EXPERIMENTAL STUDIES ON PENETRATION DEPTH IN YBa$_2$Cu$_3$O$_{7-\delta}$ 

SUPERCONDUCTORS

DISSERTATION

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By

JuYoung Lee, B.S., M.S.

* * * * *

The Ohio State University

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Dissertation Committee:

Dr. Thomas R. Lemberger  
Dr. Arthur J. Epstein  
Dr. Robert L. Mills

Approved by

Advisor  
Department of Physics
TO MY PARENTS AND MY SISTER
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VITA

Feb. 1, 1961 ................ Born, Korea
1983 ....................... B.S. in Physics, Seoul National University, Seoul, Korea
1985 ....................... M.S. in Physics, Seoul National University, Seoul, Korea
1986-1987 ................... Teaching Assistant, University of Cincinnati (no degree)
1987-1988 ................... Teaching Assistant, Department of Physics, OSU
1988-1993 ................... Research Assistant, Department of Physics, OSU

PUBLICATIONS


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Research Adviser: Thomas R. Lemberger, Professor of Physics
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CHAPTER 1

INTRODUCTION

The motivation of this work is to understand the inductive behavior of superconducting electrons in films of high-$T_c$ superconductors. Films have many potential applications. We focus on YBa$_2$Cu$_3$O$_{7-\delta}$ because it yields the highest film quality. Variations in the reported penetration depth $\lambda(T)$ measurements from film to film and lab to lab suggest grain boundary effects in their measurements. However we find that our data can be explained by $d$-wave superconductivity with a small disorder. We conjecture that the sensitivity of $d$-wave superconductivity to disorder accounts for the observed variations between groups. Thus our results bring additional support to the growing body of evidence supporting $d$-wave superconductivity and establish that fundamental studies of superconductivity can be done in films.

For a $d$-wave superconductor to indeed be accepted, it must account for $\lambda(T)$, because $\lambda(T)$ is the fundamental transport property of superconductors and because it can be measured with good precision. In the following, we begin with a discussion of the issue of the $d$-wave superconductor and several other types of measurements that support $d$-wave, and then focus on the role of $\lambda$ regarding the issue.
There is a great deal of interest in whether the high-$T_c$ cuprate superconductors are conventional ($s$-wave) or unconventional ($d$-wave). The wave function of the Cooper pairs in superconductors is the product of a spin part and an orbital part. In Knight shift measurements on high-$T_c$ superconductors, a rapid drop in the shift was observed below $T_c$, as in BCS superconductors. [1,2] This suggests that electrons are paired in spin singlets on transition so that the net magnetic field at the nuclear sites due to electrons is diminishing below $T_c$. The Pauli exclusion principle requires that the total wave function of the electron pair be antisymmetric under interchange of electrons, and therefore dictates that the orbital part of the electron pair wavefunction be symmetric in a singlet superconductor. Generally, as the first two symmetric representations of point group possess orbital angular momentum of 0 and 2 ($s$-wave and $d$-wave), the orbital wave function in singlet superconductors is categorized in the same way. Thus there are only two candidates, $s$-wave and $d$-wave, regardless of the details of the pairing mechanism. As of now, in the field of cuprate high-$T_c$ superconductors, there is no consensus, but a growing body of evidence supports $d$-wave superconductivity. Therefore, the investigation of the pair symmetry, whether it is $s$-wave or $d$-wave, becomes very important due to the fact that it is independent of details of the model and provides a robust constraint on the contending theories.

The BCS theory was so successful that it explained most aspects of conventional superconductors. In the BCS theory, superconductivity arises from the pairing of two quasiparticles. According to Landau Fermi liquid theory, the strongly-interacting bare electron is equivalent to a noninteracting dressed electron called a quasiparticle. Fermi
liquid theory is the basis for describing the normal state of a conventional superconductor. High-$T_c$ superconductors show several properties that are difficult to explain with the BCS theory. Some theories try to find the solution even outside Fermi liquid theory. [3,4] The anomalous properties observed in high-$T_c$ materials have been a puzzle, and so far there is no clear understanding of this material. We list few of these anomalous properties:

1. high $T_c$ and short coherence length;
2. linear in $T$ dc resistivity in the normal state;
3. absence of a coherence peak and a $T^3$ dependence at low $T$ in the nuclear relaxation rate;
4. rapid drop of the electron scattering rate below $T_c$ as shown by surface resistance;
5. Raman continuum;
6. midinfrared absorption;
7. anomalous dynamic structure factor $S(q,\omega)$ in polarized neutron scattering;
8. proximity to antiferromagnetic insulating phase;

Existing theories of high-$T_c$ superconductivity attempt to explain the properties listed above.

One of the most well developed theories is by Pines et al. and proposes the $d$-wave pairing. It finds spin fluctuations are a possible source of pairing in cuprate superconductors. [5] This idea originated from the fact that YBa$_2$Cu$_3$O$_{7-\delta}$ is an antiferromagnet when it is in the insulating phase ($O_\delta$), and that there is a remnant of spin fluctuations in the lattice when it is in the metallic phase upon doping ($O_T$). Pines
et al. used the NMR relaxation rate data on the normal state to make a quantitative fit to their phenomenological theory, where the imaginary part of the spin-spin correlation function peaks at the antiferromagnetic wave vector. [5] There is a temperature dependent spin fluctuation in the nearly localized Cu$^{+2}$ orbital, which has antiferromagnetic correlation in the normal state. They showed a retarded interaction between quasiparticles is induced by spin fluctuations. In their model charge carriers are described as forming an antiferromagnetic Fermi liquid. The quasiparticle scattering rate is deduced to be proportional to the maximum of either frequency or temperature, \( \max[\omega, T] \), and the quasiparticle lifetime has its origin in the quasielastic scattering of quasiparticles by the low-frequency spin fluctuations. In this model \( T_c \) is also deduced from the coupling energy between quasiparticles and spin fluctuations, which is larger than that for phonons in conventional superconductors. In \( d \)-wave pairing the coherence peak is absent because the logarithmic singularity in the density of states is weak.

The interaction potential \( V_{k,l} \) between the two conduction electrons is mediated by scattering from spin fluctuations in the lattice, and then the electron changes momentum from \( k \) to \( l \). \( V_{k,l} \) is a function of \( k-l \) only, the scattered wavevector, and is denoted by \( V_{k-l} \). It is repulsive, and peaks when \( k-l \) is equal to \( Q=(\pm \pi/a, \pm \pi/a) \); where \( Q \) is the peak in the spin-spin correlation function in antiferromagnetism, and \( a \) is the lattice constant. In spite of the repulsive interaction, the net energy in the system can be reduced when \( \Delta(k) \) changes sign upon translation by the wavevector \( Q=(\pm \pi/a, \pm \pi/a) \). Neutron scattering has revealed that the imaginary part of the copper site spin susceptibility \( Im\{\chi(Q, \omega)\} \) is peaked at wave vector \( Q \), and its frequency range spans up
to $50 \text{meV}$. [6] $\text{Im}\{\chi(Q, \omega)\}$ is proportional to the effective potential of the charge carriers, which originates from the oxygen sites. Following a weak coupling formalism, the general form of the full integral equation for the $k$-dependent order parameter in the ground state can be written as

$$\Delta_k = - \Sigma_{k,l} V_{k,l} \Delta_l / 2E_l.$$  

$\Delta_k$ is only required to change sign on translation by $k - l = Q$, where $V_{k,l}$ is most significant for the integral equation to have a nontrivial solution. This results in $d$-wave pairing with $d_{x^2-y^2}$ symmetry.

Varma et al. proposed marginal Fermi liquid theory, which starts with a broad continuum of boson excitations with which a charge carrier interacts. [3] In this picture, these excitations are observed in the electronic continuum in Raman spectra and the midinfrared absorption in the $ac$ conductivity. They deduced that the scattering rate of conduction carriers is proportional to $\max[\omega, T]$. They also explained the absence of a coherence peak in the nuclear relaxation rate and the high $T_c$. There is drop in the low energy excitation spectrum of the Raman continuum below $T_c$. It seems to be related to the rapid decrease in scattering rate observed by surface resistance measurements. It can be explained by a decrease in the inelastic scattering as the energy gap develops in the low energy excitation spectrum below $T_c$. Surface resistance experiments at higher frequencies, at terahertz, show the slow-down in the decrease of the scattering rate. [7] The cut-off energy in the bosonic excitation spectrum is large; thus the $T_c$ can be large.
Since there is no $q$ dependence in the bosonic excitation spectrum, the pairing is $s$-wave.

Anderson et al. proposed the resonance valence bond (RVB) state theory. [4] The motivation is to incorporate the fact that the superconducting phase is very close to antiferromagnetism in the insulating phase. Recently they reported a mechanism of interlayer tunneling that results in a high gap anisotropy. A high $T_c$ and no coherence peak in the nuclear magnetic relaxation rate are also derived, though the theory gives only qualitative results. [8] The resulting gap anisotropy is similar to a $d_{x^2-y^2}$ states in the sense that gap minimum is located diagonally across the Fermi surface, but the crucial difference is that the energy gap doesn't change the sign at different orientations. The pairing is highly anisotropic $s$-wave. It is indistinguishable from $d$-wave pairing in most experiments that are sensitive to the magnitude of the gap if the gap minimum is sufficiently small.

Before discussing measurements of $\lambda$ in detail, we review several other measurements that support $d$-wave pairing. They are NMR spin-lattice relaxation rate, angle-resolved photoemission, surface resistance, infrared reflectance, and $dc$ SQUID measurements with the crystal as a part of the SQUID loop. [9-16]

The NMR spin-lattice relaxation rate in a conventional superconductors shows a coherence peak just below the $T_c$ and an $\exp(-\Delta/kT)$ temperature dependence at low $T$. [17] In the cuprate superconductors, neither of these were observed, and the relaxation rate in the zero-field limit decreased like $T^3$ below $T_c$. [9] Theoretical calculation is possible in the zero field limit, thus comparison is possible. This result suggests a line node in $\Delta(k)$. 
Shen et al. measured the magnitude of the energy gap in BSCCO in different directions on the Fermi surface using the angle-resolved photoemission. [13] In this method, the leading edge of the photoemission peak coincides with the Fermi surface. As the gap opens, the position of the leading edge shifts as much as the gap energy since this energy is added to the binding energy of the photoelectron. By monitoring these shifts of the leading edge of the peak above and below \( T_c \), the magnitude of \( \Delta(k) \) is probed in various directions on the Fermi surface. According to their results on a Bi\(_2\)Sr\(_2\)CaCu\(_2\)O\(_{8+\delta}\) crystal, the minimum energy gap is located diagonally across the Fermi surface. This result is consistent with a \( d_{x^2-y^2} \) symmetry of pairing. The magnitude of the gap maximum is approximately 20meV and is located in other directions. The resolution in their measurement is about 4meV, thus it is not certain whether the minimum energy gap is zero or finite within experimental error.

Bonn et al. measured the surface resistance of YBCO crystals at microwave-frequency currents. [18,19] In this frequency range, current carried by a normal fluid is detectable, and the drive frequency is much smaller than the pair breaking threshold value \( 2\Delta/h \). The surface resistance is proportional to the real part of the conductivity in the zero frequency limit below \( T_c \). Thus the main focus of this experiment is the measurement of the scattering rate, \( 1/\tau \), below \( T_c \). In high-\( T_c \) superconductors it decreases nearly three orders of magnitude. The functional form of the \( T \)-dependence is \( \exp(T/T_0) \), and it is different from the thermally activated form \( \exp(-T_0/T) \). The result implies that there is a kind of bosonic excitation and it induces inelastic scattering, which is responsible for the linear \( T \) dependence of \( dc \) resistivity above \( T_c \). Below \( T_c \) the
excitation is depressed rapidly. Measurements of spin relaxation such as nuclear quadrupole resonance (NQR) reveals the same exponential $T$ dependence. [9] Spin fluctuations are a good candidate for both $1/\tau$ and $1/T_1$.

Infrared reflectance has been used in the search for the existence of a BCS gap in cuprate superconductors. In a conventional superconductor, at low $T$ there is a total reflection from the surface of the superconductor when the frequency of light is smaller than $2\Delta/h$, which is the threshold frequency for the pair breaking. But there is one complication. As mentioned above, the scattering rate in high-$T_c$ superconductors drops very rapidly below $T_c$ upon the superconducting transition, and it becomes very small when compared to $2\Delta/h$. Cuprate superconductors are in the clean limit below $T_c$. [15,18,19] A large part of the rise in the reflectance below $T_c$ is caused by the rapid decrease in the scattering rate, and it needs to be separated from the effect of the superconducting electrons. In order to get around this problem, preparation of dirty superconductors by doping with various impurities or irradiation was devised, and the result still showed the absence of a gap. [15,19,20] Since the above measurements are sensitive only to the magnitude and not to the sign of the gap, the above results still have room for interpretation as being anisotropic $s$-wave pairing.

Wollman et al. performed an experiment that is sensitive to the sign of $\Delta(k)$ when $k$ is in different orientations so the symmetry of the pairing state of the cuprate superconductor can be directly determined. [16] A corner of a YBCO single crystal was used to form a $dc$ YBCO-Pb SQUID. (In general, a closed loop connected by 2 pieces of superconductor at two Josephson junctions forms a SQUID.) For half of the SQUID,
a YBCO crystal is used; Pb covers the other half. In $d_2^{2+2y}$ symmetry, the current in the $a$-direction carries the opposite sign of $\Delta(k)$ to that in $b$-direction. These orientations of the current in the two Josephson junctions are designed to be in the $a$- and $b$-directions, respectively, by attaching the junctions on the two faces that meet at the corner of the crystal. There should be a phase shift of $\pi$ relative to a conventional SQUID in the plot of the critical current versus the magnetic flux in the SQUID loop if YBCO is a $d$-wave superconductor. Though there is large scattering in the data so that more improvements in the measurement are required to make an obvious determination, the result favors a phase shift of $\pi$. Also a single Josephson junction made on the corner of a crystal was tried since it should also have a similar phase shift of $\pi$ in the Fraunhofer pattern, if it is a $d$-wave superconductor, with the result favoring $d$-wave pairing.

Now we address the importance of the penetration depth measurement to the issue of $d$-wave superconductivity. The magnetic penetration depth, $\lambda$, is one of the characteristic lengths of superconductors. The change of $\lambda$, $\Delta \lambda(T) \equiv \lambda(T) - \lambda(0)$, as $T$ increases from low $T$ is roughly proportional to the number of thermally excited quasiparticles. $\lambda(0)$ is the penetration depth at zero temperature. In a conventional superconductors ($s$-wave), the energy gap $\Delta(k)$ is finite in all directions, where $k$ is a wave vector in momentum space, thus causing $\Delta \lambda$ to be an exponential function of $T$, $\exp(-\Delta/k_B T)$. $\Delta \lambda$ is nearly 0 up to a certain threshold value of $T=0.2T_c$ in weak coupling BCS, but once that value is reached, it increases rapidly. In unconventional superconductors, $\Delta \lambda$ is proportional to a power of $T$. Unconventional superconductors are characterized by the existence of a sign change in the order parameter, or
equivalently energy gap $\Delta(k)$, as a function of the orientation of $k$ on the Fermi surface. Then the existence of zeros in $\Delta(k)$ usually follows. The volume of phase space around the zeros of $\Delta(k)$ determines the number of excited quasiparticles and leads to the power law dependence of $\Delta \lambda$ on $T$. According to Goldenfeld et al., intrinsic $d$-wave superconductors of orthorhombic lattice symmetry should have $\Delta \lambda \propto T$ at low $T$ regardless of the location of the line node and the direction of current in the twinned $a$-$b$ plane. They also suggested the disorder due to impurities could alter the $T$-dependence at low $T$ from $T^1$ to $T^2$. Contrary to the earlier report on the measurements of $\lambda(T)$, the $T^2$-dependence and the linear $T$-dependence have been observed in thin films and single crystals, respectively, as the resolution was improved.

In this thesis, we report the observations of $\lambda(T)$ in films of YBCO which show intermediate behavior between crystals, which look clearly $d$-wave, and deliberately-disordered films, from which a bridge can be built. There have been theories about the effect of microscopic impurities on $d$-wave superconductivity, which was developed during the interpretation of penetration depth data on heavy-fermion superconductors [21-25]. The impurity effect is another detectable phenomenon. Generally $d$-wave superconductivity is sensitive to impurities. It accompanies the rapid drop in superfluid density and the change of $T$-dependence from linear $T$-dependence to $T^2$-dependence at low $T$. The investigation of the correlation among $\lambda(0)$, $T$-dependence at low $T$, and $T_c$ obtained from the penetration depth measurements along with the oxygen depletion of samples can lead to determination of the symmetry of pairing.
In Chapter II, we review various experiments on the penetration depth. The numerical model of a two-coil method and the experimental method, including fitting procedure, are described in Chapter III. In Chapter IV, the film data are compared with existing theory describing the impurity scattering in $d$-wave superconductors. In Chapter V, a systematic study on the oxygen depletion effect is made to probe the correlation of $T_c$, $\lambda(0)$ and the crossover behavior. Then a brief concluding summary is given in Chapter VI.

In Appendices A and B, measurements of the Bernoulli voltage in $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ and $\lambda(T)$ determined from kinetic inductance measurements in a meander line geometry are given, respectively. The same sample geometry is used for both measurements. In Appendix C, the theory of the penetration depth of BCS superconductors is introduced to cover the non-local linear response in type-I superconductors. Finally a numerical modeling of two-coil method is given in the Appendix D.
CHAPTER II

REVIEW OF EXPERIMENTS ON THE PENETRATION DEPTH

When a magnetic field is applied parallel to the surface of superconductor, the magnetic field inside the superconductor attenuates exponentially in distance from the surface due to the screening current. The attenuation is related to the intensity of the screening current: in a Drude-like picture of superconducting electrons with density \( n_s \) and mass \( m^* \), the penetration depth is \( \lambda = (\mu_0 n_s e^2/m^*)^{-1/2} \), where \( \mu_0 \) is the magnetic permeability. The decay can be non-exponential when \( \lambda < \xi_0 \), where \( \xi_0 \) is the size of the Cooper pairs, because the linear response to the magnetic field is non-local. The induced screening current at a point is a response to the averaged field over a sphere of radius \( \xi_0 \). In high-\( T_c \) superconductors, \( \lambda \geq 1400 \text{Å} \gg \xi_0 \approx 20 \text{Å} \), so the electrodynamics is local.

