1993

The effects of granularity on the microwave surface impedance of high kappa superconductors

Stephen Keith Remillard
College of William & Mary - Arts & Sciences

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The effects of granularity on the microwave surface impedance of high $\kappa$ superconductors

Remillard, Stephen K., Ph.D.
The College of William and Mary, 1993
The Effects of Granularity on the Microwave Surface Impedance of High κ Superconductors

A Dissertation
Presented to
The Faculty of the Department of Physics
The College of William and Mary in Virginia

In Partial Fulfillment
Of the Requirement for the Degree of
Doctor of Philosophy

by
Stephen K. Remillard
1993
APPROVAL SHEET

This dissertation is submitted in partial fulfillment of
the requirements for the degree of

Doctor of Philosophy

Stephen K. Remillard

Approved, November 1993

Harlan E. Schone

William J. Kossler

Dennis M. Manos

Roy L. Champion

Stuart A. Wolf
Naval Research Laboratory
To my nieces and nephews
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ABSTRACT

The microwave surface impedance of granular high temperature superconductors is an important figure of merit for technological applications. Because the behavior of the granular materials deviates significantly from that of the ideal defect free superconductors, the loss mechanisms are not fully understood. This dissertation seeks to quantify the contribution of granularity to centimeter wave and millimeter wave losses. By understanding these losses, the superconductive coupling between neighboring grains can also be understood.

The weakly coupled grain model is used as a phenomenological description of the microwave surface impedance. The granular superconducting surface is modelled as an effective resistively shunted Josephson junction. The measured surface impedance is compared to the model by plotting the normalized surface resistance versus the normalized surface reactance.

The model offers a quantitative explanation of many features observed in the surface impedance data including a local maximum in the surface reactance versus static magnetic field. The model also predicts the weaker than quadratic BCS frequency dependence of the surface resistance. The surface impedance of granular superconductors is always observed to saturate in high static magnetic fields. From analysis with the weakly coupled grain model it is concluded that the saturation is due to superconducting microshorts with properties which are independent of magnetic field.

Finally, measurement of surface resistance with an open Fabry-Perot resonator is treated within as a mini-dissertation. The loss mechanisms in the open resonator geometry are considered. The ohmic losses are computed numerically from a vector theory, and Bethe diffraction theory is used to compute a lower limit for losses arising from mode mixing.
The Effects of Granularity on the Microwave Surface Impedance of High $\kappa$ Superconductors
Chapter I

Introduction

On April 28, 1911, Heike Kamerlingh Onnes very cautiously reported to the Netherlands Royal Academy that, at two to three degrees above zero, the resistance of mercury to electrical current went to zero within the precision of his pre-World War I instruments. For 22 years this effect first seen in Hg was confused for perfect conductivity. Whereas perfect conductors offer no resistance to electricity, materials which went into Onnes’ low temperature thermodynamic phase were shown by Meissner and Ochsenfeld in 1933 to exclude all magnetic fields regardless of the magnetic history of the sample.

The new high temperature superconductors (HTSC) are ternary and quaternary ceramic materials in the BaTiO₃ perovskite family of crystal structures. Ceramics are materials composed of both metallic and non-metallic elements (usually Oxygen). Ternary and quaternary indicate three and four metallic elements respectively. Depending upon the crystal chemistry, a ternary ceramic can form multiple phases. The meaning of a phase is a particular cation stoichiometry with the corresponding anion content. For example the superconductor composed of the elements Tl, Ba, Ca, Cu and O can form Tl₂Ba₂CaCu₂O₆, TlBa₂Ca₂Cu₃O₈, and other chemical phases.

The affliction of HTSC's is multiple phase formation and small crystal size. Bulk and thick film HTSC samples are granular and often multi-phased. By means of the marvel of epitaxy, large area single crystal films are grown on single crystal...
perovskite substrates such as SrTiO$_3$. Whereas thin films on ceramic substrates may be useful in analog, digital and microwave electronics, large area superconducting devices and those including curved surfaces cannot easily be formed out of epitaxial thin films. For these applications, which are reviewed in Chapter VIII, thick film superconductors are needed. The purpose of this dissertation is to add to the understanding of the mechanisms which lead to power losses in granular HTSC's in microwave (1 GHz-100 GHz) fields. No background in superconductivity is expected of the reader, as Chapter II introduces the essential concepts of kinetic inductivity and Josephson junctions. Granular superconductivity is then introduced in Chapter III.

The microwave losses of superconductors were measured by placing them in cavity resonators and measuring the changes in cavity Q and resonant frequency. From the Q and frequency changes the surface impedance is calculated. Chapter IV discusses resonator theory and surface impedance measurement. In particular, a Fabry-Perot resonator was developed to measure surface resistance by M.E. Reeves at the Naval Research Laboratory, with contributions by the author, and is also described.

Chapter V briefly describes the numerous techniques used to manufacture samples by seven different sample contributors, including the author. The surface resistance as a function of temperature and static magnetic field was measured. The surface reactance as a function of static magnetic field was also measured. Of
interest is the result that at sufficiently high magnetic fields the surface impedance saturates. The sample is still in the superconducting state since the saturation surface resistance is lower than the normal state surface resistance.

The temperature and magnetic field dependence of the surface impedance of granular samples is dramatically different from the more ideal behavior exemplified by single crystals. For this reason a model of the grain boundary response to microwave fields is described in Chapter VI.

In Chapter VII it is shown that the grain boundary model indeed describes the surface impedance quantitatively. Furthermore, the model is used to successfully predict the surface resistance at other frequencies. In Chapter VII it is concluded, by analysis of the kinetic inductance, that the surface impedance saturates in field because the kinetic inductivity of the carriers crossing the grain boundaries saturates in field.

Chapter VIII elaborates on the device applications that partially motivate the study of granular superconductors in microwave fields. The other motivation is the purely scientific interest in the microwave response of inhomogeneous superconductors.
Chapter II

Superconductors and Superconducting Junctions

A. Introduction

Materials which offer no resistance to electrical current belong to a class of conductors called perfect conductors. Those perfect conductors which, in addition, expel all magnetic fields are called superconductors. Although in theory superconductors are but a subset of perfect conductors, all of the known perfect conductors are superconductors.

In the current state of the research field, superconductors are categorized into classical superconductors (i.e. elements and A15 compounds), exotic superconductors (heavy fermion superconductors, organics), and high temperature superconductors (HTSC). Until the discovery in 1986 by Bednorz and Müller of a cupric oxide material, La-Ba-Cu-O, with a superconducting phase transition above 30 K, superconductivity was comfortably well understood in terms of the Bardeen-Schrieffer-Cooper (BCS) theory which is described in detail by Rickayzen. In BCS theory electrons are coupled into pairs, called Cooper pairs, which can move collectively in the absence of an applied electric field. The advent of HTSC and its non-BCS like behavior has lead to a smorgasbord of new theories, modified old theories, and confusion between intrinsic and extrinsic properties.

It is the distinction between intrinsic and extrinsic properties of HTSC which
motivates this dissertation. Of interest here is energy dissipation at microwave frequencies (1GHz<f<100GHz). There exists a plethora of microwave applications of high temperature superconductors which are summarized in the last chapter. The areas of potential application range from communication to time standards to particle accelerator cavities. In this work the effect of material inhomogeneity on microwave dissipation, and ultimately on device performance, will be probed.

B. Properties of Superconductors

The expulsion of magnetic fields from superconductors, called the Meissner effect, was explained phenomenologically by F. and H. London in 1935. They used the two fluid model of Gorter and Casimir and Maxwell's equations to derive their own equation of the magnetic field in a superconductor.

Briefly, the two fluid model writes the temperature dependence of the free energy of the superconductor in terms of the population of superconducting carriers relative to normal carriers. Specifically, the total number of electrons per volume in the superconductor is \( n = n_n + n_s \), where \( n_n \) is the number of normal electrons and \( n_s \) is the number of superconducting electrons. Above the transition temperature, \( T_c \), \( n = n_n \). As a superconductor is cooled below \( T_c \), \( n_s \) continually rises from zero at the transition to \( n \) at \( T=0 \) as \( n/s = 1 - (T/T_c)^4 \). Likewise, \( n_n \) continually decreases from \( n \) at the transition to zero at \( T=0 \) as \( n_n/n = (T/T_c)^4 \). The ratio \( n_s/n \) is referred to in
Gorter-Casimir theory as the superconducting order parameter.

The magnetic field inside the superconductor is governed by the London equation

$$\vec{B} = - c \nabla \chi (L_K \vec{J})$$

(1)

where $J$ is the current density and $L_K = m/n_e e^2$ is the kinetic inductivity of the carriers to be discussed below, $m$ is the electron effective mass. What distinguishes Equation 1 from the magnetic field equation of a normal conductor is the absence of a time independent additive constant and the fact that $L_K$ contains $n_e$ instead of $n$. Using Maxwell's equations, Equation 1 can be rewritten as

$$\nabla^2 \vec{B} = \vec{B}/\lambda_L^2$$

(2)

where, in MKS units,

$$\lambda_L = \sqrt{\frac{m}{\mu_0 n_e e^2}}$$

(3)

is called the London penetration depth. $\lambda_L$ is the $e^{-1}$ penetration distance of a magnetic field into a superconductor. For the high temperature superconductors $\lambda_L \sim 10^{-7}$ m.

In the BCS theory, published in 1957, electrons of opposite momentum form phonon coupled pairs called Cooper pairs. The range of quantum mechanical phase coherence among Cooper pairs is called the coherence length, $\xi$. A physically
intuitive understanding of $\xi$ is better achieved by likening the coherence length to the spatial extent of the deBroglie wave of a conduction electron. The zero temperature coherence lengths of elemental superconductors cover over an order of magnitude from $380\ \text{Å}$ for Niobium to $16,000\ \text{Å}$ for Aluminum. Due to the anisotropy of the high temperature superconductors to be discussed later in this chapter, the coherence length of these materials depends upon orientation within the crystal. In the $c$-axis direction of $\text{YBa}_2\text{Cu}_3\text{O}_7$ (YBCO) $\xi_c \approx 3\ \text{Å}$. In the $ab$-plane $\xi_{ab} \approx 16\ \text{Å}$. In general, the high temperature superconductors are characterized by a short coherence length.

Central to the theory of superconductivity is the temperature dependent energy gap, $2\Delta(T)$, which is centered about the Fermi energy. What distinguishes this picture from that of a semiconductor is that the electrons form pairs which are bosons in a single condensate below the Fermi energy. BCS theory predicts that the transition temperature depends linearly upon the gap, $T_c \propto \Delta(0)$, and that the gap depends inversely upon the coherence length, $\Delta(T) \propto 1/\xi(T)$. Consequently, a high transition temperature corresponds to a short coherence length. Although many aspects of BCS theory do not manifest themselves in high temperature superconductors, the observed inverse correlation between $T_c$ and $\xi$ is at least consistent.

The Ginzburg-Landau parameter, $\kappa = \lambda_c / \xi$, is the ratio of London penetration depth to the coherence length. If $\kappa > 1/\sqrt{2}$ then the superconductor is said to be type II. Otherwise, the superconductor is type I. All high temperature superconductors
are type II. If a part of the superconductor were somehow driven out of the superconducting state (e.g. by application of a magnetic field) then the physical boundaries between superconducting and normal conducting regions will have a negative surface energy if $\kappa > 1/\sqrt{2}$. If part of a type II superconductor is driven out of the superconducting state by a magnetic field then it will be energetically favorable for the intruding field to maximize the normal/superconducting surface area by breaking up into small flux tubes. These flux tubes go by numerous aliases including fluxons and vortices. It is the ability to withstand high magnetic fields by forming fluxons that distinguishes type II superconductors.

A fluxon as described in the Bardeen-Stephen model is a long tube of normal conducting high magnetic field region inside the superconductor\(^{11}\). A cross section is shown in Figure 1. The diameter of the normal core region is equal to the coherence length. The order parameter, $n / n$, goes from zero at the fluxon-superconductor interface to its corresponding value for the given temperature.

\(\textbf{Figure II-1} \) Cross sectional view of a fluxon. The normal region has radius, $\xi$. 
at a distance of $\lambda_l$ from the interface. A universal characteristic of type II superconductors is that magnetic flux is quantized\(^{10}\). The magnetic flux contained in all fluxons is equal to the *flux quantum*, $\Phi_0=2.07\times10^{-7}$ Gauss-cm\(^2\). The Bardeen-Stephen picture of a fluxon is simplified from the more accurate picture which has a superconducting core with a reduced order parameter. In this model the order parameter varies continuously from zero at the center of the core to the full unsuppressed value at about two coherence lengths from the center.

The minimum magnetic field needed to introduce fluxons into the type II superconductor is called $H_{c1}$, or the lower critical field. An upper critical field, $H_{c2}$, is needed to saturate the superconductor with fluxons and drive the entire superconductor into the normal conducting state. Typical values at zero temperature for HTSC are $H_{c1}\approx10^5$ Oe and $H_{c2}\approx10^6$ Oe. Both $H_{c1}$ and $H_{c2}$ are zero at and above $T_c$. Once in the superconductor, fluxons are positioned in a hexagonally close packed arrangement in order to minimize the free energy. The fluxon arrangement will deviate from this HCP "lattice" due to the presence of material defects. It is energetically more favorable for fluxons to sit at defects, and this defect positioning is called *pinning*.

In order for a superconductor to diamagnetically exclude an applied static magnetic field, $H_{dc}$, there must be shielding currents occupying a volume at the surface one London penetration depth deep. For a type I superconductor, when the energy of magnetization, $H_{dc}^2/8\pi$, exceeds the difference in free energy between the normal and superconducting state the superconductor reverts to the normal state.
The value of $H$ at which this transition occurs is called the thermodynamic critical field, $H_c$. The density of surface eddy currents which shield a field of $H_c$ is called the critical current density, $J_c$. If a current density of $J_c$ is passed through a superconductor it will return to the normal state under the influence of its own self-field.

Because a type II superconductor is able to herd the applied field into tiny high flux vortices, the bulk critical behavior deviates significantly from the simple free energy argument above. The thermodynamic critical field is only of local relevance. On the macroscopic level, the lower and upper critical fields mark the transitions from bulk superconductivity to the mixed state, and from the mixed state to the normal state, respectively.

A macroscopically relevant critical current in type II superconductors corresponds to the amount of current which depins the vortices. At low current the vortices are fixed by material defects which establish potential wells that favor a normal region. An applied current, transverse to the fluxon axis, tilts the potential and reduces the thermal energy needed to activate the fluxons\textsuperscript{12}. When a fluxon is depinned it moves under the force of the Lorentz interaction with the supercurrent. This force leads to dissipation. The superconductor then exhibits an electrical resistivity\textsuperscript{11} $\rho = \Phi_0 B / n \eta$ where $\eta$ is the viscosity which characterizes the motion of a single fluxon. Since a moving fluxon is essentially a moving normal region, the fluxon has an inertia characterized by the kinetic inductance of unpaired carriers $\xi = m / ne^2$. 
In general, type II superconductors are more resilient to high magnetic fields and large currents. It was the discovery of type II superconductivity in the A15 compounds, such as Nb₃Sn, which lead to the realization of Kammerlingh Onnes' vision of practical superconducting magnets.

C. Josephson Junctions

Superconductivity is a collective quantum mechanical phenomenon. Cooper pairs have integer spin and collect in a condensed state. All of the pairs are in the same quantum mechanical state and are collectively described by the same wave function.

Two neighboring superconductors will have two separate condensates. If these two superconductors are brought into close proximity then the wave functions will overlap and the superconductors are said to be coupled. In 1962, Brian Josephson published a theory of the junction which exists between two superconductors in close proximity. He showed that it is theoretically

![Figure II-2 I-V characteristic of a Josephson junction. When the maximum zero voltage current is exceeded the junction goes into the voltage state. Hysteresis is indicated by the arrows.](image-url)
possible for pairs to tunnel across the junction with no potential difference established between the two individual superconductors. This can be contrasted to tunneling in normal metal films where a potential difference must exist between two conductors in close proximity if a current is to flow between them. The theory of Josephson junctions is treated in detail, for example, by Kulik and Yanson\textsuperscript{14} and by Barone and Paterno\textsuperscript{15}.

Current can pass through a Josephson junction with no applied voltage up to the junction critical current density, $J_{cJ}$. The critical current density can be exceeded only by applying a voltage, $V$. Likewise, if the junction is driven above $J_{cJ}$, a potential difference across the junction will occur. The current voltage characteristic for a Josephson junction is shown in Figure II-2, where $I$ is the current density times the junction area. The zero voltage part of the characteristic is indicated by a solid line at $V=0$. When $J_{cJ}$ is exceeded, the junction goes into the voltage state. In the voltage state there is an energy price for conduction across the junction. If $J_{cJ} \ll J_c$ then the two superconductors are said to be \textit{weakly coupled}.

There is also a corresponding penetration depth for a Josephson junction given by\textsuperscript{16}

$$\lambda_J = \sqrt{\frac{\hbar}{2eJ_{cJ}\mu(2\lambda_L+d)}}$$  \hspace{1cm} (4)

where $d$ is the thickness of the junction. This thickness is included because there is often an insulating or normal metal layer between the superconductors. $\lambda_J$ is the
depth from the surface into the junction that a magnetic field applied to the superconductor-junction-superconductor surface will penetrate. Typical values for the granular high temperature superconductors are $J_{cj} \approx 10^7$ A/m$^2$ and $\lambda_L = 1.5 \times 10^{-7}$ m yielding $\lambda_j \approx 10^{-5}$ m. In general $\lambda_L \ll \lambda_j$.

Since all of the paired carriers in the superconductor bulk are correlated quantum mechanically, all pairs are described by a collective wave function. If no phase difference exists between the two superconductors of a Josephson junction then there is no preferred tunnelling direction and no Josephson current flows. The direction of the Josephson current depends on the phase difference, $\Delta \phi$, and is governed by\(^\text{17}\)

$$J = J_{cj} \sin(\Delta \phi) . \quad (5)$$

If a current the size of $J_{cj}$ passes through a Josephson junction it is only because the phase difference is $\pi/2$ (or vice versa).

Josephson hypothesized\(^\text{18}\), and Rowell\(^\text{19}\) demonstrated experimentally, that the phase difference across a junction is position independent only in the absence of a magnetic field. When a magnetic field is applied to a junction $\Delta \phi$ varies along the junction. The maximum amount of current that can cross the junction, and the direction of the current, then varies along the junction according to Equation 5. If the magnitude of applied field is such that $\Delta \phi$ varies by exactly $2\pi$ across the junction then no net current flows and the junction is decoupled. The field needed to establish this condition is called the junction critical field, $H_{cj}$. The minimum
Figure II-3 The critical current, $I_c$, of a Josephson junction is modulated by an external magnetic field. $H_{c1J}$ is the junction critical field.

The magnetic field at which fluxons can first nucleate in the junction is called $H_{c1J}$. The maximum static current that can cross a junction versus applied field is shown in Figure II-3. Because it resembles an optical Fraunhofer diffraction pattern it is often referred to as the junction Fraunhofer pattern.

D. Kinetic Inductance

Because electrons have non-zero mass their motion under the influence of an electric field, $E$, is limited by their inertia. The limiting effect of carrier inertia upon the conductivity, $\sigma$, of good conductors is dealt with in the electron gas model by
solving the equation of motion

$$\frac{d<v_x>}{dt} + \frac{m<v_x>}{\tau} = -eE$$

(6)

where \(<v_x>\) is the average electron velocity in the x direction, \(m\) is the electron mass, and \(\tau\) is the collision relaxation time.

If the electric field has a harmonic time dependence, \(e^{int}\ (j=\sqrt{-1})\), and \(\sigma E=ne<v_x>\) is substituted into Equation 6, where \(n\) is the carrier concentration, then the Drude conductivity is acquired

$$\sigma = \frac{ne^2\tau}{m(1-j\omega\tau)}.$$  \hspace{1cm} (7)

In a perfect conductor \(\tau \rightarrow \infty\), and \(\sigma^{-1}=-jm\omega/ne^2\) is the specific impedance. This limiting impedance is the specific reactance of an inductive response to an AC field. It is equivalently written as

$$\sigma = \frac{j}{\omega L_K}$$  \hspace{1cm} (8)

where \(L_K=m/ne^2\) is called the kinetic inductivity of the carriers and has dimensions of Henry-meters. It governs the acceleration of a charge in an electric field and also results in the phase difference of 90° between the AC electric field and the resulting AC current in a perfect conductor.

In a superconductor, the kinetic inductivity is related almost entirely to the
paired carriers. This is because collisions dominate the impedance to the motion of the normal, unpaired carriers and their kinetic inductivity is consequently negligible compared to their resistivity. Thus, for a superconductor

\[ L_K = \frac{m^*}{n^2 e^*^2} \]  

(9)

where \( m^* \) and \( e^* \) are the mass and charge of a Cooper pair. From Equation 3

\[ L_k = \mu_o \lambda_L^2. \]  

(10)

Thus, the penetration of magnetic field into the surface of a superconductor depends upon the inertia and density of the carriers.

E. Surface Impedance

Maxwell's boundary conditions for electromagnetic fields require that the normal component of the magnetic induction, \( B_N \), and the transverse component of the electric field, \( E_T \), at the surface of a perfect conductor must vanish. An AC field at the surface of a perfect conductor induces AC shielding surface currents, \( K = n_x H_T \), in the conductor. At zero frequency the normal conducting electrons do no move since there is no electric field at the surface. However, at high frequency, the kinetic inductivity of the charge carriers impedes the shielding currents, and the normal conducting electrons are no longer perfectly shunted by the supercurrent. Consequently at high frequency there is a current of normal electrons dissipating
energy. If currents are impeded then an electric field parallel to the currents must exist. If the impedance is purely inductive then the electric field is 90° out of phase with the current and no power is dissipated. If the material's conductivity contains a real term then the phase between $E$ and $K$ is between 0° and 90° and the impedance is complex. Under these circumstances power is dissipated.

