Microwave Studies of High Temperature Superconductors:

Extraction of Absolute Magnetic Penetration Depth of Cuprate Superconductors from Underdoped to Overdoped

by

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Abstract

The London Penetration depth, and complex conductivity of $Tl_2Ba_2Cu_3O_{6+\delta}$ and $YBa_2Cu_3O_{6+\delta}$ were measured at microwave frequencies using a loop gap resonator. A theoretical description of cavity perturbation analysis is given with emphasis assumptions of the model. The experimental apparatus is described, both in design and function, and a discussion is given of the difficulties associated with environmental conditions under which these measurements are made. The extraction of electronic properties from measured quantities is explained for the samples of $Tl_2Ba_2Cu_3O_{6+\delta}$ that go into the thin limit regime. These properties can be understood through a model which is based on incoherent quasipartical hopping between superconducting planes. Differences in the material properties between pure, and gadolinium doped, $YBa_2Cu_3O_{6+\delta}$ are accounted for by examining the properties of a dilute spin system. Measurements taken on this material are compared with results found in other microwave bolometry experiments, and found to be in qualitative agreement.
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Preface

Data presented here in Fig. 5.5, for the a-axis penetration depth of $YBa_2Cu_3O_{6+\delta}$ ($\delta = 0.992$) was taken in the course of this thesis, however it has previously appeared in [2] and has been reproduced with permission of the authors. Surface resistance measurements included in Fig. 5.3 represent unpublished work done by Patrick Turner at the UBC superconductivity laboratory, and Fig. 5.1 is a theoretical model provided by Tami Pereg-Barnea and Richard Harris.


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Chapter 1

Introduction

1.1 The puzzle of high temperature superconductivity

Much like the Gordian knot of condensed matter physics, a solution to the conundrum of high temperature superconductivity (HTSC) has not only eluded physicists for nearly twenty years, but still has scientists and engineers alike imagining the potential rewards of its future applications. Whether for uses in power distribution and storage systems, maglev trains, supercomputers, or for any of the vast insights it may bring to many body physics, the promise of HTSC seems almost endless.

At the core of the mystery of HTSC is that although the parent compounds have a half filled electron band structure that normally leads to metallic behavior, strong coulomb repulsion of electrons in the copper oxygen planes forces the otherwise conductive electrons to be localized on the copper atoms. By doping as little as \( \frac{1}{10} \) of these sites with holes, the electron order is perturbed enough that it becomes energetically favorable for the electrons to form a collective ground state with unusual d-wave pairing symmetry (this leads to an energy gap that crosses zero, and changes sign at certain points in momentum space [3] [4]). These odd phenomena have led to theories of HTSC that are as numerous at they are different. Whether the result of spin charge separation, some unusual pairing mechanism, or some phenomenon of competing ground states, the mechanism for HTSC is certain to involve novel and fascinating physics.
Chapter 1. Introduction

The various techniques and measurements made on HTSC samples since their discovery in 1986 have provided the field with a seemingly unlimited amount of data, so one might ask why has this particular enigma not been solved? One answer to this question lies in the slow evolution of material quality. It has been shown previously [5] [6] that the effects of small amounts of impurities or crystal defects can greatly overwhelm, and mask, the true physics behind the superconductivity. The new generations of crystals being produced every year, having sharper superconducting transitions (indicating higher homogeneity), higher purity and cleaner sample surfaces, will hopefully lead to measurements that will solve the underlying mysteries of HTSC.

In this thesis, measurements of the complex conductivity of superconducting samples were taken using the experimental technique of microwave cavity perturbation. A relatively low frequency of $\approx 1\, \text{GHz}$ was achieved using a loop gap resonator (similar to the split ring resonator in [7]) The two materials studied were $YBa_2Cu_3O_{6+\delta}$ (YBCO) and $Tl_2Ba_2Cu_3O_{6+\delta}$ (Tl2201). They belong to a class of materials known as cuprate superconductors, which all contain planes of $CuO_2$. The reason for examining these two materials was to access the properties of high temperature superconductors over a large portion of the superconducting region of the cuprate phase diagram given in Fig. 1.1. In YBCO, the absolute value of the London penetration depth was determined through a technique involving a crystal grown with gadolinium impurities, and oxygen doping of $\delta = 0.993$ (crystal grown in ortho-I chain ordering). To explore the region past optimal doping, the so-called overdoped side of the phase diagram, a different material was used, namely Tl2201. With recent advancements in understanding c-axis phenomenology by [8], and given that the current literature yields no consensus of the temperature dependent penetration depth for this axis, the task of taking detailed data, on newer generation materials, was undertaken.

To begin this thesis, an introduction to the electrodynamics in the HTSC materials will be given. From here it will be explained how the magnetic screening length of
Figure 1.1: The cuprate phase diagram. The d-wave superconducting region (dSC) is of central interest to this thesis, both on the underdoped (UD), and overdoped (OD) sides. The parent phase here is antiferromagnetic (AFM), and the normal state metallic (NM). A region where it appears the superconducting order parameter is not destroyed, but merely undergoes phase decoherence [1] is shown as the pseudo gap (PG) area.

a material can be related to conductivity. Chapter 2 is mainly concerned with the extraction of the true screening length from the experimentally measured quantity. This requires the formalism of cavity perturbation which in turn requires taking explicit account of the sample geometry. The relationship of screening length to the surface resistance of samples is also explored in chapter 2 as another means of characterizing sample properties. In Chapter 3 the experimental methods used to perform the perturbation measurements are discussed with some emphasis placed on the limitations of the technique to compensate for environmental conditions such as inhomogeneous field distributions, and thermal contractions at low temperature. Finally in chapters 4 and 5, measurements on Tl2201 and YBCO are analyzed and their significance to the field of HTSC is discussed.
1.2 Microwave conductivity of cuprates

A useful concept in the description of the frequency response of superconductors is the two fluid model. It assumes that there are essentially two different species of carrier inside the superconductor. The origin of this model can be traced back to arguments used in the understanding of superfluid helium [9].

In treating the AC response of a superconductor [10] using a two fluid model, one can use the familiar Drude [11] form for the conductivity for both normal carriers and superconducting carriers. Although the Drude model does not exactly describe the proper electrodynamics of cuprate superconductivity [2] [3], it does capture some of the essential phenomenology of the quasiparticle spectrum, namely that there exists a peak in the real part of the conductivity with a width characteristic of some lifetime. The Drude conductivity is given by:

\[
\sigma(\omega) = \sigma_1(\omega) - i\sigma_2(\omega) = \frac{ne^2\tau}{m^*(1 + i\omega\tau)}
\]  

(1.1)

In Eq. 1.1, \(\sigma(\omega)\) is the conductivity as a function of the frequency \(\omega\); \(n\) the carrier density, \(e\) and \(m^*\) are the carrier charge and effective mass respectively and \(\tau\) is its lifetime. By decomposing Eq. 1.1 into real and imaginary components, \(\sigma_1\) and \(\sigma_2\), one can examine the conductivity of the normal carriers.

\[
\sigma_1 = \frac{n_ne^2\tau_n}{m^*(1 + \omega^2\tau_n^2)}
\]  

(1.2)

\[
\sigma_2 = \frac{n_ne^2\omega\tau_n^2}{m^*(1 + \omega^2\tau_n^2)}
\]  

(1.3)

The superfluid conductivity has a similar initial form, but here one sets \(\tau_s \rightarrow \infty\). Then \(\sigma_1\) collapses to a delta function at the origin, whose weight is a measure of the superfluid density [12]:

\[
\sigma_1 = \frac{\pi n_se^2\delta(\omega)}{2m^*}
\]  

(1.4)
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\[ \sigma_{2s} = \frac{n_s e^2}{m^* \omega}. \] (1.5)

It can be noted that an oscillator strength sum rule applies to the frequency integrated area of \( \sigma_{1n} \), such that the total integrated weight of \( \sigma_1 \) is temperature independent. In the microwave frequency region, \( \omega \tau_n \ll 1 \) and for \( T < T_c \), it is found that \( \sigma_{2s} \gg \sigma_{2n} \) and in general \( \sigma_2 \gg \sigma_1 \). These dominant contributions at low frequency are then added to determine the complex conductivity of the superconductor:

\[ \sigma_1 = \frac{\pi n_s e^2 \delta(\omega)}{2m^*} + \frac{n_n e^2 \tau_n}{m^*} \] (1.6)

\[ \sigma_2 = \frac{n_s e^2}{m^* \omega} \] (1.7)

In the normal state it should be noted that there are contributions to conductivity from only the normal carriers, since \( n_s \rightarrow 0 \), and so \( \sigma_2 = \frac{n_n e^2 \omega \tau_n^2}{m(1+\omega^2 \tau_n^2)} \).

Through the temperature and frequency dependence of both the real and imaginary components of the conductivity of the cuprates, fundamental mechanisms can be probed experimentally. Theories of HTSC most often attempt to describe the low temperature behavior of this conductivity. As seen above in Eq. 1.7 the imaginary part of the conductivity depends solely on \( \frac{n_s e^2}{m^*} \). This is the central quantity of interest in this thesis, as its temperature dependence can place constraints on the superconducting energy gap (see [4]).

1.3 Screening lengths

To understand how the conductivity of a material can be obtained experimentally, it is worthwhile to first consider the magnetic screening length in an arbitrary material. One begins with Ampere’s law,
\[ \nabla \times \vec{H} = \vec{J} + \frac{\partial \vec{D}}{\partial t} \]  

(1.8)

where contributions to current density are made by both the normal and superconducting carriers:

\[ \vec{J} = \vec{J}_n + \vec{J}_s. \]  

(1.9)

London’s equation (see [10]) can then be used to set \( \vec{J}_s \):

\[ \vec{E} = \frac{m^* \partial \vec{J}_s}{ne^2 \partial t}. \]  

(1.10)

Assuming \( \vec{J}_s \) has time dependence \( e^{i\omega t} \), and rearranging Eq. 1.10 one finds:

\[ \vec{J}_s = -\frac{in_0 e^2}{m^* \omega} \vec{E}. \]  

(1.11)

Ignoring for the moment the zero frequency superfluid contribution to \( \sigma_1 \) found in the previous section, it is reasonable to assume that the current contribution from the normal fluid will take the form:

\[ \vec{J}_n = \sigma_n \vec{E}. \]  

(1.12)

Using the same time dependence as \( \vec{J}_s \) for \( \vec{D} \) and substituting Eq. 1.11 and Eq. 1.12 into Eq. 1.9 and Eq. 1.8, it is easy to derive:

\[ \nabla \times \vec{H} = (\sigma_n - \frac{in_0 e^2}{m^* \omega} + i\epsilon \omega) \vec{E}. \]  

(1.13)

Taking the curl of Eq. 1.13, the left side becomes:

\[ \nabla \times \nabla \times \vec{H} = \nabla (\nabla \cdot \vec{H}) - \nabla^2 \vec{H} = -\nabla^2 \vec{H} \]  

(1.14)
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Where \( \vec{V} \cdot \vec{H} \approx \vec{V} \cdot \vec{B} = 0 \) has been taken to be zero (an assumption valid only if no magnetic material is present, or if demagnetization effects are negligible, which is the case of interest here). The right side can be related to the magnetic field through the use of Faraday's equation:

\[
\vec{V} \times \vec{E} = -\frac{\partial \vec{B}}{\partial t} = -i\omega \mu \vec{H}.
\]  

(1.15)

Using Eq. 1.14 and Eq. 1.15 the following general screening equation can be derived:

\[
\nabla^2 \vec{H} = (i\mu \omega \sigma_n + \frac{\mu n_s e^2}{m^*} - \epsilon_r \epsilon_0 \omega^2) \vec{H} = \frac{1}{\delta^2} \vec{H},
\]  

(1.16)

where \( \delta \) is the material's screening length. Below the transition temperature, and in the low frequency limit, it is usually possible to neglect the effects of displacement currents (third term in the right side of Eq. 1.16), in superconductors. Above the transition temperature \( \delta \propto \left(\frac{n_s}{n_{\text{sat}}}\right)^{1/2} \). In the limit \( \omega \tau_n \ll 1 \) this saturation value of \( \delta \) can be used as a diagnostic tool for confirming measurement accuracy (this will be explored further in chapter 4).

