0 \lambda \coth(d/\lambda) K(\rho) + \mu_0/4\pi \int K(\rho')/|\rho-\rho'| \, d\rho' = A_{\text{drive}}(\rho)
\]

Eq. (6) was derived for a film with a purely inductive response and all the variables and parameters are purely real numbers. The actual measurement is done at frequencies ranging from 500 Hz to 100 kHz, and the resistive part of the conductivity can also play a role in the analysis if it is large enough. Given a sinusoidal \( A(\mathbf{r},t) \), \( E(\mathbf{r}) = -i\omega A(\mathbf{r}) \) and eq. (7),

\[
J(\mathbf{r}) = -(1/\mu_0\lambda^2 + i\omega \sigma_j) A(\mathbf{r})
\]

replaces eq. (2) in the integral equation. A complex penetration depth can be determined from this equation and the effective thickness also becomes complex. Given this complex \( \lambda \), eq. (6) is still correct as written. This analysis allows us to retrieve both the real and imaginary parts of the conductivity of the film for use in analyzing the fluctuations.

Eq. (6) can be solved by two different methods, both of which should yield identical results. Jeanneret et al.[61] use a two-dimensional Fourier transform method to solve for the Fourier components of \( K(\rho) \). This method assumes the film is an infinite sheet, and is unable to handle finite radius films. Both Jeanneret et al. and Hebard and
Fiory [58] make the assumption that their films are infinite relative to their coil dimensions. Our films are 12mm diameter, 6 times the coil diameter, and we find that assuming an infinite film is a good approximation. After integrating over the Fourier components, the equation for the current density becomes [61]

\[ K(\rho) = I_D R_D \int_0^\infty dq_t \frac{q_t e^{-q_t h_D}}{1 + (2/\mu_0)(1/i\omega G) q_t} \times J_1(q_t R_D) J_1(q_\rho) \frac{1 - e^{-q_t\delta h_D}}{1 - e^{-q_\rho h_D}} \]  

(8)

where \( I_D \) is the current in the drive coil, \( R_D \) is the radius of the drive coil, \( h_D \) is the distance between the drive coil and the film, \( N_D \) is the number turns in the drive coil, and \( \delta h_D \) is the difference in height between two successive turns in the drive coil. \( G \) is the sheet conductivity of the film including \( d_{eff} \), and \( q_t \) is the transverse component of the Fourier wavevector. Because of the symmetry of the problem, \( K(\rho) \) depends only on the magnitude of \( \rho \), and has a component only in the \( \theta \) direction.

The voltage induced in the pick-up coil can be calculated from the integral of \(-i\omega A \cdot dl\) around the pick-up loops. Dividing out the drive current then gives the equation for the change in mutual inductance between the film being present and absent [61]

\[ \delta M = \int_0^\infty dx \frac{M(x)}{1 + (2/\mu_0 d)(1/i\omega G)x} \]  

(9)
h is the distance between the first loops in the drive and receive coils, and \( x=q_h \), a dimensionless variable of integration. \( M(x) \) is given by

\[
M(x) = \pi \mu_0 h \alpha \beta J_1(\alpha x) J_1(\beta x) e^{-\alpha^2 x^2} \frac{1-e^{-N_r^2 \beta^2}}{1-e^{-\alpha^2 x^2}} \frac{1-e^{-N_r^2 \alpha^2}}{1-e^{-\beta^2 x^2}}
\]  

where \( \alpha, \beta, \gamma, \) and \( \delta \) are the drive coil radius, receive coil radius, separation between drive coil loops, and separation between receive coil loops, all expressed in units of \( h \). \( N_r \) is the number of turns in the pick-up coil.

This solution is exact to the degree that the integral in Eq. 9 can be calculated numerically. Since a real film is not infinite, some of the flux that would be excluded from the region where \( \rho > R_{\text{film}} \) is instead allowed through, even for \( \lambda = 0 \). This background coupling, \( M_0 \), for our geometry ranges from its largest value of 15pH (out of 15 nH) at \( \lambda = 0 \), to 0 pH at \( \lambda = \infty \), because at \( \lambda = \infty \) exactly as much flux goes around the film as would if the film weren't present. The assumption of Hebard and Fiory and Jeanneret et al., which we have verified, was that this background coupling could be treated as constant and subtracted off without affecting the result. It is clear near \( T_c \) that a difference of 15pH out of 15nH is irrelevant, but at the lowest temperatures, 15pH out of \( \sim 35 \) pH could be significant. We will treat the problem of a finite film more exactly now.

The second method of solution is to transform the integral equation into a matrix equation, and invert the matrix to obtain the solution. This method is actually easier to apply to a finite radius film than an infinite one. Given the cylindrical symmetry of the
coils and film, \( A(\rho) \) and \( J(\rho) \) depend only on the magnitude of \( \rho \), and have only a \( \theta \) component. Therefore, we can treat the film as a sequence of concentric rings of width \( \Delta \rho \) and carrying a current \( K(\rho) \Delta \rho \). The vector potential \( A_\phi(\rho) \) in a single ring at radius \( \rho' \) carrying current \( I \) is given by

\[
A_\phi = \frac{\mu_0 J}{\pi k} \left( \frac{\rho'}{\rho} \right)^{1/2} \left[ \left( 1 - \frac{k^2}{2} \right) K(k) - E(k) \right],
\]

(11)

where \( K(k) \) and \( E(k) \) are the complete elliptic integrals, and

\[
k^2 = \frac{4 \rho' \rho}{\rho'^2 + \rho^2 + \delta z^2 + 2 \rho' \rho}
\]

(12)

\( \delta z \) is the distance along the z-axis between the centers of the coaxial rings.[62] Using Eq. 11 and 12, Eq. 6 can be be converted straightforwardly into a discrete matrix equation

\[
\Sigma_j [a_{ij} + \mu_0 \lambda \coth(d/\lambda) \delta_{ij}] K_j = A_{\text{drive,}i},
\]

(13)

\( A_{\text{drive,}i} \) is the vector potential at the \( i \)-th ring due to the drive coil current and \( a_{ij} \) is \( \Delta \rho \) times the vector potential at the \( i \)-th ring due to a unit current in the \( j \)-th ring. \( K_j \) is the sheet current density in the \( j \)-th ring. Inverting the matrix gives \( K_j \) in terms of the \( A_\phi \)s. Once the sheet current density is determined, the line integral of \( A \) at the pick-up coil
is straightforward to calculate. Even if the matrix inversion is done completely accurately, this method is only exact in the limit $\Delta \rho$ of the loops goes to zero. An accuracy of better than 1 part in 10000 is necessary for this method to be more accurate numerically than the infinite solution plus background subtraction. The current peaks sharply near the edge of the film, and more finely spaced rings are required to describe the current accurately there.[63]

Several coil geometries have been tried as the experiment was improved. Figure 9 shows the current distribution that would be induced in an infinite film given $\lambda = 1500 \text{Å}$, and an 18mA current in the primary coil in our most frequently used geometry. The coils are made from 26 turns of .1172mm thick wire, with the furthest 13 turns wound in the opposite sense to the nearest 13 turns. The drive coil is 1.27mm from the film because of the substrate thickness, and the receive coil is 0.254mm from the film. Figure 10 shows the current distribution that would be induced in a 12mm diameter film with this coil geometry.