A superconductor's complex conductivity possesses a real part, $\sigma_1$, which in the two fluid model is frequency dependent. At zero frequency $\sigma_1=0$. Recall that the first of the two properties of superconductors stated above is no resistance to DC current. At high frequency the conductivity of a superconductor is complex leading to non-negligible power dissipation.

The component of an electric field parallel to a conducting surface, $E_\parallel$, is linearly proportional to the surface current, $K=n\times H$, where $n$ is normal to the surface. The complex coefficient of proportionality is called the surface impedance, $Z_s = |n \times H|$. From elementary electromagnetic wave theory the characteristic impedance of a medium is $Z=(\mu/\epsilon)^{1/2}$ where $\mu$ and $\epsilon$ are the permeability and permittivity of the medium respectively. At a material interface $Z$ is the surface impedance, $Z_s$. The complex conductivity of a medium is $\sigma=j\omega\epsilon$. So the surface impedance is

$$Z_s = R_s - jX_s = \sqrt{\frac{j\omega\mu}{\sigma}}$$

(11)
where the real part is called the surface resistance and the imaginary part is the surface reactance. The physical interpretations of $R_s$ and $X_s$ are that surface resistance is a measure of energy loss and surface reactance is a measure of field penetration. The relation between surface reactance and field penetration is forged by the kinetic inductivity. If $L_k = 0$ then an infinitesimally thin eddy current layer can shield the AC field. Thus, if $L_k = 0$ then the depth of field penetration is also zero.

Surface resistance also correlates to field penetration. If the field cannot penetrate, then the conductor-vacuum boundary is defined perfectly and none of the electron gas is exposed to AC field. In fact, Equation 11 is equivalent to $Z_s = (1-j)/(\sigma \delta)$, where $\delta = (2/\omega \mu_0 \sigma)^{1/2}$ is the skin depth. For a good normal conductor, with $\tau \omega \ll 1$, $\sigma$ and $\delta$ are both real and $R_s = X_s$. Because the conductivity of a superconductor is anomalous at low frequency $\sigma$ is complex and, in general, $R_s \neq X_s$.

The surface resistance of bulk superconductors was handled in terms of BCS theory by Halbritter in 1974\textsuperscript{23}. If the superconducting material is granular, the BCS behavior is not observed due to the influence of the granularity. Macroscopic deviations from the microscopic behavior result, including smearing of the phase transition\textsuperscript{24}. This granularity contributes to enhanced power loss at the surface\textsuperscript{25}. The enhanced losses are related to the deeper field penetration, which also results from granularity as shown by Hylton et al.\textsuperscript{26}.

The complex conductivity used in Equation 11 was calculated from BCS theory by Mattis and Bardeen\textsuperscript{27}. Their calculations of $\sigma = \sigma_1 - j \sigma_2$ were performed in the extreme anomalous limit where $\xi$ is much greater than the penetration of field
into the material. At high frequencies the penetration depth is limited by the skin effect (described by the skin depth, \( \delta \)) and is consequently smaller than the London depth. When \( \delta < \xi \), the field varies significantly over one mean free path and nonlocal electrodynamics must be used. Nonlocality is not the case for HTSC at microwave frequencies\(^{28}\).

F. High Temperature Superconducting Materials

Since the early 1960's a plethora of oxide superconductors has been reported in the literature. Oxide materials which exhibit superconducting phase transitions are plagued by defects, low critical temperature and thermodynamic instability\(^{29}\). Much of the work in oxide superconductivity in the 1960's involved a family of compounds called the oxide bronzes. Many of these compounds include alkali earths and Tungsten oxide. \( \text{Sr}_{0.98}\text{WO}_3 \), for example, has a transition temperature of 4 K\(^{30}\). The highest known \( T_c \) for an oxide material before 1986 was the 13.7 K transition of \( \text{LiTi}_2\text{O}_4 \)^\(^{31}\).

Since Bednorz's and Müller's 1986 discovery of superconductivity in the \( \text{La-Ba-Cu-O} \) series, the study of Copper oxide superconductivity has resulted in higher transition temperatures, higher material qualities, and a greater understanding of metal oxide thermodynamic instability\(^{29}\). The fundamental material characteristic of high temperature superconductors is their Cu-O perovskite structure. The standard crystal model is that of cubic \( \text{BaTiO}_3 \). Although the HTSC materials typically have
an orthorhombic structure, the close lattice matching in the [001] plane renders the BaTiO₃ perovskites good substrate materials for HTSC thin film deposition. The HTSC lattices are composed of groups of neighboring Cu-O planes separated by metal oxide planes containing metals other than Copper, with planar separation of 3.2 Å. The particular materials used in this work, their Tc's, and the number of neighboring Cu-O planes is summarized in Table II-1. The Bi-Sr-Ca-Cu-O series has three superconducting phases denoted by Bi-2201, Bi-2212, and Bi-2223. In the Bi-2201 lattice all of the Cu-O planes are separated by Calcium planes. In Bi-2212 there are two neighboring Cu-O planes separated by metal oxide planes. Finally, in Bi-2223 there are three neighboring Cu-O planes. This trend of higher Tc stemming from more Cu-O planes is also seen in the Tl-Ba-Ca-Cu-O family. The direct correlation between the number of Cu-O planes and Tc continues in the Tl-series until there are more than four Cu-O planes, and in the Bi-series until there are more than three planes.

The relationship between oxygen deficiency and Tc has been an important issue. If the oxygen stoichiometry is lower than that shown in Table 1 then the Tc will be lowered. For example, the Tc of Y-123 drops continuously from 93 K to 0 K as the oxygen stoichiometry changes from O₆.₉₆ to O₆.₅. It is common practice to denote the uncertainty of the oxygen stoichiometry in samples when writing their formula. For example, one usually writes YBa₂Cu₃O₇₋₈ for Y-123.

The YBa₂Cu₃O₇₋₈ material serves as the basis for another group of materials denoted by RBa₂Cu₃O₇₋₈ where R=rare earth. Superconducting materials result for
all of the rare earths except Ce for which no compound can be formed, and Pr for which no superconducting phase transition has been observed\textsuperscript{29}.

Since the work reported here was completed the $T_c$ record has moved upward to 135 K in the Hg-Ba-Ca-Cu-O series\textsuperscript{34}. Chu et al.\textsuperscript{35} report that under high isostatic pressure the $T_c$ may be higher than 150 K for some phases. The difficulty in synthesizing the Hg compounds (Hg-1212 and Hg-1223) at ambient pressure was overcome by Chu and his coworkers via controlled vapor/solid reaction. A precursor of nominal stoichiometry Ba$_2$Ca$_{n-1}$Cu$_n$O$_x$ is prepared and sealed with HgO inside a quartz tube. Because the reaction is Hg (vapor) + precursor (solid), the decomposed Hg vapor forms the superconducting Hg-Ba-Ca-Cu-O compound above \~800°C. The $T_c$'s are 125 K for Hg-1212 and 135 K for Hg-1223. Due to the low decomposition temperature of Tl$_2$O$_3$, the controlled vapor/solid reaction is also used to prepare the Tl series materials.
<table>
<thead>
<tr>
<th>Formula</th>
<th>Notation</th>
<th># Cu-O planes</th>
<th>$T_c$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tl$_2$Ba$_2$CuO$_6$</td>
<td>Tl-2201</td>
<td>1</td>
<td>0-80 K</td>
</tr>
<tr>
<td>Tl$_2$Ba$_2$CaCu$_2$O$_6$</td>
<td>Tl-2212</td>
<td>2</td>
<td>108 K</td>
</tr>
<tr>
<td>Tl$_2$Ba$_2$Ca$_2$Cu$<em>3$O$</em>{10}$</td>
<td>Tl-2223</td>
<td>3</td>
<td>125 K</td>
</tr>
<tr>
<td>Bi$_2$Sr$_2$CuO$_6$</td>
<td>Bi-2201</td>
<td>1</td>
<td>0-20 K</td>
</tr>
<tr>
<td>Bi$_2$Sr$_2$CaCu$_2$O$_8$</td>
<td>Bi-2212</td>
<td>2</td>
<td>85 K</td>
</tr>
<tr>
<td>Bi$_2$Sr$_2$Ca$_2$Cu$<em>3$O$</em>{10}$</td>
<td>Bi-2223</td>
<td>3</td>
<td>110 K</td>
</tr>
<tr>
<td>Y$_2$Ba$_4$Cu$<em>4$O$</em>{14}$</td>
<td>Y-247</td>
<td>2</td>
<td>40 K</td>
</tr>
<tr>
<td>YBa$_2$Cu$_4$O$_8$</td>
<td>Y-124</td>
<td>2</td>
<td>80 K</td>
</tr>
<tr>
<td>YBa$_2$Cu$_3$O$_7$</td>
<td>Y-123</td>
<td>2</td>
<td>93 K</td>
</tr>
</tbody>
</table>

Table 1-1: Notation, number of neighboring Cu-O planes and transition temperatures of the different phases of the superconductor families dealt with in this work. (Taken from reference 6, Chapter 3)
Chapter III

Granular Superconductors

A. Issues of Granularity

The origin of granularity in thin films and its effect on electrical properties is reviewed by Ohring\textsuperscript{36}. In film deposition thermodynamics dictates a maximum area over which crystalline order is preserved. In the case of thin films, lattice matching to the substrate is important to guarantee large grain growth. Substrate temperature and deposition rate are also critical parameters which dictate film quality. Although properties of granular thick films and granular bulk materials are treated here, other authors have used epitaxial films to study the material and electrical properties of single grain boundaries. The results of such work are summarized below and in part B are applied to polycrystalline materials.

The Homogeneous Limit, $\xi \gg a$

The effect of granularity on the superconducting properties of a surface depends on the relative size of the grains to the coherence length. Two limiting regimes can be considered\textsuperscript{37}. In the first case, $\xi \gg a$, where $\xi_0$ is the intrinsic coherence length, and $a$ is the grain size. Here a nucleated fluxon will be larger than any grain and it will see a homogeneous superconductor with an effective coherence length, $\xi_{\text{eff}}$. $\xi_{\text{eff}}$ is considerably shorter than the intrinsic $\xi_0$ and is given by\textsuperscript{37}
\[ \xi_{\text{eff}} = \sqrt{\frac{\pi \hbar}{32e^2 N(0)\rho_n k_B (T_C - T)}} \]  

where \( N(0) \) is the normal state density of states at the Fermi level and \( \rho_n \) is the normal state resistivity of the grain boundary junction.

The effective penetration depth of a granular film, \( \lambda_{\text{eff}} \), will be determined by the Josephson penetration of the grain boundaries given by Equation 4 as well as the London penetration depth. A small Josephson critical current corresponds to a large penetration depth, \( \lambda_{\text{eff}} \propto (J_{\text{cr}})^{-1/2} \).

Aluminum has served as the primary material of study for granular low temperature superconducting films. Although bulk aluminum is a type I superconductor, if the grains are 500 Å or less in breadth then Al will exhibit type II behavior. Bulk aluminum has a zero temperature coherence length of about 1.6x10^4 Å. But a granular Al film with a ~20 Å will have a coherence length of \( \xi_{\text{eff}} \approx 20 \) Å. The reduced effective coherence length and enhanced effective penetration depth combine to give a significantly larger effective Ginzburg-Landau parameter, \( \kappa_{\text{eff}} \). It is due to this larger \( \kappa_{\text{eff}} \) that a type I bulk material can become a type II film. Because the effective coherence length is so short, it is often argued that granular LTSC films in the homogeneous limit resemble HTSC films.

In compliance with the enhanced Ginzburg-Landau parameter, granular superconducting films in the homogeneous limit typically exhibit an enhanced \( T_C \). The microscopic theory of \( \kappa \) and \( T_C \) enhancement remains controversial.
prevailing school of thought has been that the phonon modes soften in the vicinity of the grain boundaries\textsuperscript{37}. Mode softening refers to the bending of the optical modes down to the frequency of the acoustic modes at the edge of the first Brillouin zone. This results in an strengthened electron-phonon coupling which leads to an enhanced $T_C$. A second school of thought\textsuperscript{40} is that there is a reduction in the electron screening at the grain boundary resulting in stronger attractive and repulsive interactions.

A final consideration is that of fluxon motion. In the homogeneous limit the fluxons are larger than the grains. Crystalline defects are usually relied upon to pin the fluxons. When the size scale of defects, $a$, is smaller than the size scale of fluxons, $\xi_0$, then there is little to which the fluxons can pin. Thus, it is expected that granular superconducting films in the homogeneous limit should have a low critical current. This was indeed observed in the Aluminum films of Horn and Parks\textsuperscript{41}.

**The Inhomogeneous Limit, $\xi \ll a$**

For small coherence length the nucleated fluxons are much smaller than the grains and the material is in the inhomogeneous limit. This is the limit of granular high temperature superconductors. In this limit a grain boundary is long compared to $\xi_0$ and fluxons can be confined to the grain boundary regions. Because the grain size is large relative to the penetrating magnetic flux tubes, the penetration depth for a fluxon depends upon whether it is in a grain or a grain boundary. In the
inhomogeneous case, the effective penetration depth is governed by the intrinsic London depth and the Josephson depth and is

\[ \lambda_{\text{eff}} = \sqrt{\frac{\lambda_L^2 + \frac{2\lambda_L}{a} \lambda_J^2}{a}} \]  

as will be demonstrated in Chapter 6.

Because the value of \( H_{\text{C1}} \) for HTSC is considerably less than \( H_{\text{C1}} \) for the grains (~1 Oe versus ~100 Oe) fluxons nucleate much more readily, and move much more easily, in the grain boundaries. Regarding Figure II-3 it is surmised that the grains of HTSC decouple in fields on the order of 1 Oe.

The critical current of a granular superconductor is determined entirely by Josephson tunneling across the grain boundaries. In the case of any Josephson junction the critical current is that of the junction and not the intrinsic \( J_c \) of the bulk superconductor. The junction critical current density depends upon the junction length, \( a \), the junction's normal state resistance, \( R_n \), and the energy gap of the superconductor, \( \Delta \), as

\[ J_{cj} = \frac{\pi \Delta(T) \tanh\left(\frac{\Delta(T)}{2k_B T}\right)}{R_n a^2} \].

Using \( \rho_n = 10^{-4} \Omega \text{m} \), \( a = 10 \mu\text{m} \) and \( \Delta(0) = 20 \text{ meV} \), Deutscher estimates that a granular HTSC samples should have \( J_c \approx 3 \times 10^5 \text{ A/cm}^2 \). However, typical ceramic and thick film superconductors have \( J_c \) between 2000 A/cm² and 20,000 A/cm² at zero temperature. Deutscher's explanation for the discrepancy is given in reference to
the results of Mannhart et al.\textsuperscript{44} that the energy gap at the grain boundary is suppressed by as much as 50\% from its value at the center of the grain. In general

\[
\frac{\Delta_{gb}(T)}{\Delta_g(T)} \propto \sqrt{T_c - T}
\]

where the subscript $gb$ refers to the grain boundary and $g$ refers to the grain. So, the discrepancy between the assumption of uniform order parameter and the real situation grows as $T_c$ is approached. In fact, Deutscher demonstrates that order parameter suppression accounts quantitatively for the grain boundary suppressed $J_c$.

\section*{B. HTSC Grain Boundaries}

Optical micrographs illustrating the grain structure of the HTSC materials examined here will be presented in Chapter 4. The surface of a single crystal of the ceramic materials is chemically altered from that of the bulk. In particular the valency of the Copper ions at the surface will differ from the bulk due to oxygen deficiency. Goddard\textsuperscript{45} argues that there could well exist an insulating surface layer roughly 4Å thick at the grain. Consequently, a clean grain boundary could have an 8Å thick insulation layer between the grains. This is particularly likely when the $c$-axes of the two grains are not perfectly oriented and unreconstructed surfaces are consequently exposed.

The surface chemistry of the HTSC grains is very complicated and not fully understood. In general, a suppression of the density of states at the Fermi level is
observed. This leads to a thin non-superconducting layer at the grain boundaries\textsuperscript{46}. Thus, most HTSC grain boundaries are Superconductor-Normal Superconductor (SNS) or SINIS (I=Insulator) junctions.

By studying the transport properties of individual grain boundaries, Dimos et al.\textsuperscript{47} found that the superconductive coupling between grains was independent of the orientation angle between the $a$ and $b$ axes of the two neighboring grains. They found that the superconductive coupling was weak and that it was due to structural disorder at the grain boundaries.

The superconducting coupling strength is characterized by the parameter\textsuperscript{38}

\[
c = \frac{4e^2 R_n V_g N(0) k_B (T_C - T)}{\pi \hbar}
\]

where $V_g = a^3$ is the grain volume and $R_n^{-1}$ is the slope of the current-voltage curve of the grain boundary in the voltage state. The two coupling regimes are:

1. weak coupling. $c \gg 1, \xi \ll d, R_n$ large,
2. strong coupling. $c \ll 1, \xi \gg d, R_n$ small,

where $d$ is the thickness of the grain boundary layer. The granular high temperature superconductors have weakly coupled grains, as well as areas of strong coupling such as superconductive microshorts.
A. Pillbox Cavity

1. Theory of Cylindrical Cavity Resonators

The theory of cavity resonators builds naturally out of waveguide theory. One simply restricts the waveguide to a small segment with conducting endwalls. Waveguide theory is handled in detail by Lewin and by Beatty. Waves inside a waveguide are periodic in the longitudinal, $e_z$, direction. Thus, the field vectors are separable in the longitudinal and transverse directions and are written as $E(x,y)e^{jkz-j\omega t}$ and $H(x,y)e^{jkz-j\omega t}$. In light of this separability, the two-dimensional wave equation for the fields,

$$\left(\nabla^2 + \gamma^2\right)\begin{pmatrix} E \\ H \end{pmatrix} = 0,$$

where

$$\gamma = \mu \varepsilon \frac{\omega^2}{c^2} - k^2,$$

is satisfied by the $e_z$ field components alone. A resonant cavity, shown in Figure IV-1, may be physically constructed by placing metallic walls at the ends of a short waveguide segment. Such a cavity is mathematically constructed by placing boundary
conditions on the longitudinal, $\hat{e}_x$, field components. From Maxwell's equations the magnetic field, $\mathbf{H}$, can have no component normal to a perfect conductor, and the electric field, $\mathbf{E}$, can have no component parallel.

In a waveguide the transverse fields are given by the transverse gradient of the longitudinal field

$$E_t = \pm \frac{jk}{\gamma^2} \nabla_z (E_x e^{\pm jkz}) = \pm \frac{jk}{\gamma^2} \nabla_z \psi_{TM}(x,y) \quad TM \quad (19)$$

$$H_t = \pm \frac{jk}{\gamma^2} \nabla_z (H_x e^{\pm jkz}) = \pm \frac{jk}{\gamma^2} \nabla_z \psi_{TE}(x,y) \quad TE \quad (20)$$

where the TM and TE designations are for transverse magnetic and transverse electric respectively. If the longitudinal field components for the TE and TM modes are instead expressed as $\psi_{TE} e^{\pm jkz}$ and $\psi_{TM} e^{\pm jkz}$ respectively, then the fields inside a closed cavity resonator are more conveniently derived.

Inside a cavity the fields form standing waves, $A \cos(kz) + B \sin(kz)$, which
satisfy the boundary conditions

\[ E_z \big|_s = 0 \quad TM \] \hspace{1cm} (21)

\[ \frac{\partial B_z}{\partial n} \big|_s = 0 \quad TE \] \hspace{1cm} (22)

where \( n \) is the coordinate normal to the metallic surface, \( s \), and \( A \) and \( B \) can be either real or imaginary. From Maxwell's curl equations the transverse \( E \) and \( H \) fields are related by

\[ H_t = \pm \frac{\hat{e}_z \times E_i}{Z} \] \hspace{1cm} (23)

where \( Z \) is the characteristic wave impedance. Spatial modulation of \( E_i \) and \( H_i \) in the \( \hat{e}_z \) direction in Equations 19 and 20, and application of Equation 23, give
For a cylindrical resonant cavity the fields can be found exactly. Using polar coordinates, \( \psi(\rho, \phi) \), satisfies the two dimensional wave equation

\[
(\nabla_t^2 + \gamma^2) \psi(\rho, \phi) = 0
\]

or

\[
(-\frac{\partial^2}{\partial \rho^2} + \frac{1}{\rho} \frac{\partial}{\partial \rho} + \frac{1}{\rho^2} \frac{\partial^2}{\partial \phi^2} + \gamma^2) \psi(\rho, \phi) = 0.
\]

Using the usual separation of variables with \( \psi(\rho, \phi) = R(\rho) \Phi(\phi) \), the solutions are \( \Phi = e^{\pm \im \phi} \) and \( R = J_m(\gamma_{mn} R) \), where \( J_m \) is the \( m \)th order Bessel function and \( R \) is the cylinder radius. The boundary conditions, Equations 21 and 22, allow \( \gamma \) to be expressed in terms of the zeros of the Bessel functions, \( \gamma_{mn} \).
\[ TM \quad E_z|_s = 0 \quad \rightarrow \quad J_m(\gamma_{mn}R) = 0 \quad \rightarrow \quad \gamma_{mn} = \frac{X_{mn}}{R} \quad (30) \]

\[ TE \quad \frac{\partial B_z}{\partial n}|_s = 0 \quad \rightarrow \quad \frac{\partial J_m(\gamma_{mn}R)}{\partial \rho} = 0 \quad \rightarrow \quad \gamma_{mn} = \frac{X'_{mn}}{R} \quad (31) \]

where \( X'_{mn} \) indicates the \( n \)th zero of the derivative of the \( m \)th Bessel function.