In the case of a normal metal there is no contribution in Eq. 1.16 from superconducting charge carriers \( (n_s = 0) \), so an appropriate length scale for screening is given by \( \delta_{\text{skin}}(\frac{1}{2}) \) where the skin depth, \( \delta_{\text{skin}} \), is given by:

\[
\delta_{\text{skin}} = \frac{2}{\mu_0 \omega \sigma_n}.
\]  

(1.17)

For a superconductor, the contribution from the supercurrent usually overwhelms any other contributions to the screening length for all temperatures \( T < T_c \), except very near to the transition. It was found that \( \frac{n_s e^2}{m^*} \gg \omega \sigma_n \) for all frequencies measured in this report. The special name given to this screening length in the superconducting state is the London penetration depth, \( \lambda \), which is defined by:

\[
\lambda = \sqrt{\frac{m^*}{\mu n_s e^2}} = \sqrt{\frac{1}{\mu \omega \sigma_2}}.
\]  

(1.18)
Chapter 1. Introduction

It is however still necessary to consider the entire complex screening length in superconductors when looking to probe both the real and imaginary components of the conductivity. This topic will play an important role in the discussion throughout the chapters to follow.

Here one must consider how the true screening length can be extracted from the experimentally accessed quantity by taking into account the effects of sample geometry. For this one needs to understand the response of a sample to an alternating magnetic field in the situation where screening currents play a central role. This will be discussed in the next chapter under the general category of cavity perturbation.
Chapter 2

Cavity Perturbation

2.1 First principles

By introducing a small sample into an empty cavity resonator, a small change in the complex resonant frequency is induced (provided the sample does not occupy a large volume of the resonator).

\[
\frac{\Delta \tilde{\omega}_{e\rightarrow s}}{\tilde{\omega}} \approx \frac{\Delta f}{f} = \frac{\Delta f_0}{f_0} + \frac{i \Delta f_B}{2f_0}. \tag{2.1}
\]

Here \(\Delta f_0\) is the shift in the real part of the frequency of the resonator from its value when empty and \(f_B\) is the resonant bandwidth (width at half power points). In terms of the resonator \(Q\), which is the ratio of power stored in the resonator to the power lost in one cycle, one has

\[
\frac{1}{Q} = \frac{f_B}{f_0}. \tag{2.2}
\]

To describe the fields before a sample is loaded into the resonator one assumes \(\tilde{E} = \tilde{E}_0 e^{i\omega t}\) and \(\tilde{H} = \tilde{H}_0 e^{i\omega t}\), whereas afterward \(\tilde{E} = (\tilde{E}_0 + \tilde{E}_1)e^{i(\omega + \Delta \omega_{e\rightarrow s})t}\) and \(\tilde{H} = (\tilde{H}_0 + \tilde{H}_1)e^{i(\omega + \Delta \omega_{e\rightarrow s})t}\). Here \(\tilde{E}_0, \tilde{E}_1, \tilde{H}_0, \tilde{H}_1\) are all spatially varying fields. With the use of a few relatively straightforward vector identities [13] one can derive the following perturbative expression for a large change in the fields over a small volume \(V_s\):

\[
\frac{\Delta \tilde{\omega}_{e\rightarrow s}}{\tilde{\omega}} = \frac{\iiint_{V_s}((\tilde{E}_1 \cdot \tilde{D}_0 - \tilde{E}_0 \cdot \tilde{D}_1) - (\tilde{H}_1 \cdot \tilde{B}_0 - \tilde{H}_0 \cdot \tilde{B}_1)) dv}{\iiint_{V_s}(\tilde{E}_0 \cdot (\tilde{D}_0 + \tilde{D}_1) - \tilde{H}_0 \cdot (\tilde{B}_0 + \tilde{B}_1)) dv} \tag{2.3}
\]
Chapter 2. Cavity Perturbation

It is then assumed that \( E_1 \) and \( H_1 \) can be neglected in the integral over the whole volume of the cavity \( V_c \) appearing in the denominator of equation Eq. 2.3 which then simplifies to just \( 4\nu \), where \( \nu \) is the energy stored in the unperturbed cavity. To define \( D_1 \) and \( B_1 \) inside the cavity it is useful to use the identities:

\[
D_1 = \epsilon_0 (\epsilon E_0 + \tilde{E}_1) - \tilde{E}_0 = \epsilon_0 \tilde{E}_1 + \tilde{P}
\]

(2.4)

\[
\vec{B}_0 + \vec{B}_1 = \mu_0 (\vec{H}_0 + \vec{H}_1 + \vec{M}).
\]

(2.5)

From here it can be seen that the complex frequency shift is given by:

\[
\frac{\Delta \tilde{\omega}_{c-s}}{\tilde{\omega}} = \frac{1}{4\nu} \iiint_{V_s} (\vec{P} \cdot \tilde{E}_0 + \vec{M} \cdot \vec{B}_0) d^3 r.
\]

(2.6)

This expression will simplify if the sample is placed in a geometry where it only experiences a magnetic field maximum (it will be shown in chapter 2 that the ratio of the stored electric energy to the stored magnetic energy \( \approx 0.04\% \)). The electric field perturbation is then negligible (except in the case of depolarizing effects which will be covered in chapter 4), so that the first term in the above integral can be dropped. Using the definition of \( \vec{M} \) given in Eq. 2.5, and the assumption that the magnetic field strength does not change much over the volume of the sample (i.e. that \( \vec{H}_0 + \vec{H}_1 \approx \vec{H}_s \), where \( \vec{H}_s \) is the field at the sample surface) one arrives at:

\[
\frac{\Delta \tilde{\omega}_{c-s}}{\tilde{\omega}} = \frac{H_s B_0 V_s}{4\nu} \iiint_{V_s} \vec{H}_s \cdot (\vec{B}_0 + \vec{B}_1) = \frac{\Delta \tilde{\omega}_{c-p}}{\tilde{\omega}} + \frac{\Delta \tilde{\omega}_{p-s}}{\tilde{\omega}}.
\]

(2.7)

In the above expression, the term \( \frac{H_s B_0 V_s}{4\nu} \) is the shift in frequency between an empty cavity and one with perfect screening, and is labelled by \( \Delta \omega_{c-p} \).

Examining now the change induced in resonator frequency between a cavity with a perfectly screened sample, and a real material of the same shape, given in Eq. 2.7 by \( \Delta \omega_{p-s} \), the analysis can be simplified. The relevant change in frequency now becomes:
The altered $\vec{B}'_1$ now represents the total field inside the real sample, because $\vec{B}'_0 = 0$ in a perfect diamagnetic sample. In Fig. 2.1 one can see the changes produced by a sample with (b) infinite conductivity, and one with finite conductivity (c) in the magnetic field of an empty cavity (a).
2.2 Solving for field distribution in a screened sample

If the sample is to be thought of as a perturbation on the cavity fields from a state in which the sample is lossless to one where real absorption occurs it is necessary to find the fields inside the imperfect sample. The general form of the screening equation derived in the previous chapter is equally valid for a normal metal and for a superconductor. In fact this analysis will work for most materials in which screening of a magnetic field is seen and, for example, can apply to materials with diffuse magnetic spins, where account must be made for changes in magnetic susceptibility via \( \mu_0 \rightarrow \mu = \mu_0 (1 + \chi) \). Simplifications given in the present treatment of the technique will inevitably break down at higher frequencies as carrier lifetimes become comparable to \( \frac{1}{\gamma} \), and in materials that experience the anomalous skin effect, when the mean free path of carriers becomes comparable to the skin depth.

For a thin platelet of thickness \( c \) where the field is applied parallel to the flat faces and one can treat the platelet as an infinite slab, the general screening equation given in Eq. 1.16 can be reduced to the following one-dimensional case.

\[
\frac{\delta^2}{\delta^2 x} \tilde{B} = \tilde{B}
\]  

(2.9)

The general solution of Eq. 2.9 within the material is:

\[
\tilde{B} = \tilde{C}_1 e^{ikx} + \tilde{C}_2 e^{-ikx}
\]  

(2.10)

The propagation constant within the sample is given by \( \kappa \), and through the screening equation it is possible to verify that \( \kappa = \frac{i}{\gamma} \). Here \( \frac{c}{2} < x < \frac{c}{2} \) as in Fig. 2.2, where \( \tilde{B} \) for the real sample is shown to take the form of a field decaying inward from the sample faces. The coefficients \( \tilde{C}_1 \) and \( \tilde{C}_2 \) are then found to be equal by a simple symmetry argument.
Figure 2.2: The magnitude of field inside of a perfect, and a screened sample as compared with an empty cavity. Notice the slight enhancement of field outside of the sample where flux has been dispelled from the body of the material.

By imposing a boundary condition such that $\vec{B}(\pm \frac{c}{2}) = \mu_0 \vec{H}_s$, where $H_s$ is the field strength at the surface of the sample, the solution to Eq. 2.9 can be expressed:

$$\vec{B} = \mu_0 \vec{H}_s \frac{(e^{i\kappa x} + e^{-i\kappa x})}{(e^{i\frac{\kappa x}{2}} + e^{-i\frac{\kappa x}{2}})}.$$  \hspace{1cm} (2.11)

Inserting Eq. 2.11 into Eq. 2.8 the shift in resonator frequency is given by:

$$\frac{\Delta \omega_{p-s}}{\omega} = \frac{\mu_0 H_s^2}{4\nu} \frac{L_y L_z}{(e^{i\frac{\kappa x}{2}} + e^{-i\frac{\kappa x}{2}})} \int_{-\frac{c}{2}}^{\frac{c}{2}} (e^{i\kappa x} + e^{-i\kappa x}) dx. \hspace{1cm} (2.12)$$

From here, a straightforward integration yields the result:

$$\frac{\Delta \omega_{p-s}}{\omega} = 2i\mu_0 G A_{eff} \delta \tanh \left( \frac{c}{2\delta} \right). \hspace{1cm} (2.13)$$

The resonator constant $G = \frac{H_s^2}{4\nu}$ can be found by a calibration procedure using a sample of known resistivity. This takes into account the relative field strength at the
sample surface. If the reference sample is of comparable size to the superconducting samples, this calibration technique will also reduce the effect of any non-perturbative corrections. In any experiment involving large changes in temperature care should be taken to correct for expansion and contraction in both $c$ and the effective area $A_{\text{eff}} = L_x L_y$. Using the expressions for screening lengths in both metals and superconductors, given in Eq. 1.17 and Eq. 1.18, it is now possible to define the complex frequency shift in terms of the complex penetration depth.

2.3 A different perspective; considering surface impedance

It is also useful to consider the relation between the complex frequency shift and the surface impedance $Z_s$. Through this approach, the complex change in frequency is looked at from the point of view of energy absorption. To do this, one must first examine effects near the edge of the crystal, i.e. how flux expulsion from within the sample effectively modifies flux distribution $\vec{H}$ around the sample. The common treatment of these effects is expressed in terms of demagnetization factors, $N_m$, which are most easily defined for ellipsoidal samples. Following [14] and [15] the relation of $\vec{H}$ in the sample to the unperturbed $\vec{H}_0$ (relative permeability, $\mu_r$, is also required) is found to be:

$$\vec{H} = \frac{\vec{H}_0}{1 + (\mu_r - 1)N_m}. \quad (2.14)$$

When platelets are exposed to a tangential magnetic field the limiting ellipsoid becomes flat in the plane of the sample, and demagnetization is minimized, i.e. $N_m \rightarrow 0$. Were the field orientation out of the plane, maximum demagnetization would be incurred, and $N_m \rightarrow 1$. In [16] the author has shown that calculations can be performed numerically for square and rectangular samples, however the ability to make corrections for this effect in the following experiments is limited by uncertainty in the resolution of the sample dimensions. It is useful here to examine only the
limiting case where demagnetizing effects are negligible.