High-\( T_c \) superconductors are extreme type-II and in the clean limit. Thus, the measured penetration depth is identical to London penetration depth \( \lambda_L \). On the other hand, most elemental superconductors are type-I BCS superconductors, where \( \xi_0 \) is larger than \( \lambda_L \). The \( T \)-dependence of \( \lambda \) requires the full description of non-local electrodynamics. However, the phenomenologically determined form \( \lambda^2(0)/\lambda^2(T) = 1 - T^4/T_c^4 \), called Gorter-Casimir "Two-fluid" model, is sometimes reasonable approximation.
to data for strong-coupling BCS superconductors. [26]

In high-$T_c$ superconductors, there is large anisotropy in effective mass: $m^*$ in the $c$-direction is 25 times larger than that of the $a$-$b$ plane in YBCO. [27] Since $\lambda$ is defined by $\lambda = (\mu_0 t s e^2/m^*)^{1/2}$, $\lambda_a$, $\lambda$ in the $a$-direction, is associated with the screening current in the $a$-direction. In the epitaxially grown thin film where the $a$-$b$ plane is parallel to the substrate, the measurements of $\lambda$ yield $\lambda_{ab}$, the average of $1/\lambda_a^2$ and $1/\lambda_b^2$.

Historically, $\lambda$ has been measured by several methods such as via the magnetic susceptibility of colloidal powder [28], $ac$ susceptibility of a bulk sample using a mutual-inductance bridge [29], with a microwave resonance cavity at $10^9$ to $10^{10}$Hz with resolution of $2\Lambda$ [30], $ac$ susceptibility with an oscillator circuit at $10^5$Hz with the same resolution as the microwave cavity, $e/c$. [31] A large fractional change in the magnetic susceptibility is achieved as a magnetic field penetrates into a large number of the colloidal particles whose size $d$ is comparable to $\lambda$. When the distribution of particle sizes is known, an absolute $\lambda(T)$ can be deduced, but with large uncertainty. The rest of the methods measure the change in field penetration $\lambda(T)$-$\lambda(0)$ except muon spin resonance ($\mu$SR) and isotherm magnetization method, both of which came out quite recently. Both are applicable to only to bulk type-II superconductors, since they require a vortex lattice. Since $\lambda$ is much smaller than the sample size, it is difficult to devise a method that can measure the absolute value of $\lambda$ with high precision. [32]

We call the methods that can measure only the change in $\lambda$ (or $\lambda^2$ in thin film) the "relative methods", and the one that can determine $\lambda(0)$ by itself as the "absolute
method". The relative methods rely on a fitting procedure to estimate \( \lambda(0) \) and to determine the \( T \)-dependence of the superfluid density that is proportional to \( 1/\lambda^2(T) \). In other words, \( \lambda(0) \) is a fit parameter in this type of method. For BCS superconductors, an exact microscopic theory exists; thus the form of target curve for the fitting procedure is known and the estimated \( \lambda(0) \) is quite accurate. For high-\( T_c \) superconductors, the form of the target curve is as yet unknown. \( \lambda(0) \) needs to be determined from the absolute method. The advantage of relative methods is that high precision (\( \leq 0.1\% \)) can be obtained; the absolute methods lack precision in the \( T \)-dependence of \( \lambda(T) \).

In the early reports, the measurement of \( \lambda(T) \) in \( YBa_2Cu_3O_{7-\delta} \) films and crystals was interpreted to be consistent with conventional superconductivity after fitting to the BCS weak-coupling and/or two-fluid models. [33-35] Later measurements with much improved resolution conflict with the earlier reports questioning the simple BCS interpretation. Muon spin resonance (\( \mu \)SR), which is one of two methods that can determine value \( \lambda(0) \) absolutely, reported that the zero-temperature value \( \lambda(0) \) is 1400Å; and the temperature dependence follows the two-fluid model. [27,36-38] Due to both the insufficient resolution below \( 0.2T_c \) and the prevailing viewpoint at that time, the early reports failed to address the issue of \( d \)-wave superconductivity. Pond et al. measured the microwave phase velocity in a transmission line composed of parallel high-\( T_c \) films above 20K. [39] They gave a hint of the \( T^2 \) dependence of \( \Delta \lambda \) at low \( T \). Soon after this study, Goldenfeld et al. [40] reanalyzed the data produced by Fiory et al., [34] and showed that the previously ignored \( T^2 \)-dependence exists in their data. Several groups have come up with similar \( T^2 \)-dependence in films as the resolution of \( \Delta \lambda \) is improved. [41,42,18]
In contrast, Hardy et al. reported measurements of a high quality crystal where \( \lambda(T) \) increases linearly with \( T \), namely \( \delta \lambda(T) = (4\text{Å}/K)T \), between 4K to 40K, and also \( \lambda(0) = 1500\text{Å} \). [43] Since this report is exactly what is expected for intrinsic \( d \)-wave superconductors, which possesses a line node in \( \Delta(k) \) thus leading to the linear \( T \) dependence of \( \Delta \lambda(T) \), it attracted much attention. It has been known that the nonmagnetic impurities can change the \( T \) dependence of \( \Delta \lambda(T) \) at low \( T \) from \( T^{-1} \) to \( T^{-2} \) in \( d \)-wave superconductors. [21-25] The \( T^{-2} \)-dependence observed in the films can be interpreted as a disorder-induced effect due to a small amount of strong nonmagnetic impurities imbedded in the films during the film production. Hardy et al. conjectured that the linear \( T \)-dependence in crystals may be extremely sensitive to minute amounts of impurities. Contamination can easily occur by impurities in the growth process and even at an impurity concentration of 0.15%, the linear \( T \) dependence is altered. In this section and the next, we review reports on \( \lambda \) measurements and methods on bulk sample and thin film, respectively.

1. Method for Bulk samples

The advantage of single crystals is that the sample in principle can be made with the highest crystal quality so that extraneous effects are minimized. Another advantage is that \( \mu \)SR and isothermal magnetization can be applied, which are the only two methods that can determine the absolute value of \( \lambda(0) \). The accepted value of \( \lambda(0) = 1400\text{Å} \pm 100\text{Å} \) in \( \text{YBa}_2\text{Cu}_3\text{O}_{7-\delta} \) is determined by \( \mu \)SR measurements on the single crystal and ceramic samples. Recently Hardy et al. determined \( \Delta \lambda(T) \) of a clean single crystal from
microwave cavity resonance with the resolution of about $\pm 1\text{Å}$, which is the most precise measurement known so far, though the resolution in the two-coil method for thin film is comparable to that.

A. Muon Spin Rotation ($\mu$SR) [18,27,36-38]

This is one of the two methods that can determine the absolute size of $\lambda$; all the other methods measure the change in $\lambda$ or $\lambda^2$. The application of this method is limited to type II superconductors in bulk form since it requires a vortex lattice and a finite volume of the material. A magnetic field of about 1 Tesla is applied to the specimen to form a vortex lattice. Muons, with spin polarized perpendicularly to the magnetic field, are incident on the specimen. The muon stops as it gets embedded in the specimen, and its spin precesses around the magnetic field. The muon decays into a positron and two neutrinos with a lifetime of $2.2\mu s$. The positron emitted along the muon-spin direction is detected. The clock is started when the muon enters the sample. If the magnetic field were uniform, the precession of muon spins would be coherent and there would not be any depolarization. Since there is a vortex lattice, a depolarization of muon spins occurs in a certain fashion which is determined by the local magnetic field distribution $\Delta B$. The difference in the positron count between the upper and lower detectors removes background error and gives information on the time dependent muon spin polarization $P(t)$. The time evolution of the depolarization is traced back to the field distribution inside the sample. In this method, the effective magnitude of the magnetic field inside the vortex is measured.
The time dependent depolarization is given by

\[ P(t) = P(0) \int F(B_\mu) \cos(\gamma_\mu B_\mu t) dB_\mu, \] (2)

where \( F(B_\mu) \) is the probability distribution of the local field that the muons experience, and \( \gamma_\mu = 2\pi \times 135.5 \text{MHz/T} \) is the muon gyromagnetic ratio. It is a kind of Fourier transform from the time domain to the field domain. A Gaussian distribution of static internal fields \( F(B_\mu) \) leads to \( P(t) \) that also has the Gaussian form

\[ P(t) = P(0) \exp(-\sigma^2 t^2) \cos(\gamma_\mu <B_\mu > t), \] (3)

where \( <B> \) is the average of \( F(B_\mu) \), and \( \sigma \) is the depolarization rate. The width of the field distribution \( <\Delta B^2> \) has the relation

\[ <\Delta B^2> = <B^2> - <B>^2 = 2\sigma^2 / \gamma_\mu^2. \] (4)

For an ideal triangular vortex lattice, \( F(B_\mu) \) has a cusp-shaped maximum whose origin is the saddle point in \( B(r) \) between the adjacent pair of vortices. In this case, the \( <\Delta B^2> \) has the relation to the penetration depth \( \lambda \) \[44\]

\[ <\Delta B^2> = 0.00371\Phi_0^2 \lambda^{-4}, \] (5)
where $\Phi_0 = hc/2e$ (CGS) is the magnetic flux quantum. The corresponding $P(t)$ can be approximated as a Gaussian, and from equations (4) and (5), $\lambda$ is deduced. Since the measured signal $\sigma$ only depends on $\lambda$ through the vortex lattice and independent of sample size, this method can determine $\lambda(0)$ absolutely.

Pümpin et al. tried several different analyses to see the variations in the deduced $\lambda(0)$. [36] The analyses are different according to the assumed form of field distribution as listed: (1) a least square fit to a Gaussian distribution, (2) correction of the superposition of an extrinsic peak due to the muon stopping on the cryostat window, (3) the real part of the complex Fourier transform of the muon polarization. With the assumption of regular vortex lattice, the deduced $\lambda$ is insensitive to any choice of the above assumed forms of field distribution. They agree within 100Å. But if the field distribution is caused by something other than a regular vortex lattice, such as vortex pinning or loose grain boundaries, a systematic error could occur. However, even 20% error in the estimate of $<\Delta B^2>$ leads to only a 5% error in $\lambda(0)$, therefore it is a quite reliable method that can determine the absolute value $\lambda(0)$. On the other hand, 1% error in the calibration typically results in several hundred percent error in $\lambda(0)$ for the "relative methods".

The temperature dependence obtained with this method provides the exact shape of the superfluid density, which is independent of the systematic error in absolute value of $\lambda(0)$, while other relative methods require the exact value of $\lambda(0)$ to determine the shape of the superfluid density. Pümpin et al. showed that the normalized form $\lambda^2(0)/\lambda^2(T)$, which is proportional to $\sigma(T)$, fits well to $1-(T/T_c)^4$ above 0.3$T_c$. It is noted
that high-$T_c$ superconductors correspond to the clean and local limits, $\xi_0/l < 0.3$ and $\xi_0/\lambda_L < 1$. Since a ratio of $\Delta/kT_c$ larger than 2 is required to make large slope in $\lambda(0)/\lambda^2(T) = 1-(T/T_c)^4$ near $T_c$ in the local limit, it is often referred to as an approximate strong-coupling behavior.

The measurements on crystals and sintered YBa$_2$Cu$_3$O$_{7-\delta}$ ceramics by Harshman et al. yield $\lambda_{ab}(0) \approx 1415\text{\AA}$ for crystal and $\lambda_{\text{eff}}(0) \approx 1740\text{\AA}$ for ceramics, respectively, in agreement with the two-fluid model within error bars. [27] After compensating the effect of anisotropic mass in ceramic sample, they obtained $\lambda_{ab} \approx 1415\text{\AA}$. $\lambda_{\text{eff}}(0) \approx 1500\text{\AA}$, from which $\lambda_{ab} \approx 1300\text{\AA}$ is deduced, is reported in polycrystalline sample by Pümpin et al. [36] Uemura et al. reported $\lambda_{ab} \approx 1460\text{\AA}$ in the polycrystalline samples. The accepted value of $\lambda_{ab}(0) = 1400\text{\AA}$ in YBCO mainly originates from the $\mu$SR measurements. The only reliable source for $\lambda_{ab}(0)$ is the $\mu$SR measurements and the error of $\pm 100\text{\AA}$ in $\lambda_{ab}(0)$ is the best accuracy that is available so far.

B. Isotherm magnetization: [45-48]

There is a wide range $H_{c1} << H << H_{c2}$ ($H_{c2}/H_{c1} \approx 10000$) in a high-$T_c$ superconductor where the spacing $L$ between vortices is much larger than the coherence length $\xi$ and much smaller than the penetration depth $\lambda$. This fact leads to a determination of $\lambda$ from the magnetization $M$ vs $B$. The Ginzburg-Landau equations dictate that the magnetic field distribution $h(r)$ of a vortex is given by

$$h(r) = \Phi_0/(\mu_0\lambda^2)\{\ln(\lambda/r) + 0.12\} \quad \xi << r << \lambda. \quad (6)$$
The interaction energy between vortices is proportional to $\Phi_0^2/(\mu_0\lambda^2) \ln(\lambda/L)$. From the relation $\Phi_0 = BL^2$, the energy density $F$ is equal to the interaction energy divided by $L^2$ and given by

$$F = \frac{\Phi_0^2}{(\mu_0 L^2 \lambda^2)} \ln(\lambda/L) = B\Phi_0/(\mu_0 \lambda^2) \ln(\lambda/L). \quad (7)$$

This is the part of the free-energy resulting from the material only: the vortex interaction. Thus the magnetization is given by

$$M = -\frac{\partial F}{\partial B} = \frac{\Phi_0}{(2\mu_0 \lambda^2)} \ln(\lambda B/\Phi_0) + \frac{\Phi_0}{(2\mu_0 \lambda^2)}, \quad (8)$$

which has a logarithmic field dependence. The slope of the logarithmic field dependence is

$$\frac{dM}{d(\ln B)} = \frac{\Phi_0}{(2\mu_0 \lambda^2)}. \quad (9)$$

When there is an anisotropy in the effective mass, or equivalently in $\lambda$'s, and if the screening current is in both orientations of $\lambda_{\text{min}}$ and $\lambda_{\text{max}}$, the slope is modified to

$$\frac{dM}{d(\ln B)} = \frac{\Phi_0}{(2\mu_0 \lambda_{\text{min}} \lambda_{\text{max}})}. \quad (10)$$

Thus absolute and anisotropic values of $\lambda$ can be obtained with this method. When $B$
lies along the $c$-axis, then $\lambda_{\text{min}}$ and $\lambda_{\text{max}}$ are $\lambda_b$ and $\lambda_a$, where "$b$" is parallel to CuO chains.

It is noted that the reversible magnetization should be measured at thermal equilibrium. This is not trivial in high-$T_c$ materials since the measured magnetization often is hysteretic due to vortex pinning. Deviations from the regular vortex array such as a trapped field in a grain boundary can cause an error in the estimate of $\lambda$ and even a deviation from logarithmic field dependence. When a crystal sample is used, the magnetic field is required to be parallel to the wide face of crystal so that demagnetization effects at the sample edges which are not considered in the above derivation are avoided. When the magnetic field is perpendicular to the $a$-$b$ plane, the distribution of the screening current and the local magnetic requires a more rigorous description. [49]

The initial idea of this method was conceived by V. Kogan et al. [45,46] They used this method to show $\lambda^2 \propto T_c - T$ near $T_c$ up to temperatures for which $(T_c - T)/T_c = 10^{-3}$, arguing that mean-field theory is valid in that domain. Mitra et al. [46] measured $\lambda$ in randomly oriented pellets of YBCO powder and free flowing powder oriented with $c \perp H$, with the results $\lambda_{ab} = 1700\text{Å}$ and $1290\text{Å}$ respectively. No measurement has been reported on YBCO single crystals with $T_c = 90\text{K}$. $\lambda_{ab}$ of YBCO crystals with $T_c = 60\text{K}$ was reported to be $2600\text{Å}$, where the crystal size was $1 \times 1 \times 0.07\text{mm}^3$ and magnetization $M(H)$ was obtained up to $H=54\text{kG}$. [47] The reported value of $\lambda_{ab}(0)$ for BSCCO crystals in this method is $2000\text{Å}$. [48]
C. Low-field dc magnetization method

When a small magnetic field is applied parallel to a thin superconducting slab, the expression for the average magnetization, or equivalently, the susceptibility \( \chi \), is given by

\[
\chi/\chi_0 = 1 - (2\lambda/d) \tanh(d/2\lambda),
\]

(11)

where \( d \) is the slab thickness and \( \chi_0 \) is the value of the magnetic susceptibility at total flux expulsion.

In the measurement of Krusin-Elbaum et al., the YBa\(_2\)Cu\(_3\)O\(_{7-\delta}\) crystals having dimensions about 0.15mm \( \times \) 0.15mm \( \times \) 40\( \mu \)m were used with the applied magnetic field in the \( a-b \) plane (wide area), [35] so that the demagnetization factor due to finite sample size in the \( a-b \) plane is negligible. The screening current is in the \( a-b \) plane except at the sample edges where the current is in the \( c \)-direction. The contribution of edge current is negligible if sample has a large enough aspect ratio. In this measurement, as the sample mounted in the SQUID magnetometer was warmed up slowly after a small field \(( \approx 1 \text{ Oe}) \) was turned on, the zero-field-cooling (ZFC) data were obtained. They used ZFC data only, since Meissner data has a larger change in \( \chi \) due to vortices moving in and out of the sample surface, thus not representing a bulk property. Since \( \lambda_{ab} \approx 1500\text{Å} \) is much smaller than \( d = 40\mu\text{m} \), the equation (11) is approximated by

\[
\chi/\chi_0 = 1 - 2\lambda_{ab}/d.
\]

(12)
The change of $\lambda_{ab}$ at low temperature can be determined by $\Delta \chi/\chi_0 = 2 \Delta \lambda_{ab}/d$. The flux expulsion was about 90% of total expulsion, which is $-1/4\pi\text{ emu.cm}^{-3}$. Some part of the crystal is suspected to remain non-superconducting and is likely responsible for the remaining 10%. The value of $\chi_0 - \chi$, which is proportional to $\lambda$, cannot be determined to sufficient accuracy due to the large ratio of $d/\lambda$, making the determination of the absolute value of $\lambda(0)$ difficult, causing this method to be a relative one. The failure to achieve 100% flux expulsion for the nominal sample volume imposes an additional uncertainty in $\chi_0$. The conventional BCS model was assumed, and fitting was made to both BCS weak coupling and two-fluid models with the resulting value of $\lambda_{ab}(0) = 880\text{Å}$ and $1700\text{Å}$, respectively. The resolution of their data is about $100\text{Å}$, and consequently the deviation of the fit from the BCS model was obscured by the error bars.