At this point the fields in a cylindrical cavity can be written. For \( TE_{mnp} \) modes, using Equation 26,

\[ H_z = E_0 J_m \left( \frac{X'_{mn}}{R} \rho \right) \sin \left( \frac{p \pi}{d} \right) \cos m \phi \quad (32) \]

\[ H_\rho = E_0 \frac{p \pi}{d} \frac{R}{X'_{mn}} \cos \left( \frac{p \pi}{d} \right) \cos (m \phi) J'_m \left( \frac{X'_{mn}}{R} \rho \right) \quad (33) \]

\[ H_\phi = -E_0 \frac{p \pi}{d} \frac{R}{X'_{mn}} \frac{m}{\rho} \cos \left( \frac{p \pi}{d} \right) \sin (m \phi) J'_m \left( \frac{X'_{mn}}{R} \rho \right) . \quad (34) \]

Likewise, the \( TE_{mnp} \) electric fields can be found by evaluating equation 27.

Given the fields inside a cavity resonator the power dissipated in the wall material can be calculated. The power lost to an area element in a resonator is proportional to the square of the surface current, \( K = n \times H_b \) which is in turn equal (in MKS units) and normal to the surface magnetic fields,
2. Surface Resistance

With the $E$ and $H$ fields inside a cylindrical resonant cavity known from Maxwell's equations, the surface resistances, $R_s$, of the resonator materials can be measured. The lossy conductors which compose the physical resonator damp the oscillations at resonance. If a cavity resonator is constructed from a lossless material the input impedance of the resonator is pure imaginary at resonance and real and infinite at some small difference from the resonant frequency. Thus, the power transfer through the cavity is a delta function at resonance. However, if the resonator is made of a lossy conductor the electromagnetic boundaries of the cavity are not well defined due to the skin effect. This causes a finite region in frequency domain where a resonance can be supported.

The quality factor of a resonator is the resonance frequency, $f_r$, divided by the spread of the resonance in frequency domain, $\Delta f$, as depicted in figure IV-2. This

$$dP = \frac{R_s}{2} |(n \times H_y)|^2 da.$$  \hspace{1cm} (35)

$R_s$ is the surface resistance of the resonator material and has units of Ohms.

![Figure IV-2](image)
simple expression of resonator Q comes from the definition

\[ Q = 2\pi f \frac{\text{stored energy}}{\text{power dissipated}}. \]  \hspace{1cm} (36)

The stored energy, \( W \), is that of the entire resonator system. In a cylindrical resonant cavity the system is composed of the metallic boundaries and the couplers which carry the microwave radiation into the cavity. These couplers are usually small loop antennas when the operating frequency is below about 21 GHz.

Because the resonator Q depends inversely on the power dissipated, the Q can be written in terms of component Q's. So, where \( Q_L \) is the loaded or measured Q,

\[ Q_L = \omega_r \frac{W}{P} = \omega_r \frac{W}{P_{\text{cavity}} + P_{\text{coup}}} . \]  \hspace{1cm} (37)

The power dissipation in the expression for \( Q_L \) is the power dissipated throughout the entire system. The term loaded Q arises from the fact that the resonator is loaded by external circuitry. In this case the external circuitry is the couplers. In general,

\[ \frac{1}{Q_L} = \frac{1}{\omega_r W (\sum P_i)}. \]  \hspace{1cm} (38)

The cavity and coupling Q's can be analyzed separately. If the cavity is divided into top, bottom, and cylinder then one can speak of a top \( Q_t \), bottom \( Q_b \), and cylinder \( Q_{cy} \). It is important to realize that only \( Q_L \) is found from simply measuring \( f_c/\Delta f \). The component Q's in such a measurement are not associated with any bandwidth. Bandwidth is a property of the entire resonator.
The unloaded Q of the cavity, $Q_{cav}$, is

$$\frac{1}{Q_{cav}} = \frac{1}{Q_t} + \frac{1}{Q_b} + \frac{1}{Q_{cyl}}$$  \hspace{1cm} (39)

where $Q_t$ represents either a copper top or a sample top. If a value for $1/Q_b + 1/Q_{cyl}$ is known along with a measured value of $Q_{cav}$ then a $Q$ for the top is known. $1/Q_b + 1/Q_{cyl}$ can be calculated geometrically from a measured value of $Q_{cav}$ for a cavity in which the same material is used for all surfaces. If $Q_{cav}$ for the homogeneous resonator is known then

$$\eta = \frac{1}{Q_{cav}} = \frac{P_{cyl} + P_b + P_t}{P_{cyl} + P_b}.$$  \hspace{1cm} (40)

The power ratio in Equation 40 is calculated from

$$P = \frac{1}{2} R_s \int \int |H_1|^2 ds$$  \hspace{1cm} (41)

where $H_1$ is the component of the real part of the magnetic field parallel to the surface of integration. For the TE$_{011}$ mode, from equations 32, 33 and 34

$$H_\phi = 0.$$  \hspace{1cm} (42)
\[ H_p = -j \frac{2\pi R}{LX_{01}'} H_0 J_0' \left( \frac{\rho X_{01}'}{R} \right) \cos \left( \frac{\pi z}{L} \right) \]  

(43)

and

\[ H_x = -j 2H_0 J_0 \left( \frac{\rho X_{01}'}{R} \right) \sin \left( \frac{\pi z}{L} \right) \]  

(44)

so that

\[ P_{cyl} = 2\pi RH_0^2 R_s J_0^2 \left( X_{01}' \right) \]  

(45)

\[ P_b + P_t = 2\pi R^2 H_0^2 R_s \left( \frac{\pi R}{LX_{01}'} \right)^2 J_0^2 \left( X_{01}' \right). \]  

(46)

These power integrals are used to calculate \( \eta_{011} \) or, for the general \( TE_{01p} \) modes,

\[ \eta_{01p} = \frac{1 + 2 \left( \frac{R}{X_{1,1}} \right)^3}{1 + \left( \frac{R}{X_{1,1}} \right)^3} \]  

(47)

When the top is a sample, the sample \( Q, Q_s \) can now be computed from the measured unloaded \( Q, Q_s \).
\[
\frac{1}{Q_o} = \frac{1}{Q_s} + \frac{1}{Q_b} + \frac{1}{Q_{cyl}} = \frac{1}{Q_s} + \frac{1}{\epsilon Q_{cav}}. \tag{48}
\]

After rearranging Equation 48,

\[
Q_s = \frac{\eta Q_{cav} Q_o}{\eta Q_{cav} - Q_o}. \tag{49}
\]

The field energy inside the resonator is

\[
W = \frac{\mu_o}{2} \int_V |H|^2 dV \tag{50}
\]

where the integration is performed over the entire volume of the cavity. Combining equations (36), (41) and (50) the sample \(Q_s\) is

\[
Q_s = \left(\frac{1}{R_s}\right)^2 2\pi f_r \frac{\mu_o}{\int_A |H|^2 dA} = \frac{G}{R_s}. \tag{51}
\]

\(R_s\) is the surface resistance of the sample. The area integral in Equation 51 is evaluated over the sample surface. From Equation 51 it follows immediately that the component \(Q\) for the sample, \(Q_s\), is a geometrically dependent ratio of integrals of \(HH^\dagger\) divided by the surface resistance of the cavity component. \(G\) is called the geometry factor. If the component is a sample top then the sample surface resistance is \(R_s = G/Q_s\). The geometry factor depends one the mode excited and on the aspect ratio, which is length/radius, of the cylindrical cavity. For an aspect ratio of unity the
geometry factor in the TE_{011} mode is $G_s = 10,042 \ \Omega$. Likewise a geometry factor for the entire cavity, $G_{cav}$, can be calculated by integrating the denominator in Equation 51 over the entire cavity interior, and for general TE_{mn} modes is given by$^{52}$

$$G_{cav}^{mn} = \frac{Z_0}{2} \frac{[1-(\frac{m}{X_{m+1,n}})^2][X_{m+1,n}^2+(p \pi \frac{R}{L})^2]^{3/2}}{X_{m+1,n}^2+2(p \pi)^2(\frac{R}{L})^3+(\frac{mp \pi R}{X_{m+1,n}L})^2(1-\frac{2R}{L})}.$$  

For a unity aspect ratio in the TE_{011} mode, $G_{cav} = 780.7 \ \Omega$.

In sum, $R_s$ is obtained by dividing Equation 49 into the sample geometry factor. Thus, $R_s = G_s/Q_s$. $Q_{cav}$ is the unloaded Q measured with a copper endwall in place of the sample. $Q_o$ is the unloaded Q measured with a sample as the endwall.

Before $R_s$ can be measured the problem of coupling losses must be addressed. The measured, or loaded, $Q_L$, is smaller than the unloaded $Q_o$. It is essential to determine the fraction of the power that is dissipated not on the conducting surfaces, but rather in the couplers. This is accomplished by measuring the power reflected from the couplers$^{51,53}$. The experimental configuration is shown in Figure IV-3. The unloaded $Q_o$ is

$$Q_o = Q_L(1+\beta_1+\beta_2)$$  

where, for weak coupling into the cavity,

$$\beta = \frac{1-\Gamma}{1+\Gamma}.$$  

and,

$$\Gamma = \sqrt{\frac{10^{-|P_r|/10}}{10^{-|P_0|/10}}} \quad (55)$$

is the reflection coefficient at the respective coupler. $P_r$ is the reflected power in decibels at resonance and $P_0$ is the reflected power away from resonance. If absolute units of power are used (Watts) then $\Gamma = (P_r/P_0)$. Equations 53 through 55 are derived in Appendix 1. Weakly coupled means $\beta < 1$. It is also possible to be over coupled in which case $\beta = (1+\Gamma)/(1-\Gamma) > 1$. Overcoupling is not desirable as it strongly perturbs the fields. If $\beta = 1$ then the resonator is critically coupled. In this case $\Gamma = 0$ and the resonator is impedance matched to the waveguide or coaxial cable. Whereas critical coupling is desirable in power applications such as rf magnetron sputtering, it only reduces the sensitivity to actual cavity losses in these measurements and is thus

![Figure IV-3 Experimental arrangement for the measurement of reflection coefficient.](image-url)
avoided. All measurements with the systems described here are performed with weak coupling and $\beta<0.2$.

It would be tedious to measure $\beta$ each time a surface resistance measurement is to be done. For example, if $R_s$ is to be measured versus temperature, it would not be convenient to have to do a reflection measurement at each temperature since this involves successively attaching the input cable to each coupler and performing the measurement. However, the coupling depends entirely upon the geometry of the couplers and does not change with temperature. Although $\beta$ is not a direct measure of the coupling, $Q_c=Q_0(T)/\beta(T)$ is inversely proportional to the power dissipated in the coupler and is called the coupling $Q$. Because $Q_c$ depends only upon the circuitry external to the resonator, it is independent of the unloaded $Q$, and hence temperature. It follows, then, that

$$\beta(T) = \frac{\beta(T_0)Q_L(T)}{[1+\beta(T_0)]Q_L(T_0)-\beta(T_0)Q_L(T)}$$

(56)

where $\beta$ only needs to be known at one temperature, $T_0$.

3. Surface Reactance

 Whereas the surface resistance is a measure of loss, the surface reactance, $X_s$, is a measure of field penetration into the surface. For a normal conductor, below the frequency of anomalous dispersion, $X_s=R_s=1/\sigma\delta$, where $\sigma$ and $\delta$ are the conductivity and skin depth respectively. Thus, the skin depth and the microwave dissipation are
directly related. Although losses increase with increased field penetration in a superconductor, the relationship is not as simple. The two fluid model will be discussed in Chapter 6 and from it a complex conductivity will be derived.

The surface reactance of a superconductor is nevertheless a measure of field penetration with

\[ X_s = \omega \mu_0 \lambda_{\text{eff}} \]  

(57)

where \( \lambda_{\text{eff}} \) is the effective magnetic field penetration depth into the superconductor.

If the depth of field penetration changes then the effective length of the resonator changes also.

The principle of least action gives rise to Slater's theorem\cite{48,54}

\[ \omega^2 = \omega_0^2 + \frac{\mu_0 \int_{\Delta V} (H^2 - \epsilon_0 E^2) dV}{\int_{V} (\mu_0 H^2 + \epsilon_0 E^2) dV} \]  

(58)

which gives the change in frequency (\( \Delta \omega = \omega_0 - \omega \)) of the resonant mode when the resonator volume changes by \( \Delta V \). The upper integral in Equation 58 is evaluated over the perturbed volume, and the lower integral gives total energy contained in the resonator (times 4). \( \omega_0 \) is the unperturbed resonant frequency. If the length of the cylindrical resonator changes by \( \Delta \lambda \), then the change in \( \omega \) can be calculated. If no electric field is located at the sample endwall (as in the \( TE_{01p} \) modes) then, using equation 51, Slater's theorem can be rewritten
\[(\omega_o - \Delta \omega)^2 = \omega_o^2 + \frac{\omega_o^3 \mu_o \Delta \lambda}{G_t}. \quad (59)\]

Ignoring the negligible \((\Delta \omega)^2\) one arrives at the working equation for change in surface reactance:

\[\Delta X_s = -2G_t \frac{\Delta \omega}{\omega_o}. \quad (60)\]

What is measurable, then, is not the surface reactance, but merely a change in surface reactance. This measurement can be properly done when only the \(X_s\) of the sample changes. This condition exists when the sample is a superconductor and \(\lambda_{eff}\) results from a change in magnetic field.

4. Measurement of Surface Impedance

The techniques used to measure \(R_s\) and \(\Delta X_s\) have been described in the preceding discussion, and are only synopsized here. The surface impedance measurements were conducted in one of two existing cylindrical resonators using the endwall replacement technique.\(^55\) The resonators are identified by the frequency of the \(TE_{011}\) mode. A 3.8 cm diameter 11.36 GHz cavity and a 2.5 cm diameter 17.46 GHz cavity are used. The resonators are coupled by homemade loop antenna couplers and connected by semi-rigid and flexible microwave coaxial cable.

In a cylindrical cavity the \(TE_{011}\) and \(TM_{111}\) modes are degenerate (have the
same frequency). The degeneracy is separated in the above cavities by placing a mode trap on the cylindrical wall. A mode trap is a deformation of the surface in a place where the fields of the $\text{TE}_{011}$ mode are weak. In the above cases, a groove was cut into the cylindrical wall half way between the top and bottom walls. In the $\text{TE}_{011}$ mode only magnetic fields exist at this location while in the $\text{TM}_{111}$ mode only electric fields exists there. From Slater's theorem, Equation 58, since $dV<0$ for a groove in the surface, the $\text{TE}_{011}$ mode is shifted down in frequency while the $\text{TM}_{111}$ mode is shifted up. A mode splitting of approximately 100 MHz occurs in both resonators.

A Wiltron 6747B 10 MHz to 20 GHz swept frequency synthesizer is used as an rf source. The synthesizer has both discrete step sweep and continuous analog sweep capability, and is always operated in step sweep mode. This model has a 12 dBm leveled output power range and a resolution of 1 KHz.

A microwave signal is analyzed by first converting it to a DC voltage with a Wiltron model 560-7S50 diode detector. A Wiltron 562 scalar network analyzer (SNA) receives the DC signal. The SNA communicates to the synthesizer through an IEEE-488 General Purpose Interface Bus (GPIB). Frequency information comes to the SNA directly from the synthesizer over this bus line. With these two information sources the SNA plots power transmitted through the resonator versus frequency as shown in Figure IV-2. This two channel SNA is capable of measuring the -3 dB Full Width at Half Maximum which is used to calculate $Q_L$. It is also capable of measuring the depth of the power dip in reflection measurements which is used to calculate $\beta$. 
Cooling is accomplished with a CTI-Cryogenics closed cycle refrigerator. With an aluminum radiation shield, a minimum temperature of 10K is achieved. Diffusion pump pressures of ~10^{-5} torr serve as thermal insulation. Temperature is controlled by a Palm Beach Cryophysics series 4000 cryogenic thermometer/controller. One silicon diode is placed on the cold head of the closed cycle refrigerator and another on the exterior of the resonator. The controller passes current to a wire heater wound around the cold head. Using this control system, temperature remains constant to within 10 mK.

The entire surface resistance versus temperature measurement is controlled by the fortran program \textit{zstep}\textsuperscript{56}. Frequency and bandwidth are measured repeatedly at temperature steps specified by the user. The program controls the synthesizer, the SNA and the temperature controller. The program creates a data file which reports the average frequency, average bandwidth and standard error in the mean of the bandwidth

$$\delta(\Delta f) = \sqrt{\frac{\sum_{i=1}^{n} (\Delta f_i - <\Delta f>)^2}{n(n-1)}}$$

at each temperature, where \(<\Delta f>\) is the mean bandwidth at temperature, T.

Another program \textit{zmag20}\textsuperscript{56} measures \(R_s\) and \(\Delta X_s\) versus DC magnetic field at constant temperature. \(\Delta X_s\) is measured by subtracting \(f_s(H)\) from \(f_s(H=0)\). Again repeated measurements are performed at each field level and averages and standard errors are calculated. The user must increment the field manually. DC magnetic
fields of up to 120 Gauss are established by a homemade multi-turn Helmholz pair. A larger water cooled pair is occasionally used to generate fields up to 1200 Gauss.

Statistical uncertainties in measured surface resistance are determined by repeating the Q measurement five times and calculating the standard error in the mean. There is a standard error associated both with the calibration, $Q_{\text{cav}}$, and the sample measurement, $Q_0$. A similar standard error can be determined for the $\theta$'s of Equation 54. These errors are carried through the surface resistance calculation via conventional error propagation as described, for example, in Taylor$^{57}$. An uncertainty of ±1 mΩ to ±3 mΩ is usually obtained.

The uncertainty in $\Delta X_\delta$ is determined by measuring the resonant frequency fifteen times and calculating the standard error. An uncertainty of ±3 mΩ to ±8 mΩ is usually determined.

The resolutions of $R_\delta$ and $\Delta X_\delta$ are ultimately limited by the synthesizer. The Wiltron synthesizer used here has a frequency resolution of 1 KHz. From Equation 60, this limits the surface reactance resolution at 17.5 GHz to 1 mΩ for the actual sample geometry factor of 10 kΩ. The resolution in $R_\delta$ depends upon the temperature of the copper cavity. In practice, at 12 K and 17.5 GHz a surface resistance of 1 mΩ can also be resolved.
B. The Fabry-Perot Resonator

The cylindrical pillbox resonant cavity is useful for measuring the surface resistance of high R₄ films of a fixed diameter at a fixed frequency. Only the TE₀₁₁ₚ modes can be used for surface resistance measurements. In practice, only the TE₀₁₁ mode, and maybe the TE₀₁₃ mode, is within the operating frequency of the laboratory equipment.

1. Parallel Plate Open Resonator

To measure frequency dependence of R₄, the parallel plate Fabry-Perot resonator in Figure IV-4a offers a large number of useful modes. Two superconducting plates are situated facing each other with a thin dielectric spacer in between. For two identical rectangular plates the electric field, E=Eₓₑₓ is, to first approximation, 

\[ E_z = E_0 \cos(n \pi \frac{x}{L}) \cos(m \pi \frac{y}{W}) \]  \hspace{1cm} (61)

where L and W are the length and width of the two identical plates and \( \hat{e}_z \) is normal to the plates. The resonant frequencies are 

\[ f_{nm} = \frac{c}{\sqrt{\varepsilon_r}} \sqrt{\left(\frac{n}{2L}\right)^2 + \left(\frac{m}{2W}\right)^2} \]  \hspace{1cm} (62)

and the geometry factors are simply \( G_{nm} = \pi \mu_0 f_{nm} s \), where s is the plate separation.

Although a more accurate description of the fields is given by Weinstein this simple description is adequate to evaluate the applicability of the parallel plate resonator
to this study. There are three dominating sources of loss in the parallel plate open resonator: dielectric loss, diffractive loss, and Ohmic loss. The unloaded $Q$ of the resonator is

$$\frac{1}{Q_o} = \tan(\delta) + \alpha s + \frac{R_s}{G_{\text{nm}}}$$  \hspace{1cm} (63)$$

where $\tan(\delta)$ is the loss tangent of the dielectric, $1/\alpha s$ is the diffraction $Q$ caused by radiation out of the resonator, and $G_{\text{nm}}/R_s$ is the Ohmic $Q$. $\alpha$ is a constant which depends upon the frequency and the size of the plates.

To minimize dielectric and diffractive losses the separation, $s$, needs to be reduced. In practice $s$ is about $10\mu\text{m}$. The tradeoff, however, is that this reduction in $s$ corresponds to a reduction in resonator volume. Since a resonator $Q$ is linearly proportional to volume, the $Q$ of the parallel plate resonator decreases linearly with $s$. The result is that practical parallel plate resonators exhibit low $Q$'s (<20,000), and consequently weak coupling, allowing only higher quality films to be measured ($R_s<2\text{m}\Omega$ at 12.5 GHz). Because this study focuses on

![Parallel Plate Resonator](image-url)
granular surfaces with $R_s$ as large as 50 m$\Omega$ at 12.5 GHz, the parallel plate resonator would be an inappropriate choice.

2. Scaler Gaussian Wave Theory

The frequency dependence of the surface resistance was measured in this work using a modification of the above flat Fabry-Perot resonator. A larger volume cavity may be realized if one of the plates is concave. The microwave fields are then focused into a Gaussian beam with a minimum beam radius at the sample, which serves as the flat plate. To better understand this resonator, we begin with a Fabry-Perot resonator made from two identical concave mirrors as shown in Figure IV-4b. In 1961 Boyd and Gordon demonstrated that the diffraction loss with concave mirrors is orders of magnitude smaller than that with flat mirrors.