The surface impedance can be expressed as the ratio of the tangential surface electric field to the magnetic field.

\[ Z_s(r_s)H_t(r_s) = \bar{n}(r_s) \times \vec{E}_t(r_s) \]  \hspace{1cm} (2.15)

Using the method of [17] and [14] it is possible to show that the complex frequency shift can be related to the generalized power absorbed in the sample, as obtained from a simple integration of the Poynting vector over the sample surface:

\[ \frac{\Delta \tilde{\omega}_{p-s}}{\tilde{\omega}} = \frac{1}{4\mu_0} \iint_S \vec{n} \cdot (\vec{E}_t(r) \times \vec{H}_t(r)) dr^2. \]  \hspace{1cm} (2.16)

The quantity \( \vec{n} \cdot (\vec{E}_t(r) \times \vec{H}_t(r)) \) is equal to \( \vec{H}_t(r) \cdot (\vec{n} \times \vec{E}_t(r)) \) since they both represent the volume of the same parallelepiped. Given that the integration of Eq. 2.16 is only performed over the surface of the sample, Eq. 2.15 will hold and the integral can be expressed as follows, where it is assumed that the fields at the sample surface are relatively parallel to the face, so that \( \vec{H}_s = \vec{H}_t \):

\[ \frac{\Delta \tilde{\omega}_{p-s}}{\tilde{\omega}} = \frac{1}{4\mu_0} \int_S H_t(r)^2 Z_s(r) dr^2. \]  \hspace{1cm} (2.17)

Comparing to Eq. 2.13 one obtains the result:

\[ \frac{\Delta \tilde{\omega}_{p-s}}{\tilde{\omega}} = \frac{2iH_s^2 A_{eff} Z_s}{4\mu_0 G A_{eff} \delta \tanh \left( \frac{c}{2\delta} \right)}. \]  \hspace{1cm} (2.18)

An extra factor of 2 in the expression involving \( Z_s \) comes about because the effective areas to consider in the integral of Eq. 2.17, contains both the top and bottom of the sample. At this point it is useful to make a change in variables from \( \frac{\Delta \tilde{\omega}_{p-s}}{\tilde{\omega}} \) to \( \Delta \delta_{eff} \), an effective screening length:

\[ \Delta \delta_{eff} = \frac{1}{\mu_0 G A_{eff}} \left( \frac{\Delta f}{f} + i \Delta \left( \frac{1}{2Q} \right) \right). \]  \hspace{1cm} (2.19)
Where the relation of $\delta_{eff}$ to the true screening length is thus given by:

$$\delta_{eff} = \tilde{\delta} \tanh\left(\frac{c}{2\delta}\right). \quad (2.20)$$

It is possible, using Eq. 1.16, to relate the screening length to the conductivity (in the limit where displacement currents are insignificant):

$$\tilde{\sigma} = \sigma_1 - i\sigma_2 = \frac{1}{i\mu_0\omega\delta^2}. \quad (2.21)$$

Defining the real and imaginary components of the surface impedance as the surface resistance $R_s$ and surface reactance $X_s$ one has:

$$Z_s = R_s + iX_s = \mu_0\omega\tilde{\delta} \tanh\left(\frac{d}{2\delta}\right). \quad (2.22)$$

Making the approximation that the real component of the screening length, given by the London penetration depth $\lambda$, is much shorter than the imaginary contribution of the normal carriers for the case of a superconductor, it is possible to work backwards and find the surface resistance $R_s$, and the surface reactance $X_s$ in the limit of $\sigma_2 \gg \sigma_1$ as:

$$X_s \approx \mu_0\omega\lambda \quad (2.23)$$

$$R_s \approx \frac{\mu_0^2\omega^2\lambda^3\sigma_1}{2}. \quad (2.24)$$

These relations will only hold in what’s commonly referred to as the thick limit, where $\tanh\left(\frac{c}{2\delta}\right) \approx 1$, as only in such situations are $R_s$ and $Z_s$ independent of sample thickness.
2.4 Extending to two dimensions

One must be very careful in choosing $A_{\text{eff}}$, for in either isotropic, or anisotropic samples both axes for which the current runs will contribute to the overall frequency shift. For the high temperature superconductors, samples with appropriate aspect ratios can sometimes be chosen so that one set of the sample faces contributes to a greater extent than the others. However, in many situations it is extremely difficult to produce a crystal where the contribution to $R_s$ or $X_s$ from one axis dominates. For crystals with geometries where the aspect ratio mixes in-plane and out-of-plane contributions it is often necessary to solve the two dimensional screening equation:

$$\tilde{\delta}_{\text{eff}} \frac{\partial^2 \tilde{H}}{\partial x^2} + \delta_{\text{a}} \frac{\partial^2 \tilde{H}}{\partial y^2} = \tilde{H}$$

(2.25)

Using fourier series, one obtains [18]:

$$\tilde{\delta}_{\text{eff}} = \frac{ac}{a + 2\pi^2 c} \sum_{n>0} \frac{1}{n^2} \left( \frac{\tanh(\alpha_n)}{\alpha_n} + \frac{\tanh(\beta_n)}{\beta_n} \right) \tag{2.26}$$

$$\alpha_n = \frac{c}{2\delta_c} \sqrt{1 + \left( \frac{n\pi\delta_c}{a} \right)^2}$$

(2.27)

$$\beta_n = \frac{a}{2\delta_c} \sqrt{1 + \left( \frac{n\pi\delta_c}{a} \right)^2}$$

(2.28)

This solution is not used in the current thesis, but is included for completeness.

In the thick limit, where $\delta_c \ll a$ and $\delta_a \ll c$ a linear approximation can be made to Eq. 2.26, such that:

$$\lambda = \frac{1}{a + c} (a\lambda_{ab} + c\lambda_c) \approx \lambda_{ab} + \frac{c}{a} \lambda_c.$$  \tag{2.29}

Due to their growth characteristics, and magnitudes of $\lambda$, optimally and overdoped YBCO samples typically fall under this condition.
Figure 2.3: The currents induced in a screened sample are contained within a screening length of the surface. Here a sample is shown with an anisotropic screening length, typical of what is observed in HTSC materials.
Chapter 3

Experimental apparatus

3.1 Construction and design

Through the use of a loop gap resonator one can implement the field perturbation techniques derived in the previous chapter. As implied by its name, this apparatus, pictured in Figs. 3.1 and 3.2, mainly consists of a single turn of copper, electro-plated with a lead-tin \((95\% Pb : 5\% Sn)\) superconducting film, and a gap in which a 0.05mm plate of chemically pure sapphire (Crystal Systems HEMEX grade) is placed. Samples mounted within this device are exposed to a relatively uniform oscillating magnetic field applied parallel to their flat platelet faces. The principles behind and technical difficulties associated with running a resonator of this variety are similar to those addressed in [7], where the resonator instead has a split ring configuration.

The characteristics of a loop-gap device can be modelled as a simple LRC resonant circuit. The inductance and capacitance for the circuit can be roughly approximated by a single loop and a parallel plate (with sapphire dielectric), while a resistance can be traced to the absorption losses in sample and the plating of the cavity walls and some dissipation across the gap. The experiment consists of measuring the frequency and \(Q\) of the resonator as a function of the sample temperature as it is varied between \(1K \rightarrow 250K\) (the resonator is attached to a probe which can be placed in a pumped liquid helium bath). While losses associated with the resonator itself remain relatively constant, and stable over the course of an experiment, sample losses will vary strongly with temperature. Shifts in the complex resonance can then be related to sample power absorption via the Poynting vector, as discussed in the previous chapter. Slight shifts in the base frequency and \(Q\) due to thermal drift of the apparatus can be
accounted for by making regular checks at the base temperature.

The main reason for utilizing such a setup, over other more standard resonators, is that to operate at 1 GHz in a standard $TE_{011}$ mode, a 40cm diameter cylindrical cavity resonator would be required. Although such a cylindrical resonator would have a substantially higher base Q, the cavity filling factor would be extremely low for samples with typical dimensions of about 1mm. Another more practical reason for utilizing the smaller loop gap resonator is that the cost to cool to liquid helium temperature is much lower than for the 40cm diameter device.

![Resonant loop gap apparatus diagram](image)

**Figure 3.1:** The resonant loop gap apparatus in cross section. Samples are inserted into the loop (shown in detail in Fig. 3.2 on a sapphire mounting plate. The small coupling loops are located at the end of coax line on both sides of the resonator.
There are many important steps in preparing a successful resonator. The $Q$ value should not vary strongly with power coupled into the resonator. This can be achieved by ensuring that the lead tin is evenly plated over the surface of the resonator, paying particular attention to the regions where the upper loop contacts the base of the resonator. If there is a weak junction, current flowing within the Pb:Sn can be comparable to the critical current of the link, and will drive parts of the Pb:Sn film normal. Care should be taken during the plating process to keep the plating electrode in constant motion, otherwise dendritic growth may occur. The resulting uneven current and field distribution within the resonator will lead to a power dependence of the $Q$. Faulty plating may also lead to shortened film lifetime due to peeling or oxidation.

Although there is $\approx 5\%$ tin in the plating mixture, the resonator must also be quickly dried with nitrogen gas following plating to minimize any oxidation of the lead.
It should then be immediately assembled and evacuated to about ($\approx 10^{-6} \rightarrow 10^{-7}$ torr) to avoid deterioration of the film by water vapor. Any oxidation or incomplete Pb:Sn coverage of the resonator will result in a lower $Q$ for the resonator, which lowers the sensitivity and makes measurement of small crystals with low surface resistance problematic.

To prevent any rf power from being dissipated outside the region of the resonant loop-gap, all ports into the cavity are made to be much less than a quarter of the free space wavelength at the resonant frequency. Through this precaution, waves decay evanescently in the holes instead of propagating to other parts of the device.

**3.1.1 Electric field estimation**

It is a simple matter to check the validity of the assumption made in the previous chapter, that any contribution to the resonator frequency shift from electric field perturbation is negligible. A simple comparison of energy density of the electric and magnetic fields within the sample volume can be used to show that for maximum field expulsion the perturbation of the electric field is insignificant when compared to the magnetic field perturbation.

The inside of the resonator can be thought of as one side of a long rectangular waveguide, where one of the sides has been shorted by a capacitor plate. Both electric and magnetic field distributions are shown in Fig. 3.3. The largest electric fields are contained within the sapphire plate. There is a large reduction in $\vec{E}$ as one moves away from the gap, though it still radiates out to the central region of the loop. Finally, $\vec{E}$ decays to zero as one approaches the end wall of the loop.

Using Eq. 1.15, and taking an integral over the surface $S$ contained by contour $C$, it is a simple matter to derive the following:

$$\int \int_A \vec{\nabla} \times \vec{E}_0 = \omega B_0 xd. \quad (3.1)$$
Decay of $\vec{E}$ across the width of the cavity is portrayed through thinning field lines. The area of contour $C$ is given by $xd$.

Through Stoke’s theorem, this surface integral can be converted to a contour integral. Due to boundary conditions at the superconducting Pb:Sn surface, the electric field is zero at the left wall of the resonator, closest to the sample, and at the top and bottom of the contour. Over the one remaining path length $E_0$ can be roughly approximated as a constant, and therefore one finds:

$$\int \vec{E}_0 \cdot d\vec{l} = E_0d = \omega B_0 xd. \quad (3.2)$$

Dividing Eq. 3.2 by the height of the loop-gap, $d$, it is found that the magnitude of $\vec{E}_0$ varies linearly with $x$, (i.e. $E_0 = \omega B_0 x$). As a rough estimate of the contribution to $\frac{d\omega}{d\omega}$ from $\vec{E}$, a comparison can be made of the energy density of the electric field, $\frac{1}{2}\epsilon E^2$, to the energy density of the magnetic field $\frac{1}{2}\mu H^2$. 

Figure 3.3: The resonator’s magnetic (top, red) and electric (bottom, blue) fields.
Since the uncertainty of the measurement is not less than 0.04%, it usually unnecessary to include the effects of the electric field perturbation. However, a resonant depolarization effect (covered in chapter 4) does occur near the transition temperature of some superconducting samples depending on their size, shape, and superfluid density.

3.2 Operation

3.2.1 Detection methods

To both supply rf power, and access the resonant frequency of the device, the resonator is coupled inductively through small ports on either side of the loop-gap circuit (see Fig 3.1). Microwave signal is sent from a HP 83620 A synthesized sweeper into the cavity through a transmission loop which has been undercoupled to avoid any substantial reduction in the Q of the resonator; weak input coupling can be compensated for by increasing the incident power. This also reduces pulling of the frequency by the coupling. Due to geometric constraints of the present design it is in fact difficult to overcouple the device. The output coupling should also not be too far undercoupled, since one could end up in the situation where the field levels in the resonator are very high, but the signal coupled out is weak.

A 33db Avantek preamplifier is used to boost the sampled signal, and off-resonance noise is eliminated through the use of a variable frequency band pass filter. After filtering, the signal is then amplified a further 20 db. An HP 8473 crystal detector converts the rf signal to a dc voltage, which is then amplified by about 1000 x. A low pass filter is inserted after the detector, as shown in Fig. 3.4, to filter out any remaining microwave signal. The analog to digital conversion is then performed, and the output is sent via a GPIB cable to a data acquisition card on a computer.
Figure 3.4: This is a figure of the detection circuit for the experiment. Dashed lines represent GPIB connections.

Alternatively, rf transmission can be measured using a vector network analyzer. The vector network analyzer has the advantage of being a phase sensitive detector, which allows fits of the resonance in both amplitude and phase space. However its rf source is not as stable as the HP synthesizer used in the crystal detector setup.

The synthesizer is under computer control, both through GPIB connection, and an external triggering input. By fitting the output signal as a function of frequency to a Lorentzian lineshape, as one does for any harmonic oscillator, it is possible to determine the complex resonant frequency of the circuit.