D. Surface impedance

When the skin depth (or penetration depth $\lambda$ below $T_c$) of a conductor is smaller than the sample dimensions, the surface impedance is defined theoretically for a semi-infinite sample by

$$Z_s = R_s + iX_s = E_t/H_t$$

(13)

with $R_s$ and $X_s$ being the surface resistance and reactance respectively. $E_t$ and $H_t$ are the tangential components of $E$ and $H$ just inside the surface of the conductor. Experimentally, $R_s$ is inferred from the additional loss in a resonant cavity due to the
presence of the sample, and hence to the change in inverse of the quality factor $\Delta(1/Q)$, or the half-width of the resonance, upon the insertion of the sample. $X_s$ is proportional to the resonance frequency shift $\Delta f$ which occurs when the sample is placed inside the cavity.

The relation between $Z_s$ and the complex conductivity of the sample is as follows. From the equation for curl $E$ we have

$$i\omega \mu_0 H_t = \partial E_t / \partial z = i k E_t,$$

where $k$ is the complex wavevector of the $E$-field. Then

$$Z = \omega \mu_0 / k.$$  \hspace{1cm} (15)$$

By combining Maxwell’s equations for curl $E$ and curl $B$, and neglecting the displacement current, the wavevector $k$ can be expressed in the form

$$k = (i \omega \mu_0 \sigma)^{1/2},$$  \hspace{1cm} (16)$$

where $\sigma$ is the complex conductivity $\sigma = \sigma_1 + i \sigma_2$. From eq. (15) and (16), the surface impedance has the form

$$Z = \{\mu_0 \omega / (i \sigma_1 - \sigma_2)\}^{1/2}.$$  \hspace{1cm} (17)$$
In a local limit superconductor, the conductivity can be expressed as

\[ \sigma = \sigma_1 + i/(\mu_0 \omega \lambda^2). \]  \hspace{1cm} (18)

At microwave frequencies, \( \sigma_1 << \sigma_2 \) in a superconductor except extremely close to \( T_c \) where \( \lambda \) diverges. Combining eqs. (17) and (18) leads to [50]

\[ R_s = \mu_0^2 \omega^2 \lambda^3(T)\sigma_1(T)/2; \]  \hspace{1cm} (19)
\[ X_s = \mu_0 \omega \lambda. \]  \hspace{1cm} (20)

Since the reactance \( X_s \) is proportional to the flux inside the cavity, its change is also proportional to a small change in cavity volume, which in turn appears as a frequency shift \( \Delta f \).

The favorable orientation for a single crystal slab is to have the shielding current flow in the \( a-b \) plane (the broad face of the crystal). When the magnetic field is oriented in the \( a-b \) plane, there is a contribution from \( \Delta \lambda_c \) due to the shielding current in the \( c \)-direction at the edge of the crystal. However it is negligible in the wide slab geometry. \( \Delta f \) in this orientation is given by

\[ \frac{\Delta f}{f} = \frac{1}{2} \frac{V_s}{V_r} \left[ 1 - \frac{2\lambda_{ab}\tanh\left( \frac{d}{2\lambda_{ab}} \right)}{d} \right], \]  \hspace{1cm} (21)

where \( V_s \) is the volume of a crystal, \( V_r \) is the effective volume of the cavity, and \( d \) is the
thickness of crystal. For \( d \gg \lambda_{ab} \), equation (21) becomes

\[
\Delta f/f = -(V_s/2V_p)(1 - 2\lambda_{ab}/d).
\]  

The determination of \( \lambda_{ab}(T) \) from the shift of resonance frequency \( \Delta f \) upon insertion of the sample is practically impossible, since it requires measurement of \( d \) to an accuracy much smaller than \( \lambda_{ab} \). Instead, one determines \( \lambda(T) - \lambda(0) = \Delta \lambda(T) \) from the change \( \Delta f/f \) with \( T \).

Recently, Hardy et al. reported a linear \( T \)-dependence in \( \Delta \lambda(T) \), \( \Delta \lambda/T = 4\text{Å}/K \) below 0.4 \( T_c \), in single crystal \( \text{YBa}_2\text{Cu}_3\text{O}_{7-\delta} \), with the interpretation that \( d \)-wave pairing was the source. [43] They designed a split-ring type resonator working at 900MHz. The low frequency minimizes the electric field so that dissipation due to the electric field is suppressed. The shift in resonance frequency is measured as the sapphire block where the sample is mounted slides in and out of the resonator. The magnetic field is oriented parallel to the \( a-b \) plane. The contribution of \( \Delta \lambda_c \) due to the shielding current in the \( c \)-direction is negligible for a crystal for which the ratio of width to thickness is about 100. A resolution of 1Å in \( \Delta \lambda \) is achieved when the sample is as broad as 1.5 \( \times \) 1.5 \( \text{mm}^2 \). The \( Q \) of the resonator is \( 10^6 \) and its stability is 1Hz per minute. This work is responsible for a great deal of the renewed interest in \( \lambda \), including the present study.
2. Methods for thin films

The potential of thin film device applications generates great interest in thin films, and it is often required to determine $\lambda$ in thin films. Since the methods applicable to thin films are different from those applicable to bulk samples, the independent measurements on thin films and single crystals are often considered as confirmations of each other. The advantage of thin films lies in the easy manipulation of these samples, for instance oxygen depletion and patterning in a preferred geometry. Mentioning the progress made in the measurement of $\lambda$, the most precise measurements, soon after the discovery of high-$T_c$ superconductors, were made on thin films by the two-coil method of Fiory et al. [33,34] There was a $T^2$-dependence in their data at low $T$, but it was ignored because data on clean crystals with high precision like that of Hardy et al. were not available at that time. Goldenfeld et al. recognized that the $T^2$-dependence in the data of Fiory et al. could arise from $d$-wave superconductivity with a little disorder. Although the highest resolution has been achieved by microwave measurements on a single crystal, where the linear-$T$ dependence in $\Delta\lambda(T)$ was revealed, the measurements on thin films continue to supplement or confirm the previous observations.

A. Two-coil method

The two-coil method has the advantage that it is a contactless measurement so that damage to the film can be avoided. Relatively fast data acquisition with a continuous temperature sweep is another advantage over other methods like the microwave cavity. In this method, a superconducting disk film is inserted in the narrow gap between a pair
of concentric coils, the drive and the receive coil. [33,51] An applied current in the drive coil induces screening currents in the film, which attenuate the mutual inductance at the receive coil. Ideally the diameter of the film should be much larger than the coil size, to simplify numerical modeling by avoiding currents at the film edges. The screening current and the corresponding mutual inductance are numerically calculated from modeling for a given \( \lambda \). In the measurement, the mutual inductance plus stray coupling as a function of \( T \) is recorded. Since this is a relative measurement, raw data is the change in mutual inductance, \( \Delta M \), and they are proportional to \( \Delta \lambda^2 \) for the film of large diameter. The details of this method is given in Appendix D.

Fiory et al. measured both real and imaginary components of the mutual inductance for two YBa\(_2\)Cu\(_3\)O\(_{7-\delta}\) films, whose thicknesses are 500Å and 2000Å. These films were made by codeposition from three sources onto SrTiO\(_3\), followed by postannealing. The diameter of the films was about 1cm; the diameter of the coils was 1mm and separated by 1.5mm gap. The drive frequency they used was 13kHz. The resolution of their data was about \( \pm 10 \)Å. They chose their fitting parameters of \( \lambda(0) \) and \( T_c \) for a least-fit to BCS weak-coupling, and deduced values of \( \lambda(0) = 1500 \)Å and 2100Å, respectively. Although there was an apparent \( T^2 \) dependence in their data at low temperatures, it was ignored. Soon after their result, Goldenfeld et al. reanalyzed the data of Fiory et al. and made the suggestion of a \( d \)-wave order parameter disordered by an impurity effect. Fiory et al. also intentionally increased the number of defects in some of their films by high temperature annealing, and showed the enhancement of \( T^2 \)-dependence at low \( T \). They analyzed their data as extra inductance caused by vortex-
antivortex pair at pinning defect sites and claimed that the $T^2$-dependence seen in clean films was also from this "extrinsic" effect. However, the issue is the $T^2$ dependence in as clean film as possible, rather than in a heavily disordered film.

Martinoli et al. used another type of two-coil method, where both drive and receive coils are on the same side of the film. [51] The receive coil is a pair of counter wound coils so that they compensate for each other when no film is present. In this way the baseline mutual inductance is almost identical to the normal state value and it allows a close probe of $\lambda$ very near $T_c$. The radii of the drive and the receive coils are 2mm and 1.2mm, respectively. They used a Fourier transform of the integral equation for the vector potentials in order to deduce an analytic solution rather than relying on numerical modeling. Their focus was Kosterlitz-Thouless transition at near $T_c$, thus irrelevant to the issue of $d$-wave superconductivity examined by the low $T$ behavior.

We used the two-coil method similar to Fiory et al. with an improved resolution by factor of 5. We found that data from Fiory et al. are consistent with ours within the experimental errors in their data when we compare their data with ours. We also found that our data is consistent with single crystal data by Hardy et al., whose linear $T$-dependence in $\Delta\lambda$ started the issue of $d$-wave superconductor. It is also penetration depth data that formed the opinion that high-$T_c$ superconductor is a BCS superconductor in early days. Now new results in penetration depth measurements in films and crystal questions the previous interpretation and stirs the controversy of the $s$-wave vs $d$-wave order parameter in high-$T_c$ superconductors. Any careful measurement of penetration depth is very important for the investigation of the controversy.
B. Kinetic inductance

This method requires a patterning of thin films into a narrow and long line by using photolithography or electron-beam lithography. The advantage of this method is that the current path is determined by the patterned geometry, which is not the case in two-coil method near $T_c$. Sensitivity, being proportional to the number of squares in the patterned line, could be increased unlimited. The weakness of this method is that intensive patterning is required and the film properties could be altered as a result. As ac current is applied to the superconducting line, a voltage drop is developed across the end points of the line, which is purely inductive at low frequencies except very near $T_c$. It contains two contributions: one is the kinetic inductance and the other the magnetic inductance. The kinetic inductance has a purely mechanical origin: the mass inertia of electrons. On the other hand, the magnetic inductance has its origin in Maxwell’s equations. The ratio of the size of the kinetic inductance to the magnetic inductance is approximately $\lambda^2/dw$, where $d$ and $w$ are the thickness and width of the strip, respectively. Thus the magnetic inductance dominates for usual sample sizes except when $\lambda$ diverges very near $T_c$. The kinetic inductance of very a narrow strip is given by $L_k = \mu_0\lambda^2l/dw$, where $l$ is the length of the strip. Rigorous calculations for widths larger than $\lambda$ require a self-consistent solution of an integral equation, which is given in Appendix B.

Elimination of the large background signal caused by the magnetic inductance is required in practical measurements, and is done by using a variable mutual inductor, a kind of a bridge circuit applied at the stage before measurement with a lock-in amplifier.
Since a large number of squares in the strip is needed to increase the resolution, a meander line geometry is used. It also helps reduce the magnetic inductance to 1/3 that of a stretched line of the same length. The voltage drop is measured by a 4 contact method. \( \Delta \lambda \) in 1000Å thick YBa\(_2\)Cu\(_3\)O\(_{7-\delta}\) films obtained by Lee et al. shows a \( T^2 \) dependence below 40K with \( \lambda(0) \approx 3100\AA \) and 2100Å after fitting to crystal data from Hardy et al. and the function \( 1-(T/T_c)^2 \), respectively. [42] It is the one of the first couple of confirmations of the \( T^2 \)-dependence in \( \Delta \lambda(T) \) with the highest resolution at that time. [41,42]

D. Transmission line method

Here, a transmission line is composed of a dielectric layer sandwiched between two superconducting layers. When there is an impedance mismatch between the transmission line and a coaxial cable attached to the ends of the transmission line, the transmitted wave forms regularly spaced and sharply defined resonance peaks as function of a driving frequency. The phase velocity is measured from the spacing of the peaks and the length of transmission line. The phase velocity \( v_p \) is determined by \( (LC)^{-1/2} \), where the inductance per unit length \( L \) consists of magnetic and kinetic inductance, and \( C \) is the capacitance per unit length of the transmission line; \( C = \epsilon_{\text{eff}} w/d \), where \( \epsilon_{\text{eff}} \) is the permittivity of the dielectric, \( d \) is the dielectric thickness, and \( w \) is the width of the transmission line. \( L \) is given by:

\[
L = \mu_0 \{ d + \lambda_1 \coth(t_1/\lambda_1) + \lambda_2 \coth(t_2/\lambda_2) \}/w, \tag{23}
\]
where \( t \) is the thickness of the superconductor. [52] The first term represents the magnetic inductance and the last two terms are contributions from the kinetic inductance of each of the superconductors. When both superconductors are identical, \( \nu_p \) is expressed by

\[
\nu_p = c [\varepsilon_{\text{eff}} \{ 1 + 2(\lambda/d) \coth(t/\lambda) \}]^{1/2}. \tag{24}
\]

Provided \( \varepsilon_{\text{eff}} \) and \( d \) are known, this method can determine the absolute value of \( \lambda(0) \), since \( d \) can be made as small as \( \lambda \). However, this has yet to be achieved, and \( \lambda(0) \) is used as a fitting parameter.

Pond et al. prepared the trilayer YBCO-LaAlO\(_3\)-YBCO transmission line with thickness of 2500Å, 2000Å, and 2500Å respectively on a MgO substrate. [39] The frequency was scanned up to 0.6GHz. According to their report, the data points, measured above 20K, match to BCS weak-coupling with \( \lambda(0)\) = 1350Å and are inconsistent with the two-fluid model. It is also remarked that measurements below 20K are required to test the \( T^2 \) dependence, though \( \lambda^2(0)/\lambda^2(T) = 1 - (T/T_c)^2 \) fits the data with \( \lambda(0) = 1300\)Å.

Anlage et al. adopted the conventional model in the interpretation of measurements on a transmission line composed of two superconducting films separated by a 20\( \mu \)m thick dielectric spacer and clamped together. [41,53] However, the overall data cannot be described by a single BCS gap temperature dependence. Data points are listed above 12K. Low \( T \) data fit to \( 2\Delta(0)/kT_c \approx 2.5 \), while fitting of high \( T \) data
required the strong coupling limit.

One of their interpretations assumes the inductance of weak links at grain boundaries in series with that of the grains. The net penetration depth $\lambda_n$ can be shown to be

$$\lambda_n^2 = \lambda_{SC}^2(T) + \frac{\Phi_0}{\{2\pi\mu_0 a J_c(T)\}}$$

since the network of grains and weak links can be modeled as a connection in series. $\lambda_{SC}$ is $\lambda$ in bulk, and $a$ is the average spacing between weak links. $J_c(T)$ is the critical current of a weak link at $T$. The second term also approximately obeys the form of $\lambda^2$ in the BCS weak-coupling limit. Estimation of $a$ is about 1000Å according to TEM. [54] After assuming $J_c(0) = 2 \times 10^7 A/cm^2$, they obtained $2\Delta(0)/kT_c = 4.5$ and $\lambda_n(0) = 1400Å$. [55-57]

Ma et al. used a parallel plate resonator composed of a 13µm thick Teflon as a dielectric sandwiched between the two YBCO thin films grown on LaAlO$_3$ or two pieces of freshly cleaved BSCCO single crystal. [58] The operating frequency was about 10GHz, and the resonance frequency of the resonator was given by

$$f(T) = \frac{2c}{D/\varepsilon_{eff}} \frac{1}{\sqrt{1 + \frac{2\lambda}{d} \coth\left( \frac{t}{2\lambda} \right)}}$$

where $D$ is the effective length of the sample for the mode in question. Due to uncertainty in $D$ and $\varepsilon_{eff}$, they could only deduce $\Delta\lambda(T)$. The deduced $\Delta\lambda(T)$ for several
YBa$_2$Cu$_3$O$_{7-\delta}$ films and BSCCO crystals is proportional to $T^2$ below 23K with a typical resolution of about $\pm 2 \text{Å}$. We attempted to fit their data with the target curve of $\lambda^2(0)/\lambda^2(T) = 1 - T^2/T_c^2$. Usually data on the entire range of $T$ are required to fit any data. Since their data covers only below 23K, our attempt has some error. The resulting value of $\lambda(0)$ for Ma et al. is about 1600Å for the best sample and 4700Å for the poorest. It is equivalent to $\lambda(0) = 2000\text{Å}$ to 6000Å if they were fit to the crystal data of Hardy et al.; $\lambda(0)$ in our pure film is 1700Å. The $\lambda(0) = 6000\text{Å}$ seems to be too large for a pure film, making their result uncertain.

When the measurement is made with a relative method, it is important to record data for the entire range of temperatures from low $T$ to $T_c$ because $\lambda(0)$ and the shape of $1/\lambda^2(T)$ rely on comparison of data and target curve for the entire range of temperatures in the fitting procedure. Even more, there could be an anomalous kink around 40K when the LaAlO$_3$ is used as the substrates. It seems to be caused by the long grain boundaries in the film planted by substrate. LaAlO$_3$ is preferred in microwave application due to the much smaller dielectric loss than SrTiO$_3$. In our case, SrTiO$_3$ substrates and the low drive frequency (20kHz) are used. Our data covers the entire range of temperatures below $T_c$.

Summary of this chapter is the following. $\mu$SR yields $\lambda_{ab}(0) = 1400 \pm 100\text{Å}$ and $\lambda(T)/\lambda(0)$ suggesting strong-coupling BCS, but $\pm 100\text{Å}$ resolution limits its ability to distinguish among a variety of other possibilities. Hardy et al. observed $\lambda(T) - \lambda(0) \approx (4\text{Å}/K)T$ below about 40K, which would be difficult to resolve in $\mu$SR. There are no published confirmation of this. Complete sets of data, from 4K to $T_c$, on pure YBCO
films are scarce. All data show $T^2$ at low $T$, in apparent conflict with Hardy et al.

The goal of the present study is to compare high precision measurements of $\lambda(T)$ in YBCO films with the data of Hardy et al., and analyze the similarities and differences in light of recent theories of disordered $d$-wave superconductors.
CHAPTER III
EXPERIMENTAL PROCEDURES

1. Measurement of penetration depth

We used the two-coil method for measuring $\lambda(T)$. In the two-coil method, a superconducting disk film is inserted in the narrow gap between a pair of concentric coils, the drive and the receive coil. [33,34,51] As an $ac$ current $I_d$ is applied in the drive coil, the resulting magnetic field induces a screening current in the superconducting film in the direction to cancel the $z$-component of the magnetic field generated by the drive coil, resulting an attenuation of the mutual inductance of the drive and the receive coil. Equivalently, the cancellation of the magnetic dipole moment of the drive coil by its mirror image due to the superconducting film leads to a picture of the quadrupole moment as the primary magnetic moment formed by the drive coil and the diamagnetic film. The net magnetic flux in the receive coil gives rise to an $ac$ voltage, which is monitored by a two-channel lock-in amplifier. The inductive component of the pick-up coil impedance is $90^\circ$ ahead of $I_d$, and the dissipative component is in-phase with $I_d$. The dissipative component in the Meissner state is negligibly small at 100kHz.