![Figure IV-4](image)

**Figure IV-4** (b) A full Fabry-Perot resonator. (c) In a $R_s$ measurement one of the mirrors is replaced by a flat sample.
magnitudes lower than with planar mirrors. Since no dielectric is involved in the basic resonator design, the dielectric losses are eliminated. In the 1960's and early 1970's a number of authors\textsuperscript{61} published Gaussian beam eigenmodes for the concave Fabry-Perot resonator. The "quasi-optical" treatment of these microwave modes are summarized in Das\textsuperscript{62}. In their original work Goubau and Schwering\textsuperscript{63} solved the scalar wave equation

\[
\nabla^2 u(x,y,z) + k^2 u(x,y,z) = 0 \quad (64)
\]

for each cartesian component of the fields in the resonator. The periodic longitudinal dependence can be separated in the general solution

\[
u(x,y,z) = \psi(x,y,z)e^{-jkz} . \quad (65)
\]

If \(\lambda \ll D\), where \(\lambda\) is the wavelength and \(D\) is the mirror separation, then the assumption that \(\psi(x,y,z)\) is a very slowly varying function of \(z\) can be made. In this case, inserting Equation 65 into Equation 64 gives

\[
\nabla_t^2 \psi(x,y,z) - 2jk \frac{\partial \psi(x,y,z)}{\partial z} = 0 \quad (66)
\]

where \(\nabla_t\) is the transverse \((x,y)\) gradient operator. The solution of this equation
involves Laguerre polynomials and yields a large number of modes in the mode spectrum, \( \psi_{mn}(x,y,z) \), as illustrated in Figure IV-5.

The higher order \((n,m \neq 0)\) modes (HOM) are of weaker intensity since the HOM fields are weak at the center of the mirror where the coupling occurs. Because the intensity weakens with higher mode number, and there exists multiple degeneracy for all HOM's, only the \( \psi_{00}(x,y,z) \) solution will be considered for practical application. The one non-degenerate solution to Equation 66 is

\[
\psi_{00}(x,y,z) = \frac{w_0}{w(z)} e^{-\frac{p^2}{w(z)^2}} e^{-j\frac{p^2}{2R(z)}\Theta(z)}
\]

where

\[
R(z) = z(1 + \frac{z^2}{z_0^2}), \quad (68)
\]

\[
\Theta(z) = \tan^{-1}\left(\frac{z}{z_0}\right), \quad (69)
\]

\[
w(z) = w_0 \sqrt{1 + \left(\frac{z}{z_0}\right)^2}, \quad (70)
\]

\[
z_0 = \frac{1}{2}kw_0^2. \quad (71)
\]

The e\(^{-1}\) radius of the beam in the center of the resonator, \( w_0 \), is
and is called the beam waist. $R_c$ is the radius of curvature of the mirrors. Most of the
parameters defined in equations 68 through 72 are physically descriptive of some
characteristic of the resonator. $R(z)$ is the radius of curvature of the wavefront. Only
at the cavity center, $z=0$, is the wave planar. $w(z)$ is the $e^{-1}$ radius of the Gaussian
beam at any $z$. The physical meaning of $z_o$ became clear with the advent of complex
source point theory which will be introduced in the next section.

The fundamental \((m=n=0)\) eigenfunctions are found by substituting equation 67
into equation 65

\[
\mu_{00q} = \frac{w_0}{w(z)} \exp \left( -\frac{\rho^2}{w(z)^2} \right) \exp \left[ -j(kR + \frac{k\rho^2}{2R(z)} - \Theta(z)) \right]
\]

(73)

where the additional index, $q$, is the longitudinal index indicating the number of
wavelengths fitting into the resonator. $q=0$ corresponds to half a wavelength. $q=1$
corresponds to one wavelength. $q=2$ corresponds to $3/2$ wavelength, etc.

The resonance condition for even axial modes \((q \text{ even})\) is found by requiring the
real part of the eigenfunction, equivalently $\mu_{0q}$, to vanish at the spherical mirror surface.
This condition is, to first approximation,
For odd axial modes \((q \text{ odd})\) the imaginary part of the eigenfunction must vanish at the spherical mirror surface. Again to first approximation,

\[
\frac{kD}{2} - \tan^{-1}\left(\frac{D}{k\omega_o^2}\right) = \frac{(2q+1)\pi}{2}.
\]  

(74)

Combining equations 74 and 75 gives the overall resonance condition for the fundamental modes

\[
\omega_0 = \left(q + 1\right)\frac{c}{2D} + \frac{c}{2D\pi}\cos^{-1}\left(1 - \frac{D}{R_c}\right).
\]  

(76)

The resonant condition for all modes is

\[
f_{mnq} = \left(q + 1\right)\frac{c}{2D} + \frac{c}{2D\pi}(1 + m + 2n)\cos^{-1}\left(1 - \frac{D}{R_c}\right).
\]  

(77)

From this expression it is seen that the dependence of \(f_{mnq}\) is only weakly dependent upon the radius of curvature.

This analysis was referred to above as "quasi-optical" because optical resonator techniques are being applied to a microwave resonator. The resonant modes are also referred to as "quasi-TEM" modes because of their two dimensional approximation. From here on the notation TEM_{mnq} for the resonant modes will be used.

The transition from this analysis to that of a Fabry-Perot resonator with only one
concave mirror is fairly simple. With a planar metallic mirror in place of one of the concave mirrors, as in Figure IV-4c, there exists a concave image mirror behind the planar mirror. The mode pattern is not disturbed by this. However, even axial modes, with their half integer wavelength numbers are suppressed and only odd values of $q$ remain.

The fields in the cavity are given by the eigenfunction in equation 73. For even axial modes, with $Z_0 = (\mu / \varepsilon_0)^{1/2}$, the fields are

$$E_x = H_o Z_o Re[u_{00q}(x,y,z)]$$  \hspace{1cm} (78)$$

and

$$H_y = H_o Im[u_{00q}(x,y,z)].$$  \hspace{1cm} (79)$$

For the odd axial modes, which are the modes with one concave mirror and one flat mirror present,

$$E_x = H_o Z_o Im[u_{00q}(x,y,z)]$$  \hspace{1cm} (80)$$

and

$$H_y = H_o Re[u_{00q}(x,y,z)].$$  \hspace{1cm} (81)$$
Accuracy and Stability

Clearly, the modes in the concave Fabry-Perot resonator are not exactly TEM. In order to satisfy boundary conditions at the reflectors, the E and H fields will indeed contain components in all directions. This non-planar character also introduces error to equations 18-21. In argument for a more accurate theory, Cullen and Yu show that the above expressions for $E_x$ are accurate to $O\{(kw_0)^2\}$, where $w_0$ is the beam waist at the flat mirror and "O" means "order". The $E_y$ component is $O\{(kw_0)^2e^{-1}\}$ or less, and the $E_z$ component is $O\{(kw_0)^3e^{-0.5}\}$ or less. At high enough frequency $kw_0$ is large, and these higher order terms are negligible. Typically, the beam waist, $w_0$ is of order 1 cm. At 50 GHz, then, $kw_0=\sim 10$ and the errors are not so negligible.

The spot size, $w_0$, or beam radius at $z=0$, vanishes when $D=2R_c$. The resonator energy vanishes at $D=2R_c$ as well. If $D>2R_c$ the resonator is unstable and cannot contain its energy. The stability condition is written $0<\mid 1-(D/R_c) \mid<1$, and is an important design consideration. In some Fabry-Perot laser resonators it is, in fact, desirable to design in instability in order to create losses. These high losses delay population inversion and thus raise the energy extraction. This instability is often created by using one convex and one planar mirror.

3. Vector Complex Source Point (CSP) Theory

In dealing with misalignment of the mirrors in a concave Fabry-Perot cavity, Arnaud repeated the existing scalar theory, but with the radiation source positioned in complex space, $z+jz_0$. This is realized by displacing the origin by $jz_0\hat{e}_3$. It is here
Figure IV-6 Higher order components of the magnetic field calculated from CSP theory for x=y=1 cm. $H_y$ is the first order magnetic field from scalar theory.
that the physical meaning of \( z_0 \) in Equation 71 can be understood. It is the distance of the radiation source from the origin in complex space. \( z_0 \) does not carry information of the physical location of the source.

Cullen and Yu\textsuperscript{65} published the complete set of field components in 1979 for the TEM\textsubscript{00q} modes. These expressions include higher order corrections for the dominant components. The magnetic field components for odd axial modes are, to first order,

\[
H_x = \frac{2xy}{k^2w_0w^3(z)} \exp \left( -\frac{\rho^2}{w^2(z)} \right) \cos(kz-3\Theta(z)+\frac{kp^2}{2R(z)}) ,
\]

\[
H_y = \frac{w_0}{w(z)} \exp \left( -\frac{\rho^2}{w^2(z)} \right) \left[ \cos(kz-\Theta(z)+\frac{kp^2}{2R(z)}) - \frac{2}{k^2w(z)w_0} \cos(kz-2\Theta(z)+\frac{kp^2}{2R(z)}) \right]
\]

\[
H_z = \frac{2y}{kw^2(z)} \exp \left( -\frac{\rho^2}{w^2(z)} \right) \sin(kz-2\Theta+\frac{kp^2}{2R(z)})
\]

In 1985 Luk and Yu\textsuperscript{69} published expressions for all six field components for the general TEM\textsubscript{mnq} mode.

In Figure IV-6 the first order correction of the magnetic fields is shown separated into components. The lowest approximation, \( H_{y1} \) from scalar theory, is also shown for comparison. This figure shows that the CSP theory corrects the \( \hat{e}_y \) component by about 10 percent. The higher order terms in the CSP fields do not possess azimuthal symmetry. So, the curves shown in figure IV-6 are specific to the
choice of x and y. However, numerous calculations of H versus z for fixed x and y find that the CSP correction is usually below 10 percent.

4. Losses

a. Resistive and Coupling Losses

Power is dissipated in the Fabry-Perot resonator through Ohmic losses, coupling losses, scattering losses, and diffraction losses. In the closed cylindrical cavity the Ohmic losses dominate provided the experimenter is careful not to overcouple. However, other loss mechanisms arise in an open resonator, as illustrated in Figure IV-7.

As in the cylindrical cavity, the Ohmic losses are characterized by a geometry factor, G, defined in Equation 51. If the open resonator is constructed from one homogeneous material, then the unloaded Q, Q₀, is $Q_0 = G/R_\alpha$. If the curved mirror is one material (copper) and the flat mirror is another material (superconductor) then the $R_\alpha$ of the superconductor can be determined by measuring the partial Q and calculating the geometry factor for the superconductor.
With two concave mirrors, the cavity geometry factor is, from eq.51,

$$G = Z_0 \frac{\pi f D}{2 c} \left(1 + \frac{x}{\left(k\omega_o\right)^2}\right)$$  \hspace{1cm} (85)$$

where the second term results from CSP theory. The factor, $x$, in the second term has been left out in reference 68 and all other publications. The entire second term is a higher order correction which is derived from CSP theory and is frequently ignored. Geometry factor calculations were calculated by numerically integrating Equation 51 with Equations  82 through 84 using the method of Gaussian quadrature\textsuperscript{71}. The dimensions of an existing resonator ($R_c=2.46$ cm, $1.94<D<2.46$ cm) were used in this calculation. The results are in Figure (IV-8). In all cases with this resonator, the error in $G$ caused by ignoring the higher order CSP terms is between 5% and 10%. For the particular case shown in Figure (IV-8) $x=2.65$, which is significantly larger than the often assumed $<3\%$ error caused by neglecting CSP theory.

If one mirror is replaced by a planar mirror the geometry factor is reduced by
a factor of two since the volume is halved. Because the flux of power passing through a cross-section normal to $z$ is conserved, the integral $\int \mathbf{H} \cdot d\mathbf{A}$ is the same on every beam cross-section. Thus, the geometry factor of the concave mirror, $G_m$, is equal to the geometry factor of the planar mirror, $G_s$. This further implies that $G_s = G_m = 2 \times G_{\text{total}}$, so that the expression in equation 85 is in fact the sample geometry factor. Thus, for a homogeneous cavity, the resistive $Q$ is, $Q_o = G/2R_s$.

In more detail than discussed in part A of this chapter, coupling losses result from some of the energy in the resonator being coupled into dipole radiation at the coupling aperture and then being re-radiated into the waveguide. The microwave power incident upon the coupling aperture induces a magnetic dipole moment. This dipole radiates into the waveguide, into the open cavity, and out of the open cavity. Figure IV-9 illustrates the dipole in the cavity and Figure IV-7 illustrates the different sources of dissipation of its radiated power.

The total power radiated from the dipole is $P_{dp}$. Some of the power propagates back to the microwave source. A
fraction of the power is coupled into the mode. This is necessary to sustain a resonance. Finally, some of the dipole power is either attenuated in the coupling hole or coupled into other modes. This is the power lost due to the presence of an aperture with a dipole moment in the cavity. This latter power loss widens the resonance peak and is called scattering loss. So the contribution of the dipole to the overall Q is

\[ \frac{P_{\text{dip}}}{\omega W} = \frac{1}{Q_c} + \frac{1}{Q_{sc}} + \frac{P_o}{\omega W}. \] (86)

\(Q_{sc}\) is the scattering Q, \(Q_c\) is the coupling Q, and \(P_o\) is the dipole power coupled into the mode. \(W\) is the total energy contained in the resonator. The total power radiated by the magnetic dipole at the coupling aperture is\(^{72}\)

\[ P_{\text{dip}} = \frac{1}{12\pi} \mu_0 m^2 c k^4 \quad (\text{mks units}) \] (87)

where the magnetic dipole moment at the aperture is \(m=\alpha H\). The magnetic polarizability for a circular aperture is \(\alpha=(4/3)h^3\), where \(h\) is the aperture diameter\(^{73}\). Some of the dipole power radiates into the open cavity. The other half radiates in the direction of the waveguide. In resonator analysis it is conventional to regard the mode as the source exciting the dipole instead of the signal generator. Consequently the dipole is located on the cavity side of the aperture. The power associated with \(Q_c, P_c\), is half of the dipole power suppressed by attenuation in the coupling hole. Hence the coupling Q is
\[
\frac{1}{Q_c} = \frac{P_{\text{dip}}}{\omega W^2} e^{-2\alpha_{TE} d}
\]  

(88)

where \(d\) is the length of the coupling hole, or \textit{wall thickness}, and \(\alpha_{TE}\) is the waveguide attenuation constant for the coupling hole

\[
\alpha_{TE} = \sqrt{\left(\frac{X_{mn}'}{(h/2)}\right)^2 - \left(\frac{\omega}{c}\right)^2}.
\]

(89)

\(X_{mn}'\) is the \(n^{th}\) zero of the derivative of the \(m^{th}\) order Bessel function. The dominant propagation mode is the \(\text{TE}_{11}\) so \(X_{11}'=1.841\) is used. \(P_{\text{dip}}\) can be calculated, and \(Q_c\) can both be calculated from Equation 88 and measured according to the procedure of part A. So if the power coupled into the mode, \(P_o\), is known then the scattering \(Q\) can also be determined. The measured coupling \(Q\) of a four centimeter long semi-Fabry-Perot resonator (Figure IV-4c) along with Equation 88 is shown in Figure IV-10. Very close agreement between the measured coupling \(Q\) and Equation 88 is seen here.

\textbf{b. Scattering Losses}

The Coupling hole contributes to power loss in two ways. First, the microwave signal is attenuated as it passes through the hole below the cutoff frequency of the hole. Because the hole is a waveguide operated below the cutoff frequency, there is no dissipation in the hole. Any power which is not radiated out the back of the hole into the coupling waveguide is reflected back into the cavity.
Figure IV-10 Coupling Q versus frequency for a resonator with a 1 mm diameter coupling aperture. A measurement was made for each fundamental mode and compared to Equation 88.
This dipole radiation is not seen then in the coupling $Q$. Secondly, a fraction of the power radiated from the magnetic dipole at the hole is coupled into other resonator modes. Other eigenmodes are excited because the coupling aperture perturbs the dominant mode. These mixed modes are excited at the driving frequency. Hence, they are equivalent to a damped harmonic oscillator excited at a frequency other than the natural frequency. The sum of the losses due to the coupling hole is called the scattering loss, $P_{sc}$.

The dipole power scattered by the coupling hole is

$$P_{sc} = \frac{1}{2} P_{dip} (1 - e^{-2\pi rd^2})$$  \hspace{1cm} (90)

where $d$ is the aperture, or wall, thickness. Thus, scattering losses are minimized by reducing the wall thickness. The dipole radiation, $P_{dip}$, is given by Equation 87, and the magnetic dipole moment is $m = (4/3)\mu_0 H_0$, where $H_0$ is the magnetic field at the coupling aperture. For an aperture at the center of the mirror ($\rho=0, z=D/2$)

$$P_{sc} = \frac{32H_0^2}{27} \mu_0 \epsilon \pi^3 \frac{w_o^2}{w^2} \frac{w_o^2}{w^2} \cos^2(kD/2) \Theta(D/2) (1-e^{-2\pi rd^2}).$$  \hspace{1cm} (91)

Combining Equation 91 with Equation 50
The magnetic field in the presence of the aperture is a sum over all modes

$$\vec{H} = \sum_n a_n \text{Re}(\vec{u}_n)$$

where \(n=1\) corresponds to the dominant mode. The eigenmodes for the Fabry-Perot resonator are (the generalized form of Equation 73) to lowest order

$$\vec{u}_{mnq} = \frac{w_{mnq}(z)}{w_{mnq}(z)} L_m^m(\frac{2p^2}{w_{mnq}(z)^2}) \exp\left(-\frac{p^2}{w_{mnq}(z)^2}\right) \cos(k_{mnq} z + \frac{k_{mnq} p^2}{2R_{mnq}(z)} - \Theta(z))$$

where the real part is taken for the odd axial modes of the semi-Fabry-Perot resonator. \(w_{mnq}\) is the beam waist of the mode \(mnq\). \(L_m^m\) is the associated Laguerre polynomial.

One great simplification, if the coupling hole is located in the center of the mirror, is that only the \(m=0\) modes can be coupled.

\[Q_{scf} = \frac{27Dw^2(D)}{256\hbar^6\pi(\frac{f}{c})^3\cos^2(k\frac{D}{2} - \Theta(\frac{D}{2}))}(1-e^{-2a_{nq}})\]  

Figure IV-11 Lower limit of \(Q_{scf}\) versus frequency for a cavity with fixed mirror separation.
The expansion coefficients, to be used in equation 93, for hole coupling, $a_{mnq}$, were published by Bethe in 1942

$$a_{mnq} = \frac{1}{3} \frac{H_o \hbar^3}{V_{mnq} \omega_{mnq}^2} \frac{\omega_1^2}{\omega_1^2 - \omega_1^2 \Re(u_1(0)) \Re(u_{mnq}(0))}$$  \hspace{1cm} (95)$$

where $u(0)$ is evaluated at the coupling aperture. For the dominant mode $a_1=1$. $V_{mnq}$ is the normalization constant for the mode,

$$V_{mnq} = \int (\Re(u_{mnq}))^2 dV = \frac{\pi}{16} w_{mnq,o}^2 D,$$  \hspace{1cm} (96)$$

where $D$ is the separation between the flat sample and the curved mirror, and $w_{mnq,o}$ is the beam waist of the $mnq^{th}$ mode. The radial part of the volume integral is carried out to infinity. Because $m=0$, the azimuthal dependence drops out of the integral. It can also be noted that the energy contained in the $mnq^{th}$ mode is $W_{mnq} = \frac{1}{2} \mu_o V_{mnq}$. The power coupled out of the dominant mode due to mode mixing is then calculated using

$$P_{mixing} = Z_0 \pi \int_0^\infty \left[ \sum_{n=2}^\infty a_n u_n(z=0) \right]^2 r \, dr.$$  \hspace{1cm} (97)$$

Power mixed into other modes is not itself a source of dissipation. Only that power which is mixed into other modes and then dissipated contributes to peak broadening. However, Equation 97 does provide an upper limit to $P_{SC2}$. Because the losses in most of the higher order modes are dominated by diffraction Equation 97 is
a close approximation of $P_{SC2}$.

Slepian\textsuperscript{76} solved the prolate spheroidal wave equation for which the asymptotic spherical condition was applied by McCumber to the Fabry-Perot resonator. The fraction of power diffracted in the $mn^{th}$ mode (independent of $q$) is

$$
\alpha_{mn} = \frac{2\pi (8\pi N_{mn})^{1+2n+m}}{n!(n+m)!} e^{-4\pi N_{mn}(1+O(\frac{1}{N_{mn}}))} .
$$

(98)

The Fresnel number, $N_{mn}$, is the number of Fresnel zones on one mirror when viewed from the center of the other mirror\textsuperscript{62} and is

$$
N_{mn} = \frac{a_m^2}{D \lambda_{mn}} \sqrt{\frac{2D}{R_c} \left(\frac{D}{R_c}\right)^2}
$$

(99)

where $\lambda_{mn}$ is the wavelength of the $mn^{th}$ mode. $P_{SC2}$ is now a simple revision of Equation 97

$$
P_{SC2} = Z_0 \pi \int_0^\infty \sum_{n=2}^{\infty} \alpha_n [\alpha_n u_n(z=0)]^2 r \, dr .
$$

(100)

The diffraction constants, $\alpha_{mn}$, are only accurate for small $m$ and $n$. For $n$ or $m$ larger than about 4, $\alpha_{mn}$ becomes larger than unity. It is unphysical for more power to be diffracted out of a mode than to be coupled into it. Since power is coupled into the high order modes, Equation 97 is used for $P_{SC2}$ instead of Equation 100. It must be understood then that since $P_{mixing}$ is an upper limit to $P_{SC2}$, the resulting scattering $Q$
is a lower limit to $Q_{sc2}$. This lower limit on scattering $Q$ is useful for designing a resonator. Finally, the total scattered power is $P_{sc} = P_{sc1} + P_{sc2}$. The scattering $Q$ is then

$$Q_{sc} = \frac{Q_{sc1}Q_{sc2}}{Q_{sc1} + Q_{sc2}} = \frac{\omega W}{P_{sc1} + P_{sc2}},$$

(101)

where $W$ is the energy of the dominant mode.