Time domain methods

Microphonics can alter the central frequency of the loop gap resonator. For small Lorentzian bandwidths, sweeps over frequency can be badly distorted by microphon-
ics’ modulation of the resonant frequency. In this case, $Q$ is better measured by applying a pulse of power to the resonator at its resonant frequency and watching the time decay of the outcoupled power. A Tektronix TDS 520B averaging oscilloscope is utilized for data acquisition in this time domain approach to finding the cavity $Q$. It is found to be a much more stable and repeatable method for the measurement of $R_s$. The decay of energy in the cavity is insensitive to slight shifts in the resonance frequency of the cavity, whereas a fit of a Lorentzian lineshape to a frequency sweep will always yield a $Q$ that is lower than the actual $Q$.

The measurement of shifts in $Q$ might also be improved by designing future versions of this experiment to allow for the sample to be unloaded from the cavity, and reloaded, in-situ. In this way the absolute value of $R_s$ can determined without having to rely on data obtained from other experiments such as microwave bolometry.

**Robinson phase locked oscillator**

A Robinson oscillator circuit (principles of which are originally outlined in [19]) can also be used as a detector that is extremely sensitive to the real component of the resonator frequency. As seen in Fig. 3.5, the Robinson oscillator is comprised of various external room temperature electronic devices working to sustain resonant power storage in the loop-gap. Noise within the circuit will initially cause the resonator to lock into its resonant mode. This signal is amplified, and then pumped back into the resonator. Between the three amplifiers shown in fig. 3.5 a total of 79db of gain is used. A limiter is used instead of relying on the signal compression in the amplifiers to provide amplitude stability of the signal. The rf signal is sampled through a directional coupler then sent through a RHG double balanced mixer and mixed down to a frequency approximately 100kHz off of resonance. The attenuator and weak input coupling are used to keep the stored energy in the resonator at an appropriate value.
Figure 3.5: Robinson Oscillator detection circuit. The feedback loop is comprised of all elements located above the mixer on the above diagram. All GPIB connections are dashed, while coaxial connections are represented by the continuous black lines.

Adjusting the phase shifter allows one to set the oscillator frequency to the center of the loop gap resonance. The resonance signal is mixed down against the off resonance input, and a frequency counter can then be used to measure this difference in frequency. The counter is connected to a computer via GPIB cable, and by time averaging for 10s a frequency stability of $< 1Hz$ can usually be achieved. Through this method, the shift of the London penetration depth from its value at some base temperature can be performed to a level of precision generally higher than the frequency sweep method described in the preceding section.

### 3.2.2 Power dependence

While running the split ring resonator, frequent checks of the power dependence of $Q$ should be made. At the outset of the measurement, a decade of input power can
usually be chosen such that the value of the base (at \(T=1.2\text{K}\)) \(Q\) changes by no more than 10\%, while a given sample is in the superconducting state (see Fig. 3.6). Any power dissipated inside the resonator is done so in both the cavity walls and the sample. Through changing sample temperature, and hence sample absorption, current running in the resonator walls is altered slightly. If there were power dependence, this induces a change in the resonator characteristics which invalidates the assumptions of the cavity perturbation method.

![Figure 3.6: Plot of resonator Q with changing input power. Notice in the resonator with higher Q there is little variation in Q over the decade from -40dBm to -50dBm. Similar plots should also be made whenever the input or output coupling is changed.](image)

Power dependence of the \(Q\) due to problems with sample defects such as cracks, filamentary superconductivity, or otherwise is rare, however it should be considered as
a possible explanation if a previously successful loop gap apparatus becomes suddenly flawed.

3.3 Calibration

In the previous chapter on cavity perturbation, it was shown that the resonator constant $G$ was required to extract the screening length from raw frequency shift data. To calculate $G$ from first principles using only knowledge of the cavity geometry, and coupled field strength would be a daunting task for a loop gap resonator. Instead, a sample of known screening length is used as a calibration. In this particular setup a sample of lead-tin was chosen. Not only does it have the advantage of being a near perfect diamagnet (due to the Meisner effect) in its superconducting state below 7.2K, but it also behaves like a metal with large electron scattering above $T_c$. Thermal expansion effects are easily taken into account, since a wealth of data exists for this material.

For a metal with substantial scattering at low temperatures due to the 5% Sn and strong electron-phonon scattering at high temperatures, the screening length can be taken to be the classical skin depth: the low operating frequency of $1\text{GHz}$ and strong scattering avoid the anomalous skin effect. The exact DC resistivity of the composition can be taken with a simple four point probe and careful geometric measurements. A simple reexamination of Eq. 2.13 from the previous chapter gives:

$$\Delta \frac{\hat{\omega}_{p-s}}{\hat{\omega}} = \frac{\Delta f_0}{f_0} + \Delta \frac{1}{2Q} = 2i\mu_0 G A_{\text{eff}} \delta_{\text{skin}} (1 + i) \tanh \left( \frac{c(1 + i)}{2\delta_{\text{skin}}} \right)$$ (3.4)

When calibrating the resonator it is important to realize that there is no difference in RF absorption loss between an empty cavity and a cavity with a perfect conductor in it, so the best way to examine the formalism now is to think in terms of $R_s$, or $\Delta \left( \frac{1}{2Q} \right)$. By doing this one minimizes the effect of any non-perturbative correction encountered, and thermal contraction of the sample in its fully diamagnetic state will
only change the real component of the frequency shift. A plot of the calibration is given in Fig. 3.7, where the x-axis is controlled by changing the temperature of the reference sample, and \( \delta_{\text{skin}} \) is calculated from the separately measured \( \rho(T) \). Once \( G \) has been determined, it is taken to be constant over the lifetime of the resonator.

\[
\text{Im}[(1+i) A_{\text{eff}} \delta_{\text{skin}} \mu_0 \tanh \left( \frac{(1+i) c}{2 \delta_{\text{skin}}} \right)] \times 10^{18}
\]

Figure 3.7: The calibration set for PbSn with an appropriate linear fit for data taken over the temperature range \( 10K < T < 100K \). The resonator constant was determined to be \( 212.0 \pm 0.7 \times 10^{10} \) from the fit’s slope.

Using this apparatus alone it is not possible to determine the absolute value of the microwave surface resistance of the superconducting samples, since the samples cannot be inserted into the cavity in situ. By determining the shift in \( \Delta \left( \frac{1}{Q} \right) \) from base temperature, one obtains a value corresponding to the shift in \( R_s \) from base. It would be preferable to have some mechanism by which absolute \( R_s \) could be measured in situ, by either removing the sample from the cavity or by comparing it to a reference sample. Consecutive cooldowns unfortunately do not yield exactly the same base cavity \( Q \), and since the absolute value of \( R_s \) is quite low, \( \Delta \left( \frac{1}{Q} \right) \) induced
in the sample from base temperature tends to be smaller than the change in $\Delta \left( \frac{1}{Q} \right)$ between cooldowns. However if bolometric measurements on the same sample exist, the residual resistance can be determined.

### 3.3.1 Vibration

Due to microphonics which affect the sapphire sample mount, it is essential to dampen out vibrations and/or run the apparatus in the middle of the night when building traffic is at a minimum. If there is too high a level of microphonics, the Lorentzian fitting routine used will not correctly fit the acquired data, the time averaged signal being similar to a superposition of multiple Lorentzians superimposed about the many resonator central frequencies. If the resonant frequency is seen to be extremely microphonic, it is possible that the screws pulling down on the superconducting joint of the resonator are loose. In this situation the resonator should be disassembled, and re-plated.

Bags of lead shot mechanically stabilized the room temperature electronics, countering the effects of vibrations. This is especially necessary when running the Robinson oscillator circuit, since the room temperature electronics are part of the feedback loop.

### 3.4 Thermal considerations

When a material is brought down to low temperature a number of its properties are affected. As often happens, one measurement cannot be made without correcting for the convolution of these various thermal changes. The primary alteration that must be corrected for in most crystals is the effect of thermal expansion. As the dimensions contract, the sample volume, and hence the effective volume of the cavity that is screened, is reduced.
Another aspect of working at extremely low temperatures is that the experimental apparatus operates across a large thermal gradient. Because of co-axial cable and structural links, thermal variations can cause drift in the resonance frequency through various inductive and capacitive couplings. To help counteract these effects, the apparatus is arranged to sit at base temperature for some time before measurements are taken. A necessary precaution when running the Robinson oscillator is to regulate the external electronics at room temperature using a circulating water bath, as they form a part of the feedback structure. In particular, the gain and phase shifts of the amplifiers are sensitive to temperature.

3.4.1 Thermometry

Sample thermometry and regulation

To ensure that any temperature dependence of the sample conductivity measured by the apparatus is correct at low temperature, it is necessary to establish sensitive thermometry and temperature control. This is accomplished through a small resistor (≈ 1000Ω at room temperature) used for heating, and four point measurement of a Cernox 1030 resistor [20] used as a thermometer. They are situated behind the sapphire mounting platform. Unlike ruthenium oxide which has little temperature dependence above 40K and is dominated by thermal activation at low temperature, the Cernox resistor, which is based on thin film technology, has an easily measurable temperature dependence over the temperature range $T < 300K$. A sharp increase in the resistance of the cernox at low temperatures is beneficial to the experiment, because at low temperatures the thermal conductivity of sapphire, vacuum grease, and any GE varnish (used for mounting resistors) decreases sharply, and having a high Cernox resistance reduces any self heating (as resistance increases, the probe current decreased such that less power is dissipated).

Even though the Cernox resistors are designed to be stable for many thermal cycles, it is found that through extensive use, the thermometry may be offset at low
temperature, and so multiple checks are needed during the lifetime of a thermometer. Since the resistance of the Cernox does not change appreciably over the range from $4K \rightarrow 300K$, re-calibration is not necessary after an initial data set is taken.

For materials with low critical temperatures, such as very underdoped YBCO, very overdoped Tl2201 or heavy fermion superconductors, thermal precision is critical. Measurements of the thermometer resistance down to 4K can be accomplished by using a well calibrated thermometer (done at UBC utilizing a Quantum Design sample material property system). Below 4K, calibration is performed through the regulation of helium vapor pressure with a Walker regulator, and pressure vs. temperature tables. A plot of the resistance of the sample thermometer is given in Fig. 3.8.

![Vapour Pressure Calibration](image)

Figure 3.8: The resistance of the Cernox 1030 resistor rises steeply at low temperatures.
Helium Bath regulation

Helium bath temperature is kept constant by electronic regulation using a temperature sensor immersed in the bath (a nominal 100Ω Allen-Bradley resistor which has a resistance of about 32kΩ at the base temperature). Without this step, power that is dumped into the heating of the sample stage causes thermal drift to occur in the bath temperature. Data taken at higher temperature is affected by this to a larger extent, since more power is needed to raise sample temperature. To set the regulation, the sample should be brought to its highest operating temperature, and the bath allowed to equilibrate. The bath is then regulated slightly above this temperature. Thermal drift of the complex resonant frequency is still observed in this experiment, as the levels of liquid nitrogen, and helium are changing over the course of the experiment. These effects are associated with temperature gradients across the portion of the apparatus above the helium level, and are not easily avoided.
Chapter 4

C-axis measurements on

$\text{Tl}_2\text{Ba}_2\text{CuO}_{6+x}$

4.1 Why Thallium?

Of the many reasons for examining $\text{Tl}_2\text{Ba}_2\text{CuO}_{6+x}$, the most obvious is that Tl2201 is one of a limited number of materials that can be prepared on the overdoped side of the cuprate phase diagram (see Fig. 1.1). Excess oxygen is doped into interstitial sites in the thallium oxide layer located between the copper oxygen planes.

Measurements of thermal properties in materials on the overdoped side of the phase diagram [21] have indicated that as doping is raised, the magnitude of the low temperature superconducting energy gap decreases as the transition temperature decreases (microwave measurements performed here are used to test these results). It has also been shown through thermal measurements that the behavior of overdoped materials is in agreement with what is expected from d-wave BCS formalism. Under this regime, the magnitude of the superconducting energy gap goes to zero at the transition temperature. It should be noted that this is very different from the case of the underdoped region of the phase diagram. For example, in the measurements of [1] and [22] results indicate that on the underdoped side superconductivity is being destroyed by a dephasing of the order parameter.

Unlike many other cuprate superconductors Tl2201 is a single layer compound. That is, there is only one copper oxygen plane per unit cell. The well studied bilayer cuprates, such as YBCO and BSCCO have two layers of CuO$_2$ planes in each unit
cell, which can lead to even more complicated transport properties. YBCO is further complicated by a CuO chain layer which also plays a role in the electronic properties. From this structural perspective Tl2201 can be thought of as a relatively simple compound.