We now compare the results of the two methods to verify that assuming an infinite film and subtracting a background contribution is sufficiently accurate for our purposes. Lin et al.[52] have suggested that using the infinite film solution introduces errors into the determination of the $T$-dependence of $\lambda$ near $T_c$. They suggest that the normal mean field behavior [i.e. $1/\lambda^2$ linear in $(T_c - T)$] would appear to follow the 3D-XY dependence [i.e. $1/\lambda^3$ linear in $(T_c - T)$] unless the finite solution is used. Figure 11 shows the results of the infinite film calculation and the finite film calculation with background subtraction in pH. It is easily seen that the two methods agree to better than
Fig. 9. Current induced in an infinite film with $\lambda = 1500\text{Å}$
Fig. 10. Current induced in a 12mm diameter film with $\lambda = 1500\text{Å}$
Fig. 11. Comparison of the results of the infinite and finite film mutual inductance calculations.
1 part in 500 over the entire range. The error of < .2% introduced by using the infinite film approximation is utterly negligible. It introduces no discernable error into \( \lambda(T) \) near \( T_c \).

Both procedures produce a table of \( M(\lambda, \sigma_i) \). A look-up table is used to convert the measured \( M(T) \) to \( \lambda(T) \) and \( \sigma_i(T) \). The measured mutual inductance is actually the sum of three contributions. The important contribution, \( M_i \), is the part that comes from the two coils and induced current in the film. This contribution is screened when the film becomes superconducting. The second part, \( M_2 \), comes from the flux that leaks around the film, and is taken into account in the more accurate modelling. The third part, \( M_3 \), comes from the mutual inductance of other parts of the circuit. If the film is assumed to be infinite in the modelling, the second and third part together can be called \( M' \) and subtracted from the measured \( M(T) \) before using the table. All previous two-coil experiments have had the drawback that \( M' \) was not known accurately enough, either because of unknown coupling from other parts of the circuit or because the film radius and geometry was not known sufficiently well, probably because too much flux was leaking around the film and the film could not be centered sufficiently accurately. Therefore, \( M' \) was taken to be a fitting parameter, and \( M(T) - M' \) was fit to an expected penetration depth function to determine \( \lambda(0) \).

Actually, it is not necessary to eliminate \( M' \) to get the absolute value of \( \lambda \). It is only necessary that \( M' \) be well known and reproducible. The \( \lambda = \infty \) value of \( M \) is \( ~ 15 \text{nH} \), and the \( \lambda = 0 \) value of \( M \) is \( ~ 15 \text{pH} \), a factor of 1000 smaller. This shows the difficulty of removing the background contribution from other parts of the circuit. We estimate that
this background is less than 10pH and is reproducible to 1pH from run to run. The more
difficult part to account for is $M_2$, from the flux that leaks around the film. It can be
calculated accurately enough, but in practice, the amount that leaks around is very
sensitive to the exact position of the finite film.

We measured the value of $M'$ by replacing the film with a circle of thin lead foil
of the same diameter. The foil was very thick compared with $\lambda$ in lead, but still thin
compared to the coil dimensions so that previous analysis, which assumes a constant $A$
over the film thickness, is still valid. $M'$ was found to be 15 ± 2 pH. The error bars
were found by measuring $M'$ when the film was deliberately 200$\mu$m off center, an error
too large to actually occur in practice.

This uncertainty in $M'$ corresponds to a ± 500Å uncertainty in $\lambda^2/d$. In a 1000Å
thick film with $\lambda = 1500$Å, this corresponds to 150Å uncertainty in $\lambda$. The uncertainty
in $\lambda$ becomes considerably smaller as $\lambda$ increases. The 10% uncertainty in the film
thickness, which produces an additional 5% uncertainty in $\lambda$, then becomes the limiting
factor. The apparatus has been repeatedly improved throughout the course of this
investigation, and the most recent data requires no fitting to a theoretical function, and
are instead presented with error bars on the absolute value of $\lambda(0)$. Because the
uncertainty in $\lambda$ decreases as $\lambda$ increases, and $\lambda$ is largest near $T_C$, the temperature
dependence of $\lambda(T)$ near $T_C$ is extremely reliable.

The data $M(T)$ are taken by leaving the sample at a fixed location in a Helium
dewar and allowing the temperature to rise from 4.2K to 100K in the space of 2 hours.
This slow measurement ensures that the thermometer and sample are at the same
temperature to within 30mK, and that the measurement is unaffected by thermal gradients in the sample.

The noise on the data is about .2 nV/√Hz for the most recent apparatus, which corresponds to about ± 5Å in λ²/d, about ± 1.5Å in λ for a 1000Å film with λ = 1500Å, and less noise when λ increases. The data is numerically filtered later to reduce the noise even further. This combination of accuracy in λ(0) and precision on each data point makes this the best apparatus for measuring λ(T) in the world.
Chapter VI

Effect of Ni and Zn dopants on $\lambda(T)$

There are several methods by which Ni and Zn doping can affect $\lambda(T)$ in YBa$_2$(Cu$_{1-x}$M$_x$)$_3$O$_{7.4}$. First, the substitutions may change the superconducting charge carrier density. Since we are substituting Ni and Zn at the $M = .02$ to .06 level, this may change $\lambda(0)$ by a few percent. It is also possible that the addition of Ni or Zn could affect the strength of the coupling interaction that produces superconductivity in YBa$_2$Cu$_3$O$_{7.4}$, whether mediated by phonons or something more exotic. In addition, the Ni and Zn substitutions introduce scattering into the CuO$_2$ planes where the superconductivity is believed to originate. If the superconductivity is BCS $s$-wave this could push the superconductor into the dirty limit and raise $\lambda(0)$ slightly. If the pairing state is anisotropic $s$-wave, the scattering would make the state more isotropic and $\lambda(T)$ would have an $e^{\Delta/kT}$ dependence at the lowest temperatures. If the superconductivity is $d$-wave, as has been suggested by other experiments, $\lambda(0)$ would increase more quickly than other states would predict, and the linear temperature dependence observed in crystals would change to $\lambda(T) = \lambda(0) + cT^2$.

The films used for this study were codeposited and laser ablated YBa$_2$(Cu$_{1-x}$M$_x$)$_3$O$_{7.4}$ films ($M = \text{Ni, Zn}; x = .02, .04, .06$) prepared as described in chapter IV. Information on the films is shown in Table I. $1/\lambda^2(T)$ for these films is shown in
Table 1. Table of $T_c$, $\lambda(0)$ and disordered $d$-wave parameters for the doped films used in this study.