The mode mixing contribution is strongly frequency dependent with the problem becoming acutely worse at higher frequency. Sample calculations of the lower limit on $Q_{sc2}$ versus frequency and coupling aperture radius are shown in Figures IV-11 and IV-12. The fixed resonator dimensions were a concave mirror to flat mirror separation of 2 cm and a radius of curvature of 2.22 cm (7/8 inch). The scattering losses become significant at higher frequencies. In the case of figure IV-11 the losses rise rapidly above 70 GHz. For a typical resonator the scattering $Q$ must be less than $\sim 10^5$ in order to affect the measured $Q$. In the measurement of $R_s$ Scattering and diffraction losses should be more than
an order of magnitude less than the resistive losses.

When small changes are made in the coupling hole diameter, $h$, significant changes in mode mixing occur. The dependence of $Q_{SC2}$ upon $h^6$ arises from the fact that the coupling strength for the mixed modes depends on the square of the magnetic dipole moment.

From Figures IV-11 and IV-12 it is seen that with judicious choice of aperture size, the contribution of mode mixing to dissipation can be rendered negligible. Hence, $Q_{SC} \approx \omega U/P_{SC1}$. The large $Q_{SC2}$ indicates that virtually all of the power radiated by the magnetic dipole at the aperture is coupled into the dominant mode.

Mongia and Arora used Bethe diffraction theory to calculate the coupling $Q$. In their calculation mode mixing was ignored, but they included corrections to the field due to the presence of two dipoles, one on either side of the aperture.

All numerical integrations in this work were performed with the method of Gaussian quadrature using $n=48$. So, for a 3 dimensional integral a total of 110,592 mesh points were evaluated.

c. Diffraction Losses

The small amount of rf power coupled into the dominant mode, but which is not confined in the resonator due to fringing at the edges should be considered. This is referred to as diffraction loss. The fraction of power in the $mn$th mode diffracted out of the resonator is given by equation 98, so that
\[ P_D = \alpha_{mn} P_{tot} \]
\[ = \alpha_{mn} \left( \frac{1}{8} \pi \mu \omega H^2 \right) , \]  \hspace{1cm} (102)

and is expressed as a diffraction \( Q \), \( Q_D = \omega W/P_D \), by Beverini et al., who obtained

\[ Q_D = \frac{2\pi D}{\lambda \alpha_{mn}} . \] \hspace{1cm} (103)

Except for very small radius mirrors, diffraction losses are usually negligible for the fundamental \((0,0,0)\) modes. \( Q_D \) from Equation 103 is typically \( > 10^2 \). As a general rule, the diffraction losses are negligible in the fundamental modes when the Fresnel number is greater than 1.

5. Measurement of Surface Resistance

Owing to the existence of such unmeasurable losses as diffraction and scattering, the surface resistance can not be measured with an open resonator in exactly the same way as in a closed cylindrical resonator. Whereas with a cylindrical resonator the surface resistance is measured directly, an open resonator can only be properly used to measure surface resistance with respect to some known reference. This is because scattering losses are mathematically equivalent to a material inhomogeneity within the resonator. Of course, if the scattering losses were rendered negligible, then a direct measurement as described in part A of this chapter would be correct.
A disk of OFHC copper whose \( R_s \) was previously measured in a cylindrical resonator is used as the reference sample. The spherical mirror is also copper, so with the entire resonator at room temperature the unloaded Q is

\[
\frac{1}{Q_o(RT)} = \frac{R_s(RT)}{G} + \frac{1}{Q_{other}}
\]  
(104)

where \( R_s(RT) \) is the room temperature surface resistance of copper, \( Q_{other} \) is the partial Q due to the unmeasurable losses such as scattering and diffraction, and \( G \) is the resonator geometry factor. If the copper reference is replaced by a cold sample at temperature \( T \) (perhaps a superconductor at 77K) then the unloaded Q is

\[
\frac{1}{Q_o(T)} = \frac{R_s}{2G} + \frac{1}{Q_s(T)} + \frac{1}{Q_{other}}
\]  
(105)

where \( Q_s(T) \) is the sample Q, \( R_s/2G \) is the Q of the spherical mirror, and \( Q_{other} \) is unchanged from the room temperature measurement. Subtracting Equation 105 from Equation 104

\[
\frac{1}{Q_s(T)} = \frac{R_s(RT)}{2G} - \frac{1}{Q_o(T)} + \frac{1}{Q_o(RT)}
\]  
(106)

From the sample Q the surface resistance is

\[
R_s(T) = \frac{2G}{Q_s(T)}
\]  
(107)
Instrumentation

For the measurement of surface resistance the spherical mirror shown in Figure IV-4b was machined out of copper. The radius of curvature is 24.6 mm and the diameter is 30 mm. The copper surface received a fine machine finish. Polishing is accomplished with successively finer grades of sandpaper beginning with 100 µm and ending with 12 µm grit. For mechanical support the sandpaper is fixed onto a steel ball bearing with a radius of curvature of 24.6 mm. After the abrasive polishing the mirror finish is achieved by further polishing with diamond paste beginning with 9 µm grit and ending with 1 µm grit.

A single coupling aperture 0.89 mm in diameter is located at the center of the mirror. The polishing process served to reduce the wall thickness at the aperture to 0.2 mm. The wave guide is positioned behind the coupling aperture in a shaft large enough to accommodate both WR-19 (40-60 GHz) and WR-10 (75-110 GHz) waveguides. With a WR-19 waveguide coupler the cavity can be excited in the 40-60 GHz range. With a WR-10 waveguide coupler the cavity can be excited in the 75-110 GHz range. In practice, above 85 GHz the perturbation of the coupling aperture on the fields is so large that accurate $R_s$ measurements cannot be made.

The Q is determined from the reflected signal. This measurement involves only a single coupling aperture and a single waveguide. As described in Figure IV-3 the reflected signal is measured by passing the input signal backwards through a directional coupler. When the signal enters through the exit port, the directional coupler is transparent. In this arrangement the reflected signal is directed back
through the directional coupler and on to an unbiased diode where it is converted to DC for display on a scalar network analyzer. The reflected signal is fit to a Lorentzian by a program written at NRL\textsuperscript{79} in the Labview interfacing software. The Q is the frequency width at the half power point between the off-resonance reflected signal level and the on-resonance reflected signal level.

Figure IV-13 shows $R_s$ versus frequency of polished brass measured at room temperature in this Fabry-Perot resonator. The two lowest points were measured in the large cylindrical cavity described in part A of this chapter using the TE$_{011}$ mode at 11.3 GHz and TE$_{013}$ mode at 16.5 GHz\textsuperscript{80}. The line through the data indicates the expected square root frequency dependence extrapolated from 11.3 GHz. That the measurements in the cylindrical cavity are consistent with those in the experimental Fabry-Perot resonator is indicative of the accuracy of the Fabry-Perot technique.

Figure IV-14 shows the surface resistance versus temperature of an epitaxial YBCO film deposited onto an MgO substrate by laser ablation at NRL. The complete phase transition is not visible because as the superconductor approaches $T_c$, its penetration depth becomes comparable to the film's thickness of 300 nm. This results in mode damping due to a substantial contribution of the substrate to the losses. If the film were thicker, or if the sample geometry factor were large compared to the cavity geometry factor then a resonance could have been observed above $T_c$ because the substrate would have no effect. In cylindrical resonators this indeed occurs. Many authors perform a corrective calculation based on the substrate loss tangent then to compensate for the substrate effect\textsuperscript{81}. Work is presently underway at NRL to
Figure IV-13 The $R_s$ of a polished brass plate was measured at 11.3 GHz and 16.5 GHz in the cylindrical resonator and between 44 GHz and 110 GHz in the Fabry-Perot resonator. Square root frequency dependence is observed.
Figure IV-14 The surface resistance of an epitaxial YBCO film was measured at 55 GHz in the Fabry-Perot resonator.
include these corrections in thin films tested in the Fabry-Perot resonator.

At the time of this writing, efforts are underway in other laboratories to use Fabry-Perot resonators to measure surface resistance$^{82,83,84}$. Historically, Fabry-Perot resonators have been used as laser cavities$^{62,85,86}$ as well as for loss tangent measurement.$^{87,88}$ Measurement of loss tangent has been carried out successfully with the resonator reported here and is intended to be used increasingly for that purpose in the future$^{89}$.

C. The Coaxial Resonator

Coaxial resonators were considered at the onset of this work. The resonator structure shown in figure IV-15 consists of a long hollow conducting tube enclosing a short center conductor of length L.

The Q of a coaxial resonator is optimized if the ratio of the diameter of the outer tube to that of the center conductor is 3.6$^{74}$. The resonator is excited in a half-wave resonant mode. Thus, the wavelength is $\lambda=2L/n$ where $n$ is an integer mode number. The losses in the half-wave resonant modes are concentrated on the center conductor if its diameter is much smaller than that of the tube$^{90}$. In this case the geometry factor of the center conductor is small compared to that of the tube. This allows for a sensitive surface resistance measurement of the center conductor which could be a superconducting wire sample. Since numerous modes can be excited, the frequency dependence of the surface resistance can be studied.
A coaxial resonator 10 cm in length and 2.5 cm in diameter was constructed out of a copper tube\textsuperscript{91}. The cavity was hermetically sealed and Helium was used as an exchange gas to cool the center conductor which was a YBCO wire suspended by PTFE sample holders. The resonator was operational between 1 and 6 GHz and was limited by coupling into non-half-wave resonant TE modes in the higher frequencies. Preliminary measurements made by D.B. Opie\textsuperscript{91} found that the surface resistance of a YBCO wire was quadratic in frequency. The coaxial resonator was not used in this work due to time constraints and the need to go to higher frequencies. For this reason the Fabry-Perot resonator was used for frequency dependence studies.

Figure IV-13 The coaxial resonator used by Opie to measure $R_s$ of wires. The dashed line shows the field profile of the n=2 half-wave mode.
Chapter V

The Surface Impedance of Granular Superconductors: Experiment

A. Sample Preparation

The superconducting samples used in this work were synthesized by a diverse selection of techniques and by numerous individuals in different laboratories. Samples are categorized by material and form. All samples used here were either in bulk or thick film form.

For presentation purposes the eleven key samples are summarized in Table V-1. Two thick films of Tl-Ba-Ca-Cu-O (TBCCO) were magnetron sputtered from targets of nominal stoichiometry 2212 (i.e. Tl_2Ba_2CaCu_2O_x) onto Consil 995 substrates by Paul Arendt at Los Alamos National Laboratory. TBCCO#1 and TBCCO#2 were sputtered onto a BaF_2 buffer layer and annealed at 860°C for six minutes in a Tl overpressure. TBCCO#3 was sputtered onto a CaF_2 buffer layer and melted at 910°C for 2 minutes followed by a slow cool. The Tl overpressure anneals were needed because Tl has a low vapor pressure and evolves rapidly from the material above 500°C.

Single phase Bi_2Sr_2CaCu_2O_y (BSCCO) powder was synthesized from correct proportions of Bi_2O_3, SrCO_3, CaCO_3, and CuO by Kevin C. Ott at Los Alamos National Laboratory. In order to get high phase purity it was necessary to sinter the

* Consil 995 is an alloy composed of 99.5% wt Ag, 0.25% wt Mg, and 0.25% wt Ni.
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<td>author</td>
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<td>2212</td>
<td>e-phor</td>
<td>film</td>
<td>40 μm</td>
<td>NA</td>
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<td>20x88a</td>
<td>2223/Pb</td>
<td>sinter</td>
<td>film</td>
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<td>SSC-A</td>
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<td>spray</td>
<td>bulk</td>
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<td>SSC-B</td>
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<td>bulk</td>
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<td>10 μm</td>
</tr>
<tr>
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<td>SSC-C</td>
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<td>123</td>
<td>e-phor</td>
<td>film</td>
<td>12 μm</td>
<td>10 μm</td>
</tr>
</tbody>
</table>

**Table V-1** Summary of the key superconducting samples used in this work. d is the grain size.
powder within 1°C below its melting point. After a brief anneal at 850°C the melting point was determined by analyzing a small portion of the powder with a Perkin-Elmer Differential Thermal Analyzer. The powder was then heat treated near this melting point. The powder was used to prepare samples BSCCO#1 and BSCCO#2 by electrophoretic deposition. BSCCO#1 was deposited onto a 1 inch diameter Consil 995 substrate. It was melted at 870°C for 1 minute prior to anneals of eight hours at 805°C. BSCCO#2 was also deposited onto a 1 inch diameter Consil substrate and annealed at 805°C for eight hours. BSCCO#2 was not melted. Because all films deposited by electrophoresis were prepared by the author, a description of the process will follow over the next few pages.

High phase purity is difficult to achieve with the higher T_c phase Bi_2Sr_2Ca_2Cu_3O_{10}. Early observations showed that the Bi-2223 phase occurred in small unconnected pockets surrounded by the Bi-2212, Bi2201 phases as well as CuO impurities. Attempts to produce Bi-2223 usually resulted in suppressed T_c's of 75-80 K rather than the 110 K transition temperature of the pure phase. Doping the Bi-2223 material with lead was found by Sunshine et al. to be conducive to high phase purity. The lead substitutes the Bi in the lattice and transition temperatures as large as 107 K have been observed in bulk and thick film samples.

A (Bi_{2-x}Pb_x)Sr_2Ca_2Cu_3O_{8+δ} thick film, BSCCO#3, was prepared by Nan Chen of Illinois Superconductor Corporation. The film was sintered in 8% Oxygen at 825°C on an MgO substrate. No buffer layer was used between the BSCCO and the MgO. Although the sample was not melt processed the highly granular film was ð-
axis oriented with rocking angle peak widths around 5°. BSCCO#3, by the way, exhibited the 107 K transition temperature observed in lead doped materials.

High phase purity bulk YBa$_2$Cu$_3$O$_{7.5}$ pellets were synthesized by Seattle Specialty Ceramics (SSC) using their own patented spray pyrolysis technique$^{98}$. Y, Ba and Cu salts were mixed in solution with a proprietary chemical. Very small droplets, which were formed by an atomizer, were dehydrated and heated. The heat fueled an exothermic reaction which resulted in stoichiometrically correct (1 Yttrium, 2 Barium and 3 Copper) granules composed of Y$_2$O$_3$, BaCO$_3$, and CuO. After calcining, the powder was then pressed into pellets and annealed. The pellets YBCO#1, YBCO#2 and YBCO#3 had bulk densities of 5.3g/cm$^3$ which is 84% of the theoretical value. YBCO#1 and YBCO#2 were one inch in diameter. YBCO#3 was two inches in diameter.

Sample YBCO#4 was screen printed onto a 3% yttria stabilized zirconia substrate by Tim Button at ICI, Advanced Materials in Runcorn, England. A proprietary ink containing YBCO was applied to the substrate. The film was then annealed above the Y-Ba-Cu peritectic temperature resulting in oriented films with large grain growth$^{99}$. A peritectic is an isotherm on the phase diagram above which liquid phase and solid phase coexist.

Sample YBCO#5 was a thick film electrophoretically deposited onto annealed silver. YBCO#6 (not in table) was deposited onto a 25 mm diameter and 0.25 mm thick substrate.
Electrodeposition

Superconducting thick films are electrodeposited using a process described by other authors\textsuperscript{25,100,101,102,103} and depicted in Figure V-1. The electrodeposition process and the subsequent heat treatment was optimized for maximum grain size and orientation by Hein\textsuperscript{104}. Superconducting or unreacted precursor powder suspended in a polar medium will form charged crystallites which will migrate in an electrostatic field. This migration is called \textit{electrophoresis}. A potential difference is established between two electrodes in the suspension, one of which is an annealed metallic substrate. The actual flow of current from electrode to electrode is \textit{electrolysis}. The simultaneous occurrence of these two processes are involved in the electrodeposition of thick ceramic films.

Films of uniform thickness can be electrodeposited onto substrates of any geometry\textsuperscript{105}. The uniformity of deposition, or high throwing power, is exploited in the deposition of paint onto automobile bodies and of superconducting films onto
curved surfaces such as maser electrodes. Uniform thickness is achieved because as areas of the arbitrarily shaped substrate closest to the counterelectrode become coated, the accumulation of the resistive film directs the current towards the as yet uncoated areas farther away.

As the entire substrate coats the current begins to drop indicating the gradually increasing electrical isolation of the substrate. A typical current versus time curve is shown in Figure V-2. The large current drop due to the growing film thickness is clearly visible. Taking the bath resistance to be constant during

![Figure V-2](image)  
*Figure V-2  Current vs. time for the electrodeposition of a BSCCO thick film. Electrode area was 1 cm² and electrode separation was 1 cm.*
deposition, the wet film resistance can be estimated from

\[ R_f = V \left[ \frac{I_0 - I}{I_0 I} \right] \]  \hspace{1cm} (108)

where \( I_0 \) is the current when the power is first turned on. The final film resistance calculated from Figure V-2 is approximately 1 M\( \Omega \). As time carries on, the current levels off at some value greater than zero. At this time no more deposition is taking place and all of the current is due to electrolysis.

The optimal method of electrodeposition depends on the ceramic. The most complete study of deposition parameters was performed by Hein. The average grain size and orientation depend upon the granule size in the starting powder and the purity of the starting powder. It was also found that a large magnetic field (\(-5\) Tesla) applied normal to the substrate during deposition resulted in highly oriented films. In the samples reported here small, unoriented grains were desired in order to enhance the effect of granularity. The preparation of these samples is described in the following.

Single phase Bi-2212 powder was ball milled for five minutes then suspended in acetone with a concentration of 3.5g/50ml. A ten minute dispersion process in an ultrasonic cleaner was followed by a six minute sedimentation. Deposition lasted 90 seconds in a 12,000 V/m field. Three depositions were completed on a single one inch diameter substrate each followed by eight hour anneals at 805°C. Surface profilometer measurements show final film thickness of 40 \( \mu \)m and surface
roughness of 10 μm.

Unreacted YBa₂Cu₃ precursor powder was prepared using plasma spray pyrolysis by SSC. The resulting powder is composed of stoichiometrically correct particles of unreacted Y-Ba-Cu. The powder was added to reagent grade acetone with a concentration of 2 grams/liter. The suspension was dispersed for ten minutes and a 90 second deposition followed a one minute sedimentation. The suspension was again dispersed and another 90 second deposition followed. Samples YBCO#5 and YBCO#6 (not in Table V-1) were then annealed for two hours at 880°C. Sample YBCO#6 then received another identical set of depositions. Both samples were then annealed at 915°C followed by a slow cool (10°C/hr) down to 860°C and another slow cooling (10°C/hr) between 500°C and 400°C. The former slow cool is to allow grain growth aided by a liquid phase BaCu flux with an 890°C eutectic. The latter slow cool is to allow the YBCO to be oxygenated. YBCO#5 was 12 μm thick and ybco#6 was about 10μm thick.

**B. Material Characterization**

The materials properties of the samples used in this work were evaluated by optical microscope and X-Ray diffraction (XRD). A 500x optical microscope was used to determine grain size. For thick film and bulk materials it was found that the images yielded by light microscopes lead to easier identification of grain boundaries than those of electron microscopes. A GE electron diffractometer was used to determine phase purity of the samples. The diffraction patterns for YBCO,
BSCCO and TBCCO using Cu Kα radiation have been published and were used to identify the dominant phase(s)\textsuperscript{110,111,112}.

TBCCO\textsuperscript{#1} and TBCCO\textsuperscript{#2}, annealed at 860°C, were small grained (average 5 to 10 μm across). All three TBCCO films were 7 μm thick. The XRD pattern for TBCCO\textsuperscript{#1} is shown in Figure V-3a. This film had three preferred orientations and a high degree of phase purity with Tl-2212 dominating. There is also a small Silver peak due to the Consil substrate. Most thick films on Silver or Consil substrates exhibited this small Silver peak.

The XRD pattern for TBCCO\textsuperscript{#3} is shown in Figure V-3b. The grains were well oriented and mostly of the Tl-2212 phase. Tl-2223 and impurity phases (e.g. CaO) were also present. The Optical microscope revealed very small grains no larger than 10 μm across situated between large grains between 100 μm and 200 μm across, as shown in Figure V-4. Thus, the large grains were weakly connected by three or four small grains.

XRD of the melt textured and non-melt textured Bi-2212 thick films (BSCCO\textsuperscript{#1} & BSCCO\textsuperscript{#2} respectively) demonstrates that melting enhances c-axis orientation. Figure V-5a reveals slight c-axis orientation in the non-melt textured film, whereas Figure V-5b shows only [00f] peaks to the precision of the diffractometer. Traces of Bi-2201 can be identified in the melt textured sample since the low angle [002] peaks are enhanced by orientation. Traces of CuO can also be identified in the melted sample.

The three YBCO pellets, YBCO\textsuperscript{#1}, YBCO\textsuperscript{#2}, YBCO\textsuperscript{#3}, had very high phase
purity with grains about 10 \( \mu \text{m} \) per side. XRD revealed a very large [013/103/011] composite peak indicating no preferred orientation. The XRD pattern of YBCO#2 is in Figure V-6a. An optical microscope photograph is shown in Figure V-4.