The schematics of our implementation of the two-coil method is shown in Fig. 1. The induced $ac$ voltage in the receive coil is measured by a PAR 5208 two-channel
Figure 1. Schematic circuit diagram of our two-coil apparatus. The radius of the film is at least twice that of the drive coil. The reference of lock-in is in-phase with the current in the drive coil.
lock-in amplifier. Both the inductive and the dissipative components are monitored. \( I_d \) is monitored from the voltage drop across the standard resistor \( R_{std} \) and its phase is used for the reference of the lock-in amplifiers via the reference generator. The primary coil is solenoidal with 84 turns; its inner and outer diameters are 3\( \text{mm} \) and 7\( \text{mm} \), and its length is 3\( \text{mm} \). The secondary coil has 300 turns with the same dimensions. There is a 2\( \text{mm} \) gap between the coils. The coils are mounted in a frame made of plexiglas. The \( ac \) magnetic field at the film corresponding to 0.04\( MA/cm^2 \) is less than 1\( mG \) perpendicular to film, even at the film edges, and less than 0.2\( G \) parallel to the film, so one would not expect the field to induce vortices.

Numerical modeling described below, shows that the induced current is largest at the drive coil radius (Fig. 2). When the film size is finite, the magnetic field at the film edges will be enhanced due to a large demagnetization factor in a flat superconducting geometry when the field is applied perpendicular to the plane. In our numerical modeling, the finite size effect is accounted for. The corresponding screening current at the film edge is enhanced as shown in Fig. 2. Its effect on the mutual inductance is less than 2% in our apparatus.

Our three laser ablated films, \( A, B, \) and \( C \) were grown on \( \text{SrTiO}_3 \) (100) substrates epitaxially with the \( a-b \) plane lying parallel to the substrate. These nominally identical films were made in the same facility (Los Alamos National Lab). The films were circular with diameters of 2.2\( cm \) for films \( A \) and \( C \), and 2.0\( cm \) for film \( B \); and thicknesses of 330 ± 30\( \text{Å} \). Their transitions were at 88K, and were 1K for film \( A \) and \( C \), and 0.5K wide for film \( B \). After measurements on fully oxygenated films, we
Figure 2. Radial dependence of the induced current density, $J_s(r)$, in the film and the vector potential associated with the drive coil, $A_{ext}(r)$, at the plane of film with $I_d=0.8mA$. Radius of film is 1.1cm, and $\lambda^{2}/d = 1.2\mu m$. The finite size effect appears as the enhancement of shielding current at film edges, which is accounted for in the modeling. The shielding current peaks just below the drive coil.
annealed the films in 1 atm of Ar at 250°C systematically to lower the oxygen content and re-measured λ.

The critical current density $J_c$ was determined by increasing $I_d$ until dissipation was detected. We found $J_c = 10^6 \text{A/cm}^2$ and $10^7 \text{A/cm}^2$ for films A and B, respectively. The pair breaking $J_c$ in high-$T_c$ superconductor is estimated to be about $10^9 \text{A/cm}^2$ due to the short coherence length $\xi_0 \approx 10\text{Å}$. The observed $J_c$’s are much smaller than that, and seem to be limited by effects such as grain boundaries or vortex-antivortex pair formation. The almost identical $1/\lambda^2(T)$ in both film A and B, in spite of the large difference in $J_c$’s, implies that the extraneous factors are negligible in the low current limit.

We determined $\lambda(T)$ from the mutual inductance $M(T)$ of concentric coils of the copper wire on opposite sides of the film, following Fiory et al. [33,34] The measuring current was at 20kHz; identical data were obtained at other frequencies. As shown in Fig. 2, an ac current in the drive coil of 0.8mA induces a current density in the film of 0.04 MA/cm²; identical data were obtained with currents that were ten times smaller, which indicated the absence of nonlinear response to the current amplitude. The current-voltage linearity was also confirmed until $J_c$ was reached at 4.2K.

The measured $M(T)$ for film A is shown in Fig. 3. Within a few Kelvin of $T_c$, in the vicinity of the Kosterlitz-Thouless transition, the conductivity has a resistive component. For the temperatures of interest here, namely below 0.98$T_c$, the film conductivity is purely inductive. Modeling extracts both components of the conductivity. The numerical model describes the coils as numerous loops of current coaxial with a
Figure 3. The mutual inductance $M$ vs $T$ for film A. (a): Inductive (upper) and dissipative (lower) components of $M$. (b): The inductive component of $M$ is magnified.
circular superconducting film. The number of loops, their radii, and the distance from the superconducting film are appropriately modeled. With the geometry fixed, $M$ depends on $T$ only through the $T$-dependence of the currents induced in the superconducting film, i.e., $M$ is a function only of $\lambda(T)$. $M$ versus $\lambda$ is determined by calculating the supercurrent density $J_s$ induced in the film and then the flux linking each loop of the secondary coil. While we use the full numerical model to obtain $\lambda$, we note that there is a simple and accurate approximation over a wide $T$ range in which the film conductance is purely inductive. When the film diameter is large enough relative to that of the coils, $M$ is a function of only $\lambda^2/d_{\text{eff}} : M(T) = \alpha \lambda^2(T)/d_{\text{eff}} + \beta$. [59] $d_{\text{eff}}$ is the effective thickness and given by $d_{\text{eff}} = \lambda \tanh(d/\lambda)$ to a good approximation, where $d$ is the actual film thickness. Effectively the current is regarded to be uniform through the thickness $d_{\text{eff}}$ and zero elsewhere. It indicates that screening current exists only in the depth of $\lambda$ in thick film superconductor. $\alpha$ is calculated to $\pm 5\%$ from the model, and $\beta$ is a constant which includes the unknown stray coupling from other parts of the circuit.

$\alpha$ is affected by the geometry, including the coil dimension, and can be estimated by numerical modeling as accurately as the measurement of the geometry. When $T \leq 0.9T_c$, the mutual inductance $M$ drops by a factor of several thousand and becomes comparable to the stray coupling. Even 1% error in the empty-coil inductance propagates as several hundred percent error in the baseline of $M$, and also the uncertainty due to stray coupling adds to it. Thus the baseline of $M$ is practically unknown, and this method is only accurate in the change in $M$. The baseline of $M$, or equally $\lambda(0)$, is the only unknown parameter. For the films discussed here, $d << \lambda$, so $J_s$ varies only
slightly through the film thickness, and $M = \alpha \lambda^2(T)/d + \beta$, so that in essence, we determine $\lambda^2(T) - \lambda^2(0)$ from $M(T) - M(0)$ in analogy to the microwave cavity measurements which determine $\lambda(T') - \lambda(0)$ from the resonant frequency $f(T)-f(0)$. [43]

There are two main sources of uncertainty. First, the ±10% uncertainty in the film thickness $d$ and the ±5% uncertainty in $\alpha$ translate into an ±11% uncertainty in $\lambda^2$. This uncertainty affects the magnitude of $\lambda$ but not its dependence on $T$. Secondly, the choice of $\lambda(0)$ significantly influences the dependence of $\lambda$ on $T$ at $T/T_c < 0.6$ where $\lambda(T)$ is not much larger than $\lambda(0)$, but has little effect above about $0.9T_c$. Thus, the slope of $\lambda^2(T)$ near $T_c$ is well defined.

2. Fitting procedure and analysis

Data analysis begins with a fitting procedure for determining $\lambda(0)$. The procedure used by other groups has been to choose $\lambda(0)$ and $T_c$ for the film to obtain a "best-fit" of $\lambda^2(0)/\lambda^2(T/T_c)$ to an analytic function like $1-T^2/T_c^2$, or $1-T^4/T_c^4$, or the weak-coupling BCS result. Then they discuss the important features of the data. Ideally, several theories would predict $\lambda(T/T_c)$, and one could see which theory produced the best "best-fit" to the data. Theories are not yet capable of such predictions.

We take a more experimental approach which focuses on the validity of the recent measurements of $\lambda(T)$ in YBCO crystals by Hardy et al. These results, which have not yet been reproduced in other labs, show that $\lambda(T)-\lambda(0)$ is linear in $T$ at low temperatures, suggesting a $d$-wave superconducting state. There has been a great deal of skepticism because most other measurements, including ours, show a quadratic $T$-
dependence at low \( T \). Our fitting procedure is to choose \( \lambda(0) \) and \( T_c \) for our film to obtain a "best-fit" to the data of Hardy et al. The procedure is justified a posteriori by the excellent agreement we obtain. Quantitative and qualitative differences will be shown to be consistent with \( d \)-wave superconductivity with a little disorder in the film.

We have also used the same template curve for the oxygen depleted thin films. If oxygen depletion basically affects the material by reduction of the charge carrier concentration rather than an increase of the disorder in film, then theory anticipates that the shape of \( 1/\lambda^2(T/T_c) \) might not change due to oxygen depletion.

To find a best-fit of film data to crystal, we must minimize some function. We choose to minimize the difference in values of \( \lambda^2(T/T_c) \) because the random noise in \( \lambda^2 \) is uniform for the entire range of temperatures in our measurements. The size of the error bar at each data point should be considered in weighting the data points to determine the best-fit. \( \Delta M(T) = M(T) - M(0) \) is what we measure, and as mentioned in the previous section, \( \Delta M(T) \) is proportional to \( \Delta \lambda^2 = \lambda^2(T) - \lambda^2(0) \). The random background noise is simply added to \( \Delta M(T) \). This procedure emphasizes the best agreement near \( T_c \) where \( \lambda^2 \) is large and most accurately measured. We used the downhill simplex method for the minimization algorithm. [60]

The first step in the procedure is to choose a value of \( \lambda_x(0) \) for the data of Hardy et al., who actually measured \( \lambda_x(T) - \lambda_x(0) \). ("x" denotes crystal) Any value of \( \lambda(0) \) from 1300Å to 1700Å would work; important numbers from these fits are in Table 1.

There are 3 fitting parameters: \( \lambda_{film}(0) \) for the film, and \( T_c \) and a scale factor for the crystal data. The significance of these parameters is best understood from
Table 1. Best-fit parameters of film A for different choices of $\lambda_x(0)$ of crystal for $0.25 \leq T/T_c \leq 0.93$.

<table>
<thead>
<tr>
<th>$\lambda_x(0)$</th>
<th>r.m.s. value of $\delta\lambda^2(T/T_c)$ (x10^4Å^2)</th>
<th>$\lambda_A(0)$ of film</th>
<th>$T_c$</th>
<th>The intercept of template curve</th>
</tr>
</thead>
<tbody>
<tr>
<td>1300Å</td>
<td>2.56</td>
<td>1633 ± 50Å</td>
<td>87.62K</td>
<td>1563Å</td>
</tr>
<tr>
<td>1400Å</td>
<td>2.40</td>
<td>1698 ± 50Å</td>
<td>87.48K</td>
<td>1630Å</td>
</tr>
<tr>
<td>1500Å</td>
<td>2.35</td>
<td>1770 ± 55Å</td>
<td>87.42K</td>
<td>1701Å</td>
</tr>
<tr>
<td>1700Å</td>
<td>2.14</td>
<td>1884 ± 55Å</td>
<td>87.15K</td>
<td>1874Å</td>
</tr>
</tbody>
</table>
examination of the best-fit from $0.25T_c$ to $0.93T_c$ for film $A$, with $\lambda_A(0)=1700\text{Å}$ and $\lambda_x(0)=1400\text{Å}$, shown in Fig. 4. There is excellent agreement between $\lambda_A^2(0)/\lambda_A^2(T/T_c)$ and $1.08\lambda_x^2/\lambda_x^2(T/T_c)$. The upper panel shows the absolute differences between these curves. The larger $\lambda(0)$ in the film and the quadratic behavior at low-$T$ are quantitatively explained in the context of $d$-wave superconductivity, discussed below.

Table 1 lists the best fit parameters for film $A$ for several choices of $\lambda_x(0)$ between $1300\text{Å}$ and $1700\text{Å}$. The corresponding "best fit" values of $\lambda_A(0)$ are $1630 \pm 50\text{Å}$ and $1880 \pm 55\text{Å}$. Thus, values of $\lambda_{\text{film}}(0)$ are $20\%$ to $40\%$ larger than for clean crystals. The size of the error in the best-fit value of $\lambda_{\text{film}}(0)$ is deduced by finding the range of the initial fit parameter value that leads to the same best r.m.s. value of $\delta \lambda^2(T/T_c)$.

Fig. 4 shows the fitting when $\lambda_x(0) = 1400\text{Å}$ is assumed for the crystal, which is consistent with $\mu$SR data. Excellent agreement is shown. The difference of about 0.002 shown in the fig. 4 (a) is the size of the error in digitizing the data of Hardy et al. Note that this procedure weights the high-$T$ data much more heavily than low-$T$ data, so the excellent agreement all the way to $T/T_c$ of 0.25 is unlikely to be coincidental. In the next chapter, this will be discussed more in detail. The quality of the fit deteriorates rapidly if we try to extend to below $0.25T_c$ or above $0.93T_c$.

To emphasize the significance of the excellent fit of films to crystals, best fits of the same data of film $A$ to $1-(T/T_c)^2$, BCS, and two-fluid are illustrated from Fig. 5 to Fig. 7 for $T/T_c \leq 0.93$. In the figures, the dashed curve is the template curve and the noisy curve is the best-fit of film $A$ to it. The scale of the vertical axis is normalized by
Figure 4. Best-fit of $\lambda_A^2(0)/\lambda_x^2(T/T_c)$ for film A to $1.08\lambda_x^2(0)/\lambda_x^2(T/T_c)$ for the crystal of Hardy et al. Difference between the curves in (b). The r.m.s. difference from 0.25 to 0.95$T/T_c$ is 0.002, only slightly larger than the experimental noise.
the best-fit value of $\lambda_A(0)$ for convenience. The horizontal bars in the upper panel are at $\pm 0.01$. Best fit parameters for the corresponding choice of the template curve are given in Table 2. Their agreements are poor when compared to that of the best-fit to the single crystal data; the differences are well outside of the experimental noise. In fig. 5, the best-fit to $1-T^2/T_c^2$ occurs at $\lambda(0) = 1310\text{Å}$ and $T_c = 88.3\text{K}$. The difference between these curves, shown in the upper panel, is about 5% of $\lambda_A^2(0)/\lambda_A^2(T/T_c)$ which is much larger than that of the fit between films and crystals. Previously, a phenomenological $1-(T/T_c)^2$ curve frequently was used to match the $T^2$-behavior at low temperatures that was observed by several other groups. The improved resolution in our measurement (about $\pm 1\text{Å}$) and the temperature range of data from 4.2K to $T_c$ reveals that the deviation is much larger than the noise level in the data, showing that $1-T^2/T_c^2$ curve is not adequate for fitting. Two-fluid model and BCS fit show similar deviations up to 6% and 10% of $\lambda_A^2(0)/\lambda_A^2(T/T_c)$, respectively. But the r.m.s. value of $\delta\lambda^2(T/T_c)$ of two-fluid fit is the largest due to the largest value of $\lambda_{\text{film}}(0)$, meaning that two-fluid model is the most inadequate.

The excellent agreement with the crystal data is a posteriori justification of our assumption that the dependence of $\lambda(T)/\lambda(0)$ on $T/T_c$ in films and crystals are the same. The adjustable fit parameters $\lambda(0)$ and $T_c$ do not allow one to fit other, similar $T$-dependencies to anywhere near the experimental precision. The agreement in spite of the difference between the type of sample and experimental method strongly supports that both our measurement and that of Hardy et al. reveal an intrinsic property of these materials.
Table 2. Best-fit parameters of film A for different choices of template curve for $T/T_c \leq 0.93$. Best-fit is determined by minimizing the difference in $\lambda^2$, denoted as $\delta \lambda^2$. Two-fluid model is the worst choice for the template curve.

<table>
<thead>
<tr>
<th>models</th>
<th>r.m.s. value of $\delta \lambda^2(T/T_c)$ ($\times 10^4\text{Å}^2$)</th>
<th>$\lambda_A(0)$ of film</th>
<th>$T_c$</th>
<th>The intercept of template curve</th>
</tr>
</thead>
<tbody>
<tr>
<td>crystal $\lambda_x(0)=1400\text{Å}$</td>
<td>2.4</td>
<td>$1698 \pm 50\text{Å}$</td>
<td>87.48K</td>
<td>1630Å</td>
</tr>
<tr>
<td>$1-T^2/T_c^2$</td>
<td>6.4</td>
<td>$1308 \pm 40\text{Å}$</td>
<td>88.27K</td>
<td>1333Å</td>
</tr>
<tr>
<td>two-fluid</td>
<td>20.4</td>
<td>$2058 \pm 60\text{Å}$</td>
<td>89.24K</td>
<td>2121Å</td>
</tr>
<tr>
<td>BCS</td>
<td>14.7</td>
<td>$1298 \pm 40\text{Å}$</td>
<td>88.60K</td>
<td>1374Å</td>
</tr>
</tbody>
</table>
Figure 5. The best-fit of $\lambda_A^2(0)/\lambda_A^2(T/T_c)$ of film A (solid curve) to $1-(T/T_c)^2$ (dashed curve). Difference between the curves in (b) is shown in (a).
Figure 6. The best-fit of $\lambda_A^2(0)/\lambda_A^2(T/T_c)$ of film $A$ (solid curve) to two fluid model, $1-(T/T_c)^4$ (dashed curve). Difference between the curves in (b) is shown in (a).
Figure 7. The best-fit of $\lambda_A^2(0)/\lambda_A^2(T/T_c)$ of film A (solid curve) to BCS weak-coupling (dashed curve). Difference between the curves in (b) is shown in (a).
CHAPTER IV

EXPERIMENTAL RESULTS AND DISCUSSION

In this chapter, we report on the comparison of our results of precision measurement on films with clean crystal data by Hardy *et al.* [43] and also the comparison of our film and the $\mu$SR result on bulk by Uemura *et al.* [37,38] in an oxygen depletion study. There are a couple of calculations of $d$-wave theory by Hirschfeld *et al.* and Kim *et al.*, [25,61] which are valid at low $T$. We compare our low $T$ data with these calculations. The data at higher temperatures are analyzed in terms of our phenomenological model of $d$-wave superconductors. Oxygen depletion study enables the comparison of film with $\mu$SR result of bulk sample. It verifies that our data are associated with the YBCO and not grain boundaries or other defects.