Another interesting aspect to working with Tl2201 is that crystals grown with higher $T_c$'s appear to be tetragonal, and those with lower $T_c$ exhibit only slight orthorhombicity (anisotropy between a and b axes of $< 1\%$). This eliminates some complications, such as twinning, that arise when analyzing other HTSC materials.

4.2 Why c-axis?

The interpretation of c-axis superfluid stiffness measurements in the HTSC materials has long been a point of dispute. There have been reports, and explanations of all types of low temperature dependencies ranging from $\rho_s \propto T \rightarrow \rho_s \propto T^5$ (see [23] and [24]). Though many results exist for magnetically oriented grains, and thin films, there are few results for single crystals (see [25] [22] for more recent data). In the microwave frequency limit, there is little data on the overdoped side of the phase diagram. This may well be because sufficiently thick single crystals of this material have until now been difficult to grow and anneal homogeneously.

4.3 Sample preparation and manipulation

Samples are initially grown by a self-flux method, and initial oxygen content places them in the far overdoped region of the phase diagram with a $T_c$ of approximately 5K (for more information on the quality and preparation of these samples see work done at UBC by [26]). Extra oxygens sit in interstitial sites between the CuO$_2$ planes. By annealing them in a constant, reduced oxygen atmosphere it is possible to lower their doping from Tl$_2$Ba$_2$CuO$_{6+\delta}$, closer to the stoichiometric compound Tl$_2$Ba$_2$CuO$_6$ and hence increase $T_c$. The constant oxygen partial pressure helps to ensure that crystals
remain homogenous on the whole. This annealing technique has been refined over many trials and results have shown that it is preferable over other methods, such as vacuum annealing. In the crystals measured for this work $\Delta T_c \sim 1 - 2K$. There were two dopings, one annealed from a $T_c$ of 5K to a $T_c$ of 65 K, and one to a 25K final transition temperature.

Clean Tl2201 platelets are a rarity, as they generally tend to clump together during the growth process, or are subjected to flux spotting. They also do not seem to cleave well along any plane parallel to the c-axis, however some success has been made in cutting these crystals with a fine gauge wire saw (wire diameter = 25$\mu$m). Uneven edges result in a certain amount of uncertainty when measuring the crystal dimensions, since the irregularities of the crystal face on one side of the crystal do not usually line up with the irregularities of the crystal face on the other side of the crystal. This may be less of a bother in future generations of crystals grown by the UBC group, since advances in furnace design will allow crystals to be grown and separated from surrounding flux while the flux is still liquid. Crystals examined "as-grown" generally seem to have better faces out of the plane than those cut or cleaved. As will be shown later, ill-defined sample dimensions can considerably increase the difficulty of determining the zero temperature c-axis superfluid stiffness.

Crystals are also difficult to cleave perpendicular to the (001) direction. Throughout all manipulation of these samples, even during rigorous cleaning, no planar cleavage was ever seen. It is standard and relatively straightforward to cleave $Bi_2Sr_2CaCu_2O_{8+\delta}$ samples in situ for experiments in which sample surface quality is critical such as Angle Resolved Photo-Emission Spectroscopy (ARPES) and Scanning Tunnelling Microscopy (STM) [27]. With the absence of an interplane chain layer in Tl2201, there had been some hope of finding easy cleavage for these, and other experiments.

Single crystals of Tl2201 were placed in the 0.95GHz loop gap resonator with the oscillating magnetic field applied parallel to the $CuO_2$ planes. Currents induced in the sample would then run around the axis of the field (see fig. 4.4). To show that
this is truly a c-axis measurements, one needs to consider that the aspect ratio of the crystals' c-axis thickness to their average width was in the range of 1 : 2 → 1 : 10 whereas the typical literature values of the c-axis penetration depth [28] to the in-plane penetration depth [29] are approximately 17μm : 0.1μm.

4.4 Extraction of $\lambda_c$

Since only changes from the base temperature value of the effective screening length can be observed through the use of this particular loop-gap resonator apparatus, certain constraints must be placed on the experimentally observed screening length, to extract the true conductivity.

Through Eq. 1.16 it can easily be seen that for a sample in its normal state, $n_s = 0$ and therefore $\Re\{\frac{1}{\delta}\} = -\epsilon_r\epsilon_0\mu_0\omega^2$. Since this value is in practice smaller than the resolution of the experiment, $\Re\{\frac{1}{\delta}\}$ is usually set to zero in the limit $T > T_c$. 
4.4.1 Plasma edge crossing in c-axis measurements

A strong contribution to the absorption of these samples comes from a plasma edge crossing, which occurs when the real part of the dielectric constant passes through zero with changing superfluid density. This enhanced absorption loss in $R_s$ can be explained most clearly in terms of a depolarization effect. Although the contribution to Eq. 2.6 from $P \cdot \mathbf{E}$ was previously shown to be negligible, it is now seen that in the limit of $\Re\{\epsilon\} \rightarrow 0$, a resonant absorption occurs, because of the c-axis Josephson plasmon.

To examine this effect it is first useful to define the polarization, $\bar{P}$, of an ellipsoidal sample in an electric field (following [30]) as:

$$\bar{P} = \epsilon_0 (\epsilon_r - 1) \bar{E}$$

(4.1)

where $\bar{E}$ is the field within the sample. It can also be shown that $\bar{P}$ is related to $\bar{E}_1$ via a depolarization factor $N$.

$$\bar{E} = \bar{E}_0 + \bar{E}_1 = E_0 - \frac{N \bar{P}}{\epsilon_0}$$

(4.2)

where $\bar{E}_0$ is the externally applied electric field. Here it is assumed that $\bar{E}_0$ is along a principle axis of the ellipsoid and $N$ is the appropriate depolarizing factor. Combining Eq. 4.1 and Eq. 4.2, one is able to relate sample polarization solely to the external electric field.

$$\bar{P} = \frac{\epsilon_0 (\epsilon_r - 1)}{1 + N (\epsilon_r - 1)} \bar{E}_0$$

(4.3)

Taking an ellipsoidal sample in the limiting form where it resembles a thin slab, with an electric field perpendicular to the plane, the depolarization is clearly maximized and it is found that $N = 1$ (see [30]). Whereas a sample geometry could also be chosen such that the electric field lies in the plane of the thin sample, and
the limiting case brings $N \to 0$. As the samples utilized for this experiment are not ellipsoidal, this treatment is not entirely accurate, and $N$ cannot be easily defined. However, phenomenological features in the measurements can be better understood by including this depolarization effect. Due to sample aspect ratios encountered, and the fact that the electric fields are typically perpendicular to the broad crystal face, it is expected that all crystals examined had depolarization factors greater than that of a sphere, for which $N = \frac{1}{3}$.

Now that the polarization has been defined as a function of the external field it is necessary to re-examine Eq. 2.6.

$$\frac{\Delta \tilde{\omega}_{e \to s}}{\tilde{\omega}} = \frac{A}{4u} \iiint_{V_s} \left( \vec{F} \cdot \vec{E}_0 + \vec{M} \cdot \vec{B}_0 \right) d\tau^3$$  \hspace{1cm} (4.4)

The contribution to the first term in the above integration may no longer be negligible, since the polarization can become very large when $\Re\{1 + N(\varepsilon_r - 1)\} \to 0$. The shift in resonator frequency due to depolarization is now:

$$\frac{\Delta \tilde{\omega}_{\text{depol}}}{\tilde{\omega}} = \frac{A}{4u} \iint_{V_s} \vec{P}(\vec{r}) \cdot \vec{E}_0(\vec{r}) d\tau^3 = \frac{e_0(\varepsilon_r - 1)ac}{4u(1 + N(\varepsilon_r - 1))} \sum_{i=1}^{n} \frac{1}{n} \int_{\frac{b}{2}}^{b} E_0^2(x) dx. \hspace{1cm} (4.5)$$

The analysis in chapter 3 showed that the electric field varies approximately linearly with the distance along the crystal’s width. Here, not only are the sample’s widths different, but sample location in the field changes slightly between mountings. Thus $G_2 = \frac{\text{con}}{4u} \sum_{i=1}^{n} \frac{1}{n} \int_{\frac{b}{2}}^{b} E_0^2(x) dx$, cannot be taken to be a well-determined resonator constant. In terms of $G_2$ one finds:

$$\frac{\Delta \tilde{\omega}_{\text{depol}}}{\tilde{\omega}} = \frac{G_2(\varepsilon_r - 1)}{1 + N(\varepsilon_r - 1)} = \frac{G_2}{N} - \frac{G_2}{N^2} \frac{1}{1 + N(\varepsilon_r - 1)}. \hspace{1cm} (4.6)$$

Examining $\frac{\Delta \tilde{\omega}_{\text{depol}}}{\tilde{\omega}}$ closely, one realizes that in a perfectly conducting material $\varepsilon_r \to \infty$ so that much like the magnetic effects treated in chapter two, this shift can be thought of as two separate contributions. The first term $\frac{G_2}{N}$ represents the shift
from an empty cavity to a perfect conductor, and the other term, $\frac{C}{N} \frac{1}{1+N(\varepsilon_r-1)}$, can be interpreted as a shift from a perfect conductor to a real sample. To fully comprehend how this depolarization contributes to the frequency shift, it is necessary to first examine the dielectric constant of the material.

### 4.4.2 The relative dielectric constant

Examining [11], one finds that the dielectric function for the optical properties of an insulator can be written:

$$\varepsilon_r(\omega) = \varepsilon_r^0(\omega) + \frac{i\sigma}{\varepsilon_0\omega}. \quad (4.7)$$

This treatment is also applicable to the case of a superconductor, where $\varepsilon_r^0$ is a constant value at microwave frequencies (one should take note that in [11] the $e^{-i\omega t}$ convention gives $\sigma = \sigma_1 + i\sigma_2$, as opposed to the $e^{i\omega t}$ convention used in this thesis which requires $\sigma = \sigma_1 - i\sigma_2$). In infrared measurements $\varepsilon^0(\omega)$ is described using a saturation value at high frequency, $\varepsilon_r^\infty$, and then accounting for all the phonon mode contributions by summing Lorentz oscillators:

$$\varepsilon_r^0(\omega) = \varepsilon_r^\infty + \sum_j \frac{S_j\omega_{T_j}^2}{\omega_{T_j}^2 - \omega^2 - i\omega\gamma_j}. \quad (4.8)$$

However, since microwave measurements are performed at frequencies far below any of the phonon modes, one can make the simplification that the sum in the above equation is taken only over the oscillator weights $S_j$. From grazing incidence reflectivity measurements reported in [31] it is possible to find values of $S_j$ taken on a single crystal of Tl2201 with a $T_c$ of 82K and a $\Delta T_c$ of 13K. Although crystals used in the present cavity perturbation experiment are of better homogeneity, (shown by sharper superconducting transitions), oscillator weight is generally independent of phonon mode width. Hopefully $S_j$ will not be very different in the lower $T_c$ sample, although as more measurements are performed, this might be checked explicitly. Relating the microwave $\varepsilon_r^0$ to the infrared data it is found that:
\[ \varepsilon_r^0 = \varepsilon_r^\infty + \sum_j S_j \approx 9.8. \] (4.9)

The values of \( S_j \) given in [31] also appear to be temperature independent having a summed value which is the same at room temperature as at 8K.