YBCO#4 was composed of large grains 500 \( \mu \text{m} \) to 1000 \( \mu \text{m} \) per side. However, with layers of small grains no larger than 5 \( \mu \text{m} \) per side separating these large grains, YBCO#4 had a surface morphology similar to TBCCO#3. The XRD pattern of YBCO#4 in Figure V-6b revealed high phase purity and a visible but suppressed composite peak indicating partial orientation.

The particular granular structures of TBCCO#3 and YBCO#4 is characteristic of melt processing. The formation of large grains results from the fact that small particles melt faster than large particles. The large particles serve as grain growth sites. The smaller grains nucleate out of the melt during the cool down. Lewis et al.\textsuperscript{113} found the large grains of YBCO only in films which were partially melted. If the film is held in the furnace above the melting temperature long enough for a total melt then there are no favored grain growth sites in the melt. This is consistent with the early finding of Licci, Scheel and Besagni\textsuperscript{114} that single crystals of YBCO could not be grown from a total melt. All melt textured films in this dissertation were partially melted.

YBCO#5 was also highly phase pure. It possessed small grains 10 \( \mu \text{m} \) per side. A small degree of porosity was seen with the optical microscope.
C. Temperature Dependence of the Surface Resistance

The temperature dependencies of $R_s$ for the eleven key samples of Table V-1 are presented in Figures V-7 through V-10. The warm-ups were performed both with and without a static magnetic field applied parallel to the film (or pellet) surface. All measurements, except YBCO#3 and YBCO#4, were carried out at 17.5 GHz. YBCO#3 and YBCO#4 were measured at 11.3 GHz.

$R_s(T)$ for TBCCO#1 is in Figure V-7a and for TBCCO#2 is in Figure V-7b. The sensitivity to the static field is greater at low temperature. When a field of 1000 Oe (0.1 T) is applied very little temperature dependence in $R_s$ is observed. The temperature dependence is lost because all of the grains have been decoupled by the shielding currents. This will be discussed later as the first evidence for grain boundary dominated microwave losses.

The large grained TBCCO#3 is seen in Figure V-7c to be less sensitive to the magnetic field than the smaller grained samples. Because the shielding currents have fewer grain boundary junctions to decouple the field has less of an effect on the losses. This will also be discussed later as evidence for grain boundary dominated microwave losses.

BSCCO#1 had a suppressed $T_c$ of 65 K as shown in Figure V-8. Bi-2212 usually has a transition at 85 K. This suppression is caused by the presence of Ni in the Consil substrate. Ni can replace Cu in small amounts and significantly lower the $T_c$. For this reason Consil has been abandoned as a substrate in the electrophoresis program at William and Mary. BSCCO#2, which was not melt
textured exhibited no superconducting phase transition.

Observations of magnetically suppressed temperature dependence of \( R_s \) can also be made by comparing YBCO#2 of Figure V-9a with the melt textured YBCO#4 of Figure V-9b. The small grained pellet was much more sensitive to the field than the large grained film. Low surface resistance of films prepared by the screen printing/melt processing technique used to prepare sample YBCO#4 have been reported by the supplier of YBCO#4\textsuperscript{116}.

A number of the samples tested in this work had negative temperature coefficients of surface resistance, and likewise resistivity, in the normal state. This is shown in Figure V-10 for YBCO#5. This is not to be interpreted as semiconductivity which is a property of the band structure. The negative coefficient is rather a consequence of the granularity and is due to thermally activated tunnelling across boundaries between conducting grains\textsuperscript{117}.

As discussed in Chapter IV surface reactance cannot be directly measured versus temperature with a cavity resonator. However, Orbach\textsuperscript{118} obtained \( \lambda(T) \) curves for epitaxial films using a Gorter Casimir fit.

D. Magnetic Field Dependence of the Surface Impedance

Numerous researchers\textsuperscript{119,120,121,122} report an increase in both \( R_s \) and \( X_s \) as an applied static magnetic field is increased. This behavior is also observed here as
shown in Figures V-11 through V-13. As the growing shielding currents decouple more grains the surface resistance increases. Eventually all of the grains are decoupled and the surface resistance stops increasing. This saturation is seen in Figure V-11 for TBCCO#1. The surface reactance saturates in the same manner.

Figure V-11 may lead one to believe that the shielding currents caused by the field affect $R_s$ in the same manner as they affect $X_s$. However examination of the same measurements on the pellet YBCO#2 in Figure V-12 indicates that a more complicated process must be governing the field penetration. An important peculiarity is observed in $\Delta \lambda_{\text{eff}}$. As the field is increased at 77 K the effective rf penetration depth (equivalently surface reactance) actually begins to decrease above 12 Oe. At 86 K the surface reactance begins to decrease immediately upon application of a magnetic field. It is this apparent improvement in one of the superconductor's properties with increasing field that is at the heart of this work.

The large grained TBCCO#3 is much more resilient in a magnetic field than the small grained samples. Whereas application of a 60 Oe field at 12 K caused a 500% increase in $R_s$ for TBCCO#1 only a 70% increase resulted for TBCCO#3. A 30 Oe field caused an 800% increase in $R_s$ of YBCO#2 but only a 50% increase for the large grained YBCO#4. No negative magnetic field coefficient of the effective rf penetration depth was observed in the large grained samples.

In a separate experiment the surface impedance of TBCCO#1 and TBCCO#3 was measured at 12 K to a higher field of 1200 Oe. This allows the observation in Figure V-13 that even in saturation at low temperature there exists a very small slope
in both $R_s$ and $X_s$. Although the large grained TBCCO#3 had a zero field residual surface resistance which was similar to the small grained TBCCO#1, it saturated at a much lower value of $R_s$.

As the static magnetic field is ramped back down, hysteresis is observed in both the surface resistance and the surface reactance. Because flux can remain trapped in the grain boundary junctions after the field is removed, some of the grain boundary junctions remain in the voltage state\textsuperscript{129}. Hysteresis in both the $R_s(H)$ and $\Delta X_s(H)$ of TBCCO#1 at 12 K is shown in Figure V-14. Of interest is the observation that the surface resistance and surface reactance have identical hysteresis. For any given value of $R_s$ there exists only one corresponding value of $X_s$ regardless of whether there is a history of applied magnetic field.

E. Frequency Dependence of the Surface Resistance

Measurement of surface resistance versus frequency, $f$, poses a significant technical challenge. Most conventional cavity resonators are design to measure $R_s$ of films in one or two modes only. For example the cylindrical cavity described in Chapter IV can be operated in the $TE_{011}$ mode at 11.3 GHz and in the $TE_{013}$ mode at 16.5 GHz. It is typically necessary to use a variety of cavities if one wants to measure $R_s(f)$ over a large microwave frequency range. It was the goal in designing and building the Fabry-Perot resonator, with its wide tuning range, to measure $R_s(f)$ with only one resonator. Measurements made with the Fabry-Perot resonator will
be presented in Chapter VI.

Woodall et al.\textsuperscript{124} measured the surface resistance of bulk YBCO wires which were used as the center conductor in a coaxial resonator. Their resonator supported 17 modes between 1 and 20 GHz. Measurements performed in this frequency range found that the surface resistance of the granular YBCO depended upon frequency approximately as $R_s \propto f^{1.4}$. An important figure of merit is the \textit{cross-over frequency} with copper. Because the frequency dependence of the $R_s$ of a superconductor is stronger than the $R_s \propto f^4$ of a normal metal, there exists some frequency above which the superconductor is more lossy than copper. For HTSC the cross-over frequency lies between 10 GHz, for low quality granular materials, and 80 GHz, for high quality epitaxial films. The determination of the cross-over frequency is one important application of the Fabry-Perot resonator.

Delayen and Bohn\textsuperscript{125} measured the frequency dependence of YBCO wires as well with a coaxial resonator. Their measurements were performed between 4.2 K and 92 K and between 243 MHz and 1.041 GHz. Quadratic frequency dependence of the surface resistance was found at all temperatures in their experiments. However, this only indicates that the low frequency surface resistance depends quadratically upon the frequency. In Chapter VII weaker frequency dependence in the millimeter wave regime will be demonstrated.
Figure V-3 X-Ray diffraction pattern of (a) TBCCO#1 and (b) TBCCO#3.

Figure V-4a Optical micrograph of sample TBCCO#3 at 100x. The scale is 1000 µm from the left to the right edge of the photo.
Figure V-4b  Optical microscope photograph of sample TBCCO#3 at 500x illustrating the layers of small grains between the large grains. The scale is 200 μm from the left to the right edge of the photo.

Figure V-4c  Optical microscope of sample YBCO#2 at 500x.
Figure V-5a X-Ray diffraction pattern of the melt textured sample BSCCO#. Unlike the melt textured sample of Figure V-5a, this sample exhibits more than just [0,0,l] peaks.
Figure V-6 X-Ray diffraction pattern of YBCO#4 (top) and YBCO#2 (bottom).
Figure V-7a  Temperature dependence of the surface resistance of sample TBCCO#1 in various static magnetic fields applied parallel to the sample surface.
Figure V-7b  Surface resistance versus temperature of sample TBCCO#2.
Figure V-7c  Temperature dependence of the surface resistance of the large grained sample TBCCO#3. Because of the larger average grain size and higher degree of orientation, the sample is more resilient under a static magnetic field.
Figure V-8 Temperature dependence of the surface resistance of the sample BSCCO#1.
Figure V-9a Temperature dependence of the surface resistance of sample YBCO#2 in various static magnetic fields applied parallel to the surface.
Figure V-9b Temperature dependence of the surface resistance of sample YBCO#4. Because of the larger average grain size and higher degree of orientation, the sample is more resilient under a static magnetic field.
Figure V-10 Temperature dependence of the surface resistance of sample YBCO#5. The negative temperature coefficient is indicative of granularity.
Figure V-11 Static magnetic field dependence of (a) the surface resistance and (b) the surface reactance of sample TBCCO#1 at 17.3 GHz ($T_c=101$ K). The field was applied parallel to the sample surface.
Figure V-12 Static magnetic field dependence of (a) the surface resistance and (b) the surface reactance of sample YBCO#3 at 11.3 GHz ($T_c=92$ K). At higher temperature the effective penetration depth is reduced with additional field. The field was applied parallel to the sample surface.
Figure V-13 Static magnetic field dependence of the surface resistance of the small grained TBCCO#1 and the large grained TBCCO#3 at 17.5 GHz and 12 K ($T_c$=101 K for both). The field was applied parallel to the sample surface.
Figure V-14 Static magnetic field dependence of (a) the surface resistance and (b) the surface reactance of the small grained sample TBCCO#1 at 17.5 GHz. The direction of hysteresis is indicated by the arrows.
Chapter VI

The Surface Impedance of Granular Superconductors: Theory

A. The Two Fluid Model and Mattis Bardeen Theory

The superconducting state was described phenomenologically by Gorter and fluids. The fluid of normal conducting electrons exists at all temperatures greater than T=0. A second fluid of superconducting electrons has zero density above the critical temperature, T_c. Empirical results of the specific heat of superconductors lead to the conclusion that between T=0 and T=T_c the density of the normal fluid, n_n, relative to that of the superfluid, n_s, varies continuously with temperature from zero to unity as

\[ \frac{n_s}{n} = 1 - \left( \frac{T}{T_c} \right)^4 \]  \hspace{1cm} (109)

where n is the total electron density, n=n_s+n_n.

In the relaxation time approximation, the equation of motion of the normal electrons in an AC electric field, E, is

\[ \frac{d\nu_n}{dt} + \frac{\nu_n}{\tau} = -\frac{eE}{m_e} \]  \hspace{1cm} (110)

where \( \nu_n \) is the velocity of the normal fluid and \( \tau \) is the collision relaxation time. Both the electric field and the electron velocity have harmonic time dependence with frequency \( \omega \). Because the superelectrons conduct without collisions (e.g. \( \tau \to \infty \)), the
The superfluid equation of motion is

\[
\frac{dv_s}{dt} = -\frac{eE}{m_e}.
\] (111)

The total electron current, \( J = J_s + J_n = -n_e e v_e - n_n e v_n = \sigma E \), can be calculated from Equations 110 and 111.

Inserting the solutions to Equation 109 and Equation 110, for harmonic time dependence, into Equation 111, rearranging, and solving for \( \sigma \) gives

\[
\sigma = \frac{e^2 n_e \tau}{m(1 + \omega^2 \tau^2)} - j\frac{n_e e^2}{\omega m} - j\frac{e^2 n_n (\omega \tau)^2}{m \omega (1 + (\omega \tau)^2)}
\] (112)

or,

\[
\sigma = \sigma_1 - j\sigma_2
\] (113)

where \( \sigma_1 \) and \( \sigma_2 \) are the real and imaginary parts of the conductivity. Thus the conductivity of a superconductor is always complex and frequency dependent. As \( \omega \to 0 \), \( \sigma_2 \) is proportional to \( j/\omega \) and \( \sigma_1 \) is constant. At high frequency \( \sigma_1 \propto 1/\omega^2 \) and \( \sigma_2 \propto j/\omega \). But between the low and high frequency extremes \( \sigma_1 \) is finite. One important note is that \( \sigma \) is often written as \( \sigma = \sigma_s + \sigma_n \) where \( \sigma_n \) is complex and represents the terms of \( \sigma \) containing \( n_n \). At low frequency \( \sigma_n \) is real and nonanomalous. \( \sigma_s \) represents the term of \( \sigma \) containing \( n_s \).

If the complex conductivity is inserted into the surface impedance,

\[
Z_s = (j \omega \mu_s / (\sigma_s + \sigma_n))^{1/2}
\]

the two-fluid model surface impedance for \( \omega \tau \ll 1 \) results.
where the London penetration depth is \( \lambda_L = (\frac{m}{2\mu_0 n_e e^2})^{1/2} \). If \( \omega \ll 1 \) (e.g. microwave frequencies or lower) then \( \sigma_n \) is frequency independent. This yields the very important results that \( R_s \propto \omega^2 \lambda_L^3 \) and \( X_s = \omega \mu_0 \lambda_L \). The result that the surface resistance should depend quadratically upon the frequency is of fundamental importance to this work.

In Chapter V the frequency dependence of \( R_s \) as measured by other authors was reported. In Chapter VII new results obtained with the Fabry-Perot resonator will be presented. The frequently observed weaker than quadratic frequency dependence is a signature of granularity. Surface reactance, on the other hand, is by definition \( \omega \mu_0 \lambda_{\text{eff}}(\omega) \), where \( \lambda_{\text{eff}}(\omega) \) is the effective rf penetration depth, which can be frequency dependent.

This model of Gorter and Casimir is approximately applicable to classical superconductors and, in many cases, gives good qualitative agreement. However, in the presence of a material discontinuity, such as a Josephson junction, the impedance to the supercurrent becomes very reactive\(^{128}\) and the resistance to the normal current becomes significant. Although, there are two fluids crossing the material interface, the impedance picture must be altered. This is the case for a granular superconductor composed of an array of superconducting crystallites bordered by Josephson junctions.

Müller\(^{129}\) has suggested that perhaps for HTSC \( \sigma_1 \) has a temperature independent (or weakly temperature dependent) residual term, \( \sigma_{\text{res}} \). A consequence of such a
modification of the two-fluid model is that there remains an excess of unpaired charge carriers greater than $n(T/T_c)^6$. This explains why the rf losses in HTSC appear to be limited intrinsically to something greater than the BCS prediction. It would also result in a frequency dependence less than $\omega^2$, which is consistent with measurements of polycrystalline films. However, it is the goal of this work to demonstrate that residual losses in granular superconductors are dominated by grain boundary dissipation.

**Mattis-Bardeen Theory**

If the mean free path, $l$, of electrons in the superconductor is much smaller than $\xi$ then the superconductor is said to be in the *BCS dirty limit*\(^2^8\). If $l > \xi$ (as it is for HTSC) then the superconductor is in the *BCS clean limit*. In the clean limit $\sigma_i \ll \sigma_2$ and $\sigma_2 = 2n_s e^2/\omega m$ as given by the two fluid model. The BCS result for $\sigma_i$, worked out by Mattis and Bardeen\(^2^7\) (M-B), is the same in both the clean and dirty limits. M-B predict a bump in $\sigma_1$ just below $T_c$. Although measurements of $\sigma_2$ for YBCO resemble the BCS clean limit, the BCS bump is usually missing from $\sigma_1$ measurements\(^2^8\).

The M-B conductivity was used to calculate the surface impedance of superconductors by P.Miller\(^1^3^0\). Agreement between Miller's calculations and $Z_s$ of superconducting Aluminum and Tin was demonstrated. Due to the possibility of strong BCS coupling (e.g. $2\Delta(0)/k_BT_c > 3.5$) in HTSC materials, M-B theory has not always rendered an accurate description of the microwave properties of HTSC\(^1^3^1\). However, an important prediction of M-B theory for weak BCS coupling is that as the angular frequency is increased at low temperature there should be a rapid rise in $R_s$ at
\[ -3.5k_B T_C / \hbar \] For YBCO, \( T_C = 93 \) K, there should be an absorption edge in the infrared region \( \sim 10^{13} \) Hz which has been observed in numerous optical experiments. Because of the high transition temperature, \( \hbar \omega \) for microwave frequencies is much lower than the energy gaps of the HTSC materials, and large microwave absorption is avoided until much higher frequencies than for LTSC materials.

B. The Weakly Coupled Grain Model

1. Experimental Evidence for Granular Losses

The need to consider the contribution of granularity to the microwave losses in polycrystalline HTS has been established by the results of Chapter V. The significant contribution by granularity is evidenced by the following four details of the data.

- **There exists a large low temperature residual surface resistance,** \( R_{\text{res}} \). A good epitaxial film at 17.5 GHz will have \( R_{\text{res}} \sim 100 \) m\( \Omega \). The polycrystalline bulk and thick film samples exhibit \( R_{\text{res}} \) from 1 to 50 m\( \Omega \).

- **The surface resistance loses its temperature dependence when a large magnetic field is applied.** Mannhart found that HTSC Josephson junctions often have a temperature independent \( R_N \) while in the high voltage regime \( (V \sim 2mV) \). In a large magnetic field the grain boundaries are in a high voltage state with a temperature independent resistance.

- **The normal state surface resistance of many samples decreases with increasing temperature.** Conductivity across grain boundaries is thermally activated as described in the theory by Abeles. For the cases of high angle
grain boundaries and poor $\hat{c}$-axis orientation, the thermal activation energy is even larger. If the grain boundaries are able to dominate losses, then there exists the potential for them to enhance the conductivity at higher temperature.

The surface impedance of the above granular superconductors is sensitive to DC magnetic fields which are more than an order of magnitude smaller than the bulk $H_{c1}$. In the HTSC's $H_{c1}$ is $\sim 10^2$ to $10^3$ Oe for $H$ parallel to the $\hat{c}$-axis and $\sim 40$ to 100 Oe for $H$ normal to the $\hat{c}$-axis$^{135}$.

That the second observation is indicative of granularity depends upon the premise that HTSC grain boundaries are Josephson junctions. Mannhart, as well as a number of other authors such as Marcon, et al.$^{136}$ and Vad, et al.$^{137}$, have found clear evidence of the Josephson effects in HTSC grain boundaries. Mannhart directly measured the I-V characteristics of YBCO bicrystalline films. Marcon et al. successfully applied a Josephson junction array model to the microwave absorption of polycrystalline YBCO samples. Their results yielded values for $H_{c1}$ between 3 and 6 Oe. Vad, et al. did similar work with the BSCCO materials.

Another useful clue to the role of granularity was the finding by Küpfner et al.$^{138}$ that the coupling between the grains of YBCO, TBCCO and possibly BSCCO is weak. From a practical standpoint that means that the intergrain critical current is much lower than the intragrain critical current. The weak coupling manifested itself in a second large hysteresis peak in the imaginary susceptibility below $T_C$. This second peak corresponded to intergrain losses and occurred not immediately below $T_C$, but rather just
below the temperature at which grains become phase locked.

Chaudhari et al.\textsuperscript{139} found that grain boundaries artificially patterned with an excimer laser into high quality epitaxial films of Y-123 exhibited $J_c$'s which were considerably lower than those of individual grains. Furthermore, they found that the high quality grain boundaries had regions of strong coupling and regions of weak coupling.

The results of Mannhart, Marcon and Küpfer lead to a picture of the granular superconductor with a large Ginsburg-Landau parameter which is accurately depicted as a three dimensional array of Josephson junctions. In most cases only the surface of the superconductor is exposed to the field and a two dimensional array is a more appropriate picture of the superconductor. These junctions fill a range of junction widths and thicknesses, as well as a range of bicrystal orientations.

Cooke et al.\textsuperscript{140} summarize this variety of junctions by modelling the

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{josephson_coupled_block_model.pdf}
\caption{Josephson coupled block model of Clem. DC pair transport across the block boundaries is treated by Clem with this model.\textsuperscript{142}}
\end{figure}
superconducting surface as an array of junctions with a distribution of saturation fields. The saturation field is defined as that applied rf field above which the junctions described by that field contribute no more additional rf loss. This treatment is appropriate for high rf power since it is essentially an rf critical state problem.

When a granular superconducting surface, with $\xi \ll a$, is exposed to a static magnetic field shielding currents are established and some of the grain boundary Josephson junctions go into the voltage state. The junction is then resistive to quasiparticle transport. Paired carriers maintain phase coherence as they tunnel across the junction and are met only by the junction kinetic inductance.

When a finite frequency field is applied to the surface the paired electrons crossing the grain boundary are met by an inductive reactance. Because this reactance is finite, the normal carriers are not perfectly shunted and are able to cross the junction as well. This normal, or quasiparticle, conduction is resistive$^{141}$. Thus, under this circumstance, the inductive pair tunnelling is resistively shunted.