<table>
<thead>
<tr>
<th>Dopant</th>
<th>Method</th>
<th>Thickness (Å)</th>
<th>$T_c$ (± 0.7K)</th>
<th>$\lambda(0)$ (Å)</th>
<th>$1/\tau$</th>
<th>$T_\phi$</th>
<th>$\lambda_{gap}(0)$ (Å)</th>
</tr>
</thead>
<tbody>
<tr>
<td>2% Ni</td>
<td>Codep.</td>
<td>1000</td>
<td>79</td>
<td>2900±300</td>
<td>88K</td>
<td>115K</td>
<td>2400</td>
</tr>
<tr>
<td>4% Ni</td>
<td>Laser</td>
<td>1000</td>
<td>70</td>
<td>3920±60</td>
<td>185K</td>
<td>150K</td>
<td>3000</td>
</tr>
<tr>
<td>6% Ni</td>
<td>Codep.</td>
<td>3000</td>
<td>68</td>
<td>12440±60</td>
<td>273K</td>
<td>190K</td>
<td>3700</td>
</tr>
<tr>
<td>2% Zn</td>
<td>Laser</td>
<td>3200</td>
<td>62</td>
<td>3900±400</td>
<td>72K</td>
<td>90K</td>
<td>2400</td>
</tr>
<tr>
<td>4% Zn</td>
<td>Laser</td>
<td>2300</td>
<td>43</td>
<td>7800±800</td>
<td>144K</td>
<td>110K</td>
<td>3000</td>
</tr>
</tbody>
</table>
figure 12. The 4% and 6% Ni films were measured on the most recent apparatus. The error bars in figure 12 reflect the 500Å uncertainty in $\lambda^2/d$. The 2% Ni as well as the 2% and 4% Zn films were measured on earlier versions of the apparatus, hence the larger noise on the 2% Ni data. They were destroyed in the course of other investigations and could not be remeasured. The mutual inductance $M'$ from other parts of the circuit was therefore not known for these films.

Since $M'$ influences the dependence of $\lambda$ on $T$, the procedure used commonly with several experimental techniques that suffer from this problem is to choose $M'$ so that $\lambda(T)/\lambda(0)$ is close to a target function of $T/T_c$.[43,58,61] The 4% and 6% data, which do not suffer from this problem, provide the target function for the earlier data. Specifically, $1/\lambda^2(T)$ just below $T_c$ in the 4% and 6% Ni films extrapolates to $T=0$ at about twice the experimental value of $1/\lambda^2(0)$, so we choose $M'$ for the 2% Ni, 2% Zn, and 4% Zn films to do the same.

At the level of detail appropriate in this chapter, all of the $1/\lambda^2$ curves in figure 12 have the same shape, namely, $1/\lambda^2(T)$ decreases monotonically and has a negative curvature everywhere. $1/\lambda^2$ generally flattens off as $T/T_c \rightarrow 0$. Doping introduces no dramatic changes in the dependence of $\lambda^2(0)/\lambda^2(T/T_c)$ on $T/T_c$. The magnitude of $\lambda$ is another story. The superfluid densities at $T=0$, $n_s(0) \propto 1/\lambda^2(0)$, for 2% Ni, 4% Ni, and 6% Ni are lower than for pure YBCO films by factors of 3, 6, and 60, respectively. For 2% and 4% Zn films, $n_s(0)$ is lower by factors of 5 and 25, respectively. Within the $\pm 0.005$ uncertainty in $x$ and the uncertainties in $\lambda^2(0)$, these numbers represent a factor of 2 reduction in $n_s(0)$ for each percent of dopant, for both Ni and Zn.
Fig. 12. Experimental $1/\lambda^2(T)$ for Ni and Zn-doped YBa$_2$Cu$_3$O$_{7-\delta}$ films.
The strong decrease in $n_s(0)$ with a relatively minor decrease in $T_c$ stands in stark contrast to the correlation of $T_c$ with $n_s(0)$ found by Uemura et al. [64] for a variety of high-$T_c$ superconductors, including Pr-doped and oxygen depleted YBa$_2$Cu$_3$O$_{7.4}$, suggesting that substitutions for Cu behave differently than Pr doping and oxygen depletion. It appears that Ni and Zn act primarily to induce disorder, while Pr doping and O depletion act primarily to affect carrier density. Since the decrease in $n_s$ occurs for both laser ablated and codeposited films, it seems unlikely that it is associated with film microstructure, and more likely that it is an intrinsic effect. We note that infrared measurements on irradiated crystals of Bi$_2$Sr$_2$CaCu$_2$O$_{8+}$[26] and YBa$_2$Cu$_3$O$_{7.4}$[25] show a dramatic decrease in $n_s(0)$ with irradiation, which generates point disorder, but not grain boundaries.

Our experimental results raise numerous questions. Are the suppressions in $n_s(0)$ and $T_c$ accounted for by $d$-wave theories, given the scattering rates from infrared measurements? How does the superconducting density of states, $N_s(E)$, evolve with doping? Are dramatic reductions in $n_s(0)$ consistent with the relatively minor changes in the dependence of $\lambda/\lambda(0)$ on $T/T_c$?

Before getting into specific models, we want to discuss conclusions one can draw from the data, independent of model. The decrease in $n_s(0)$ could signify changes in the density $n$ and effective mass $m^*$ of electrons, i.e., the plasma frequency $\omega_p^2 = ne^2/m^*\varepsilon_0$ (mks units). Since doping is less than 9% of the planar Cu sites, one might expect $\omega_p^2$ to change by no more than about 9%. Infrared measurements on these same films support
this expectation.\cite{23,24} The decrease in $n_s(0)$ could signify that disorder pushes spectral weight [\textit{i.e.}, area in $\sigma_1(\omega)$ vs. $\omega$] to frequencies much higher than the "gap" frequency, so there is less conductivity available at low frequencies to condense into the $\delta$ function at $\omega=0$. This is the primary mechanism for reduction of $n_s$ with disorder in $s$-wave models. For the most heavily doped films, 4\% Zn and 6\% Ni, $1/\tau$ (Table I) is about $4kT_c/\hbar$, so that roughly half of the spectral weight lies above the gap frequency. This would account for about a factor of 2 reduction in $n_s$, which is much smaller than the observed reductions of 25 (4\% Zn) and 60 (6\% Ni). The reduction in $n_s$ must be due almost entirely to modifications in the superconducting density of states. Quite rigorously, the large reduction in $n_s$ means that the area missing from the real part of the optical conductivity, $\sigma_{1s}(\omega)$ vs. $\omega$, in the superconducting state is reduced by the same large factor. $\sigma_{1s}(\omega)$ in the doped films must be very close to $\sigma_{1n}(\omega)$; within the uncertainty, infrared measurements find no difference at all. In other words, disorder activates photon absorption processes that are forbidden in the undoped material. The obvious physical interpretation is that disorder moves the superconducting density of states $N_s(E)$ toward the normal-state density of states $N_n(E)$ by filling in $N_s$ at low energies. Then the number of quasiparticles available to absorb photons, and the density of final states for the quasiparticles to scatter into are only slightly smaller than in the normal state. The precise dependence of $N_s(E)$ on $E$ for a particular level of disorder is model dependent, but the fact that $N_s(E)$ is not much different from $N_n(E)$ is not model dependent.
It is interesting to consider our results within models of disordered $d$-wave superconductivity.\cite{13,34} In these models, the intrinsic anisotropy of the order parameter combined with strongly scattering disorder leads to creation of an "impurity band" in $N_s$ centered at $E=0$, which dramatically decreases $n_s$ and $T_c$. For very small concentrations of impurities, the impurity band changes the dependence of $\lambda$ on $T$ from linear to quadratic below a cutoff temperature proportional to the width of the impurity band. This crossover may have been observed in Zn-doped $\text{YBa}_2\text{Cu}_3\text{O}_{7.8}$ crystals (but not in Ni-doped crystals),\cite{48} and in very thin $\text{YBa}_2\text{Cu}_3\text{O}_{7.8}$ films.\cite{9} Based on theory and these measurements, disorder in the present samples is much too large to expect to observe the crossover.