In section 1, we discuss the comparison between the data on pure YBCO. In section 2, the different low-$T$ behavior of films and crystals are interpreted within a $d$-wave model with disorder due to strong impurities. In section 3, the possibility of additional inductances caused by grain boundaries and the vortex-antivortex pair formation is discussed. The purpose of this section is to draw conclusion negating the extrinsic effects in our measurements. In section 4, result of oxygen depletion is discussed. Finally in section 5, our result of $\lambda^2(0)/\lambda^2(T)$ is compared with isotropic $s$-
wave to show that they are incompatible.

1. Comparison between film and crystal

There is excellent agreement between our film data and Hardy's crystal data for $0.25 \leq T/T_c \leq 0.93$. [43] Figure 4 illustrates the agreement between our films and Hardy's crystals. In the Fig. 4 (b), the dashed curve is a numerical interpolation of $1.08\lambda_x^2(0)/\lambda_x^2(T/T_c)$ for the crystal, with $\lambda_x(0) = 1400\text{Å}$. [That is, $\lambda_x(T) = \delta \lambda_x(T) + 1400\text{Å}.]$. The noisy curve is data for film $A$, with $\lambda_A(0) = 1700 \pm 50\text{Å}$ and $T_c = 87.5\text{K}$. Between $0.22T_c$ and $0.95T_c$, the curves are within $0.5\%$ of each other, as shown in Fig. 4 (a), where the horizontal lines denote $\pm 0.01$.

We found that data from early reports by Fiory et al. also fits to the crystal data curve very well, i.e., to within the experimental noise of about $\pm 0.01$. [33,34] The resulting change in $\lambda(0)$'s are from the reported value $1500\text{Å}$, $1660\text{Å}$, and $2100\text{Å}$ from their best-fit to BCS to $1770\text{Å}$, $1910\text{Å}$, and $2700\text{Å}$ for a best fit to Hardy's data. While the large value $\lambda(0) = 2700\text{Å}$ calls the results for the $2000\text{Å}$ thick film into question, there is an obvious consistency among the independent sets of measurements on thinner films. Our data have noise 5 times smaller, so the agreement with Hardy is even more impressive.

Having shown the excellent agreement above $0.25T_c$, we now discuss the differences between films and crystal data below $0.25T_c$. There are two differences. First the $T^2$-dependence in the film data is clearly in contrast to the linear behavior of crystal data. The observation that $\Delta \lambda(T) \propto T^2$ for our films is insensitive to the choice
of $\lambda(0)$. The measured mutual inductance $M$ depends on $T$ only because it is proportional to $\lambda^2$, so that at $T=0$ a small change in $M$ is proportional to a small change in $\lambda$: $dM/dT = [dM/d\lambda]_{\lambda(0)}d\lambda/dT$. Second, $\lambda_{\text{film}}(0)$ is significantly larger than $\lambda_{\chi}(0)$.

The comparison of film and crystal data constitutes the gradual crossover behavior: the smooth connection of the similarity at high $T$ and the difference at low $T$. In the next section, we explain this phenomena with the analysis of the $d$-wave models, which are consistent with both low $T$ and high $T$ data.

2. Crossover behavior

It is important to investigate the underlying physics for the crossover: the excellent agreement in the functional form of $\lambda(T/T_c)/\lambda(0)$ above $0.25T_c$ and the difference below $0.25T_c$. Physically, a change in the sample from pure material due to disorder in the CuO$_2$ planes will affect three measurable quantities, namely: the $T$-dependence of $\lambda(T/T_c)/\lambda(0)$ at low $T$, the value $\lambda(0)$, and $T_c$. The correct theory will describe all of these changes. In this section we compare the low $T$ behavior with a theory by Hirschfeld $et$ $al$. Then we make a simple calculation to fit the data in the high $T$ region by adopting a disorder-broadened $d$-wave density of states. Though this model is very simple, the fact the high $T$ behavior is insensitive to the change of low $T$ behavior is the basis for the justification.

Hirschfeld $et$ $al$. recently considered a simple model of a disordered, two-dimensional $d$-wave superconductor with resonant scattering from impurities. [25] Complications like dispersion along the $c$-axis and a possible van Hove singularity in the
density of states were set aside; disorder affects the superconducting state through its
competition with the anisotropic $d_{x^2-y^2}$ order parameter, and not through its possible
influence on normal-state parameters like the electron density of states and
antiferromagnetic spin fluctuations. In their theory, the superfluid density $\propto \lambda(0)^2$ is
a monotonically decreasing function of the scattering rate, and also the curvature of $\lambda(T)$
at low $T$ scales proportionally with it. The theory calculates the suppression in $T_c$ with
disorder, too, but since the measured $T_c$ likely is influenced by many effects not included
in the theory, the best comparison is made by using only low-$T$ quantities: the reduction
of superfluid in reduced form, $\lambda(0)^2/\lambda_0^2 - 1$, and the increase in curvature in $\lambda^2(T)/\lambda_0^2$,
where $\lambda_0$ is $\lambda(0)$ for the pure material. These quantities are independent of such details
as band structure, strong coupling, Fermi liquid effects, etc., since disorder induced
changes in these quantities cancel out in the reduced form $\lambda(0)/\lambda_0$. Thus the comparison
with experiment is quite meaningful at low $T$.

The theory describes the disorder effect due to the low concentration of strong
scattering in $d$-wave. It is valid for $T/T_c << 1$ and for disorder parameter (=scattering
rate) $\gamma << \Delta_0$, where $\Delta_0$ is the amplitude of order parameter. It predicts that disorder
increases $\lambda(0)$ and decreases the curvature in $\lambda^2(T)/\lambda^2(0)$:

$$\frac{\lambda_0^2}{\lambda^2(T)} = 1 - \frac{2\gamma}{\pi \Delta_0} \ln \left( \frac{4\Delta_0}{\gamma} \right) - \frac{\pi}{3\Delta_0 \gamma} T^2$$

or,

$$= 1 - c_0 - \frac{\pi}{3\Delta_0 \gamma} T^2$$

(28)
\[
\frac{\lambda^2(0)}{\lambda^2(T/T_c)} = 1 - \frac{\pi T_c^2}{3\Delta_0 \gamma(1-c_0)} \frac{T^2}{T_c^2}
\]

\[
= 1 - c_2 \frac{T^2}{T_c^2},
\]

(29)

where \( \lambda(T) \) is the penetration depth in the weakly disordered material. From Eq. (28), \( c_0 \) is the fractional change in \( \lambda^{-2}(0) \) from disorder for fixed \( \lambda_0 \); \( c_2 \) is the curvature of \( \lambda^2(T/T_c)/\lambda^2(0) \). Assuming that \( \Delta_0/kT_c \) is a weak function of disorder, then theory finds that \( c_0 = (2\pi/3)\ln(12/\pi x) \) and \( c_2 = T_c^2/[\Delta_0^2 x(1 - c_0)] \) are known functions of the disorder parameter, \( x = 3\gamma/\pi \Delta_0 \).

Figure 8 shows the calculated \( 1/c_2 \) vs \( c_0 \) for \( \Delta_0/kT_c = 3 \) and 3.5 in the solid and dashed curve, respectively. \( \Delta_0/kT_c = 3 \) is suggested by our pheneomenological analysis, below. The three points in Fig. 11 are deduced from the data on the fully oxygenated films \( A0, B0, \) and \( C0 \). From data, \( c_0 \) is given by \( c_0 = 1 - \lambda_x^2(0)/\lambda_{film}^2(0) \), where \( \lambda_x(0) = 1400Å \) is used, \textit{i.e.}, the same total charge carrier density as that of Hardy's crystal is used; \( c_2 \) is given by \( \lambda_{film}^2(0)/\lambda_{film}^2(T) = 1 - c_2 T^2/T_c^2 \). The disorder parameter \( x = 3\gamma/\pi \Delta_0 \) is marked in the upper horizontal axis. Table 3 lists the corresponding parameters: \( T_c, \lambda(0), c_0, \) and \( 1/c_2 \). Hardy's single crystal is located at the origin: the curvature is infinite \( (1/c_2 = 0) \) and superfluid density is equal to the total charge carrier density. The infinite curvature means the low-\( T \) parabolic behavior is as sharp as the corner of a triangle, which makes the linear \( T \)-dependence of \( \Delta \lambda \). As disorder increases for the fixed total charge carrier density, the \( 1/c_2 \) vs \( c_0 \) progresses along the curve of Hirschfeld's calculation. Their calculation is not accurate for large impurity density,
Figure 8. $1/c_2$ vs $c_0$ for $\Delta_0/kT_c = 3$ and 3.5, from the theory of Hirschfeld and Goldenfeld, which is valid only for small impurity concentration. Points represent data from the fully oxygenated samples A0, B0, and C0 with $\lambda_0 = 1400\text{Å}$. Hardy's crystal corresponds to $1/c_2 = c_0 = 0$. $c_2$ is the curvature of $\lambda^2(0)/\lambda^2(T/T_c)$ as $T/T_c \rightarrow 0$; $c_0$ is the fractional reduction in $\lambda^2(0)$ in film relative to crystals. The size of disorder parameter $3\gamma/\pi\Delta_0$ is marked in the upper horizontal axis.
Table 3. $T_c$, $\lambda(0)$, $c_0$, and $1/c_2$ for YBCO films $A$, $B$, and $C$ at various oxygen contents.

<table>
<thead>
<tr>
<th>sample</th>
<th>$T_c$ (K)</th>
<th>$\lambda(0)$</th>
<th>$c_0$</th>
<th>$1/c_2$ (±5%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$B0$</td>
<td>88.3</td>
<td>1707 ± 50Å</td>
<td>0.33</td>
<td>0.95</td>
</tr>
<tr>
<td>$B1$</td>
<td>86.7</td>
<td>1840 ± 55Å</td>
<td>0.42</td>
<td>1.02</td>
</tr>
<tr>
<td>$B2$</td>
<td>86.7</td>
<td>1878 ± 55Å</td>
<td>0.44</td>
<td>0.98</td>
</tr>
<tr>
<td>$B4$</td>
<td>77.5</td>
<td>2294 ± 65Å</td>
<td>0.63</td>
<td>0.99</td>
</tr>
<tr>
<td>$B6$</td>
<td>72.2</td>
<td>2370 ± 70Å</td>
<td>0.65</td>
<td>0.99</td>
</tr>
<tr>
<td>$B7$</td>
<td>64.9</td>
<td>2592 ± 75Å</td>
<td>0.71</td>
<td>1.07</td>
</tr>
<tr>
<td>$B8$</td>
<td>60.1</td>
<td>2711 ± 80Å</td>
<td>0.73</td>
<td>1.04</td>
</tr>
<tr>
<td>$B9$</td>
<td>56.5</td>
<td>3028 ± 90Å</td>
<td>0.79</td>
<td>1.07</td>
</tr>
<tr>
<td>$B10$</td>
<td>53.7</td>
<td>3220 ± 95Å</td>
<td>0.81</td>
<td>1.02</td>
</tr>
<tr>
<td>$B11$</td>
<td>51.6</td>
<td>3540 ± 100Å</td>
<td>0.84</td>
<td>1.03</td>
</tr>
<tr>
<td>$B12$</td>
<td>47.8</td>
<td>4827 ± 140Å</td>
<td>0.911</td>
<td>1.08</td>
</tr>
<tr>
<td>$A0$</td>
<td>87.5</td>
<td>1698 ± 50Å</td>
<td>0.32</td>
<td>1.07</td>
</tr>
<tr>
<td>$A1$</td>
<td>82.3</td>
<td>2396 ± 70Å</td>
<td>0.66</td>
<td>1.18</td>
</tr>
<tr>
<td>$A2$</td>
<td>65.2</td>
<td>2586 ± 75Å</td>
<td>0.71</td>
<td>1.27</td>
</tr>
<tr>
<td>$C0$</td>
<td>86.7</td>
<td>1630 ± 50Å</td>
<td>0.26</td>
<td>1.02</td>
</tr>
<tr>
<td>$C1$</td>
<td>86.7</td>
<td>1828 ± 50Å</td>
<td>0.41</td>
<td>1.19</td>
</tr>
<tr>
<td>$C2$</td>
<td>87.1</td>
<td>1896 ± 55Å</td>
<td>0.46</td>
<td>1.14</td>
</tr>
</tbody>
</table>
namely $3\gamma/\pi\Delta_0 > 0.05$. The location of film data agrees well with Hirschfeld's calculation when $\Delta_0/kT_c \approx 3 - 3.5$. Thus, we attribute the difference between crystal and films at low $T$ to the disorder in pure film.

The above model of Hirschfeld et al. is valid only at low $T$. To compare the full $\lambda^{-2}(T)$ vs $T$ data at least roughly with $d$-wave theory, we use an approximate model for disordered $d$-wave superconductors. [62] We assume a circular Fermi surface on the CuO$_2$ plane. The angular dependence of the $d$-wave order parameter $\Delta_k$ on Fermi surface is given by $\Delta_k = \Delta_0 (\cos^2 \theta - \sin^2 \theta)$, where $k$ is the wavevector on Fermi surface and $\theta$ is angle between $k$ and x-axis. The density of states in a hypothetical clean film is given by

$$N_s(E) = N_n(0) < \text{Re} \{ E/(E^2 - \Delta_k^2)^{1/2} \} >_k,$$  \hspace{1cm} (30)

where $<>$ means angular average over directions of $k$ on a circle. $N_n(0)$ denotes the density of states at Fermi energy in normal state. $N_s(E)$ has a logarithmic singularity at $E = \Delta_0$ and its form is shown in Fig. 9 for $\Gamma = 0$. We determine $\Delta_0(T)$, the $T$-dependence of $\Delta_0$, by fitting to the $\lambda(T)$ of Hardy's single crystal data with $\lambda(0) = 1400\text{Å}$. In the simplest model, the penetration depth is given by: [63]

$$\frac{\lambda_0^2}{\lambda^2(T)} = 1 - 2 \int_0^\infty \left( -\frac{\partial f}{\partial E} \right) \frac{N_s(E)}{N_n(0)} dE.$$  \hspace{1cm} (31)

With $\Delta_0(T)/kT_c$ as given in Fig. 10, we obtain the excellent fit to Hardy's data in Fig.
Figure 9. Disordered $d_{x^2-y^2}$ densities of states used to fit the crystal ($\Gamma/\Delta_0 = 0$) and pure film data ($\Gamma/\Delta_0 = 0.13$). $\Gamma$ is the scattering rate. A circular Fermi surface and the angular dependence of $\Delta(k) = \Delta_0 \cos(2\theta)$ are assumed.
Figure 10. $\Delta_0(T)$ used to fit the $\lambda(T)$ data of Hardy et al. The BCS weak coupling temperature dependence, scaled to a maximum $\Delta_0$ of $2.8kT_c$, is shown for comparison.
Figure 11. Clean $d$-wave fit to the crystal data of Hardy et al. (triangles) and disordered $d$-wave fit to the pure film data (dots) from the density of states in Fig. 9 and order parameter in Fig. 10.
11. As shown in Fig. 10, we obtained $\Delta_0/kT_c \approx 2.8$. The form of $\Delta(T)/\Delta_0$ is not much different from that of the BCS weak coupling model. In other words, a BCS-like $\Delta(T)/\Delta_0$ can produce the $1/\lambda^2(T)$ observed in single crystals. The obtained $\Delta(T)/\Delta_0$ does not leave much room for anomalously large $T$-dependencies of quantities like charge carrier density, scattering rate, effective mass, effective attractive electron-electron interaction, etc.

In order to include disorder we broaden the density of states with the algorithm first proposed by Dynes et al. in conventional superconductors: [64-66]

$$N_s(E) = N_n(0) < Re[(E-i\Gamma)/(E-i\Gamma)^2-\Delta_k^2]^{1/2}>_k,$$

(30)

where $\Gamma$ is the scattering rate. Scattering has the largest effect for directions in $k$-space where $\Gamma \geq \Delta_k$. With $\Gamma/\Delta_0 = 0.13$, we obtain the curve in Fig. 9. For the same $\Delta(T)/\Delta_0$ and $\lambda_0$ as for the crystal of Hardy et al., we fit the film data by adjusting the fit parameters $\Delta_0/kT_c$, $\Gamma$, and $\lambda_B(0)$. The result of fitting to the fully oxygenated film $B$ is shown by dots in fig. 11, with the best-fit parameters $\Gamma \approx 0.13\Delta_0$, $\Delta_0 \approx 2.85 \pm 0.5 kT_c$, and $\lambda_B(0) \approx 1750\text{Å}$. Thus, the reduction in $\lambda(0)^{-2}$ is due to disorder for a fixed total charge carrier concentration.

We conclude that disordered $d$-wave superconductivity is capable of accounting for the similarities and differences between films and crystals of YBCO on the basis of a small amount of disorder in the films. The disorder is so small that it is difficult to estimate from other measurements, such as extrapolating resistivity $\rho(T)$ to $T=0$. 
3. Extrinsic effects

It is sometimes argued that it is not possible to observe the intrinsic penetration depth of YBCO in films. Films are usually suspected to be different from crystals, namely because of more grain boundaries and more defects in films than in crystals. Also some variations in the \(\lambda\) measurement from lab to lab causes doubt whether thin film is proper at all for transport measurement. But we found some films in substantial agreement with crystals. In this section, we discuss the extrinsic effects of grain boundary and vortex-antivortex pair formation in detail, and provide arguments that they are negligible in high quality films. In the next section, supporting evidence for the absence of extrinsic effect in some high quality films will be given from oxygen depletion.

A. Grain boundary

As mentioned before, there is the crossover behavior in films. Somehow the differences between films and crystals are negligible above \(0.25T_c\) and appear only below \(0.25T_c\). Based on this observation, the possible scenario falls into either one of two cases. First, \(\lambda(T)\) in high quality films is in fact dominated by the grains rather than grain boundaries and possible defects. Secondly, the inductance of weak links at grain boundaries is important in \(\lambda(T)\) in films, but its \(T\)-dependence coincidentally has the same functional dependence on \(T\) as that of the intrinsic grains except below \(0.25T_c\). So far the characteristic \(T\)-dependence of the effective \(\lambda\) of a network of weak links has not been determined in high-\(T_c\) superconductors. While the former case seems more likely, it is
worthwhile to consider the latter in a little more detail because of the inevitable high density of grain boundaries in films.