2. Theoretical Prelude to the Weakly Coupled Grain Model

In 1989 J.R. Clem published a phenomenological theory of layered superconductors$^{142}$ which describes the large magnetic penetration depth and resistivity due to the layered structure of the HTS materials. These property enhancements occur in Clem's model when the superconductor is subjected to a magnetic field. The superconducting layers (e.g. a-b planes) are divided further into rectangular blocks (Figure IV-1) which are Josephson coupled. When in the voltage state the blocks have
an effective resistivity

\[ \rho_i = \rho_{lo} \frac{a_i' A_i}{a_{i'} A_{i'}} + R_{in} \frac{A_i}{a_i} \]  \hspace{1cm} (115)

where \( \rho_{lo} \) is the intrinsic normal state resistivity of the superconductor. \( R_{in} \) is the tunnel resistance between two blocks. As depicted in Figure VI-1, \( A_i = a_j a_k \) and \( A_i' = a_j' a_k' \) are the areas of the sides of the blocks with and without the junction, respectively.

Although Clem's model was not intended to describe granularity, the implications are clear. Clem's goal was to describe the intrinsic anisotropy of the HTS materials. The model does indeed demonstrate that it is possible for a layered superconductor to behave as if it were in the BCS clean limit along one direction and in the dirty limit along another. But it is the notion of a Josephson coupled array of superconducting grains which is applicable to granular materials.

In a theoretical investigation into the Hamiltonian of a phase locked array of Josephson junctions, Zagrodnzinski\textsuperscript{143} described an array of coupled superconducting grains as a material characterized by a maximum magnetic field which destroys the lossless flow of supercurrents. Using the AC Josephson effect, phase locked Josephson arrays have been exploited since the early 70's as submillimeter wave sources\textsuperscript{144}. In recent years this work has begun to yield high power levels.
3. The Model

Hylton et al.\textsuperscript{145} were the first to realize the relevance to the microwave surface impedance of the above mentioned resistively shunted Josephson junction. It was only a matter of time before the rf specific impedance of HTSC would be modeled by a kinetic inductivity shunted by a lossy resistivity.

The intrinsic conductivity for a defect free superconductor with a unity order parameter (i.e. at zero temperature) is \( \sigma_0 = -j/\omega L_G \), where \( L_G = \mu_0 \lambda_L^2 \) is the kinetic inductivity of the intrinsic superconductor. The G subscript indicates grain as it will later be used to represent the grains of a polycrystalline surface. This conductivity, when inserted into Equation 11, yields a purely imaginary, or reactive, surface impedance. If the current is harmonic with time dependence \( e^{j\omega t} \), then the reactivity of the superconductor is \( x_L = +j\omega L_G \). At finite temperature the Mattis-Bardeen conductivity, or the Gorter-Casimir conductivity, can be invoked to describe the resistive

\textbf{Figure VI-2} Superconducting block picture. For clean contact, the separation of decoupled grains is \( 2\lambda_L \).
term for the intrinsic impedance. This resistivity will be regarded as negligible compared to the resistivity of the grain boundary.

The weakly coupled grain model describes the response of grain boundary junctions to rf currents by a resistively shunted kinetic inductance. Paired electrons cross the junction with an inductive electrodynamics described by $L_j=\mu_0\lambda_j^2$, which has dimensions of Henry-meters. $\lambda_j$ is the Josephson penetration depth of the grain boundary junction. The parallel combination of the junction inductivity, $L_j$, and resistivity, $R_j$ (dimensions Ω-m), yields a junction conductivity

$$\sigma_j = \frac{1}{R_j} - \frac{j}{\omega \mu_0 \lambda_j^2}$$

(116)

The conductivity of the grain is in series with the grain boundary, reflecting the temporal difference between the grain transit and the boundary transit of each carrier. Therefore, the conductivities of a grain boundary and its bordering grains combine as ($L_G$ and $R_j$ are inductivity and resistivity, respectively. See above.)

$$\frac{1}{\sigma} = j\omega L_G + \frac{j R_j \omega \mu_0 \lambda_j^2}{R_j + j\omega \mu_0 \lambda_j^2} \cdot \frac{d}{a}$$

(117)

where $j\omega L_G$ is the only significant part of the grain's conductivity. The grain-grain boundary system is depicted as an equivalent circuit in Figure VI-3.

One additional conductivity mechanism was included in the picture by Portis, et
Figure VI-3 Equivalent circuit for the weakly coupled grain model. The grain and shunt inductivities are also depicted.

al.\textsuperscript{140} to account for the possibility of carriers bypassing the junction. A shunt inductivity, $L_s$, represents those carriers which find their way around the defects. Because the short is simply composed of the superconducting material, the specific kinetic inductivities, $L_G$ and $L_s$, are equivalent. The Josephson penetration depth, $\lambda_j$, is much larger than the London penetration depth. Consequently it is also true that $L_j \gg L_s$. Thus, when the shorts do occur the effective inductivity is significantly reduced. This becomes an issue in high quality epitaxial films where currents often find shorts around the junction defects\textsuperscript{146}. When granularity dominates the material continuity then Portis argues that these grain boundary shorts can be neglected.
In another paper, Portis solves the sine-Gordon equation for a Josephson junction with a small microwave field superimposed upon a large DC magnetic field. He uses the $\lambda_j$ large limit and the assumption that the superconducting phase varies slowly along the junction to derive a wave vector which to first order describes a resistively shunted inductor.

4. Effective Medium Parameters

It must be emphasized that the above discussion was for a single grain boundary and its two neighboring grains. In 1981 Ioffe and Larkin used percolation theory to describe the smeared superconducting phase transition which resulted from material inhomogeneities. They reduced the distribution of inhomogeneity properties to one single effective inhomogeneity. This same effective medium approach was followed by Hylton et al. in 1988 to reduce the grain boundary array to a single grain boundary. The previous discussion is, in fact, adequate to perform model analysis. All of the features of the data discussed in the previous chapter can now be explained. Furthermore, the frequency dependence can now be predicted.

The results are the same regardless of whether $L_j$ and $R_j$ are understood to be junction parameters or effective medium parameters. The relation between the two parameter sets was determined by Portis and Hein using a simple geometrical argument. The matrix of grains is shown in Figure VI-2. For a clean contact between
grains, the junction thickness is simply its magnetic thickness, \(2\lambda_L\). The resistivity and kinetic inductivity of the effective medium of junctions are \(\rho_j\) and \(\ell_j\). The kinetic inductivity relates the current to the electric field\(^{149}\) by \(\ell_j(dJ/dt) = E_j\). For the individual grain and for the effective medium we have respectively

\[
L_j \frac{dJ_j}{dt} = E_j
\]  
(118)

and

\[
\ell_j \frac{d<J>}{dt} = <E>
\]  
(119)

where \(J_j\) and \(E_j\) are the microwave current and microwave field in the junction. \(<J>\) and \(<E>\) are the average microwave current and microwave field at the surface. The two averaging assumptions are that \(<J>=J_j\) and that \(<E>=(2\lambda_L/a)E_j\) where \(a\) is the linear dimension of the grain. The field is scaled linearly in the ratio of grain boundary area to total sample area in order to average the inertia of the carriers over the entire medium. Combining Equations 118 and 119, the averaging assumptions result in

\[
\ell_j = \frac{2\lambda_L L_j}{a}
\]  
(120)

and

\[
\rho_j = \frac{2\lambda_L R_j}{a}.
\]  
(121)
From hereon the symbols for the effective medium, $\ell_j$ and $\rho_j$, will be used.

Hylton et al.\textsuperscript{145} wrote the complex conductivity, Equation 117, in terms of the $I_cR$ value of the Josephson junction. From Equation 10 in Chapter 1 the average junction kinetic inductivity is

$$l = \mu_0 \lambda_f^2$$

$$= \frac{\hbar}{2edI_c}$$

(122)

where $I_c$ is the average junction critical current. Hylton, et al.\textsuperscript{145} get

$$\frac{1}{\sigma} = j \mu_0 \omega (\lambda_L^2 + \lambda_J^2) + \omega^2 \mu_0 \lambda_J^2 \frac{\hbar}{2eI_cR}$$

(123)

expressed in terms of the effective $I_cR$ value.

5. Surface Impedance from the Weakly Coupled Grain Model

At this point the surface impedance can be written in terms of the equivalent circuit elements. It is the intent of this work to study highly granular materials. Therefore, the limit $\ell_o \ll \ell_j$ will be used. The effective medium conductivity, $\sigma_{eff}$, is expressed by replacing $R_j$ and $L_j$ in Equation 116 with the effective medium symbols $\rho_j$ and $\ell_j$, resulting in

$$\frac{1}{\sigma_{eff}} = \rho_j \frac{j \omega^{-1} + 1}{1 + \omega^{-2}}$$

(124)
where

\[ x = \frac{\omega l_j}{\rho_J} . \]  

(125)

Inserting Equation 124 into Equation 11 gives

\[ R_s = R_C \sqrt{\frac{x}{\sqrt{1+x^2}} - \frac{x}{1+x^2}} \]  

(126)

and

\[ X_s = R_C \sqrt{\frac{x}{\sqrt{1+x^2}} + \frac{x}{1+x^2}} , \]  

(127)

where

\[ R_C = \sqrt{\frac{1}{2} \omega \mu_0 \rho_J} \]  

(128)

is called the classical surface resistance.

\( R_C \) is not expected to be strongly temperature dependent. Likewise it is independent of DC magnetic field. The very weak temperature dependence of \( R_j \) of the grain boundary Josephson junctions is the only potential source of variation in \( R_C \). As previously mentioned such a variation is usually not observed in published results.

In the limit of small \( x \), we have for the surface reactance
where $x$ is small, but not so small that $\ell_s$ is significant. Using Equation 10 in Chapter II, we arrive at the useful result for small $x$

\[ X_s \approx \omega \mu_0 \lambda_f. \]  

(130)

Because both $\rho_s$ and $\ell_s$ are frequency independent, $x$ is linear in frequency. If $x$ and $R_s$ can be determined at one frequency then the surface resistance at any other frequency can be predicted using Equation 126. In the limit of small $x$ (good superconductor) the dependence $R_s \propto \omega^2$ is recovered. Likewise, in the classical limit of large $x$ (normal-like conductor) the dependence $R_s \propto \omega^{\frac{1}{2}}$ is recovered. Hein has determined the frequency dependent frequency exponents for $R_s \propto \omega^n$

\[ n(\omega) = 1 + \frac{x^2}{2(1+x^2)} \cdot \left[ \frac{1}{1-(1+x^2)^{-\frac{1}{2}}} - 2 \right]. \]  

(131)

The model parameter, $x$, is the important variable. It depends monotonically on temperature, frequency, static magnetic field and microwave power in a complicated manner. A more complete physical understanding of $x$ is achieved by considering the wave vector in the superconductor

\[ k^2 = \frac{1}{\lambda^2} + \frac{2j}{\delta^2} \]  

(132)

which is equivalent to $k^2 = j\omega \mu_0 \sigma$. Equating these two expressions for $k$, and using the
effective medium version of Equation 124, gives

\[ x = \frac{2\lambda^2}{\delta^2} \quad (133) \]

where \( \lambda \) is the effective superconducting microwave penetration depth which goes to infinity at \( T_C \). \( \lambda \) is the microwave field penetration corresponding to the combined effective medium inductivities \( \ell_0, \ell_s \) and \( \ell_j \) depicted in Figure VI-3. For the highly granular materials in this work the shunt and granular inductivities are negligible and \( \lambda = (\ell_j/\ell_0)^{1/4} \).

It is important to note that Equations 125 and 133 are defining the same parameter, \( x \). The important point here is that \( \omega \ell_j/\rho_j \) is equivalent to \( 2\lambda^2/\delta^2 \). Thus, the observed temperature, magnetic field and frequency dependence of the surface impedance depends on the ratio of superconducting penetration depth to skin depth.

At low temperature, frequency and magnetic field \( x \) is small. As these quantities rise, \( x \) also rises. Close to \( T_C \) \( x \) approaches infinity and \( R_s \approx X_s \approx R_C \). The classical surface resistance is not to be confused with the value of \( R_s \) at \( T_C \). Rather it corresponds roughly to the value of the surface resistance at the temperature where magnetic field dependence vanishes. The field dependence of \( R_s \) vanishes at the temperature, \( T_{ch} \), where all of the grains become thermally decoupled. This grain decoupling is observed in Figures V-7 and V-9 to occur within a few kelvin of \( T_C \).

The temperature, magnetic field and frequency dependence of \( x \) can be predicted. Since \( \rho_j \) is at most very weakly temperature dependent, the kinetic inductivity, which
increases with temperature, governs the temperature dependence of $x$. $\rho_f$ is also
independent of magnetic field. From Equation 4 the Josephson kinetic inductivity is
$\ell_J(H) = \Phi_0 / 2\pi J_{cJ}(H)d$. Also recall at sufficiently high field ($H > H_{c1}$)
$J_{cJ}(H) = J_o / [1 + (H/H_o)]$, where $J_o$ and $H_o$ are field independent constants. This leaves

$$\ell_J(H) = \ell_J(0) + \alpha H$$  \hspace{1cm} (134)

which can be tested experimentally. Finally, since resistivity and inductivity are
frequency independent properties, $x$ is predicted to depend linearly upon frequency.

The linearity of Equation 134 cannot be measured in high magnetic fields
because the surface impedance becomes field independent. This saturation in field was
shown in Figure V-13. The junction kinetic inductivity continues to increase as the field
is ramped up. However, when the magnitude of $\ell_J$ approaches that of $\ell_s$, the response
of the surface impedance to the magnetic field weakens. This is because the shunt
kinetic inductivity is field independent and dominates the grain boundary in high
magnetic field. Thus, if $\rho_f$ has been determined, then the saturation value of $R_s$ gives
$\ell_s$ directly from Equation 126 where $x_M = M/\rho_f$.

Crucial to testing the weakly coupled grain model is the determination of the
slope $dR_s/dX_s$. In Figure VI-4 $R_s/R_c$ is plotted against $X_s/R_c$ from Equations 126 and
127. The surface resistance and surface reactance are normalized to $R_c$ in order to
make $x$ the only implicit parameter. The arrow indicates the direction of increasing $x$.
Of course, temperature, magnetic field, frequency and rf power are all implicit
parameters governing $x$. For $x$ small $dR_s/dX_s$ is very small but positive. As $x \to \infty$,
Figure VI-4 The normalized surface resistance of Equation 126 is seen here to be double valued in the normalized surface reactance of Equation 127.
dR_s/dX_s\to -1. When x=1.728, dR_s/dX_s\to \infty for increasing x, and dR_s/dX_s\to -\infty for decreasing x. Thus, the surface resistance is double valued in surface reactance. More importantly, the surface reactance is double valued in magnetic field, temperature, frequency and microwave power. This is the first direct corroboration between the weakly coupled grain model and the data from Chapter V. Figure V-12 shows a surface reactance which is indeed double valued in magnetic field.

At this point a brief discussion of the distinction between penetration depth and skin depth is in order. The microwave surface reactance, \(X_s=\omega \mu_o \lambda_{\text{eff}}\), is a measure of the effective penetration, \(\lambda_{\text{eff}}\), of the microwave field into the superconductor. \(\lambda_{\text{eff}}\) is governed by both the skin effect and the superconductive shielding. From Equation 132

\[
\alpha = -j \frac{1}{\mu_o \omega} \left[ \frac{1}{\lambda^2} + \frac{2j}{\delta^2} \right].
\]  

(135)

If the skin effect were ignored, then inserting Equation 135 into Equation 11 from Chapter II would yield a purely reactive surface impedance. It is due to the simultaneous occurrence of the skin effect and superconductive shielding that the surface resistance of a superconductor is non-zero. For an ideal superconducting material the London depth and the skin depth calculated from BCS theory would be used in Equation 135.
C. Contribution of Flux Flow to the Surface Impedance

At low magnetic fields fluxons remain pinned to material defects. When a superconductor is subjected to a large magnetic field, or equivalently to a large current, the fluxons can be depinned by the current/fluxon Lorentz force interaction\(^1\). Fluxon can also be depinned by thermal activation. This is a process known as \textit{flux creep} where fluxons hop between pinning sites. In either case the fluxon must acquire enough energy to overcome the pinning energy.

Energy is dissipated when fluxons move. Flux motion can be thought of as a bundle of quasiparticles being dragged across the superconductor. There are other loss mechanisms such as magnetic relaxation. When a magnetic field, \(B\), moves with velocity, \(v\), there is an electric field \(E=\mathbf{B} \times \mathbf{v}\). Since \(v\) is parallel to the Lorentz force, \(F_L=\mathbf{J} \times \mathbf{B}\), \(E\) is parallel to the supercurrent, \(J\). Thus, flux motion induced electric field is dissipative\(^1\).

The force acting on the charge carriers moving with velocity \(v\) in the presence of flux motion is then a combination of the electric field force, \(eE\), and the Lorentz force, \(ev \times B\). This gives an equation of motion\(^1\)

\[
m \frac{d\vec{v}}{dt} = e(\vec{E} + \vec{v} \times \vec{B}) .
\]  

(136)

If the fluxon velocity is rewritten in terms of its viscosity, \(\eta=-ev\Phi_0/V\), and the fields are harmonic in time, then Portis and Hein\(^1\) solve for the impedance to the current, \(1/\sigma_{ff}=E/J=E/nev\),

\[
1/\sigma_{ff}=E/J=E/nev ,
\]
This leads us to the equivalent circuit representation for flux flow impedance of a resistivity in series with an inductivity. Inserting Equation 137 into Equation 11 gives

\[
\frac{1}{\sigma_{ff}} = \frac{\Phi_0 B}{\eta n^2} - j\omega m. \tag{137}
\]

where \(x = \omega \ell_{ff}/\rho_{ff}\) and \(R_0 = (\omega \mu_0 \rho_{ff}/2)^{1/2}\). \(\ell_{ff}\) and \(\rho_{ff}\) are the flux flow inductivity and resistivity deduced from Equation 137.

A plot Equation 138 versus Equation 139 reveals a single valued curve. This is important because if flux flow dominates the microwave losses then the surface reactance will never be double valued in magnetic field. Pambianchi et al.\(^{152}\) found that flux flow indeed dominates the losses in their epitaxial thin films which are studied in DC magnetic fields with a parallel plate resonator. It will be concluded in the next chapter that flux flow is not the dominant loss mechanism in the granular samples studied here.
In a more complete parameterization, Coffey and Clem\textsuperscript{153} account separately for the losses due to flux flow and flux creep. They write the complex penetration depth, \( \lambda(\omega, B, T) \) in terms of the London depth, the normal fluid skin depth, the flux flow resistivity, and the flux creep factor which indicates the portion of flux motion losses which result from thermal activation. The surface impedance is then calculated from \( Z_s = j\omega \mu_0 \lambda(\omega, B, T) \). Because grain boundaries are not included in the model, only intragranular flux is considered. Thus, this model is only directly applicable if \( H > H_{c1} \) (\( \sim 10^2 \) Oe) and granularity is not an issue. Pambianchi, et al\textsuperscript{152} found that the Coffey-Clem parameterization offered an accurate description of their \( Z_s \) measurements of epitaxial films in high field (\( \sim 10^3 \) Oe).

D. The Stripline Model

In order to accomodate flux flow, granular and intrinsic losses in one equivalent circuit model, Portis and Cooke\textsuperscript{154} model the grain boundaries as superconducting striplines. Whereas, the effective medium model assumes a uniform wave vector throughout the material, the stripline model describes waves propagating down the grain boundaries. The grain boundary transmission line is composed of two superconducting walls which are Josephson coupled. This is to be contrasted to a conventional transmission line with normal conducting walls which are capacitively coupled. Flux flow results in higher wall impedance. Through the stripline model flux flow losses which are induced by high microwave power are
accomodated in the same model as grain boundary losses\textsuperscript{155,156}. If the stripline model is carried to the limit of zero flux flow losses, the results of the weakly coupled grain model are recovered. Application of this model is still in its infancy, and more will be heard on it in the future.
Chapter VII

Fit to the Weakly Coupled Grain Model

A. Algorithm for Mapping the Data onto the Normalized Model Curve.

Because absolute $X_s$ cannot be measured, the $R_s$ versus $\Delta X_s$ data cannot be plotted directly onto the theoretical curve of Figure VI-4. However, because the slope of the data is unique, a mapping scheme is employed as illustrated in Figure VII-1. The fortran code *fitter* was written to conduct the mapping. The program is in Appendix 2, and a flow chart of it is in Figure VII-2.

A trial value of $R_c$ is first divided into $R_s$. Next the value of $X_s(H_{dc}=0)$ which can be added to $\Delta X_s(H)$ to result in the point being placed on the model curve is determined. Within the program *fitter*, a lookup table containing $x$, $R_s/R_c$ and $X_s/R_c$ is referenced to find the necessary value of $X_s$. All points in the $[\Delta X_s(H), R_s(H)]$ data set must have the same value of $X_s(H_{dc}=0)$. The choice of $R_c$ which results in the lowest standard deviation of $X_s(H_{dc}=0)$ among the points in the data set is taken to be the correct value. A typical graph of the standard deviation of $X_s(H_{dc}=0)$ for sample YBCO#2 at 18 K and 17.5 GHz is shown in Figure VII-3. There is clearly little ambiguity in the choice of $R_c$. The standard deviation of $X_s(H_{dc}=0)$ will be quoted throughout this chapter as the uncertainty in $X_s(H_{dc}=0)$. It must be understood that this is not a measurement uncertainty in the surface reactance. Instead it is an uncertainty in the fit.
Figure VII-1 (a) The model curve with an arrow indicating the direction of increasing $x$, and (b) the mapping scheme employed to plot the $Z_s$ data on the model curve.
Figure VII-2 Mapping of the surface impedance data onto the model curve.
Figure VII-3 The standard deviation of the zero field offset in surface reactance for sample YBCO#2 at 18 K and 17.5 GHz. The std. dev. was calculated over all of the \([R_s, \Delta X_s]\) points' offsets, which ideally are all the same. The minimum point occurs at the best choice of \(R_c\).
From the mapping, \( R_C \) and \( X_s(H_{DC}=0) \) are readily determined. Likewise \( x_o=x(H_{DC}=0) \) is determined since any pair of \((R_s/R_C, X_s/R_C)\) points has a unique value of \( x \). Having determined \( x_o \) and \( R_C \) at one frequency, the frequency dependence of \( R_s(\omega) \) at constant temperature and static magnetic field can be predicted from Equation 126.