Kim, Preosti and Muzikar (KPM)\cite{34} present a $d$-wave calculation of $n_s(0)$ and $T_c$ which extends the Hirschfeld and Goldenfeld theory to high concentrations of both weakly and strongly scattering impurities. The parameters of the KPM theory are the scattering rate $1/\tau$, the strength of scattering from each impurity which ranges from "Born" to "unitarity," a hypothetical transition temperature $T_{co}$ of the "pure" material, and the $T_c$ of the disordered material. We assume unitarity scattering in the following discussion. With $1/\tau$ from infrared measurements and $T_{co}$ fixed at 90K, KPM predict $T_c$ suppressions about the same as observed for 2\% Zn, a little larger than for 4\% Zn, and much larger than observed for the Ni-doped films. Indeed, with $T_{co}$ fixed, KPM predict $T_c = 0$, and therefore $n_s = 0$, for 4\% and 6\% Ni. To compare KPM with $n_s(0)$ vs. $1/\tau$, we assume that Ni and probably Zn affect the mechanism for superconductivity, and hence $T_{co}$. For example, if we take $T_{co}$ of the 4\% Zn film to be 110K, then $h/2\tau kT_{co} =$
0.65, and KPM predict $T_c = 0.38 T_{co} = 43$K, which matches the measured $T_c$. Given \( \hbar/2\pi kT_{co} = 0.65 \), KPM find $n_s(0)$ reduced by a factor of 4, which is considerably less than the observed reduction of 25. The assumed values of $T_{co}$ and the predicted values of $\lambda(0)$ found by the same procedure are given in Table I. Overall, given our assumption about the dependence of $T_{co}$ on disorder, KPM underestimate the reduction in $n_s(0)$. The discrepancy grows from roughly a factor of 2 at 2% Zn and Ni to a factor of 6 for 4% Zn and 10 for 6% Ni. However, considering the simplicity of the theory, the extreme sensitivity to disorder and the experimental uncertainty in $1/\tau$, even a factor of 10 discrepancy is not too discouraging. The values of $T_{co}$ larger than the 90K value for the pure material may imply that Ni (and to a lesser extent Zn) increases the effectiveness of the pairing mechanism in addition to increasing the scattering rate.

Next, we want to get an idea of how $N_S(E)$ evolves as $x$ increases, and to address the question of whether the $T$ dependence of $\lambda^2(0)/\lambda^2(T)$ is consistent with a strong filling-in of the superconducting density of states at low energies. We must construct an ad hoc phenomenological model which relates a disorder-broadened $N_s(E)$ to $\lambda(T)$, since there is no tractable theory available. We base the model on $d$-wave theories, largely because we can then compare our numerical results for $\lambda(0)$ vs. $1/\tau$ with the analytic KPM theory as a check on the model.

Our ad hoc model begins with a generic $d$-wave density of states $N_{s,p}(E)$ for pure YBa$_2$Cu$_3$O$_{x\delta}$ generated by summing all of the BCS densities of states corresponding to the $d_{x^2-r^2}$ gap function, $\Delta_x = \Delta_0[\cos^2(k_x) - \cos^2(k_y)]$, over a cylindrical Fermi surface. This density of states is a universal function of $E/\Delta_0$. We then find the order parameter
\( \Delta_0(T) \) that results in the calculated \( \lambda_p(T) \) in perfect agreement with the data of Hardy et al.\[6\] on pure YBa\(_2\)Cu\(_3\)O\(_{7-d}\) crystals, except within about 3K of \( T_c \) where 3D-XY fluctuations may affect \( \lambda(T) \).\[14\] \( \lambda_p^2(0)/\lambda_p^2(T) \) is related to \( \Delta_0(T) \) through\[65\]

\[
\frac{\lambda_p^2(0)}{\lambda_p^2(T)} = 1 - \frac{(\beta/\pi)\Delta_0}{\int_{-\infty}^{\infty} dx \text{sech}^2(\beta \Delta_0 x/2)} \\
\times [\theta(x-1)K(x^2) + \theta(1-x)xK(x)],
\]

(14)

where \( \theta \) is a step function, \( K \) is a complete elliptic integral, and \( \beta = 1/kT \). The resulting \( \Delta_0(T) \) is shown figure 13. \( \Delta_0(0) \) is \((3.0 \pm 0.1) kT_c \) and \( \Delta_0(T)/\Delta_0(0) \) is similar to BCS, i.e., it is flat at low temperatures and approaches \( T_c \) as \((2.25)(1 - T/T_c)^{1/2} \), with a coefficient only 30% larger than the BCS value of 1.74.

We assume that \( \lambda(T) \) changes with doping because \( N_s(E) \) fills in at low energies. To model changes in \( N_s(E) \) with disorder, we broaden the density of states \( N_{s,k}(E) \) corresponding to each \( k \) direction with the relation,\[66\]

\[
N_{s,k}(E) = \text{Re} \{ (E + i\tau)/[(E + i\tau)^2 - \Delta_k(T)^2]^{1/2} \}
\]

(15)

and then sum over all directions in \( k \) space. We assume that \( \Delta_0(0)/kT_c = 3.0 \), even as doping suppresses \( T_c \), and that the dependence of \( \Delta_0(T)/\Delta_0(0) \) is also unaffected by doping. This broadening algorithm has the important property that it conserves states. It also has the undesirable property that the peak in \( N_s(E) \) occurs at \( E > \Delta_0 \) when disorder is present. Therefore, after calculating \( N_s(E) \) with a particular disorder parameter, \( \hbar/\tau \Delta_0 \),
Fig. 13. $\Delta_0(T/T_c)$ used to fit $\lambda(T/T_c)$ for pure YBa$_2$Cu$_3$O$_{7-\delta}$ crystals from Hardy et al. The BCS temperature dependence is shown for comparison.
we rescale the energy axis so that the peak in $N_s(E,T)$ occurs at $\Delta_0(T)$. The densities of states at $T=0$ corresponding to the various samples are shown in figure 14. As $T$ increases, $\Delta_0(T)$ decreases, so disorder effects are stronger. The peak in $N_s(E)$ drops below $\Delta_0(0)$ and structure in $N_s(E)$ disappears rapidly. The rapid broadening of $N_s$ as $T$ increases could, in principle, lead to qualitative changes in the dependence of $\lambda(T)$ on $T$ as disorder increases. In the following, we find that this is not the case.