In BCS theory, the Ambegaokar-Baratoff form for $I_c$ of a junction with a tunnel barrier, i.e., a S-I-S tunnel junction, is given by $I_c = [\pi \Delta(T)/2eR_n] \tanh(\Delta(T)/2kT)$, where $R_n$ is the tunneling resistance per unit area of the junction when both metals are in the normal state. [67] The inductance of a Josephson junction is given by $L_J = \Phi_0/(2\pi I_c \cos\phi)$, where $\phi$ is the phase difference across the junction. The inductance of a network of junctions is proportional to the average of $1/I_c$. Thus its $T$-dependence is the same as the $\lambda^2(T)$ of superconductor. The same thing might apply to unconventional superconductors. However, it is noted that S-I-S or S-I-N tunnel junctions are hard to form due to the short coherence length $\xi_{ab} \approx 10\AA$. The remaining possibility, the $T$-dependence of a S-N-S or weak-link Josephson junction, is very different from that of a S-I-S tunnel junctions. Experiments on the $T$-dependence of $I_c$ in S-N-S type junctions tells us that $I_c$ is proportional to $\exp(-T/T_0)$, where $T_0$ is a constant, showing a positive curvature in the $T$-dependence. [68,69] Similar $T$ dependence is also observed in S-semiconductor-S junctions. [70] On the other hand, $\lambda^{-2}$ in bulk superconductors should have a negative curvature. Thus it is highly unlikely that what we observed in $\lambda^{-2}(T)$ of YBa$_2$Cu$_3$O$_{7-\delta}$ films is caused by a network of grain boundaries rather than a bulk property.
B. Vortex-antivortex pair

In the previous section, the extrinsic effects due to grain boundaries are discussed and their possibility was ruled out. In this section another possibility of extrinsic effects is described: vortex-antivortex pair creation at defect sites. This phenomenon has been proposed as the explanation of the $T^2$-dependence of $\lambda(T)$ in films at low $T$. The presentation of an argument negating this possibility is the purpose of this section.

It is possible to generate a $T^2$ dependence of $\lambda$ at low $T$ even in a BCS superconductor if the shielding current in the film is large enough to create vortex-antivortex pairs at flaws in the film. [71,72] When there are defects such as slot-like apertures and the shielding current is perpendicular to the slots, the shielding current tends to concentrate at the ends of the slots. If the current is high enough, a pair of counter circulating vortices nucleates at the ends of the slot. The underlying physics is that elastic vortex motion in a pinning potential causes extra inductance in the system. The harmonic pinning potential exerts a restoring force $-kx$ on the vortex, where $x$ is the displacement from the equilibrium position. There is also a damping force, $\eta dx/dt$, where $\eta$ is the damping coefficient. The equation of motion is $\eta dx/dt + kx = dJ\Phi_0$, where $d$ is film thickness, $J$ the current density, and $\Phi_0$ the flux quantum. At zero frequency, $\eta dx/dt$ is negligible compared to $kx$. The current density $J$ is given by $J = kx/(d\Phi_0)$. The pinning energy is maximum due to the displacement of a vortex from its pinning center, when $x = \xi$. The maximum of the pinning energy $k\xi^2/2$ is the same as the condensation energy lost in the core area, which is given by the product of the energy density $\mu_0H_c^2/2$ and effective core volume $\pi\xi^2d$. Thus $k = H_c^2d/4$. The inductance due
to a vortex is the energy stored in pinning divided by the $2I^2$. It is given by $L_p = k_0^2/2(Jdw)^2 = k_0^2\Phi_0^2/2k_0^2\xi^2w^2 = \Phi_0^2/2k_0w^2$, where $w$ is the width of the sample. The sheet inductance due to $N_v$ vortices per unit area is given by $\Phi_0^2N_v/k$. Empirically it is known that $H_c = H_c(0)(1 - T^2/T_c^2)$. The penetration depth due to vortex motion is related to $L_p$ by $\lambda_p^2 = dL_p/\mu_0$, and leads to the form

$$\lambda_p^2 = d\Phi_0^2N_v/[\mu_0k(0)(1-T^2/T_c^2)^2] \tag{27}$$

Provided the shielding current is large enough, $N_v$ is twice of the number of defects in the film in zero-field limit, since a vortex-antivortex pair is formed at each defect. If one applies a large magnetic field, the number of vortices is simply $B/\Phi_0$. Eq. (50) leads to the $T^2$ dependence at low $T$, however, $1/\lambda^2(T) \propto H_c(0)^2(1-T^2/T_c^2)^2 \propto (1-T/T_c)^2$ near $T_c$ should be curved upward, and an inflection point in $1/\lambda(T)^2$ occurs whose location depends on the size of the effect if vortex-antivortex pairs make a significant contribution to $L_k$. We observe $1/\lambda^2(T) \propto (1-T/T_c)$ near $T_c$, indicating that vortex-antivortex pairs are not significant in our samples.

4. Penetration depth in oxygen depleted YBa$_2$Cu$_3$O$_{7-\delta}$ thin films

There are several reasons why one would like to know how the penetration depth in films is influenced by oxygen depletion. For one thing, the level of oxygen of our "fully-oxygenated" films is uncertain, so that it is important to know that the conclusions drawn from them are insensitive to the precise level of oxygenation. More
fundamentally, oxygen depletion is a means to change the effective total charge carrier density, by removing holes from the CuO$_2$ planes, with the introduction of very little disorder in the planes because the oxygens come from the CuO chains. This allows an additional point of comparison with the disordered $d$-wave theory of Hirschfeld et al. A technical reason is that one might expect the influence of grain boundaries to be enhanced if oxygen depletion is particularly damaging to the intergrain Josephson coupling through grain boundaries. In this case, the $T$-dependence of $\lambda$ would change dramatically.

There have been several studies of the effect of oxygen depletion on transport in the high-$T_c$ superconductors, most of which are on the normal state properties of resistivity or Hall coefficient. [73-75] Reports of measurements of oxygen depletion effects on the penetration depth are rare. The best measurement was made by Uemura et al. from $\mu$SR, where the muon spin depolarization rate $\sigma$ and $T_c$ are given. Recently they provided the relation $\lambda_{ab} = 2700/\sigma^{1/2}$ for their data, where $\lambda_{ab}$ is in the unit ofÅ, $\sigma$ in $\mu$s$^{-1}$. [76] There is a plateau at 90K in $T_c$ versus $1/\lambda_{ab}^2(0)$, [77-79] and a universal linear dependence of $T_c$ on $\lambda_{ab}^{-2}(0)$ at lower $T_c$'s. [37,38] They suggested that the linear dependence of $T_c$ was due to local pairing and Bose condensation of the pairs. [37,38] Lee et al. reported $\lambda_{ab}(T)$ of a YBa$_2$Cu$_3$O$_{7-\delta}$ crystal of $T_c=60K$ from isotherm magnetization method and obtained $\lambda_{ab}(0) = 2600\text{Å}$. [47] In this chapter, we report the high precision measurements of $\lambda_{ab}(T)$ of YBa$_2$Cu$_3$O$_{7-\delta}$ thin films whose $T_c$ ranges from 90K to 40K by varying the oxygen content in systematic way. The results agree quite well with the measurements on bulk samples just cited.
A reduction of oxygen content by a small amount and measurement of $\lambda_{ab}(T)$ are made repeatedly by annealing for several minutes at a fixed annealing temperature, like 250C for sample B and C or 280C for sample A, in 1atm Ar. Each data curve in Fig. 12 is the result of one trial. We provide a complete list of $T_c$ vs $\lambda_{ab}(0)$ in Table 3. The rest of this section deals exclusively with $\lambda_{ab}$, so we drop the subscript.

Fig. 12 shows $\lambda^2(T)$ for three films A, B and C at various oxygen contents represented by dashed, fine solid, and thick solid curve, respectively, as well as the crystal data (dots) in real scale. The top 3 data sets are fully oxygenated films C0, B0, and A0, respectively. It turns out that every curve in Fig. 12 has nearly the same shape; namely, the same $\lambda^2(0)/\lambda^2(T/T_c)$ vs $T/T_c$ for different $T_c$'s and $\lambda(0)$'s. Fig. 13 shows the overlap of 6 different normalized curves $\lambda^2(0)/\lambda^2(T/T_c)$ for sample B, which is the best one of the three samples, at various oxygen contents with different $T_c$'s and $\lambda(0)$'s.

$1/\lambda^2(T)$ in film A is almost the same as that of film B when they are fully oxygenated. When the oxygen content is lowered in film A, a long concave tail occurs near $T_c$, as shown by the bottom dashed curve in contrast to other curves from film B. Inhomogeneity in film A may be responsible for that behavior and might be relevant to the difference in $J_c$ of film A and B; $J_c$ of film A is $10^6 A/cm^2$, being ten times smaller than that of film B. Fully oxygenated film C has the largest $1/\lambda^2(0)$ in films. Since there is about $\pm 10\%$ error in the film thickness, $1/\lambda^2(0)$ has the same size of error. The difference in $1/\lambda^2(0)$ among the films is close to the error bound. Film C has a sharp drop near $T_c$ and a small concave tail at higher temperatures. The drop indicates that the shielding current in film reached critical current at that temperature. Unfortunately,
Figure 12. $1/\lambda^2(T)$ vs $T$ for the crystal of Hardy et al. (solid circles) and for our films $A$, $B$ and $C$ at various oxygen contents (dashed, fine solid, and thick solid curve, respectively). The top 3 data sets are fully oxygenated films $C0$, $B0$, and $A0$, respectively.
Figure 13. $\lambda^2(0)/\lambda^2(T/T_c)$ vs $T/T_c$ for several oxygen depleted YBCO films (films B0, B1, B6, B7, B10, and B12). There is good overlap of $\lambda^2(0)/\lambda^2(T/T_c)$ among the curves even though $T_c$ varies from 88K to 48K.
sample C fractured after two anneals. The absence of the long concave tail and a sharp
drop in film B near $T_c$ tells the good quality of film B.

From Fig. 12, we see that $1/\lambda^2(0)$ decreases as the oxygen is reduced. At first,
$1/\lambda^2(0)$ decreases for the fixed $T_c$ of about 88K, forming the plateau of $T_c$ vs $1/\lambda^2(0)$.
Beyond that both $T_c$ and $1/\lambda^2(0)$ decrease. This is clearly shown by $T_c$ vs $1/\lambda^2(0)$ in Fig.
14. Our films' data are given by the hollow circles, triangles, and squares; the $\mu$SR
data of bulk YBCO by Uemura et al. [37,38] are indicated by diamonds connected by
solid lines. We notice that the plateau in $T_c$ vs $1/\lambda^2(0)$ and the initial linear decrease of
$T_c$ with decreasing $\lambda^2(0)$ reproduce the results of Uemura et al. From this piece of
information, we conclude that superfluid density in the films is about 33% smaller than
that of bulk samples which have the same total charge carrier density, i.e., oxygen
concentration. Equivalently superfluid density is smaller than the total charge carrier
density by 33% in films, i.e., $\rho_s/\rho_0 \approx 2/3$. Data below 50K suggests the slight sign of
a second plateau. It has been known that there are two plateaus at 90K and 60K in $T_c$
vs $\delta$, the oxygen content, from several studies. [77,78,79]

Oxygen depletion also accompanies a slight increase of disorder, though it mostly
causes the reduction of total charge carrier density. In Fig. 13, there is about 2% 
discrepancy at $T = 0.5T_c$ from the top (oxygen-depleted) to bottom (fully oxygenated)
sample films. As the oxygen content is reduced, the linear portion between 0.25$T_c$ and
0.4$T_c$ disappears and takes on a round shape. This could happen as the crossover
temperature moves up due to a slight increase of disorder. We expect a slight increase
of disorder due to the oxygen depletion. In order to see the significance of the 2%
Figure 14. $T_c$ vs $1/\lambda^2(0)$ for our three films at various oxygen content. $\mu$SR data of bulk YBCO from Uemura et al. are presented by diamonds. $1/\lambda^2(0)$ of films is about 30% smaller than for bulk samples with the same $T_c$. 
discrepancy at $0.5 T_c$, we simulated the effect of the increased disorder by increasing the scattering rate from $\Gamma = 0.13 \Delta_0$ to $0.3 \Delta_0$ in the model of broadened density of states described in section 2. The $\lambda^2(0)/\lambda^2(T/T_c)$ vs $T/T_c$ for $\Gamma/\Delta_0 = 0.13$ and 0.3 are shown Fig. 15 (a) in solid curve and dashed curve, respectively. The discrepancy is about the same size as that of film B shown in Fig. 13. Fig. 15 (b) shows $\lambda_0^2/\lambda^2(T/T_c)$ vs $T/T_c$ for the same result of the simulation. The 2% increase of $\lambda^2(0)/\lambda^2(T/T_c)$ at $0.5 T_c$ is equivalent to the 20% increase of $\lambda^2(0)$. The reduction in $1/\lambda^2(0)$ due to oxygen depletion in film B12 is a factor of 8, which is much larger than the part contributed from the increased disorder.

In our determination of $\lambda(0)$ from the best-fit, we used the same template curve extract from Hardy et al. for all oxygen depleted films, because the exact form of template curve for YBCO at each oxygen concentration is not known. As the oxygen content is lowered, $1/\lambda^2(T)$ is expected to be more curved. The more round shaped template curve leads to the larger nominal value of $\lambda(0)$. The template curve we use is linear below $0.4 T_c$. Thus our nominal value of $\lambda(0)$ is deviated a little from the actual value. But it is certain that the actual $\lambda^2(0)$ is larger than the nominal value by less than 20%. The slight larger value of actual $\lambda(0)$ also influences the comparison of our data with the calculation of Hirschfeld et al. Data points in Fig. 16 represent the reciprocal of curvature, $1/c_2 = \lambda^2(0)/\delta \lambda^2(T)$, at low $T$ and the fractional reduction of superfluid density $c_0$ in the same way as for the fully oxygenated films in Fig. 8. The increase of $c_0$ is mostly attributed to the reduction of total charge carrier density. A slight increase of $1/c_2$ was expected because of the increase of disorder due to oxygen depletion. For
Figure 15. The simulation of disorder effect in (a) $\lambda^2(0)/\lambda^2(T/T_c)$ vs $T/T_c$ and (b) $\lambda_0^2/\lambda^2(T/T_c)$ vs $T/T_c$. The difference in (a) of about 2.5% is equivalent to 25% difference in $\lambda^2(0)$. 
Figure 16. $1/c_2$ vs $c_0$ for YBCO films $A$, $B$, and $C$ at various oxygen contents. The increase of $c_0$ with fixed $1/c_2 \approx 1$ in sample $B$ implies that oxygen depletion mainly causes reduction of the total charge carrier density without causing disorder, according to the theory of Hirschfeld et al.
films A and C, there is a slight increase of $1/c_2$ as much as 20%. For film B, $1/c_2 \approx 1$, being almost constant. In the determination of $1/c_2$, one needs to know the value of $\lambda(0)$. The actual $\lambda^2(0)$ of film B12 could be 20% larger, so does the actual value of $1/c_2$; the actual $c_0$ for film B12 shifts slightly to the righthand side since $1-c_0$ decreases by the same fraction. So the actual $1/c_2$ of film B could increase with increasing $c_0$.

The values of $c_0$ and $1/c_2$ are listed in Table 3. The size of random error in $c_2$ is about $\pm 5\%$, which is much larger than the random error of 0.2% in $c_0$. This is because $c_2$ is a kind of curvature, which is proportional to the small change from the zero temperature value. Note that $c_2$ is nearly constant at unity, so the low-$T$ behavior is close to $1-T^2/T_c^2$. This is contrasted to the reports from a couple of other groups, where $\lambda(T)$ is flat at low $T$ so that their $1/c_2$ could be much larger than unity. [58,80]

The summary of this section is the following. The nearly identical shape of $\lambda^2(0)/\lambda^2(T/T_c)$ vs $T/T_c$ at various oxygen contents and the comparison of $T_c$ vs $1/\lambda^2(0)$ with $\mu$SR data reveal that our measurement is an intrinsic property of superconducting grains. Thus our films are as reliable as single crystals. $1/\lambda^2(0)$ of the fully oxygenated sample is about $35\mu m^{-2}$, compared with $50\mu m^{-2}$ for Hardy's single crystal. Also from the comparison of $T_c$ vs $1/\lambda^2(0)$ between our data and Uemura et al., $1/\lambda^2(0)$ in film is about 30% smaller than that of bulk probed by $\mu$SR. The difference in $1/\lambda^2(0)$ between the two is likely due to the disorder in the film. In the next section, we will discuss the implication of $\rho_s/\rho_0 = 2/3$. This will be another supporting argument for $d$-wave pairing.
5. Comparison with isotropic s-wave

In this section, we will compare d-wave and isotropic s-wave models as explanation of the reduction of the superfluid density by 30% from the total charge carrier density. Since impurity effects on the penetration depth of anisotropic s-wave models proposed especially for oxide superconductors is not known, we have to rely on isotropic s-wave as an alternative.

Kim et al. [61] recently made calculation of nonmagnetic impurity scattering effects in a d-wave superconductor at zero $T$. Their calculation is valid for the entire range of impurity concentration, but only provides the increase of $\lambda(0)$ as a function of scattering rate. They made comparison among the cases of the strong impurity potential and the weak impurity potential in d-wave and the isotropic s-wave. The isotropic s-wave is insensitive to the strength of the scattering potential. On the other hand, d-wave is strongly dependent on the strength of the scattering potential.

The scattering rate $1/\tau$ in YBCO film is about $2 \times 10^{13}$ rad/sec at 100K. Correspondingly, $\hbar/\tau \Delta_0$ is about 0.9. At zero $T$, since only the residual scattering rate remains, $1/\tau$ is expected to be much smaller. Surface resistance measurements on single crystals show $1/\tau$ drops to below 1% of normal state value at low $T$. For thin films, the real part of the conductivity ranges from $3 \times 10^6$ to $3 \times 10^7 (\Omega m)^{-1}$ at 4.2K. [58,80] The corresponding $1/\tau$ is about from $3 \times 10^{12}$ to $3 \times 10^{11}$ rad/sec. Thus $\hbar/\tau \Delta_0$ ranges about from 0.01 to 0.1. Then we find from the calculation of Kim et al. that the ratio $\rho_s/\rho_0$ is from 0.65 to 0.9 for strong scattering in d-wave, from 0.85 to 0.97 for weak scattering in d-wave, and from 0.93 to 0.97 for isotropic s-wave for the estimated range of $1/\tau$. 
[61] Our data dictates $\rho_s/\rho_0 = 2/3$. Thus the most suitable model is $d$-wave with strong scattering impurities.

For another way to estimate the increase of $\lambda(0)$, note that it is approximately given by $\lambda(0) = \lambda_L(0)(1 + \xi_0/\ell)^{1/2}$ in $s$-wave superconductor, where $\ell$ is the mean-free-path of the charge carrier and $\xi_0 = 10\text{Å}$ is the coherence length in the $a$-$b$ plane. Even if the residual resistivity of $60\mu\Omega cm$ in the fully oxygenated film is assumed, $\ell$ is at least $50\text{Å}$. The largest possible increase in $\lambda(0)$ would be from $1400\text{Å}$ to $1530\text{Å}$ if it were an isotropic $s$-wave superconductor. This increase is much too small when compared with the value $\lambda_A(0) = 1700\text{Å}$ in the film.