Inserting the superconductor's conductivity (derived in chapter 1) into Eq. 4.7 it is possible to arrive at the following useful expression:

\[ \varepsilon_r(\omega) = \varepsilon_r^0 - \frac{1}{\mu_0\varepsilon_0\omega^2\lambda_c(T)} + \frac{i\sigma_1}{\varepsilon_0\omega}. \] (4.10)

Separating out real and imaginary contributions to the complex frequency shift;

\[ \Re\left\{ \Delta\omega_{\text{dopol}} \right\} = \frac{G_2}{N^2} \left[ N + \frac{-\varepsilon_r^0 + \frac{1}{\mu_0\varepsilon_0\omega^2\lambda_c(T)} - \frac{1}{N} + 1}{\left(\varepsilon_r^0 - \frac{1}{\mu_0\varepsilon_0\omega^2\lambda_c(T)} + \frac{1}{N} - 1\right)^2 + \left(\frac{\sigma_1}{\varepsilon_0\omega}\right)^2} \right] \] (4.11)

\[ \Im\left\{ \Delta\omega_{\text{dopol}} \right\} = \frac{G_2}{N^2} \left[ \frac{\frac{\sigma_1}{\varepsilon_0\omega}}{\left(\varepsilon_r^0 - \frac{1}{\mu_0\varepsilon_0\omega^2\lambda_c(T)} + \frac{1}{N} - 1\right)^2 + \left(\frac{\sigma_1}{\varepsilon_0\omega}\right)^2} \right]. \] (4.12)

A large absorption peak is found in \( \Im\left\{ \Delta\omega_{\text{dopol}} \right\} \), which can be seen clearly in measurements of \( R_s \). A resonance effect appears in the complex frequency shift, when \( \frac{1}{\mu_0\varepsilon_0\omega^2\lambda_c(T)} = \varepsilon_r^0 + \frac{1}{N} - 1 \). If one were to sweep the frequency, and fix the temperature it is also clear that the resonance would be seen, near the plasma frequency \( \omega_p = \sqrt{\frac{n_s\varepsilon^2}{\varepsilon_0\varepsilon_r^2m^*}} \). However, in these measurements, the temperature is swept at fixed frequency, so the maximum in \( R_s \) occurs when \( \omega_{\text{meas}} = \omega_p \)

By comparing the data with the model proposed, it appears that there is an extra contribution to the width of the peak in \( R_s(T) \). Any inhomogeneity in the sample's superfluid density would essentially cause different parts of the crystal to experience the plasmon enhancement at different temperatures. Since it is more likely that a continuous distribution of superfluid density rather than distinct regions exists, one expects to see a smooth broadening of the resonant effect.
Chapter 4. C-axis measurements on Tl$_2$Ba$_2$CuO$_{6+z}$

Figure 4.2: Real and imaginary components of the effective penetration depth of samples in the $T_c = 65K$ doping. Above $T_c$, the real component is seen to saturate at a value given by the average sample width. The plasmon peak of crystal 1 is given in the inset.

A factor that may contribute to the difference between experimentally observed plasmon peaks, and the resonant absorption effect predicted from examining the superfluid density, is that the depolarization constant is not well defined in this sample geometry. In fact the analysis is only exact in the case of an ellipsoid, and so the effective depolarizing factor in this situation is actually varying spatially. One can easily see that as $N$ decreases, the center of the plasmon will shift toward lower temperature, and the overall scaling of the peak is increased, so a spatially dependent depolarization factor would certainly affect the resultant plasmon.

Inserting Eq. 1.16 into Eqs. 4.11 and 4.12 it can be shown that:
Figure 4.3: Real and imaginary components of the effective penetration depth of samples in the $T_c = 25K$ doping. Above $T_c$, the real component is again seen to saturate at a value given by the average sample width. The plasmon peak of crystal 3 is now given in the inset. Differences in the normal state of the imaginary component can likely be attributed to flux spots or dirt on the surface of crystal #4, and the fact that it is much larger than crystal #3.

\[
\frac{\Delta \tilde{\omega}_{depol}}{\tilde{\omega}} = \frac{G_2}{N^2} \left[ \frac{1}{\mu_0 \omega^2 \delta^2} - \frac{1}{N} + 1 \right] \left[ \Re \left\{ \frac{1}{\mu_0 \omega^2 \delta^2} \right\} + \frac{1}{N} - 1 \right] + \left( \Im \left\{ \frac{1}{\mu_0 \omega^2 \delta^2} \right\} \right)^2.
\]  

(4.13)

In the limit of $T > T_c$ there will be a saturation value of $\Re \{ \frac{\Delta \tilde{\omega}_{depol}}{\tilde{\omega}} \}$. However if the value of $N$ is assumed to be larger than that corresponding to a sphere, then any contribution can be shown to be far less than the resolution of the saturation value of $\tilde{\delta}_{eff}$. Since $G_2$ is very small, this effect should contribute very little to the surface impedance well away from the transition temperature.
4.4.3 The thin limit saturation of $\mathcal{R}\{\tilde{\delta}_{\text{eff}}\}$

As previously mentioned, crystal platelets of Tl2201 often have fairly uniform thickness in the $c$ direction, but are otherwise misshapen (see Fig. 4.4). This can result from their preparation, or the process of cleaving. In order to accurately extract the penetration depth one must first consider how to treat $\Delta \tilde{\delta}$.

The approach taken here is to approximate the crystal shape as a series of small slivers perpendicular to $\vec{H}_{rf}$, as shown in Fig. 4.4. Each sliver can be treated as an individual crystal using Eq. 2.13, and by then summing the contributions from all slivers, the proper screening length can be extracted using the expression:

$$
\tilde{\delta}_{\text{eff}} = \sum_{i=1}^{n} \frac{\tilde{\delta}}{n} \tanh \left( \frac{F(a(i-0.5))}{2\delta} \right).
$$

In Eq. 4.14 $a$ is the crystal dimension running parallel to the magnetic field, $F(x)$ is an interpolated function of the measured crystal width at point $x$, and $n$ is an arbitrarily high number set so that further increases do not limit the precision of the extraction.

Figure 4.4: This is a figure of an oddly shaped crystal that can be easily modelled using the $c$-axis technique.
Examining $\delta_{\text{eff}}$ as the screening length approaches the width of a crystal sliver (i.e. $\delta < F(x_i)$) it can be seen that a saturation of the effective penetration depth occurs where $\tanh\left(\frac{F(x_i)}{2\delta}\right) \approx \frac{F(x_i)}{2\delta}$. The maximum value that the real component can reach is $\frac{F(x_i)}{2}$. By applying this principle to Eq. 4.14 it is easy to show that for a series of slivers, that $\Re\{\delta\}$ approaches the average width in this thin limit.

$$\Re\{\delta_{\text{eff}}\} \rightarrow \sum_{i=1}^{n} \frac{1}{n} \frac{F(x_i)}{2} = \frac{\langle F(x) \rangle}{2}$$ (4.15)

Here it is also necessary to note that depolarization effects analyzed in the previous section will also have a saturation value in the limit of $T > T_c$. However a quick calculation of the contribution given in Eq. 4.11 places it on the order of nanometers, far less than even the resolution of the saturation value.

By then shifting the experimental saturation value of $\Re\{\Delta \delta_{\text{eff}}\}$ to the average width, it is possible to extrapolate down to the zero temperature value of $\delta_{\text{eff}}$. To obtain the absolute value of the penetration depth it is then a simple matter of using Eq. 4.14.

Reasonable values for the sample dimensions can be obtained using an optical microscope. A graticule is used to calibrate the microscope eyepiece divisions for each magnification. Photos are taken using a 4 megapixel digital camera. The program Scion Image can then be used to analyze digital pictures of the crystals.

When measuring the width function $F(x)$ it is important to both take measurements of the crystal width along the field axis, and take an average width by measuring the area of the sample, and dividing by the length. Integrating the interpolated function and dividing by the length should give an answer close to average width, although it will inevitably be slightly different due to the slight uncertainty in each of the measurements. A re-scaling of the interpolation function, by the ratio of average width to the integrated mean value, is then used to ensure the correct saturation of $\Delta \delta_{\text{eff}}$. 
A final step in checking the validity of this saturation assumption is to calculate the screening length that can be expected from displacement currents alone. If the value of $\varepsilon^0 = 9.84$ is taken to be correct, then the corresponding length can be found to be $\delta_{\text{displace}} = 15.9\text{mm}$, which is far greater than the maximum width of any measured sample.

4.5 Results and Discussion

It has been shown that the value of the effective screening length can be constrained in the following three ways:

- $\Re\{\frac{1}{\delta^2}\} \rightarrow -\varepsilon_\infty \mu_0 \omega^2$ for $T \gg T_c$.

- Depolarization absorption peak at zero crossing at $\Re\{\varepsilon\}$

- $\Re\{\Delta\delta_{\text{eff}}\} \rightarrow \frac{\langle F(x) \rangle}{2}$ for $\delta \gg c$.

It is important to understand the limitations, as well as the successes, of each of these constraints before attempting to analyze data. The first of the aforementioned conditions can be used to guarantee that an appropriate saturation value is chosen for $\delta_{\text{eff}}$. Generally speaking, the value obtained for $\Re\{\frac{1}{\delta^2}\}$ is smaller than the precision of the apparatus, and so it would appear to saturate at zero as the temperature increases above the transition.

A value of the offset in $R_s$ was obtained from bolometric measurements for one of the crystals at the $T_c = 65K$ doping. This allowed for a more careful analysis of the plasmon peak observed at this doping. In this study, and one done for the $T_c = 25K$ doping it has proved impossible to utilize the properties of the plasmon peak to achieve a value of the zero-crossing of $\sigma_2$, or the depolarization. Although agreement could not be obtained between the plasma edge crossing and the peak in $R_s$ seen in the inset of Fig. 4.2, the phenomenological behavior is seen to be correct. It is believed that the problem can be attributed to an inhomogeneity of the
superfluid within the sample due to a gradient of the oxygen doping. Oxygen content is set in the crystal by annealing it in a constant oxygen atmosphere, which if left long enough should eventually achieve some steady state concentration. Incomplete oxygen equilibration would leave the sample in a state where the doping in the central region of the crystal would be higher (corresponding to a lower $T_c$) than the outer parts of the crystal. Alternatively, the problem of inhomogeneity might be due to a varying ratio of thallium to copper inside the crystal. Since Cu is apt to substitute into the Tl site, such an inhomogeneity may be due to a problem during the growth process, not the anneal. However, electron probe micro-analysis (EPMA) performed at the UBC Earth and Ocean Sciences department shows that the thallium to copper ratio is fairly constant throughout the Tl2201 samples, although a polishing process is required for these tests. The EPMA is not as sensitive to light atoms, so oxygen concentration gradients could not be determined. Viewing these results, it would appear that oxygen is the more likely candidate for this particular problem.

It can be noted that the depolarization analysis outlined above may be better applied to measurements taken on the c-axis properties of materials such as very underdoped YBCO, where any inhomogeneity is on a much smaller length scale than the penetration depth.

Finally, the true conductivity can be extracted using a root finding algorithm in Mathematica, solving Eq. 4.14 for each measurement of the effective screening length. Figs. 4.6 & 4.7, give the superfluid fraction, $f_s = \frac{\lambda(0)^2}{\lambda(T)^2}$, as it was extracted from both dopings.

The measured $\lambda_0$ value found for the $T_c = 25K$ doping was $14 \pm 2\mu m$, while the lower doping yielded a $\lambda_0 = 15 \pm 2\mu m$. The uncertainty in these values is dominated by the inability to measure the physical dimensions of the samples to high precision, and is amplified through the extraction process.
Figure 4.5: As the sample has oxygen removed during the annealing process, a gradient of doping can be formed between the inner and outer sample material. As the screening length presses further into the crystal, superfluid corresponding to superconductor with a higher oxygen content (and therefore lower $T_c$), disappears rapidly.

Unlike the results given in [24] which reports a $c$-axis temperature dependence of $\sigma_2 \propto T^5$ in aligned grains of $HgBa_2CuO_{4+\delta}$, it was found that $\sigma_2$ behaved with a temperature dependence of $\approx T^2$ for temperatures at less than a third of the transition temperature. In preliminary fitting using a technique recently developed by Sheehy et al. [8] it is seen that the temperature dependence of $\sigma_2$ can be modelled quite effectively by allowing for three separate power law regimes.

The general scheme can be explained by considering incoherence in the tunnelling mechanism between planes. A coherent hopping model of the $c$-axis transport properties would predict a linear temperature dependence of the superfluid density which is not observed in careful microwave measurements of high quality samples.
Figure 4.6: From two pieces cleaved off a single crystal, the fraction of superfluid carriers, \( f_s \), was extracted for the \( T_c = 65 \) doping of Tl2201. Data obtained through the use the Robinson oscillator technique outlined in chapter 3 agrees well with that taken through the transmission method. In the inset, oscillator measurements are seen to be very stable.

By easing the constraint that in-plane momentum of carriers is conserved when quasiparticle hopping occurs from one CuO plane to another, it is possible to claim that the final in-plane momentum will differ from the initial by only a small vector, such that \( \vec{q} = \vec{k}_f - \vec{k}_i \). If \( \vec{q} \) is decomposed into a component in the nodal direction, \( q_1 \), and another in the antinodal direction, \( q_2 \), then a natural re-scaling can be performed utilizing the quasiparticale energy \( E = \sqrt{v_F^2 q_1^2 + v_\Delta^2 q_2^2} \).

Now the new small momentum difference can be given by components \( \vec{q}_1 = v_F q_1 \) and \( \vec{q}_2 = v_\Delta q_2 \). Since it is found that \( v_F \gg v_\Delta \), this re-scaled vector difference is very small in the antinodal direction, while being nearly unconstrained in the nodal direction.
Figure 4.7: $f_s$ is shown for the $T_c = 25$ doping of Tl2201. Here is an example of two crystals with large differences in homogeneity near $T_c$. A slight departure of the two curves over the mid range can probably be attributed to demagnetization effects, thermal expansion, and a difference in homogeneity at temperatures near $T_c$.