We determine $\lambda(T)$ by calculating the area missing from $\sigma_{1s}(\omega)$ vs. $\omega$. $\sigma_{1s}(\omega)$ is proportional to the rate at which photons with energy $\hbar\omega$ are absorbed by quasiparticles. It is an integral over the initial-state energy of the product of the densities of initial and final states, the appropriate Fermi functions and the Drude conductivity, $\sigma_{1D}(\omega) = \omega_p^2 \tau \epsilon_0 / (1 + \omega^2 \tau^2)$. [23,24,67] Coherence factors do not play a significant role in the strongly disordered state, although they do contribute to the pure (and weakly disordered) $d$-wave result [Eq. 14]. In our model

$$\frac{\lambda^2_0(T)}{\lambda^2(T)} = \frac{2\tau}{\pi} \int_0^\infty \frac{d\omega}{1+\omega^2\tau^2} \frac{1}{\hbar \omega} \times \int_{\infty}^{\infty} [1-N_s(E)N_s(E+\hbar\omega)] \left[ f(E) - f(E+\hbar\omega) \right] dE$$

(16)

with the densities of states in figure 14. We fix $\lambda_0(0) = 1500\AA$, which corresponds to assuming that $\omega_p = c/\lambda_0(0)$ is unaffected by doping. $1/\tau$ and $\omega_p$ are assumed independent of $T$. The results of the calculation are shown in figure 15.

We can compare the calculated increase in $\lambda(0)$ with results found above for the KPM theory. The comparison is admittedly rough, since the KPM theory provides a wide
Fig. 14. Disorder-broadened $d$-wave densities of states deduced from the phenomenological model. $\Delta_0(0) = 3kT_c$. 
Fig. 15. Calculations of $1/\lambda^2(T)$ from our model, Eq. (16), for various Ni and Zn concentrations, given the parameters in Table I.
range of predictions for $n$, depending on the assumed dependence of $T_c$ on disorder, but
nevertheless, for 4% Zn, for example, our calculation yields a sixfold reduction in $n$, where the KPM theory as applied above, finds a fourfold reduction. Thus, our simple model is not obviously implausible. Next, we examine the calculated shape of $\lambda^2(0)/\lambda^2(T)$ vs $T/T_c$, Fig. 15, and find that it is not strongly affected by doping. To emphasize this point, Fig. 16 shows the surprisingly good agreement with the 4% Ni film, when $1/\tau$ used in the calculation is 30% larger than the value in Table I, which is within the experimental uncertainty in $1/\tau$. Near $T_c$, the model correctly reproduces the mean-field linear $T$ dependence. The discrepancy near $T_c$ in figure 16 likely includes non-mean-field fluctuations not included in the model.[14] The very slight discrepancy below 10K occurs because the model calculation produces $T^2$ dependence in $1/\lambda^2(T)$ at the lowest temperatures, and there is actually a small linear term in the Taylor expansion of the data at the lowest temperatures.[68] This may be due to phase fluctuations in the order parameter.[50,69-71] These discrepancies will be dealt with in Chapters VII and VIII. Similar agreement can be found for all except the 6% Ni sample, which requires a $1/\tau$ much larger than found in infrared measurements. To show the similarity in the results obtained in films made by different techniques, figure 17 shows the data from the 4% Ni film shown in figure 16, and a codeposited 4% Ni film. $M'$ was not known for the codeposited data, so $M'$ was adjusted slightly to give the best agreement. These comparisons with $d$-wave theories and models indicate that $d$-wave theories likely will be able to account for our experimental results. Of course, they do not rule out other models.
Fig. 16. Data and model calculations for the 4% Ni film. $1/\tau = 240$K is the best fit scattering rate.
Fig. 17. Comparison of $1/\lambda^2(T)$ for laser ablated and codeposited 4% Ni films.
Having generated approximations to $N_s(E=0)/N(0)$ for each doping level, we can compare with the specific heat data of Loram et al.\[40\] on Zn-doped $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ bulk sintered samples, which also indicate a gapless density of states. The normal state density of states ratio, $N_s(0)/N_{np}(0)$, is about unity for Zn concentrations up to 10\%, which we assumed in our model when we assumed $\lambda_0(0) = 1500\AA$ for all films. More importantly, 3\% Zn increases $N_s(0)$ to about 60\% of $N_s(0)$, a significant filling-in of the gap. We estimate that $N_s(0)$ is about 80\% of $N_s(0)$ at 2\% Zn. This apparently larger effect could be explained by uncertainties in the interpretations of the two experiments or by larger disorder nucleated by dopants in films, which cannot be annealed as thoroughly as bulk samples. In the end, both measurements indicate a substantial filling-in of the density of states at low energy at small doping levels. Figure 18 shows the specific heat expected for the Ni-doped films, given the density of states corresponding to the infrared $1/\tau$. The specific heat jump quickly becomes significantly smaller, in agreement with data on Ni-doped crystals\[39\] as well as data on other dopants\[35-38\]. Also, the low-temperature specific heat becomes linear, with a slope that increases as the dopant concentration increases.

Finally, we note that measurements on $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ crystals doped with much smaller concentrations of Ni (up to 0.75\%) and Zn (up to 0.31\%) find that Ni and Zn have different effects on the low-temperature behavior of $\lambda(T) - \lambda(0)$, at least at small concentrations.\[48\] 0.75\% Ni does not significantly alter the low-temperature linear term, $\lambda(T) - \lambda(0) \propto T$, while 0.15\% Zn is sufficient to make the low-temperature term quadratic, \textit{i.e.,} $\lambda(T) - \lambda(0) \propto T^2$. In apparent contrast, we find that Zn and Ni have
Fig. 18. Model specific heat vs. $T$ for the Ni-doped films in the study.
similar effects. The apparent contradiction may not be real, since these authors measured the difference $\lambda(T) - \lambda(0)$, but not the absolute value of $\lambda(0)$, so a direct comparison with our results is not possible. In addition, only the 4% and 6% Ni films have precise enough data at the lowest temperatures to determine whether $\lambda(T) - \lambda(0)$ is linear or quadratic. This question is the subject of Chapter VII.
CHAPTER VII

Effect of thermal phase fluctuations on $\lambda(T)$

As has been mentioned before, many experiments point to the existence of an order parameter with $d$-wave symmetry in the high-$T_C$ superconductors. Some are directly sensitive to the phase of the order parameter, but others are only sensitive to the existence or non-existence of nodes in the order parameter, and therefore cannot distinguish between $d$-wave and anisotropic $s$-wave states. Although measurements of the low-temperature penetration depth $\lambda(T)$ do not determine the directional dependence of the phase of the order parameter, the linear $T$-dependence of $\lambda(T) - \lambda(0)$ has been interpreted as evidence for nodes in the gap function.

Several recent theoretical papers[50,68-70] question the $d$-wave interpretation by pointing out that thermal fluctuations in the phase of the order parameter could give a linear $T$-dependence comparable in magnitude to that observed, with favorable values for the important parameters, e.g. mass anisotropy $m_c/m_{ab}$ and $\lambda(0)$. With conventional values for these parameters, $m_c/m_{ab} = 25$ and $\lambda(0) = 1400\AA$, thermal fluctuations are calculated to be about 10% of the observed effect. While each of the theoretical papers uses a different framework and makes different approximations, all are basically Ginzburg-Landau theories of anisotropic, homogeneous superconductors combined with a model of the dynamics of phase fluctuations. All agree on the important result:

63
\[
\frac{\lambda^2(T)}{\lambda^2(0)} \approx 1 - \eta k T \mu_0 \lambda^2(0) / b (\phi_0/2\pi)^2 ,
\]

leading to:

\[
\frac{1}{\lambda} \frac{d\lambda}{dT} = \frac{d\ln(\lambda)}{dT} = \frac{\eta k \mu_0 \lambda^2(0)}{2b (\phi_0/2\pi)^2}.
\]  

b is a microscopic length, such as the spacing between CuO$_2$ bilayers, or the c-axis coherence length, or the $ab$-plane coherence length, all of which are about 10Å. $\eta$ is of order unity, and it varies from one theory to another, but its specific value is not relevant here because of the large discrepancy between theory and experiment. Below, we offer a quick derivation that contains the essential concept, and agrees to within constants on the order of $\pi$ with the more exact calculations.