This comparison implies the reduction of $\rho_s/\rho_0$ is too large to explain with isotropic $s$-wave. Since there is no understanding of anisotropic $s$-wave, we could not make comparison with anisotropic $s$-wave and the above comparison is not complete. However, it is possible that the reduction of $\rho_s/\rho_0$ in anisotropic $s$-wave would be smaller than that of isotropic $s$-wave, since gap anisotropy decreases as it becomes more disordered and the gap minimum increases. The above analysis in combination with the quadratic and linear $T$-dependence at low $T$ strongly supports $d$-wave superconductivity.
CHAPTER V
CONCLUDING SUMMARY

We have performed the first extensive experiments of the penetration depth of YBa$_2$Cu$_3$O$_{7-\delta}$ thin films. Their greatest significance lies in the investigation of the pairing symmetry of high-$T_c$ superconductors. A current theory proposed by Pines and collaborators, where pairing is caused by spin fluctuations in antiferromagnetic order, attracts peoples’ attention. The basic feature of this model is that the pairing has a $d$-wave orbital wave function. Anderson proposed a theory, which is the counterpart to Pines model, where pairing is $s$-wave but is highly anisotropic. By only focusing on the gap anisotropy at different orientations on the Fermi surface, the results of these two models look similar, making it difficult for most of experiments to distinguish the difference between the two. An experiment that is sensitive to the sign of the energy gap was devised to determine the better theory, since $d$-wave superconductivity produces a change of sign at different orientations on the Fermi surface while $s$-wave does not. Though the result favors $d$-wave pairing, it is not conclusive due to the limited resolution.

There has been a theory about the effect of microscopic impurities on $d$-wave superconductivity, which was developed during the interpretation of penetration depth
data on heavy-fermion superconductors. The impurity effect in $d$-wave superconductor is another detectable phenomenon. Generally $d$-wave superconductivity is sensitive to the impurities. In particular, a small level of disorder causes a rapid drop in superfluid density and a change of $T$-dependence from linear $T$-dependence to $T^2$-dependence at low $T$.

The penetration depth is a measure of the number of quasiparticle excitations, which depends on the available phase space at the fermi surface. In an $s$-wave superconductor, there is a finite energy gap at the fermi surface, and it leads to an exponential $T$-dependence of $\Delta\lambda(T)$. The energy gap in a $d$-wave superconductor has line nodes as a result of the sign change for different orientations on the Fermi surface, giving the linear $T$-dependence of $\Delta\lambda(T)$. These days, high resolution is available in penetration depth measurements, making them a good test for determining the minimum magnitude of the energy gap.

In early days, the results of penetration depth measurements were used to conclude that high-$T_c$ superconductors have a conventional energy gap like BCS superconductors. Since the resolution was poor at that time, large errors in the data masked the discrepancy between the data and BCS results. Since then the resolution has been improved by a factor of about 100. Recently it was reported by Hardy et al. that a clean single crystal shows a linear $T$-dependence of $\Delta\lambda(T)$ at low $T$. This observation is interpreted as the result of a $d$-wave energy gap. Several other groups have reported $T^2$ dependence of $\Delta\lambda(T)$. The observed $T^2$ dependence in film can be interpreted as an impurity effect in $d$-wave superconductor. On the other hand, the linear $T$-dependence
is thought to be observed because they are effectively "clean" at higher temperatures. Then there should be a crossover between linear $T$-dependence and $T^2$-dependence if the impurity concentration in the sample is between that of a clean crystal and a thin film.

We have performed intensive penetration depth measurements on high quality YBa$_2$Cu$_3$O$_{7-\delta}$ thin films made by laser ablation. A systematic oxygen depletion of the films and measurements of $\lambda(T)$ were made repeatedly. There is a close overlap of the $\lambda^2(0)/\lambda^2(T/T_c)$ curves from the samples of different $T_c$ and different $\lambda(0)$, which are induced by the partial oxygen depletion. Since $T_c$ is determined by superconductor grain, this proves that the measured $1/\lambda^2(T)$ in a high quality film is the properties of the superconducting grain, not the extraneous effects such as grain boundary weak links.

The data obtained from films look similar to the crystal data by Hardy et al. above a crossover temperature, which separates $T^2$ and $T^1$ dependence. Our measurement on high quality films has the usual $T^2$ dependence below the crossover temperature. We used a two-coil method that has higher resolution than other results performed with this method. Careful numerical modeling of the two-coil apparatus is achieved. We used a fitting procedure, which is common to every method except $\mu$SR and isotherm magnetization. We used a proper fitting procedure by adopting the template curve extracted from the single-crystal data of Hardy et al. The goodness of our fitting procedure is justified by excellent agreement between our data and the template curve. We also analyzed the results of the assumed grain-boundary weak links and vortex-antivortex pair formation at defects sites, and found that their implications were incompatible to our result.
We quantified the crossover behavior from the data of the fully oxygenated films. The low $T$ data was compared with the calculation of Hirschfeld et al. for the disordered $d$-wave. With $\Delta_0/kT_c \approx 3$, they agree well. The information in high $T$ data is used for the analysis with the simple model of broadened density of states. They also resulted in $\Delta_0/kT_c = 2.8 \pm 0.5$. From the oxygen depletion study, $T_c$ vs $1/\lambda^2(0)$ reproduces the results of Uemura et al. We found that superfluid density is about 30% smaller than total charge carrier density from the comparison of $T_c$ vs $1/\lambda^2(0)$ of our data and the $\mu$SR data from Uemura et al. The most well developed and the closest model to explain our penetration depth results, the $T^2$ dependence at low $T$ and the large reduction of superfluid density, is $d$-wave pairing with strong impurity scattering. Still there is a room for highly anisotropic $s$-wave with the gap ratio of maximum to minimum being about 20, but that is only because such $s$-wave is not understood.

We have introduced several results from other experiments supporting $d$-wave pairing, but not a single experiment made a decisive proof of $d$-wave pairing. The collection of the consistent results from independent methods only strongly supports $d$-wave pairing. There are still some reports coming out of penetration depth measurements that claim anisotropic $s$-wave pairing. Penetration depth is distinguished from other methods because of its high precision. As the $d$-wave issue started after the penetration depth results came out, further tests of this $d$-wave issue are dependent on the high precision measurement of penetration depth. Our result supports $d$-wave pairing, and we hope future results lead to the clear understanding of this material.
APPENDIX A

MEASUREMENT OF BERNOULLI VOLTAGE $V_B$ IN A HIGH-$T_c$ SUPERCONDUCTOR

Measurement of the Bernoulli voltage can determine the sign of supercurrent charge carriers, or equivalently the sign of their effective mass. The difference in charge carrier velocities at different positions inside a superconductor on the application of a current gives rise to an electrostatic field inside the superconductor, in analogy to the pressure gradient corresponding to velocity difference in an ordinary fluid. This is the Bernoulli effect in superconductors. The direction of electric field is determined only by the sign of the charge carrier; it is independent of the direction of current. If the charge carrier velocity is made large by making the sample have a narrow center, there is acceleration upon the incoming of the charged particle and a deceleration upon outgoing. The direction of the force points toward center. Thus for a positive charge carrier (negative effective mass), the electric field direction is toward the narrow center, and vice versa. A proper design of sample geometry such as narrow strip with wide ends or a simple disk is required. [81,82] The effect gives information on the supercurrent effective mass and its sign, and the correlation of $T_c$ and charge carrier density $n$. The effective mass $m^*$ probed by $V_B$ is equal to the inverse of the average curvature of $\epsilon(k)$
on the Fermi surface, where $\epsilon(\mathbf{k})$ is the energy-momentum dispersion relation. Other methods which measure effective mass often measure different physical quantities such as the ratio of velocity to momentum at the Fermi surface.

$V_B$ was measured for the conventional superconductor Pb and In a long time ago. [83-86] No measurement has been made on a high-$T_c$ superconductor. In high-$T_c$ superconductor, where Hall coefficient has strong $T$-dependence in contrast to the constant behavior in metal, another means to investigate charge carrier density has significance. A combination of $\chi^2(0)$ and $V_B$ reveals $n$ and $m^*$.

In thermodynamic equilibrium, the Helmholtz free energy $F$ can be written as a function of volume $V$, temperature $T$, charge carrier density, and superfluid momentum $p_s$. [87] If current is weak, $F$ can be expanded in powers of $p_s$, and terms higher than second order can be neglected. Since $F$ is minimum when $p_s = 0$, a linear term in $p_s$ is absent, and

$$F(V,T,n,p_s) = F_0(p_s=0) + \frac{1}{2} \bar{p}_s \cdot \bar{\alpha} \cdot \bar{p}_s,$$  (A-1)

where the tensor $\bar{\alpha}$ is the coefficient of expansion, and is related to the superfluid density $n_s$ and the effective mass $m^*$. In isotropic system, it reduces to $\alpha = n_s/m^*$ from the relation for the supercurrent $J_s = e\alpha p_s$. The electrochemical potential, which is the sum of chemical potential $\mu$ and electrostatic potential $eV_B$, is uniform in the sample in thermodynamic equilibrium. Therefore,
\[-eV_B = \delta \mu = \frac{\partial F}{\partial n} = \frac{\partial \alpha}{\partial n} \frac{P_s^2}{2}. \] (A-2)

At \(T=0\),
\[
\frac{\partial \alpha}{\partial n} \left< \frac{1}{m^*} \right>_{f.s.} = \left< \frac{\partial^2 \varepsilon}{\partial k^2} \right>_{f.s.} \] (A-3)

from the relation \(1/m^* = \partial^2 \varepsilon(k) / \partial k^2\), where the bracket represents average over the Fermi surface. For an isotropic superconductor,
\[
-eV_B = \left( \frac{\partial n_s}{\partial n} \right) \frac{P_s^2}{2m^*} = \left( \frac{n_s}{n} \frac{\partial n_s}{\partial T_c} \frac{\partial T_c}{\partial n} \right) \frac{p_s^2}{2m^*}. \] (A-4)

The implicit dependence of \(T_c\) on \(n\) is responsible for the second term in the right side of equation (A-4). Near \(T_c\) \(n_s/n=0\), and \(dn_s/dT_c = n/T_c\) since \(n_s = n(1-T/T_c)\), being linear in \(1-T/T_c\) in a second order mean field transition. Information on \(dT_c/dn\) can be obtained from the measured \(V_B\) near \(T_c\). \(V_B\) can be also expressed by
\[
-eV_B = \frac{\lambda^2(0)\mu_0 J_s^2}{n} \frac{J_s^2}{2} \] (A-5)

for a known current density \(J_s\) at low \(T\).

Since \(V_B\) is proportional to \(J_s^2\), \(V_B\) is the second harmonic of the drive current. Since the electrochemical potential is constant in thermodynamic equilibrium, \(V_B\) cannot be measured by a voltmeter, which measures the difference in electrochemical potential across the lead contacts. The contact potential difference between sample and contact leads exactly cancels the Bernoulli voltage, whose origin is in the chemical potential.
difference at two different position inside the sample. Thus a capacitive contact has to be used as shown Fig. 17. Since a curl-free closed loop should have zero potential difference, there is an electrostatic potential difference between the capacitor electrodes which cancels $V_B$ upon applying current. As an $ac$ current is applied to the sample, the contact capacitor goes through the charging and discharging process in repetition. The corresponding flow of electrons occurs in the lead connected to the capacitor. When a passive device like a resistor or inductor is connected to the capacitor, a voltage drop develops across its terminals. As shown in Fig. 17, we used a meander line geometry to provide a large area for the capacitor electrode. The width and thickness of the line are $80 \mu m$ and $1000 \AA$ respectively, and its stretched length is $17 cm$. It is the same sample used for the kinetic inductance measurement. The capacitance of the contact is required to be larger than $1 nF$ to overcome the attenuation caused by stray capacitance of the cable attached to sample and instrument. We also tried the deposition of counter-electrode on the backside of the substrate to use SrTiO$_3$ as dielectric because SrTiO$_3$ is a ferroelectric and has large dielectric constant of several thousand at low $T$. But we have not obtained reproducible result.

Deposition of a $1 \mu m$ thick SiO layer as a dielectric material on the meander line and subsequent deposition of $3000 \AA$ thick Ag counter-electrode were made to form a capacitor. Since the resulting voltage is very small, on the order of $1 nV$, and the capacitance of the contact is small, about $1 nF$, the passive amplification by an $LC$ resonator circuit is adopted in order to increase the signal to noise ratio by tuning at resonance frequency. The resonance circuit also behaves as a band-pass filter with a
Figure 17. Schematic circuit diagram for Bernoulli voltage measurement. Center tap of the meander line is connected to ground to average out the impurity signal over the capacitive contact C.
bandwidth of $f_0/Q$, thus only a signal of the tuned frequency is amplified, where $f_0$ is the resonance frequency and $Q$ is the quality value of resonator. An inductor of 2.6 henry with a ferrite pot core manufactured by Phillips is connected to the capacitive contact to form an $LC$ resonator circuit. The inductor is shielded by $\mu$-metal can and is kept at room temperature. Amplification by the factor of $Q$, which is about 100, was achieved by measuring the voltage drop across the inductor. Ground is attached to the center tap of the meander line so that impurity in signal due to second harmonics in the applied current is averaged out over the capacitive contact. A typical $f_0$ is 1.5kHz, and the frequency of applied current is half of that.

Measurements at 4.2K are shown in Fig. 18. The signal voltage is proportional to the square of the applied current. The sign is consistent with positive charge carriers. The resultant $m^*$ is $20m_e$, using $\lambda(0)=2000\text{Å}$. It is rather large compared to the reported value of 7 found in de Haas-van Alphen experiments [88] and 10 from photoemission [89]. The discrepancy could be accounted by a possible non-superconducting layer of normal metal on top of the sample surface in the capacitive contact. In that case, the electric field between the Ag contact and superconducting layer can be screened by a normal-metal layer as thick as the Thomas-Fermi screening length, on the order of 10Å in the high-$T_c$ materials. Good surface quality is required to prevent the formation of a conducting layer extrinsic to the bulk. If what we measured is a property intrinsic to bulk, then the result can be interpreted as probing the small curvature of $\varepsilon(k)$ at the Fermi surface: a larger effective mass than other methods can probe. The possibility is that the inflection point of $\varepsilon(k)$ is close to Fermi surface.
Figure 18. Bernoulli voltage, $V_B$ vs current density squared, $J_s^2$. $V_B$ is predicted to be proportional to $J_s^2$. There is offset of about 0.7nV in the signal due to the lock-in amplifier.
APPENDIX B

$\lambda(T)$ OF $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ DETERMINED BY KINETIC INDUCTANCE

In this method the kinetic inductance of a narrow superconducting strip is measured by a four-contact method. A small linewidth $w$ enhances the kinetic inductance to magnetic inductance ratio, which is approximately $\lambda^2/dw$, where $d$ is the sample thickness. A large number of squares increases the sensitivity of the measurement. A meander line geometry is adopted to accommodate a large number of squares in a limited substrate area.

Our films are prepared by codeposition of Y, BaF$_2$, and Cu onto a SrTiO$_3$ (100) substrate, with a postanneal in wet oxygen at 800°C for 90 minutes in a tube furnace. The sample is patterned into a meander line ($100nm \times 80\mu m \times 17.5cm$) by standard photolithography and etching with dilute nitric acid ($<0.1\%$). Silver contacts are then deposited on the film and annealed under flowing oxygen at 450°C for an hour. Leads are attached with pressed indium. The critical current density $J_c$ is typically higher than $10^6A/cm^2$ at 4.2K, and $T_c(R=0) \approx 88K$. The resistivity is $\rho=130\mu\Omega cm$ at $T=100K$ and extrapolates linearly to less than $10\mu\Omega cm$ at $T=0$.

The line consists of 26 folds of $0.67cm$ long segments spaced by $80\mu m$, as shown in Fig. 19(a). Current and voltage leads connect to the lower contact pads, as in Fig.
20, with an additional voltage lead (not shown) at the upper pad for diagnostic purposes. The schematic sample geometry of a 6 fold line is shown in Fig. 19 (a) for clarity. A current density from 750 to $2 \times 10^4 \text{A/cm}^2$ is applied at 30kHz; it is monitored by the voltage drop across the standard resistor, (Fig. 20). In this current range, the magnetic field at the film edges is below $H_{c1}$. Small current densities allow measurements close to $T_c$; large current densities amplify the signal to noise at the lowest temperatures. The voltage is 90° ahead of current, as measured by a PAR 5208 two channel lock-in amplifier; no dissipation is observed.

The measured voltage has two contributions. One, the kinetic inductance $L_k$, is from the electric field that accelerates the supercarriers inside the film. The other, the magnetic inductance $L_m + L_M$, is from the changing magnetic flux $d\Phi/dt$ picked up in the voltage measuring circuit from the ac currents in the sample and in the adjustable mutual inductor, respectively.

It is important to include in the analysis the nonuniform supercurrent density $J(r)$ within the film. It is sufficiently accurate to neglect variations in $J(r)$ along the length of each of the 26 long narrow segments, and to ignore the very short end segments completely. The slight variation in $J(r)$ through the film thickness is accounted by considering the effective thickness $d_{\text{eff}} = \lambda \tanh(d/\lambda)$, through which the $J(r)$ is regarded uniform. [59] The normalized current density $J(x)/J_0$ is calculated numerically from [90,91]

$$-\mu_0 \lambda^2 J(r) = \nabla \chi + \mu_0/4\pi \int J(r')/|r-r'| \, dr'.$$ (B-1)
Figure 19. Meander line geometry and current density inside the line. (a) Left: schematic diagram of meander line geometry. Right: sample geometry. (b) Normalized current density $|J(x)/J_0|$ of 6 fold 80$\mu$m wide 0.67cm long line for $\lambda^2/d=400$ nm is plotted against $x$ along the dashed line indicated in (a).
Figure 20. Schematic circuit diagram. 4 contacts are attached to the meander line. 99\% of the magnetic inductance of the sample is canceled with the adjustable mutual inductance.
This equation is derived from combining London’s equation, \( J(r) = -1/(\mu_0 \lambda^2) A(r) \) with the general solution to Maxwell’s equations, \( A(r) = \nabla \chi + \mu_0/4\pi \int J(r')/|r-r'|dr' \), where \( \chi \) is a function determined by boundary conditions. In the integral, we neglect currents outside of the sample. In the limit of a long line, the above equation leads to

\[
-\mu_0 \lambda^2 J(x_i) = \nabla \chi_i + \sum_j \mu_0/4\pi \int J(x_j) \ln(L/|x_i-x_j|)dx_j, \tag{B-2}
\]

where \( x \) is the coordinate across the segments. The contribution of the \( j \)-th segment on the \( i \)-th segment is incorporated by the summation on \( j \). With no external applied magnetic field, \( \nabla \chi_i \) is constant within each segment, and its size is determined by the fixed total current.