To properly apply this treatment to the overdoped region of the phase diagram, account must be made for a temperature dependent gap function. This is unlike the analysis used for underdoped c-axis measurements, where a relatively constant gap may be assumed (as mentioned previously superconductivity is likely destroyed due to dephasing of the order parameter). Examining the results of thermal transport measurements reported in [21], where the anisotropy ratio, $\frac{\nu_E}{\nu_\Delta}$, was determined to be a relatively high value of 270 (for a sample of Tl2201 overdoped to 15K), Tl2201 appears to be an ideal candidate to test the model. As one examines samples with lower $T_c$ (increasing doping), the ratio of $\frac{\nu_E}{\nu_\Delta}$ should increase, as $\nu_\Delta$ should decrease roughly with $T_c$ (in the ideal case of a BCS d-wave superconductor). It
would also appear that in the theory of Sheehy et al. the normal fluid density, which is proportional to $\frac{1}{\lambda(0)^2} - \frac{1}{\lambda(T)^2}$ should at low temperature go as $\sim (\frac{T^2}{v_g})$ for this relatively high anisotropy ratio. By fitting the low temperature measurements of the normal fluid density to a simple quadratic (see Fig. 4.8 and Fig. 4.9), it was possible to confirm that the gap velocity is decreasing from the $T_c = 65K$ doping to the one with $T_c = 25K$, assuming a relatively doping independent $v_F$. [21].

![Figure 4.8: The temperature dependence of the normal fluid density is fit to a quadratic power law for $T < \frac{T_c}{6}$ for the lower doping.](image)

A slight downturn is also observed in the superfluid density near $T_c$. This is likely due to the same problems encountered during the analysis of the depolarization resonance, and is attributed to an incomplete annealing process. As the screened portion of the sample becomes smaller near $T_c$, and the superfluid density within the crystal would correspond to a higher oxygen doping, the resulting behavior would appear as...
Figure 4.9: The temperature dependence of the normal fluid density is fit to a quadratic power law for $T < \frac{T_c}{6}$ for the higher doping.

It is also worthwhile to mention that this sudden drop in superfluid density is predicted from some theoretical models of HTSC. A Kosterlitz-Thouless transition is characterized by a discontinuity at $T_c$, as the superfluid falls to zero. One should also be aware that fluctuation effects near the superconducting transition, due to forming pockets of superfluid, will greatly increase the effective conductivity of normal carriers, and causes a peak in the real component of the conductivity, however these effects can be complex in the c-axis. This should not be confused with the plasmon peak, in $R_s$, and fluctuation effects would only appear clearly in the extracted conductivity. The downturn might also be attributed to an enhancement to $X_s$, because of depolarization. The peaks shown in the insets of Fig. 4.2 and Fig. 4.3 would have corresponding features in the real component of $\tilde{\varepsilon}_{eff}$, but it can be inferred from
Eq. 4.13 that there is zero contribution at the peak of the plasmon.

As stated earlier in chapter 2, the effect of demagnetization is not accounted for in this experimental analysis. It can be reasoned that for this field orientation, thinner crystals minimize demagnetization effects. However, the uncertainty incurred from measurements of the thickness, c, is larger in the thinner crystals. All crystals are dimensioned to the same precision, so the percentage error will be lower in the thicker platelets. As these two tradeoffs are mutually exclusive, a balance must be found when attempting to choose crystals with appropriate dimensions.

Leaving the sample out of the desiccator for a long period has been found to have no adverse affect on the surface quality of these crystals, even as they are expected to have a high affinity for water. Throughout the cleaning and cutting processes the crystals were exposed to n-heptane, and for brief periods acetone. In the case of the $T_c = 65K$ doping where two pieces of a single crystal were exposed to varying amounts of these conditions, there is very little discrepancy between the conductivity measurements.

Since the thermal expansion data for Tl2201 has not yet been taken, exact expansion corrections are impossible to calculate. Since the effect of thermal expansion will be worse in larger, and thicker crystals, the more accurate dataset should be taken as the crystal with the smaller thickness. However, the curves in Fig. 4.6 agree quite well over all temperatures below the transition, which is not surprising, as most materials have very little thermal expansion below 50K. Uncertainty in the room temperature measurements of the crystal dimensions is also larger than the expansion in typical anisotropic oxide ceramics [32]. It is possible that the slight differences between the data sets of crystals belonging to the same doping are due to some geometric consideration, be it demagnetization or thermal expansion.

The precision of this technique is limited mainly due to the optical measurements of crystal geometry. The exact alignment of a crystal in the magnetic field is also
uncertain, and slight changes in rotation about the out of plane axis produce orien-
tations with varying average widths (some crystals are clearly affected more than
others due to their shape). Another area of concern is the calculation of screening
saturation width, as often the edges appear rough under a simple optical microscope.
In the future, when a larger array of samples are available, an alternative may be to
use thicker samples with smaller widths relative to their length. These were found to
be the best candidates for the analysis, since the effect of variations in the measure-
ment of \( c \) is reduced if \( c \) is large. Samples that go thin after only a small amount of
penetration have slightly less uncertainty relative to wider ones. Ideally it is best to
have at the very least, a small sample to set \( \delta_{\text{eff}}(0) \) and a sample with greater loss to
nail down \( \sigma_1 \) and \( \sigma_2 \). This is a result of the fact that the resolution of the experiment
to measure frequency and bandwidth is relatively constant, while the magnitude of
the signal loss is largely dependent on sample size. Thicker crystals grown by the
methods outlined above also tend to be prone to flux spotting, or inclusions of other
crystallites, which can greatly affect measurements of \( R_s \). Work is currently being
done at present in the UBC group to eliminate these problems by removing Tl2201
crystals from the melt during the growth process.
Chapter 5

Absolute $\lambda$ measurements on Gd doped $YBa_2Cu_3O_{6+\delta}$

5.1 A new approach

The goal of this experiment was to confirm that the intrinsic electrodynamics of YBCO are not altered by doping in a small amount of gadolinium paramagnetic impurities. It is part of a recent attempt to determine the absolute value of $\lambda$ using only microwave bolometry, and resonator techniques.

A limitation of cavity perturbation experiments in general, is that one can not usually arrive at an absolute value for the penetration depth, although the shift in $\lambda$ can be measured to high precision and accuracy. The particular case of dealing with samples that go into the thin limit regime was discussed in the previous chapter, and works moderately well if the sample is clean, however in most cases microwave spectroscopy relies on other experiments such as far infrared (Meisner state), $\mu SR$ (in the vortex state), or perhaps in the future through scanning Hall probe measurements (also in the vortex state) to set the value of the zero temperature penetration depth.

Measurements of the surface impedance made on single crystals of $YBa_2Cu_3O_{6+\delta}$ doped with $\approx 1\%$ gadolinium atoms (grown in the same manner as [33]) express features from which the true penetration depth can be extracted. As a greater portion of the experiment is performed via broadband microwave bolometry (for greater detail see [34]), it is not the objective of this thesis to reveal final results, however a brief synopsis will be given on the general technique.
Gadolinium is a spin $\frac{7}{2}$ atom which acts as a magnetic field probe in a crystal, experiencing any local fields, with an associated zero field quadrupole energy level splitting. The atoms substitute for Yttrium which is located at a site directly between the Copper oxygen planes. Since most of the electronic transport is located within the CuO planes, measurements were taken to determine the extent to which the gadolinium atoms can be thought of as passive impurities.

There are three dominant motivations to measurement of $Z_s$ in these crystals using fixed frequency resonator. The first reason is to determine the temperature dependence of $\lambda$ in these materials, as this sets how the zero frequency delta function weight of $\sigma_1$ changes with temperature. From this slope, the oscillator sum rule for $\sigma_1$ can be checked explicitly. Secondly, a check was required to determine if measurements taken on pure YBCO crystals (i.e. with no gadolinium doping) were different from those with the gadolinium. To do this one needs to examine the scattering peak in $R_s$ below $T_c$ that is characteristic of high purity YBCO. Impurity studies performed on YBCO [35] have indicated that in general even small concentrations of foreign atoms can alter the scattering characteristics of a HTSC.

Thirdly, through measurements taken in $R_s$ near the superconducting transition, crystal homogeneity can be probed easily. By examining measurements made near the superconducting transition of a crystal it is possible to make qualitative judgements

### 5.2 Finding $\chi_\nu$

To determine how gadolinium impurities might contribute to surface resistance in a HTSC sample, one must first examine the power absorption $P$ (see [36]), of a system of spins in an oscillating magnetic field, which is given by:

$$P = \frac{dE}{dt} = -\vec{M} \cdot \frac{d\vec{H}}{dt}$$  \hspace{1cm} (5.1)
By following through the definition of the magnetic susceptibility \( \chi = \chi H \) it is possible to show that the power dissipation can be simplified to:

\[
\frac{dE}{dt} = (\chi' H_0^2 \cos(\omega t) \sin(\omega t) + \chi'' H_0^2 \omega \sin^2(\omega t)) = \frac{1}{2} \chi'' H_0^2 \omega. \tag{5.2}
\]

The relevant Hamiltonian for a spin in an rf magnetic field is:

\[
H = H_0 + H' = H_0 + \vec{m} \cdot \vec{H}_0, \tag{5.3}
\]

where \( \vec{m} \) is the magnetic moment of a gadolinium atom which is then simply given by \( \mu_B \vec{g} \vec{S} \). Choosing the direction of the applied field to lie in the x-axis, the perturbation becomes simply \( \mu_B g H_0 S_x = \mu_B g H_0 \frac{S_+ + S_-}{2} \). Fermi’s golden rule then yields the transition rate between spin states:

\[
W_{a \rightarrow b} = \frac{2\pi}{\hbar} |V_{ab}|^2 \delta(E_a - E_b) \tag{5.4}
\]

In Eq. 5.4, \( |V_{ab}| \) can be taken to be \( \mu_B g \langle m_a | \frac{S_+ + S_-}{2} | m_b \rangle \), from the definition of the perturbation. This term shows explicitly that the only allowed transitions are ones where \( m_a \) and \( m_b \) differ by one.

To determine the total power absorbed by all the spins, one sums over all spins and transitions. To do this one needs to directly account for the population of each state using Boltzmann statistics (this also gives temperature dependence). Therefore the number of spins in a particular energy level is given by:

\[
N_i = N \frac{e^{E_i/k_B T}}{\sum_j e^{E_j/k_B T}}, \tag{5.5}
\]

where \( N \) is the total number of spins, and \( E_i \) is the energy of a state. By knowing the probability of a transition occurring, the state populations and the energy splitting \( (E_a - E_b = \hbar \omega) \) one can define the absorption of each allowed transition as:

\[
P_{ab} = W_{a \rightarrow b}(N_a - N_b)\hbar \omega. \tag{5.6}
\]
Inserting Eq. 5.4 and Eq. 5.5 into Eq. 5.6, and comparing with Eq. 5.2 it is a simple matter to find that each transition contributes to the susceptibility via a term:

\[ \chi''_{ab} = \frac{\pi N \mu_B^2 g^2}{\hbar} \langle m_a | \frac{S_+ + S_-}{2} | m_b \rangle^2 \delta(\hbar \omega) \frac{e^{E_B^a}}{E_B^a} \left( 1 - e^{-\frac{\hbar \omega}{E_B^a}} \right). \tag{5.7} \]

As a diffuse spin system, the gadolinium atoms will inevitably interact with each other on some length scale determined by their concentration in the material. Although this measurement is performed with no external DC field, dipole fields of spins will inevitably alter the local field at the location of any near neighbors, and these effects lead to a broadening of the delta function. To deal with this broadening, a phenomenological shape function is used instead of a delta function, which in practice appears to be a Lorentzian (given by \( L(\omega) \)). So the final form of \( \chi'' \) for transitions between spin states \( m_a \) and \( m_b \) is given by:

\[ \chi''_{ab} = \frac{\pi N \mu_B^2 g^2}{\hbar} \langle m_a | \frac{S_+ + S_-}{2} | m_b \rangle^2 L(\omega) \frac{e^{E_B^a}}{E_B^a} \left( 1 - e^{-\frac{\hbar \omega}{E_B^a}} \right). \tag{5.8} \]

To find the total \( \chi'' \) is necessary to sum over all transitions, taking the local field into account explicitly the crystal field parameters (a nice discussion of this is given in [37]).