The basic concept is that at low temperatures fluctuations in the magnitude of the order parameter cost too much energy, and fluctuations in the phase are the primary mechanism for reducing $n_s$. Since $\nabla \phi$ is proportional to the supercurrent density, this is equivalent to saying that the fluctuations involve flowing supercurrents, rather than fluctuations in the density of superconducting carriers. Two-dimensional supercurrent fluctuations can be modelled by assuming that all of the carriers in a volume $\xi \times \xi \times b$ move as a unit. $\xi$ is the coherence length in the film, and $b$ is a length that depends on the coupling between the superconducting layers. The total mass of the superconducting electrons in this volume is then $n_s \xi^2 b m^*$. Using the equipartition theorem in two dimensions:
\[ \left< v_i^2 \right> = \frac{2kT}{n_m \xi^2 b}. \] (19)

These thermal supercurrents cause a reduction in the equilibrium \( |\psi| \) just as an externally generated supercurrent would\[71-72\], so

\[ \frac{\delta \lambda}{\lambda} = \frac{1}{2} \frac{\delta n}{n} = \frac{m^* kT}{\hbar^2 n b}. \] (20)

Given that \( 1/\lambda^2 = \mu_0 n_e^2/m^* \), and \( \phi_0 = \hbar/2e \),

\[ \frac{\delta \lambda}{\lambda} = \frac{\pi^2 \mu_0}{\phi_0^2} \left( \frac{\lambda^2}{b} \right) kT. \] (21)

This is a factor of \( 2\pi/ln2 \) (\( \approx 9 \)) larger than the expression given by Coffey. The change is linear in \( T \) and \( dln(\lambda)/dT \) increases with \( \lambda^2 \), features that all models have in common. Equations 17 and 21 show that the fractional change in \( n_s \), which is proportional to \( \lambda^2 \), is expressed as the ratio of the thermal energy, \( kT \), to a characteristic superconducting energy, \( (\phi_0/2\pi)^2/[\mu_0 \lambda^2/b] \).

This theoretical result is robust. It is insensitive to parameters such as the anisotropy, \( m_\parallel/m_{ab} \), in the effective mass. The same GL theory would apply for the anisotropic-\( s \) and \( d_x^2d_y^2 \) order parameters. The detailed theoretical results are that \( dln(\lambda)/dT \) can increase more rapidly than \( \lambda^2(0) \), but probably not less rapidly. One wonders whether quantum effects could "freeze out" the classical thermal fluctuations,
but theory finds that they do not, as long as the Coulomb interaction is adequately screened[69]. Screening is adequate when the $ab$-plane resistivity does not exceed some limit, which is at least several mΩ-cm. According to theory, when the resistivity exceeds this amount, quantum fluctuations become so strong that $T_c$ is suppressed to zero, so this limit is not a concern for the films discussed herein. We emphasize that the theory of Roddick and Stroud[50] demonstrates that this result holds whether the film is homogeneous and uniform, or granular. In addition, these fluctuations have been observed at the expected magnitude well below $T_c \approx 2$K in thin ($d<\xi$) Al films, through their effect on the relaxation of a nonequilibrium quasiparticle charge imbalance[72]. Hence, they surely exist and contribute to the $T$-dependence of $\lambda$. The question is, what fraction of the observed effect is due to fluctuations? This must be answered experimentally.

Intentionally disordered films provide a good test of whether this mechanism is the likely cause of the observed linear dependence in crystals. Measurements of $\lambda(T)$ in films have typically shown $\lambda(T) - \lambda(0) \propto T^2$, which has been attributed both qualitatively and quantitatively to slight disorder affecting the $d$-wave density of states.[9,13] This slight disorder barely affects the values of $\lambda(0)$ and $\xi$, and a 3.5 Å/K linear term in $\lambda(T)$ should be visible, but is not. More recent experiments, including pure YBa$_2$Cu$_3$O$_{7-\delta}$ films in this thesis, have seen $\lambda(T) - \lambda(0) \propto T$ with slopes near 4 Å/K.[7,8] Films intentionally doped with Ni and Zn have significantly higher values of $\lambda(0)$, ranging up to 1.2 μm for 6% Ni. In addition, removing oxygen from the films changes the carrier density and therefore $n_s$, and can also increase $\lambda(0)$ to ~1μm. The
theory of Roddick and Stroud predicts that \( d\lambda/dT \) for films with \( \lambda(0) = 1\mu m \) should be around 100 Å/K. Slopes this large should be clearly visible.

The films we study are the same as in the previous chapter. The data on the Zn-doped films is insufficient to draw any conclusions because \( T_c \) is low and the data was not taken at low enough temperatures. The 2% Ni data is too noisy to draw any conclusion on the low-temperature \( \lambda(T) \). This leaves the laser ablated 4% Ni film, the 6% Ni film, and the codeposited 4% Ni film used for comparison in figure 17. In addition, we removed oxygen from the laser ablated 4% Ni film to increase \( \lambda(0) \). All of the data shown was measured with \( \lambda^3/d \) accurate to \( \pm 500\) Å except the codeposited 4% Ni data, whose baseline \( M' \) was adjusted slightly to give good agreement with the 4% Ni laser data.

Chapter VI describes how \( \lambda(0) \) depends on dopant in these films, but here we focus on \( \lambda(T) \) at low temperatures. Although the absolute accuracy in the measurement of \( \lambda(T) \) is about 150Å in the pure film, and smaller in the doped ones, the precision in the temperature dependence \( \lambda(T) \) can be as small as \( \pm 1\) Å on top of this. It is therefore quite easy to determine whether the low-temperature dependence is linear or quadratic, as this does not depend in any way on the actual value of \( \lambda(0) \). In addition, the coefficient of any linear term (i.e. \( d\lambda/dT \) or quadratic term (i.e. \( 1/2 d^2\lambda/dT^2 \)) is relatively insensitive to the chosen value of \( \lambda(0) \) within the error bars. As \( \lambda(0) \) is accurate to better than 10%, these coefficients are also accurate to better than 10%. A linear term of \( \sim 100\) Å/K would be impossible to miss.
We now attempt to fit the data below 20K in the films a Taylor expansion: \( \lambda(T) = \lambda(0) + c_1T + c_2T^2 \) to see whether the lowest temperature data is linear or quadratic and to set bounds on the size of \( c_1 \) that is consistent with the data. Figure 19 shows \( \lambda(T) \) for a pure \( \text{YBa}_2\text{Cu}_3\text{O}_{7-\delta} \) film. \( \lambda(4K) \) is 1820 ± 130Å, and the linear dependence at low temperatures is clearly visible. The dashed line is a best fit to \( \lambda(T) = \lambda(0) + c_1T \), \( \lambda(0) = 1805\text{Å} \) and \( c_1 = 7.4\text{Å/K} \), both values slightly larger than the data of Hardy et al. The dotted line is the best fit to \( \lambda(T) = \lambda(0) + c_2T^2 \), \( \lambda(0) = 1850\text{Å} \), \( c_2 = .28\text{Å/K}^2 \), which obviously does not fit the data. From examination of fits with \( c_2 \) in the range \( 0 < c_2 < 0.28\text{Å/K}^2 \), we estimate \( c_1 = 7 ± 2\text{Å/K} \). Interestingly, we find both \( d\lambda/dT \) and \( d\ln(\lambda)/dT \) (0.0033/K ± 0.001/K) to be somewhat larger than found in crystals. Observation of \( T \)-linear behavior in pure \( \text{YBa}_2\text{Cu}_3\text{O}_{7-\delta} \) films establishes that there is nothing about the thin film geometry \textit{per se} which reduces the \( T \)-linear behavior seen in crystals.