The contribution of the sample, \( L_k + L_m \), to the measured voltage comes entirely from the \( \nabla \chi \) term. The magnetic inductance \( L_m + L_M \) is from the line integral of \( A \) around the voltage measuring loop, while the kinetic inductance comes from the line integral of \( J \) throughout the film from one contact pad (1) to the other (2). Therefore, the total sample inductance \( L_S \) is:

\[
L_S = (\int \nabla \cdot d\ell) / I = \{\chi(r_2) - \chi(r_1)\} / I, \tag{B-3}
\]

where \( I \) is the total current. From Eqs. (B-1) and (B-3), \( L_S \) is:

\[
L_S = L_m + L_k \approx \alpha \mu_0 \ell + \mu_0 \ell \lambda^2 / wd. \tag{B-4}
\]
Here, \( \alpha \approx 1 \), depending on the sample geometry. Also, \( l, w, \) and \( d \) are the length, width, and thickness of the sample, respectively. If \( J \) were uniform across the film width, then \( L_k \) would equal \( \mu_0 \lambda^2 l/wd \) and \( L_m \) would be independent of \( \lambda \). The nonuniform current density modifies the dependence of \( L_k \) on \( \lambda \) and makes \( L_m \) weakly dependent on \( \lambda \). Above about \( 0.8T_c \), \( L_k \) diverges and thereby dominates the \( T \) dependence of \( L_S \), so that \( \lambda \) is determined unambiguously in that range of temperatures.

The mutual inductance \( L_M \) is adjusted to cancel about 99\% of \( L_m \), i.e., \( L_M + L_m \approx 0.01L_m \), thus increasing the sensitivity in the voltage measurement at low \( T \). For \( l=10cm, w=80 \mu m, d=1000 \AA, \) and \( \lambda=2000 \AA, L_m \) is 40nH and \( L_k \) is about 600pH. Because of the less constricted geometry of supercurrents in the two-coil method for measuring \( \lambda \), the effective kinetic inductance in this method could be made larger, thus the resolution, than that of the two-coil method by increasing the number of squares in the line.

The one fitting parameter is the size of the "noncanceled" inductance, \( L_M + L_m(0) \). In general, this quantity is about equal to the kinetic inductance at \( T=0 \). Figure 21 shows \( 1/\lambda^2 \) vs \( T \) with \( L_M + L_m(0) \) chosen for a best-fit to the template curve extracted from crystal data by Hardy et al. No choice of \( L_M + L_m(0) \) gives the characteristic flatness of the BCS and two-fluid formulas for \( \lambda(T) \) for \( T < 0.3T_c \). We see that \( \lambda/\lambda(0) - 1 \propto T^2 \) for \( 0.06T_c < T < 0.4T_c \) from the plot of \( \lambda(T) \) vs \( T^2 \) in Fig. 22. The dependence on \( T^2 \) is robust, experimentally. \( L_S \) depends on \( T \) only because \( L_S \) depends on \( \lambda \), so that at low \( T \), \( dL_S/dT = [dL_S/d\lambda]_{\lambda(0)} d\lambda/dT \).
Figure 21. $1/\lambda^2$ vs $T$ for YBa$_2$Cu$_3$O$_{7-\delta}$. The value of $\lambda(0)$ is determined from the best-fit to crystal data from Hardy \textit{et al.} which is indicated by dashed curve. A larger measuring current, $J = 16000\, \text{A/cm}^2$, is used at low $T$ to increase the signal to noise ratio.
Figure 22. $\lambda(T)$ vs $(T/T_c)^2$, showing that $\lambda(T)-\lambda(0) \propto T^2$ below $0.4T_c$. 

$100\text{nm} \times 80\mu\text{m} \times 8.75\text{cm}$

$J=1.6 \times 10^4 \text{A/cm}^2$
It is possible for a $T^2$ dependence of $\lambda$ to be generated even in a BCS superconductor if the applied current is large enough to create vortex-antivortex pairs at flaws in the film. [71,72] However, the additional inductance generated by the vortex motion scales with spring constant of the pinning potential energy, which is about $H_c^2(T)\mu_0d$. Thus, $1/\lambda^2(T) = H_c(0)^2(1-T^2/T_c^2)^2 = (1-T/T_c)^2$ near $T_c$ should be curved upward if vortex-antivortex pairs make a significant contribution to $L_k$. We observe $1/\lambda^2(T) \propto (1-T/T_c)$ near $T_c$, indicating that vortex-antivortex pairs are not significant in our samples.

In summary, we achieve a high resolution of $\pm 5\text{Å}$ in the determination of $\lambda(T)$ from measurements of the inductance, kinetic and magnetic, of a long meander line. Accurate numerical modeling of the nonuniform current density across the film width is made. We find a clear $T^2$ dependence of $\lambda/\lambda(0) - 1$ at low $T$. 
APPENDIX C

PENETRATION DEPTH IN BCS SUPERCONDUCTORS

The current response to various Fourier components of a vector potential $A(q)$ in superconductor is generally given by

$$J(q) = -K(q,T) A(q),$$  \hspace{1cm} (C-1)

where $-K(q,T)$ is the linear response kernel. The expression for the London penetration depth, $\lambda_L$, in Drude form is $(\mu_0 n_s e^2 / m^*)^{-1/2}$, where $n_s$ is superfluid density and $m^*$ is the effective mass. In the BCS model, $\lambda_L^{-2}(T)$ is defined to be equal to $K(0,T)$, the kernel in the infinite wavelength limit, and its full expression is given by

$$K(0,T) = \lambda_L^{-2}(T) = \lambda_L^{-2}(0) \left[ 1 - 2 \int_{\Delta}^{\infty} \left( \frac{-\partial f}{\partial E} \right) \frac{E}{\sqrt{E^2 - \Delta^2}} dE \right],$$  \hspace{1cm} (C-2)

where $f(E)$ is fermi function. Another form of $\lambda_L(T)$ is given by

$$\lambda_L(T) = \lambda(0) \left[ 1 - \frac{\partial \ln \Delta(T)}{\partial \ln T} \right]^{-1/2},$$  \hspace{1cm} (C-3)

and its numerical value can be found in Mühlschlegel’s tabulation. \cite{92} It also can be
approximated by the simple analytic form,

$$\lambda_L(T) = \{\Delta(T)/\Delta(0)\tanh(\Delta(T)/2kT)\}^{-1/2}. \quad (C-4)$$

The penetration depth defined by $\lambda = \int_0^\infty B(z)dz/B(0)$ is what is measured in bulk samples, where $B(0)$ is the magnetic field at the infinite plane of the sample surface at $z=0$. Maxwell's equations determine the relation

$$\nabla^2 A = -\text{curl}\,\text{curl}\,A = -\text{curl}\,B = -\mu_0 J_{\text{total}} = -\mu_0(J_{\text{ext}} + J_{\text{med}}), \quad (C-5)$$

where $J_{\text{ext}}$ is the external current, and $J_{\text{med}}$ is the current in the superconducting medium. Fourier transform of eq. (C-5) is

$$q^2 a(q) = \mu_0 J_{\text{ext}}(q) - K(q)a(q). \quad (C-6)$$

In solving for $a(q)$, the result is given by

$$a(q) = J_{\text{ext}}(q)/\{K(q) + q^2\}. \quad (C-7)$$

We consider an infinite medium of superconductor with a current sheet $J_{x,\text{ext}} = -1/\mu_0 B_0\delta(z)$. The $q$-th component of $B_y$ is given by $B_y(q) = iqa(q)$ from $B = \text{curl}\,A$. Integrating over all the Fourier component yields
\[ h(z) = \frac{B_0}{i\pi} \int_{-\infty}^{\infty} \frac{qe^{iqz}}{K(q)+q^2} dq = \frac{2B_0}{\pi} \int_{0}^{\infty} \frac{q\sin(qz)}{K(q)+q^2} dq. \tag{C-8} \]

The penetration depth defined as above is given by

\[ \lambda = B_0^{-1} \int_{0}^{\infty} h(z)dz = \frac{2}{\pi} \int_{0}^{\infty} \frac{q\sin(qz) dq}{K(q)+q^2}. \tag{C-9} \]

where \( K(q) \) is a function of \( q\xi_0 \) so that \( K(q) \) becomes \( K(q\xi_0) \). The asymptotic form of \( K(q) \) is given by \( K(q) = 1/\lambda L^2 \) for \( q=0 \); and \( K(q) = 3\pi/(4\xi_0 q\lambda L^2) \) for \( q\xi_0 \to \infty \), where \( \xi_0 \) is the size of Cooper pair, defined by \( \xi_0 = \hbar v_F /\{\pi\Delta(0)\} \) where \( v_F \) is the Fermi velocity.

When \( \xi_0 < < \lambda L \), the field variation over the distance of \( \xi_0 \) is negligible so that the average of the field inside a sphere of radius \( \xi_0 \) is simply the value at the center. It is named the local limit. The linear response is effectively weighted in the infinite wavelength limit, since \( K(q\xi_0) \) remains as \( K(0) \) in the range of \( q \) where the integrand in eq. (C-9) has significant values. From eq. (C-9), \( \lambda \) is equal to \( \lambda_L \).

The above result is only for a clean metal. The dirty limit is characterized by the short mean free-path \( \ell \) due to impurity scattering, so that \( \ell < \xi_0 \). The real-space expression of the non-local linear response is given by
\[ J(r) = -\frac{1}{\xi_0 \lambda_L^2(T)} \int \frac{R(r')}{R^4} J(R, T) dr', \]  
(C-10)

where \( R = r - r' \) and \( J(R, T) \) is the real-space kernel obtained by the Fourier transform of \( K(q, T) \). It has a similar form to \( \exp(-R/\xi_0) \) and its \( T \) dependence is very weak. The impurity effect is approximated by multiplying the kernel \( J(R, T) \) by the factor \( e^{-R/\ell} \).

When \( \ell < \xi_0 < \lambda_L \), it is in local limit; thus, \( \lambda \) is given by \( K(q=0)^{-1/2} \), but it is the modified kernel due to short \( \ell \). It is denoted by \( K(0, T, \ell) \). The effective penetration depth \( \lambda_{\text{eff}}(\ell, T) \) is given by

\[ \frac{\lambda_L^2(T)}{\lambda_{\text{eff}}^2(T)} = \frac{K(0, T, \ell)}{K(0, T, \infty)} = \int J(R, T) e^{-R\ell}/\xi_0 \, dR. \]  
(C-11)

The resultant form is

\[ \lambda(T) = \lambda_L(T) \{1 + \xi_0/\{J(0, T)\ell\}\}^{1/2}. \]  
(C-12)

\( J(0, T) \) is 1 at \( T=0 \) and 1.33 at \( T_c \). For dirty superconductors such as alloys, the local limit is a good approximation, and \( \lambda = \lambda_L(\xi_0/\ell)^{1/2} \).

When \( \xi_0 > \lambda_L \), it is called an extreme anomalous limit. In this limit, \( \xi_0 q \) is large in the range of \( q \) where the integration in eq. (C-9) is effective. \( K(q \xi_0) \) is approximated by \( 3\pi/(4\xi_0' q \lambda_L^2) \), where \( \xi_0' = \xi_0/J(0, T) \). The resultant form of eq. (C-9) is \( 0.58(\lambda_L^2 \xi_0')^{1/3} \). In most pure-element superconductors, \( \xi_0 > \lambda_L \); thus the extreme
anomalous limit fits better. Every superconductor is in the local limit near $T_c$ since $\lambda_L > \xi_0$. Thus there will be a change in $T$-dependence from one limit to the other when $\lambda_L = \xi_0$. The $T$-dependence of $\lambda$ in the entire range of temperature requires the full description of a microscopic model.
APPENDIX D

Numerical modeling in the two-coil method

Modeling of the apparatus can be described by classical electromagnetism, which requires the self-consistent solution of an integral equation for the shielding current density in the superconducting film. Since the displacement current $\varepsilon_0 \partial D/\partial t$ is negligible at low frequencies ($\omega = 10^6 \text{rad./s}$), Ampere's law, $\nabla \times B = \mu_0 J$ is a good approximation from which the vector potential $A(r)$ can be derived. The induced vector potential $A(r)$ due to $J(r)$ in film is

$$A(r) = \frac{\mu_0}{4\pi} \int \frac{J(r')}{|r-r'|} \, dr',$$  \hspace{1cm} (D-1)

where $r'$ is the coordinate in the film. The total vector potential in film is given by

$$A(r) = \frac{\mu_0}{4\pi} \int \frac{J(r')}{|r-r'|} \, dr' + A_{\text{ext}},$$  \hspace{1cm} (D-2)

where $A_{\text{ext}}$ is the contribution from $I_d$. The general expression for the screening current density $J(r)$ for a given $A(r)$ inside the superconductor is
\[ J(r) = \frac{1}{V} \int K(r,r') A(r') \, dr', \quad (D-3) \]

where \( V \) is the volume for normalization, and \( K \) is kernel of the integral.

Since the high-\( T_c \) superconductor is in the clean and local limits, it is reduced to the London equation

\[ J(r) = K(r) A(r) = \frac{1}{\mu_0 \lambda^2} A(r). \quad (D-4) \]

The combination of eq. (D-1) and (D-3) leads to the gauge-invariant form of the integral equation for the self-consistent solution \( J(r) \),

\[ \mu_0 \lambda^2 J(r) + \frac{\mu_0}{4\pi} \int \frac{J(r') \, dr'}{|r-r'|} = A_{\text{ext}}(r). \quad (D-5) \]

The first term represents the kinetic inductance, whose origin is mass inertia of superconducting electrons; the second term, the magnetic inductance, has purely electromagnetic origin and is usually several thousand times as large as the first term.

The slight current variation in the \( z \)-direction can be incorporated by using an effective thickness \( d_{\text{eff}} = \lambda \tanh(d/\lambda) \), to a good approximation, where \( d \) is film thickness. Effectively the current is regarded uniform throughout the thickness \( d_{\text{eff}} \), and zero elsewhere. The corresponding kinetic inductance of the sheet is modified to \( L_k = \mu_0 \lambda^2 / d_{\text{eff}} = \mu_0 \lambda \coth(d/\lambda). \) [59] When the film thickness is much smaller than \( \lambda \), the current variation in the \( z \)-direction is negligible, and the relation \( L_k = \mu_0 \lambda^2 / d \) is
recovered.

Under the assumption of a circular current distribution in film, the model has axial symmetry. There is only an azimuthal component of the vector potential. The axial symmetry further simplifies eq. (D-5) to an one dimensional integral equation. The vector potential distribution $A_\phi$ around a single annular ring carrying current $I_d$ has the expression

$$A_\phi = \frac{\mu_0 l_d}{\pi k} \left( \frac{b}{x} \right)^{1/2} \left[ \left( 1 - \frac{k^2}{2} \right) K(k) - E(k) \right], \quad (D-6)$$

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

$$k^2 = \frac{4br \sin \theta}{b^2 + r^2 + 2br \sin \theta} = \frac{4bx}{b^2 + x^2 + h^2 + 2hx} \quad (D-7)$$

in spherical and polar coordinates respectively. [93] In this equation, $x=r \sin \theta$, $h=r \cos \theta$, $b$ is the radius of ring, $r$ is the distance from the center of the ring, and $\theta$ is angle from the $z$-axis in spherical coordinates; $x$ is the distance from the $z$-axis and $h$ the height from the ring in polar coordinates.

We divide the disk film into concentric annular rings of width $\Delta x$ to obtain a numerical solution for $J(r)$. Then eq. (D-5) leads to the following matrix form:

$$L_k J_i + \Sigma_j a_{ij} J_j = A_{ext,i}, \quad (D-8)$$

where $J_i$ is the total current in $i$-th annular ring, and $A_{ext,i}$ is the vector potential at the $i$-th ring due to $I_d$. Here, $a_{ij}$ is the vector potential at $i$-th ring due to a unit current in
\( j \)-th ring, which is governed by the same form as eq. (D-6) with \( h=0 \). Extension of \( L_k \) to complex numbers is often required to incorporate the dissipation due to the screening current in the film. The phase of \( L_k \) and \( J \) is defined relative to \( A_{ext} \). By multiplying \((L_k I + a)^{-1}\) to left of both sides of eq. (D-8), \( J(r) \) is given by

\[
J = (L_k I + a)^{-1} A_{ext},
\]

(D-9)

where \( I \) is unit matrix. Fig. 2 displays \( J(r) \) and \( A_{ext}(r) \) in film.

Once \( J(r) \) is obtained, the net vector potential at any position can be obtained. The net magnetic flux in the pick-up coil is the sum of contributions from the drive coil, \( J(r) \) in the film, and an unknown amount of stray magnetic field. The mutual inductance \( M \) of the pick-up coil is given by

\[
M = \Sigma_i (2\pi r_i) A_{p,i}/I_d,
\]

(D-10)

where \( r_i \) is the radius of \( i \)-th turn of the pickup coil, and \( A_{p,i} \) is the vector potential at the same position. \( A_{p,i} \) can be divided into

\[
A_{p,i} = A_{stray,i} + I_d \Sigma_d a(z_d + z_i, r_i, r_d) + \Sigma_j a(z_i, r_i, r_j) J_j,
\]

(D-11)

where \( a(z, r_i, r_j) \) is the vector potential at \( r_i \) produced by a unit current loop of radius \( r_j \) at a distance \( z \) along the axis. The first term in right side of eq. (D-11) represents the
contribution from the stray coupling between wires attached to coils, and the last two terms are the vector potentials from the drive coil and from film, respectively. Here, \( z_d \) and \( z_i \) are the distances between the drive coil and film and the pick-up coil and film, respectively. Also \( r_d \) and \( r_i \) are radii of the drive and pick-up coils, respectively. The direction of \( J \) is opposite to that of \( I_d \) to account for the diamagnetism of superconducting film.

The mutual inductance \( M \) is calculated for a given \( \lambda \). In an experiment, \( M \) is measured as a function of temperature. After eliminating \( M \), the common variable, \( \lambda \) is obtained as a function of \( T \). However, since last two terms in eq. (D-11) almost cancel each other, \( A_{p,i} \) is several thousand times smaller than either terms. In practice, this causes complete uncertainty in estimating the location of the baseline of \( M \), since even small errors in estimation of either the second or third term in eq. (D-11) leads to a large error in the baseline of \( M \). This is the reason why this method provides a relative measurement. Only the change in \( M \) can be measured, and the location of the baseline is treated as a fitting parameter.
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