### 5.3 Relating magnetic susceptibility to surface impedance

Since the magnetic susceptibility is a causal response function (for a thorough discussion of causality see [38]), one can relate the real and imaginary components via the Kramers Kronig relations:

\[ \chi'(\omega) - \chi_\infty = \frac{1}{\pi} P \int_{-\infty}^{\infty} \frac{\chi''(\omega') d\omega'}{\omega' - \omega} \tag{5.9} \]
Chapter 5. Absolute $\lambda$ measurements on Gd doped $YBa_2Cu_3O_{6+\delta}$

$$\chi''(\omega) = -\frac{1}{\pi} P \int_{-\infty}^{\infty} \frac{(\chi' - \chi_{\infty})(\omega')d\omega'}{\omega' - \omega}$$ \hspace{1cm} (5.10)

To first determine a value for $\chi_{\infty}$, it is necessary only to realize that at high frequency, spins would be unable to respond to an oscillating field, such that $\chi_{\infty} = 0$. One must also ensure that in the integral of Eq. 5.9 the full $\chi''$ is integrated. This means that since $\chi(\omega)$ is a physical quantity, it must obey the symmetry relation $\chi(-\omega) = \chi^*(\omega)$. Examining the real and complex parts of this statement it is found that $\chi'$ must be even, and $\chi''$ must be odd.

Using the method outlined in Appendix 1, one finds that for a Lorentzian peak occurring at $\omega_0$, the imaginary part of the magnetic susceptibility is given by:

$$\chi''(\omega) = \frac{A}{\pi} \frac{\Gamma}{2} \frac{\Gamma}{2} - \frac{A}{\pi} \frac{\Gamma}{2} \frac{\Gamma}{2},$$ \hspace{1cm} (5.11)

and the corresponding real component is found to be:

$$\chi'(\omega) = \frac{2A\mu}{\pi} \left( \frac{\omega_0^2 - \omega^2 - \frac{\Gamma^2}{2}}{(\omega - \omega_0)^2 + \left(\frac{\Gamma}{2}\right)^2} \right).$$ \hspace{1cm} (5.12)

In this experiment $\chi''(\omega)$ can be written as a sum of Lorentzian peaks in the frequency spectrum, and so a superposition of these functions can be used to represent the entire magnetic susceptibility (as seen in Fig. 5.1 and Fig. 5.2). Through broadband bolometry measurements, the energy splitting of the transitions is set, and crystal field parameters are found using the technique of [37].

It is next possible to determine the amplitude of the contribution to $X_s$ by employing the relation:

$$Z_s^2 = \frac{i\omega\mu_0(1 + \chi' - i\chi'')}{\sigma_1 - i\sigma_2} = R_s^2 - 2iX_sR_s - X_s^2.$$ \hspace{1cm} (5.13)
Expanding Eq. 5.13, and making the assumptions that $\sigma_1 \ll \sigma_2$ and $R_s \ll X_s$, it is possible to show that

$$X_s = \mu_0 \omega \lambda (1 + \chi')^{\frac{1}{2}}$$  \hspace{1cm} (5.14)

$$R_s = \frac{\mu_0^2 \omega^2 \lambda^3 \sigma_1}{2} (1 + \chi')^{\frac{1}{2}} + \frac{\mu_0 \omega \lambda \chi''}{2(1 + \chi')^{\frac{1}{2}}}$$  \hspace{1cm} (5.15)

In the analysis of bolometry measurements, a smooth function has been fitted to the first term in Eq. 5.15, whereas it is actually seen to have some resonant features due to $\chi'$. In future analysis this effect will be compensated for, however it should be mentioned that for the present this turns out to adversely affect the analysis of the resonator data.
Figure 5.2: This is a figure of the real part of the susceptibility, as calculated using the above technique to find the contribution from each Lorentzian in fig. 5.1.

Checks were made of the power dependence of the absorption in both the bolometry apparatus, and the loop-gap, to verify that the crystal remains in a linear response regime throughout the course of the experiment. In particular, the lineshapes observed in $\chi''$ should not change with the strength of the applied oscillating field. An EPMA was used to measure the gadolinium concentration in the sample of $Y_x Gd_x Ba_2 Cu_3 O_{6+\delta}$, and it was determined that $x = 0.0015$.

5.4 Results

Examining Fig. 5.4 and Fig. 5.5, in the low temperature region of $R_s$ and $\Delta \lambda$ one sees an anomaly that increases roughly as $\frac{1}{T}$. This is precisely what one would expect for a paramagnetic impurity such as gadolinium.
A first step in the analysis was to take the fit parameters from bolometry data collected on the crystal and calculate $\chi'$ and $\chi''$ for the Gd spins to see whether they can account for the anomalous features. However, the operating frequency of 0.95 GHz used in this experiment happens to fall on the edge of the low frequency resonant peak at 1.3 GHz (Fig. 5.1), a feature that is not well determined in the fits to bolometry data (note, the fit of the crystal field parameters is dominated by features at higher frequency). In the absence of time to do a detailed analysis of the low frequency peak, the values of $\chi'$ and $\chi''$ were just scaled independently so as to remove the anomalies in the low temperature resonator data. This required $\chi'$ to be 0.4 times the bolometer fit value and $\chi''$ to be 1.7 times the fit value. As these values fall within a factor of 2 of the values taken from the initial fits, the technique seems to work reasonably well. The temperature dependence of $\chi'$ and $\chi''$ appears to be correct, since the anomalies in $R_s$ and $\Delta \lambda$ can be made to disappear, to within the accuracy of the data.

By re-analysing the background fit for the bolometric data set (given by the first term in Eq. 5.15), and by making slight alterations to the crystal field parameters, it is likely that a better agreement between the resonator, and bolometry data.

It should also be noted that the sample used for this experiment was slightly twinned ($\approx 87\% a$ axis, $13\% b$ axis), and an effective penetration depth was taken to be 120 nm, which corresponds to an value taken from the initial analysis of the bolometry measurements for this particular crystal.

It can first be noted that the surface resistance of Gd doped YBCO is seen to be similar to that observed in pure YBCO (see Fig. 5.4). This is a very important finding, as it shows that the Gd does not otherwise affect the microwave electrodynamics.

In Fig. 5.5 it is possible to see that with the Gd contribution subtracted, temperature dependence of the penetration depth in the Gd doped YBCO remains linear at low temperature, and the value of the corrected slope was found to be $0.41 \pm 0.01 \text{[nmK}^{-1}]$, which is what one would expect for a undoped YBCO sample that is
Figure 5.3: $R_s$ Data taken with the bolometer shows the ESR spectrum of this sample. The crystal field fitting process is not sensitive to exactly modelling the weight of smaller peaks, due to a large background contribution.

twinned in by the same amount (in Fig. 5.5 a-axis dependence = 0.42±0.01[nmK$^{-1}$], b-axis = 0.38 ± 0.02[nmK$^{-1}$], so for a : b=87 : 13, one expects 4.1 ± 0.01[nmK$^{-1}$]).

The scattering peak in the surface resistance of YBCO is also seen to be similar to that observed in pure YBCO (see Fig. 5.4), and it can be shown that the intrinsic scattering rate [2] in the crystals is relatively unaffected by doping them with gadolinium. As samples become chemically purer and have more perfect crystallinity the scattering peak seen in Fig. 5.4 becomes more prominent, and so it is common to observe differences in this feature between samples, especially if they are not from the same growth run, as in this case.

Since a limited number of samples were grown with the gadolinium impurities, and multiple annealing runs were required to produce samples over a wide range of oxygen content, some samples used in the bolometry experiments had problems with
Figure 5.4: The corrected surface resistance data is shown along with a-axis data taken on pure YBCO. The apparatus lacked the sensitivity to measure b-axis $R_s$ on pure YBCO.

surface quality. Although this will not greatly affect the background subtraction of the first term in Eq. 5.15 for the bolometry based analysis, it will certainly affect the resonator technique. The effect most often seen in YBCO crystals with poor surface quality is an upturn of the low temperature surface resistance, away from the linear regime shown in [5]. It is possible that the upturn in $R_s$ has a contribution from surface contamination, so that the rescaling of $\chi'$ given earlier may not have to be quite as high.

This experiment also pushes the limits of the loop gap resonator to measure extremely low loss samples with precision. It can be observed that the noise in the data corresponds to changes of less than one thousand in a $Q$ of roughly one million, the maximum stability of the device. It was because of this reason that b-axis $R_s$
Figure 5.5: The enhancement, and correction of the base temperature shift in penetration depth from Gd atoms. Shown also are the $a$ and $b$ axis base temperature shifts in penetration depth.

data was not collected. Since there is less microwave absorption in the $b$-axis of the crystal, the $R_s$ contribution from $b$-axis twined regions in the sample were taken to be negligible.

The state of homogeneity of these samples is reflected in the measurements of $R_s$ near the transition from the normal to the superconducting state. In the detailed plot given in Fig. 5.6 it is possible to see that the superconducting transition is clearly kinked, with two regions. The first region to the right represents the overdoped part of the crystal, which has a sharp clean transition at approximately 89K. It is postulated that the second region, which is the long slow climb from 89K to 93K, that the inner region of the crystal is seen to be doped more likely towards optimal doping. One guesses that the crystal was probably not annealed for long enough in the constant oxygen atmosphere. As the crystal becomes penetrated deeper and
Figure 5.6: This is an example of the surface resistance seen in an inhomogeneous sample. The sharp portion of the transition to the left is indicative of a YBCO sample that has been overdoped, while the broader transition to the right would appear to be indicative of material with optimal doping ($T_c$ for optimal doping is $\approx 93K$). As the YBCO anneal is performed at relatively low temperature, diffusion times can be long, and difficulties can arise when attempting to produce a fully homogenous doping.

Deeper with magnetic fields the less doped section begins to play more of a role in the absorption.

This slight inhomogeneity only shows up very near the transition temperature, and at low temperatures where the effects due to changes in the magnetic susceptibility are most prominent in the bolometric and resonator measurements, the field only penetrates into the outer, homogenous part of the crystal.
Bibliography


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Appendix A

Kramers-Kronig calculation for a simple Lorentzian

The following is a derivation of the contribution to real part of the magnetic susceptibility for a Lorentzian shape in the imaginary component.

\[ \chi(\omega) = \chi'(\omega) - i\chi''(\omega) \]  

(A.1)

By knowing that the \( \chi' \) and \( \chi'' \) are just Hilbert transforms of each other (using Kramers-Kronig relations) and that since the susceptibility is a real response function \( (\chi(-\omega) = \chi^*(\omega)) \), one can impose the condition:

\[
\chi'(\omega_0) = \frac{2P}{\pi} \int_0^\infty \frac{\omega d\omega}{\omega^2 - \omega_0^2} \left[ \frac{1}{(\omega - \mu)^2 + (\frac{1}{2})^2} - \frac{1}{(\omega + \mu)^2 + (\frac{1}{2})^2} \right]
\]  

(A.2)

Where P just represents the Cauchy principle value of the integral. Some simple algebra will now reduce the integral to:

\[
\chi'(\omega_0) = \frac{4\mu}{\pi} P \int_{-\infty}^{\infty} \frac{\omega d\omega}{(\omega - \omega_0)(\omega + \omega_0)(\omega + \mu + i\frac{1}{2})(\omega + \mu - i\frac{1}{2})(\omega - \mu + i\frac{1}{2})(\omega - \mu - i\frac{1}{2})}
\]  

(A.3)

Expanding the integration to cover the contour given in Fig. A.1 it is easy to see that the integrand goes to zero faster than \( \frac{1}{\omega} \) in the upper integration path. In taking the contour over the upper half plane it should be apparent that the third and fifth poles in Eq. A.3 do not contribute residues to the integral. The contribution of the
principle value of the first two poles is also found to cancel and by summing up the contribution of the remaining two poles one should arrive at:

\[
\chi'(\omega_0) = \frac{4\mu}{\pi} 2\pi i \left\{ \frac{(-\mu + i \frac{\Gamma}{2})^2}{-2i\mu((-\mu + i \frac{\Gamma}{2})^2 - \omega_0^2)(-2\mu + i\Gamma)} - \frac{(\mu + i \frac{\Gamma}{2})^2}{2i\mu((\mu + i \frac{\Gamma}{2})^2 - \omega_0^2)(2\mu + i\Gamma)} \right\}
\]

Finally one arrives at the solution, which just corresponds to taking the complex part of the two initial Lorentzians (positive and negative frequency)

\[
\chi'(\omega_0) = \frac{2A\mu}{\pi} \left[ \frac{\mu^2 - \omega_0^2 + \frac{\Gamma^2}{4}}{((\omega_0 - \mu)^2 + \frac{\Gamma^2}{4})((\omega_0 + \mu)^2 + \frac{\Gamma^2}{4})} \right]
\]