Figure 20 shows \( \lambda(T) \) for the laser ablated 4% Ni film. \( \lambda(4K) \) is 3920 Å and the data is still linear at the lowest temperatures. The dashed line shows the best fit to \( \lambda(T) = \lambda(0) + c_1T + c_2T^2 \) below 20 K, \( \lambda(0) = 3874\text{Å} \), \( c_1 = 9.1\text{Å/K} \), \( c_2 = .149 \text{Å/K}^2 \). A line with \( c_1 = 12 \text{Å/K} \), \( c_2 = 0 \) would also be a reasonable fit. The dotted line shows the best fit to a pure \( T^2 \) law with no linear term. It is not a reasonable fit to the data. The 4% Ni film definitely has a linear term in \( \lambda(T) \) at the lowest temperatures with a coefficient of 9 ± 3 Å/K. Interestingly, it is the same as found in undoped \( \text{YBa}_2\text{Cu}_3\text{O}_{7-\delta} \) films, and is therefore in complete agreement with the observation in crystals that \( d\lambda/dT \) is unaffected by Ni doping\[49\]. Thus, \( d\ln(\lambda)/dT = 0.0018 ± 0.0005/K \) in 4% Ni-doped \( \text{YBa}_2\text{Cu}_3\text{O}_{7-\delta} \) films, about half the value found in pure films, even though \( \lambda^2(0) \) is larger
Fig. 19. $\lambda(T)$ for the 1000Å thick pure YBa$_2$Cu$_3$O$_{7.8}$ film. The dashed line is a linear fit with a slope of 7.4Å/K, and the dotted line is a $T^2$ fit.
Fig. 20. $\lambda(T)$ for the 1000Å thick 4\% Ni-doped film. The dashed line is a fit to a Taylor expansion. The dotted line is a $T^2$ fit.
by a factor of 4.6. This is a factor of 9 lower than predicted, which is well outside the range of experimental uncertainty.

Similar results are obtained on the codeposited 4% Ni film. $\lambda(T)$ is linear in $T$ at the lowest temperatures with $d\lambda/dT \approx 10 \text{ Å}/\text{K}$, just as for the laser ablated film. Figure 21 shows the codeposited 4% Ni data. The dotted line is a $T^2$ fit, and the dashed line is a linear fit with $d\lambda/dT = 12 \text{ Å}/\text{K}$. The laser ablated 4% Ni data is shown for comparison.

Figure 22 shows $\lambda(T)$ for the 6% Ni film. $\lambda(4K) = 12440 \pm 60 \text{ Å}$, and the data have no obvious linear term at low temperatures. The dotted line shows the best fit to $\lambda(T) = \lambda(0) + c_1 T + c_2 T^2$ with $c_1$ fixed at 0 Å/K. $\lambda(0) = 11569\text{Å}$, $c_2 = 2.0 \text{ Å}/\text{K}^2$. The dashed line shows the best fit with $c_1$ fixed at 20 Å/K. The data is fitted perfectly well with no linear term at all, we estimate that $c = 5\text{Å}/\text{K} \pm 5\text{Å}/\text{K}$, so that $d\ln(\lambda)/dT = 0.0004/\text{K} \pm 0.0004/\text{K}$. This is one-eighth of the value in pure films, even though $\lambda^2(0)$ has increased by a factor of 47. This experimental value is a factor of 380 lower than predicted for thermal phase fluctuations.

The fluctuation predictions depend only on the value of $\lambda(0)$ and not on how that value of $\lambda(0)$ was obtained. Specifically, the predictions do not depend on whether $\lambda(0)$ is large because of the large scattering rate which we suspect to be the cause in the 6% Ni film, or because of a smaller superconducting carrier density. Therefore, we removed oxygen from the laser ablated 4% Ni film to remove carriers and increase $\lambda(0)$ to approximately 1μm. The effect of phase fluctuations should be the same in both films. It would be surprising to discover a linear term in the deoxygenated 4% Ni sample that
Fig. 21. Comparison of low-temperature $\lambda(T)$ for laser ablated and codeposited 4% Ni films.
Fig. 22. $\lambda(T)$ for the 3000Å thick 6% Ni-doped film. The dotted line is a $T^2$ fit and the dashed line is a fit to the Taylor expansion with $c_1 = 20$ Å/K.
was not present in the 6% Ni film.

Figure 23 shows $\lambda(T)$ for the deoxygenated 4% Ni sample. The temperature was lowered to 1.7K by pumping on the liquid He until the vapor pressure was below 20 Torr. $\lambda(1.7K)$ is 1.0008 $\mu$m, and the data is incredibly flat at low temperatures. There is no discernable linear term, and $\lambda(T)$ is even flatter than the best fit to $\lambda(T) = \lambda(0) + c_2 T^2$, shown by the dashed line. The results for both films with $\lambda(0) \approx 1 \mu$m are the same. No linear term is evident in $\lambda(T)$, even though $d\lambda/dT$ as small as 5 Å/K could be detected. Certainly there is no linear term $\sim 100$ Å/K. The data on the two films with $\lambda(0) \approx 1 \mu$m are consistent with each other and with the absence of $T$-linear behavior, $d\ln(\lambda)/dT = 0$. As noted above, theory predicts $d\ln(\lambda)/dT = 0.15/K$ when $\lambda(0) = 1 \mu$m. The discrepancy is roughly 400 times larger than the experimental uncertainty.

We can safely conclude that most of the linear $T$ dependence in pure YBa$_2$Cu$_3$O$_{7.8}$ is not from thermal phase fluctuations. Regardless of film fabrication technique or method of increasing $\lambda(0)$, the slope $d\ln(\lambda)/dT$ decreases while theory predicts an increase. Invoking granularity does not help.

This still leaves the issue of where the thermal fluctuations are. Suppose that the fluctuations are 10% of the $T$-linear behavior in pure YBa$_2$Cu$_3$O$_{7.8}$, as expected from theory when conventional values for materials parameters are used, and suppose that the rest of the effect is from nodes in the order parameter, which is strongly reduced by disorder. Thermal fluctuations should yield $d\ln(\lambda)/dT \approx 0.015/K$ when $\lambda(0) = 1 \mu$m, so that $\lambda^2(T)$ would extrapolate to zero at 30K, well below the measured $T_c$'s. The experimental upper limit on $d\ln(\lambda)/dT$ at low $T$ is less than 10% of this.
Fig. 23. $\lambda(T)$ for the deoxygenated 4% Ni film. The dashed line is a $T^2$ fit.
We can only conjecture. It is possible that the length, $b$, that enters the theories through the two-dimensional penetration depth, $\lambda^2/b$, is larger than its assumed value of about 10Å in pure YBa$_2$Cu$_3$O$_{7-\delta}$. The picture would be that the Coulomb interaction couples phase fluctuations in different CuO$_2$ bilayers. The upper limit on $b$ would be the film thickness, which would be large enough to account for a reduction in the expected effect by a factor of up to several hundred for the films studied here. This would put the effect below the experimental resolution.

Emery and Kivelson[69] mention that their model describes phase ordering in the X-Y universality class. Therefore, another consequence of their model is the existence of a large region near $T_c$ where critical phase fluctuations dominate. The penetration depth near $T_c$ would no longer have mean-field behavior, $\lambda(T) \propto (T_c - T)^{1/2}$, but instead would follow the 3D-XY model predictions, $\lambda(T) \propto (T_c - T)^{1/3}$. Emery and Kivelson suggest that the $T_c$ in pure YBa$_2$Cu$_3$O$_{7-\delta}$ is nearly as large as is theoretically possible for a metal this poor, and bears no relation to the mean-field $T_c$, which could be considerably higher. Again, this prediction can be checked by measuring $\lambda(T)$ near $T_c$ to see if it displays mean-field or 3D-XY behavior.

Lobb[5] has estimated the size of the critical region in which the Ginzburg-Landau mean-field theory is no longer expected to be valid as

$$|T - T_{c0}| < 1.07 \times 10^{-9} \frac{G/K^2}{\kappa^4 T_c^3 / H_{c2}(0)}$$

(22)
The large values of $\kappa$ and $T_c$ in YBa$_2$Cu$_3$O$_{7-\delta}$ suggest a large critical region, but the best estimates of these parameters gives a critical region less than .2K wide, although a 2K critical region is not inconceivable. Kamal et al.[14] find that $1/\lambda^3$ is linear over ~20K, to within 1mK of $T_c$. Agreement over a range this large is likely accidental, as one of the assumptions of both the 3D-XY and mean-field calculations is that $T$ is very near $T_c$. If $T$ is even a few degrees below $T_c$, mean-field behavior can look linear in $1/\lambda^3$. Therefore, one needs to induce very small currents to avoid exceeding the critical current near $T_c$ in the films.

Figure 24 shows both the resistive and inductive parts of the signal in the pick-up coil with a drive coil current of 250\mu A in a pure YBa$_2$Cu$_3$O$_{7-\delta}$ film. The transition is extremely sharp, less than .2K wide, and the signal does not change when the drive current is reduced by a factor of ten, implying that the film response is in the linear regime. We obtain the real and imaginary parts of the conductivity in this film, which is shown in figure 25. The "steps" in the figure are due to discreetness in the digital signal from the lock-in amplifier being read by the computer. $\sigma_2$ is proportional to $1/\lambda_2$, and appears linear in $(T_c - T)$ to within .2K of $T_c$, which is about the width of the critical region to be expected from the calculation of Lobb[5]. We do not see evidence of 3D-XY fluctuations outside this region. Figure 26 shows $1/\lambda^3$ vs. $T$ for this film. It also appears linear to within a few degrees of $T_c$, but the data deviate obviously from a line at a temperature well below $T_c$.

Figure 27 shows the complex conductivity in a SmBa$_2$Cu$_3$O$_{7-\delta}$ film made by "MBE", with a transition region about .7K wide. Again, $1/\lambda^2$ is linear to within .7K of
Fig. 24. Inductive and resistive pick-up coil signal near $T_c$ in a pure $\text{YBa}_2\text{Cu}_3\text{O}_{7.8}$ film.
Fig. 25. Both components of the complex conductivity near $T_C$ for a pure YBa$_2$Cu$_3$O$_{7.5}$ film.
Fig. 26. $1/\lambda^3$ for a pure $\text{YBa}_2\text{Cu}_3\text{O}_{7.4}$ film.
Fig. 27. Both components of the complex conductivity near $T_c$ for a SmBa$_2$Cu$_3$O$_{7.5}$ film.
$T_c$, implying a critical region smaller than .7K. Figure 28 shows $1/\lambda^3$ near $T_c$, again showing the deviation from linearity a few degrees below $T_c$.

The small critical regions near $T_c$ in these films are consistent with the lack of fluctuation effects in the low-temperature $\lambda(T)$. We conclude that although fluctuations are expected to play a larger role in YBa$_2$Cu$_3$O$_{7-\delta}$ than in the lower-$T_c$ superconductors, their effects are still negligible until the temperature is within $\sim .2$K of the transition.
Fig. 28. $1/\lambda^3$ for a SmBa$_2$Cu$_3$O$_{7+4}$ film.
CHAPTER VIII
Concluding Summary

We have measured the penetration depth $\lambda(T)$ in several pure, Ni-doped, and Zn-doped $\text{YBa}_2(\text{Cu}_x\text{M}_{1-x})_3\text{O}_{7.4}$ films ($x = .02-.06$) to determine the effect of moderate amounts of disorder on the electronic density of states. We find that the penetration depth increases drastically with dopant concentration, about a factor of 2 for every 2% dopant. This is roughly consistent with what one would expect from scattering in a $d$-wave superconductor, but inconsistent with expectations for either isotropic or anisotropic $s$-wave superconductors. We show that a phenomenological model based on a broadening of the density of states with disorder can account for both the rapid increase in $\lambda(T)$ and the shape of $1/\lambda^2(T)$.

The linear temperature dependence in $\lambda(T)$ at low temperatures in pure YBCO crystals and some pure YBCO films is often considered evidence for $d$-wave pairing in YBCO. If this linear dependence were due to phase fluctuations, as some have suggested,[50,68-70], the effect of thermal phase fluctuations should be even more clearly visible in films with large values of $\lambda(0)$ (e.g. the Ni- and Zn-doped films, as well as oxygen depleted Ni-doped films). The fact that they are not is strong evidence that phase fluctuations are not responsible for the observed linear dependence in pure YBCO. Even $d$-wave superconductors should have phase fluctuation effects at low $T$,\n
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though. The undetectability of these fluctuations remains to be explained. They must be present below our ability to detect them.

During the course of this investigation, I developed an apparatus for measuring $\lambda(T)$ from the change in the mutual inductance of two coils on opposite sides of the superconducting film that is currently the only method for measuring both the absolute value of $\lambda(0)$ and the temperature dependence $\lambda(T)$ to good precision. This probe is currently the best in the world for measuring $\lambda$, if you are willing to tolerate measurements in films rather than crystals.

Our results are strong evidence that $d$-wave pairing is responsible for superconductivity in YBa$_2$Cu$_3$O$_{7-\delta}$, and that the effect of thermal fluctuations is negligible over the entire temperature range, except within perhaps .2K of the transition.
REFERENCES


52. S.M. Anlage, to be published in Proc. of Conf. on Spec. in Novel Superconductors, Stanford (USA), March 15-18, 1995.