The mapping procedure is carried out for each \([\Delta X_s(H), R_s(H)]\) data set taken at constant temperature. The mapping should, and does, yield the same value of \( R_C \) at all temperatures. However, the value of \( X_s(H=0) \) is different at each temperature, reflecting the temperature dependence of \( \epsilon_f \).

The arrow in Figure VII-1 indicates the direction of increasing \( x \). This is implicitly the direction of increasing temperature, magnetic field, microwave power or frequency. The arrow also indicates the direction of increasing \( \lambda/\delta \). The reduction in surface reactance, or \( \lambda_{\text{eff}} \) with increasing field as seen in Figure V-12 is now accounted for in terms of the weakly coupled grain model.

**B. Temperature and Static Magnetic Field Dependence of \( Z_s \)**

The mapping algorithm described in Figure VII-2 is performed with surface impedance data taken at constant temperature. Results from granular samples of the three material families presented in Chapter II, TBCCO, BSCCO, and YBCO, will be presented here. Although grain size and grain orientation have not entered quantitatively into this analysis, attempts to fit the model to large grained, melt textured films will also be summarized.
1. TBCCO

In Figure VII-4a the change in surface resistance versus the change in surface reactance from the zero field values of sample TBCCO#1 at 12 K, 76 K and 92 K, with magnetic field as the implicit variable, is shown to be monotonically increasing and everywhere to be concave up ($d^2R_s/dX_s^2 > 1$). These data were taken from Figure V-11. Figure VII-4b shows the result of the mapping procedure of sample TBCCO#1 at these same temperatures. The solid line is the model curve. The lowest value for $x$ at each temperature is indicated on the curve. At 92 K and

<table>
<thead>
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<th>Temp (K)</th>
<th>$x_0$</th>
<th>$R_C$ (mΩ)</th>
<th>$\rho_s$ (Ω-m)</th>
<th>$\delta$ (μm)</th>
<th>$\lambda_{eq}$ (H=0)</th>
<th>$\xi$ (H-m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>12.0</td>
<td>0.125</td>
<td>161</td>
<td>3.8x10^{-7}</td>
<td>2.34</td>
<td>0.59μm</td>
<td>4.3x10^{-19}</td>
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<tr>
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<td>161</td>
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<td>0.60</td>
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<tr>
<td>35.0</td>
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<td>164</td>
<td>3.9x10^{-7}</td>
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<td>0.70</td>
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<td>0.185</td>
<td>170</td>
<td>4.2x10^{-7}</td>
<td>2.47</td>
<td>0.77</td>
<td>7.1x10^{-19}</td>
</tr>
<tr>
<td>54.0</td>
<td>0.190</td>
<td>165</td>
<td>3.9x10^{-7}</td>
<td>2.39</td>
<td>0.82</td>
<td>6.8x10^{-19}</td>
</tr>
<tr>
<td>64.0</td>
<td>0.255</td>
<td>170</td>
<td>4.2x10^{-7}</td>
<td>2.47</td>
<td>0.87</td>
<td>9.8x10^{-19}</td>
</tr>
<tr>
<td>71.4</td>
<td>0.320</td>
<td>167</td>
<td>4.0x10^{-7}</td>
<td>2.42</td>
<td>0.91</td>
<td>1.2x10^{-18}</td>
</tr>
<tr>
<td>76.0</td>
<td>0.340</td>
<td>176</td>
<td>4.5x10^{-7}</td>
<td>2.55</td>
<td>1.00</td>
<td>1.4x10^{-18}</td>
</tr>
<tr>
<td>82.0</td>
<td>0.400</td>
<td>167</td>
<td>4.0x10^{-7}</td>
<td>2.42</td>
<td>x.xx</td>
<td>1.5x10^{-18}</td>
</tr>
<tr>
<td>92.0</td>
<td>0.731</td>
<td>156</td>
<td>3.5x10^{-7}</td>
<td>2.26</td>
<td>1.20</td>
<td>2.3x10^{-18}</td>
</tr>
<tr>
<td>95.0</td>
<td>0.966</td>
<td>172</td>
<td>4.3x10^{-7}</td>
<td>2.50</td>
<td>1.23</td>
<td>3.8x10^{-18}</td>
</tr>
</tbody>
</table>
H > 10 Oe the surface reactance in Figure V-11 became field independent while the surface resistance still varied with field. In a plot of $dR_s/dX_s$ versus $H$ this would be a singularity. By comparing this observation with the model curve, it is seen that this infinite slope is predicted by the weakly coupled grain model. In Figure VII-4b the infinite slope fits the model curve quite well.

Table VII-1 summarizes the physical properties of sample TBCCO#1 which were determined from the model. The classical surface resistance indeed is temperature independent. It is important to realize that a strong temperature dependence could have resulted as well. But because the normal state resistance of HTS Josephson junctions is virtually temperature independent, the $R_C(T) =$ Constant result is reassuring. With $R_C$ and $x(H=0)$ known from the mapping, the zero field values of $\ell_j$ and $\rho_j$ can be calculated. The effective medium junction kinetic inductivity, $\ell_j$, is seen to be weakly temperature dependent at low temperature. As $T_C$ is approached $\ell_j$ increases dramatically. It must be remembered that by denoting the inductivity which is solved for from the value of $x_o$ with the symbol $\ell_o$ it is assumed that $\ell_j$ is completely shunting $\ell_o$. In high magnetic fields this assumption cannot be made.

The value of $\lambda_{\text{eff}}$ in Table VII-1 is determined from $\lambda_{\text{eff}} = \omega_0 \mu_j / X_s(H=0)$. Recalling the discussion surrounding Equation 130 if $x < 0.4$ then $\lambda_{\text{eff}} = \lambda_j^{\text{eff}}$. The effective medium $\lambda_j^{\text{eff}}$ is related to the actual Josephson penetration depth using Equation 120. Since
\[ \lambda_{J}^{\text{eff}} = \sqrt{\frac{2\lambda_{L}}{d}} \lambda_{J} \]  

(140)

then using \(d\approx10\ \mu\text{m}\) and \(\lambda_{L}\approx0.15\ \mu\text{m}\), we get for TBCCO\#1 at 12 K that \(\lambda_{J}\approx3.4\ \mu\text{m}\).

The small values of \(x\) as well as the large values of \(\lambda_{\text{eff}}\) indicate that the grain boundaries are dominating over the shunt and intrinsic impedances. From this it is justifiable to first order to ignore \(\lambda_{L}\) in the analysis.

For all of the measurements performed with TBCCO\#1 the magnetic field was oriented parallel to the sample. It was verified that the weakly coupled grain model is satisfied independently of field orientation by performing measurements on TBCCO\#2 with the field applied perpendicular to the sample surface. The model plot is shown in Figure VII-5. At 77 K, \(R_{c}=195\ \text{m}\Omega\), \(X_{S}(H=0)=152\pm2\ \text{m}\Omega\), and \(x(H=0)=0.340\). This result will be used later to predict the frequency dependence of the surface resistance of this sample.

In very strong magnetic fields (\(H>500\ \text{Oe}\)) the surface impedance of granular HTSC samples saturates as shown in Figure V-13. The \(\Delta Z_{S}(H)\) results at 12 K for sample TBCCO\#1 measured up to 1,200 Oe were mapped onto the model curve and are shown in Figure VII-6. Since \(\xi\) grows very large in strong fields, the value of \(\xi\) at saturation is \(\xi_{S}\). The result is that \(x_{\text{saturaion}}(12K)=0.69\) which gives \(\xi_{S}(12K)=2.4\times10^{18}\ \text{H}\cdot\text{m}\), which is a factor of 5 larger than the value of \(\xi_{J}\). The values in the \(\xi\) column of Table VII-1 are actually the parallel combination of \(\xi_{S}\) and \(\xi_{J}\). The low value of \(\xi_{S}\) indicates that these values for \(\xi_{J}\) are only approximate.
2. BSCCO

The field dependence of the surface impedance of BSCCO#1 was measured at 11K. The slope of $R_s$ versus $\Delta X_s$ is close to unity ($0.82 < \frac{dR_s}{dX_s} < 0.96$) and only weakly field dependent between zero and 50 Oe$^\text{107}$. If the slope is weakly field dependent then a very large value of $R_C$ is needed to accomplish a mapping onto the model curve. Indeed, a rather large $R_C$ of 291 m$\Omega$ was found. The field penetration was also quite large with $X_s(H=0)=256\pm2$ m$\Omega$. However a field independent slope of unity is also indicative of flux flow dissipation. Because the BSCCO compounds are characterized by high flux flow losses in low magnetic fields$^{157,158}$, it is possible that the losses in BSCCO#1 are dominated by fluxon motion.

The field dependence of the surface impedance of BSCCO#3 was measured at 50 K and 17.5 GHz. The results of the mapping onto the model curve are shown in Figure VII-7. Of all of the samples studied in this work, BSCCO#3 had the lowest zero field residual surface resistance. However, its surface impedance was very sensitive to the static magnetic field. The slope, $\frac{dR_s}{dX_s}$ varied from 0.19 at 0 Oe to 0.922 at 70 Oe. $R_C$ was a more modest 208 m$\Omega$ and $X_s$ was 100±4 m$\Omega$. The $R_s$ versus $\Delta X_s$ data mapped onto a large range of the model curve indicating that the grain boundary model adequately described sample BSCCO#3.
3. YBCO

Model analysis was performed with the surface impedance of the bulk YBCO samples. All three of the bulk YBCO samples fit the model remarkably well. The mapping to the model curve of $\Delta Z_s(H)$ for sample YBCO#3 at 11.3 GHz is shown in Figure VII-8. The peak in $X_s(H)$ that was seen in Chapter 4 maps onto the double valued $R_s/R_C$ versus $X_s/R_C$ curve. Although their measurements never reached the $R_s=X_s=R_C$ condition observed in Figure VII-8, Hein et al.\textsuperscript{159}, as well, saw the double valued nature of the surface impedance in YBCO thick films. At 86 K, as the magnetic field increases, the slope of $R_s$ versus $X_s$ approaches -1. At the point of $R_s=X_s$ the surface resistance becomes magnetic field independent. Because $x$ goes to infinity in high field for the bulk YBCO samples the kinetic inductivity is dominated by $\chi_f$. Finally, the large value of $\lambda_{\text{eff}}$ indicates that grain boundaries

<table>
<thead>
<tr>
<th>Temp(K)</th>
<th>$x(H=0)$</th>
<th>$R_C$(m$\Omega$)</th>
<th>$\rho_x$(Ωm)</th>
<th>$\delta$(μm)</th>
<th>$\lambda_{\text{eff}}(H=0)$ (μm)</th>
<th>$\xi$(H-m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>16.0</td>
<td>0.249</td>
<td>321</td>
<td>2.3x10$^{-6}$</td>
<td>7.2</td>
<td>2.3</td>
<td>8.08x10$^{-14}$</td>
</tr>
<tr>
<td>51.0</td>
<td>0.364</td>
<td>321</td>
<td>2.3x10$^{-6}$</td>
<td>7.2</td>
<td>2.9</td>
<td>1.18x10$^{-17}$</td>
</tr>
<tr>
<td>75.0</td>
<td>0.585</td>
<td>316</td>
<td>2.2x10$^{-6}$</td>
<td>7.0</td>
<td>3.5</td>
<td>1.90x10$^{-17}$</td>
</tr>
<tr>
<td>80.0</td>
<td>0.739</td>
<td>362</td>
<td>2.9x10$^{-6}$</td>
<td>9.0</td>
<td>4.5</td>
<td>3.02x10$^{-17}$</td>
</tr>
<tr>
<td>85.9</td>
<td>1.390</td>
<td>356</td>
<td>2.8x10$^{-6}$</td>
<td>8.0</td>
<td>4.7</td>
<td>5.49x10$^{-17}$</td>
</tr>
</tbody>
</table>
dominate the rf field penetration. The zero field values of $\rho$, and $\ell$, versus temperature are shown in table VII-2.

The degree to which the model parameter, $x=\omega \ell / \rho$, is linear in magnetic field is a measure of the junction contribution to the surface impedance. As argued in Chapter VI, if $\ell \ll \ell_0$, then the effect of junction shunting by superconducting microbridges is negligible, and from Figure VI-3, the $\ell$ in $x$ is consequently $\ell_0$. The linear field dependence of $\ell_0$ is then reflected in $x$. In samples where the granularity makes less of a contribution to the losses $\ell_i$ and $\ell_b$ are more similar in magnitude. In this case the field independent $\ell_b$ begins to influence, and eventually dominate, the effective kinetic inductivity at higher fields. The values of $x=\omega (H) \rho / \omega$ in elevated fields at 76 K for the samples YBCO#3 and TBCCO#2 are in Figure VII-9. Linearity of $x$ in field is observed for the bulk YBCO sample. The TBCCO film is linear in field only at low fields. As the field is increased $x$ begins to saturate. In no samples has $x$ been observed to have a stronger than linear field dependence.

- The saturation in kinetic inductivity is proposed as the mechanism behind the saturation in field of the surface impedance of granular superconductors. By decoupling the grains, as shown in Figure II-3, the magnetic field induces a rise in the effective kinetic inductivity of the transport current in the grain boundary. Saturation corresponds to superconducting transport occurring entirely via percolation across superconducting microbridges.
In brief summary, the bulk YBCO samples provide the best examples of granular superconductors available in this work. The contribution of the grains to the losses is completely masked by the junction array.

4. Universality of the Model

The applicability of the weakly coupled grain model to superconducting samples from the TBCCO, BSCCO and YBCO families of materials has been demonstrated. As a final display of this material universality, the model maps of samples TBCCO#1, BSCCO#3, and YBCO#2 at 17.5 GHz are presented together in Figure VII-10. Only the YBCO samples are driven into the extreme granularity limit at this frequency. The BSCCO film at 50 K is observed to go the farthest into the good superconductor limit.

5. Large Grained Samples

The field dependence of the surface impedance of the large grained sample TBCCO#3 is very weak as seen in Figure V-13. The surface resistance versus surface reactance of TBCCO#3 at 15 K is shown in Figure VII-11. The field was ramped up to 250 Oe. From Figure VI-4 it is clear that the weakly coupled grain model predicts that in the good superconductor limit, \( x << 1 \), and for grains large, the condition \( \frac{d^2R_s}{dX_s^2} > 1 \) must hold. That the opposite condition exists in Figure VII-11 indicates that granularity is not dominating the losses in this sample. This is not unexpected since the grains in this sample are as large as 0.5 mm.
C. Frequency Dependence of the Surface Resistance

In an experiment involving the Fabry-Perot resonator described in Chapter IV, the frequency, \( f \), dependence of \( R_s \) of sample TBCCO#2 at 77 K and zero magnetic field was measured between 17 GHz and 82 GHz. The field dependence of \( Z_s \) was measured at 77 K and 17.5 GHz. The mapping of this data onto the model curve was presented in Figure VII-5 and yielded \( R_c = 195 \) m\( \Omega \) and \( x(H=0) = 0.340 \). Using these values in Equation 126, the surface resistance at other frequencies is known. The complicated frequency dependence given by the weakly coupled grain model is tested by comparing the calculated \( R_s(f) \) to the measured \( R_s(f) \) in Figure VII-12. The measurements between 44 GHz and 82 GHz were made with the Fabry-Perot resonator.

Frequency exponents have also been calculated using equation 131. The curve of Figure VII-12 is shown again in Figure VII-13 along with the frequency dependent frequency exponent, \( n(f) \). At low frequency and 77 K the surface resistance is nearly quadratic with \( R_s \propto f^4 \). At 90 GHz the surface resistance depends linearly upon frequency. With this result, the frequency dependence of the surface impedance of granular superconductors is accounted for quantitatively by the weakly coupled grain model. For the bulk YBCO samples \( R_s \) depends upon the square root of frequency at 77 K above 60 GHz.

Although frequency dependent measurements could only be performed at 77 K, the frequency exponent could nonetheless be calculated at any temperature given the surface resistance at that temperature along with the sample's characteristic value.
of $R_C$. The values of the frequency exponent versus temperature at 17.5 GHz and 60 GHz for sample TBCCO#1 is shown in Figure VII-14. These two curves were generated using Equation 131 and the data of Table VII-1.

Finally it should be mentioned that Nguyen, Oates, et al.\textsuperscript{160} studied the power dependence of the surface impedance of epitaxial thin films. They found their measurements to be in accordance with the weakly coupled grain model for rf surface fields $<50$ Oe. Miller et al.\textsuperscript{161} found the weakly coupled grain model to describe losses in YBCO thin films deep into the submillimeter range ($10$ GHz $< f < 3\times10^4$ GHz). Furthermore, they demonstrated that the weakly coupled grain model is equivalently a two-fluid model. This is seen here by comparing Equations 112 and 116 and realizing that both describe resistively shunted kinetic inductivities.
Figure VII-4a: Change in the surface resistance from its zero field value versus change in surface reactance from its zero field value of sample TBCCO#1 at 12 K, 76 K, 92 K, 95 K, and 17.5 GHz.
Figure VII-4b Result of mapping the surface impedance of sample TBCCO#1 onto the model curve at 12 K, 76 K, and 95 K.
Figure VII-5  Results of mapping the surface impedance of sample TBCCO#2 onto the model curve at 15 K and 70.1 K. In this case the orientation of the field is normal to the film.
Figure VII-6 Results of mapping the high field surface impedance of sample TBCCO#1 onto the model curve at 12 K. The field was ramped up to 0.12 T.
Figure VII-7 Results of mapping the surface impedance of sample BSCCO\#3 onto the model curve at 50 K. $T_c = 107$ K.
Figure VII-8 Results of mapping the surface impedance of sample YBCO#3 onto the model curve at 16 K, 75 K, 86 K.
Figure VII-9  Magnetic field dependence of the kinetic inductivity, $\frac{\ell}{\omega} = x p / \omega$, for samples TBCCO#1 and YBCO#3 at 76 K. The TBCCO sample saturates in field due to the presence of microshorts.
Figure VII-10 Samples TBCCO#1, BSCCO#3 and YBCO#2 all at 17.5 GHz have been mapped onto the model curve, and are shown together in order to emphasize the universal applicability of the weakly coupled grain model to the HTSC ceramics.
Figure VII-11 The surface resistance versus the change in surface reactance for the large grained sample TBCCO#3 at 15 K. The field, which was ramped up to 250 Oe, is the implicit parameter. The concave down nature of the curve indicates that the weakly coupled grain model does not describe this sample's surface impedance.
Figure VII-12  The surface resistance versus frequency of sample TBCCO#2 was measured and compared to the prediction of the weakly coupled grain model. The dashed line is the quadratic extrapolation of the two fluid model.
Figure VII-13  The model predicted frequency dependence of $R_s$ of sample TBCCO#2 at 77 K is shown again along with the value of the frequency exponent.
Figure VII-14 Temperature dependence of the frequency dependent frequency exponent of the surface resistance of sample TBCCO#1. $T_c = 101$ K.
Chapter VIII
Applications of Bulk and Thick Film Superconductors

This dissertation describes the microwave losses caused by granularity. If the HTSC materials are to be technologically applicable it will be necessary to minimize the losses. A plethora of rf applications has arisen in the past five years. Summarized here are: antennas, stripline resonators, cellular communication technology, the hydrogen maser, and accelerator cavities. Because the author devoted considerable time toward static magnetic shielding, it too will be reviewed.

Antennas are used either as receivers or transmitters, and the analysis of an antenna's efficiency does not depend upon which application is intended. The radiation resistance, \( R_{\text{rad}} = P_{\text{rad}}/I_{\text{rms}}^2 \), of an antenna is an effective resistance which dissipates the same amount of power as is radiated. \( P_{\text{rad}} \) is the dipole power radiated by the antenna dipole. In general, a large \( R_{\text{rad}} \) corresponds to an efficient radiator. The Ohmic resistance, \( R_r \), corresponds directly to the surface resistance of the antenna materials. The efficiency is then defined as

\[
\epsilon = \frac{R_{\text{rad}}}{R_{\text{rad}} + R_r}
\]  

and is unity if \( R_r \ll R_{\text{rad}} \).

Khamas, et al. describe a short electric dipole antenna made out of a YBCO wire formed on a teflon substrate by a polymer composite process.
Briefly, YBaCu precursor is sintered and ground to a 0.3 μm average particle size. It is then mixed with a proprietary (ICI, Runcorn) nonaqueous polymer which results in a plastic mixture. The mixture is then formed into wires by a ram extruder. Upon further sintering the polymer is removed and pure YBCO remains.

An electrically short antenna suffers from the affliction of low $R_{\text{rad}}$. For small magnetic dipole antennas the radiation resistance depends upon the $4^{\text{th}}$ power of the antenna size. The $R_{\text{rad}}$ of a small electric dipole antenna depends upon the square of the antenna size. It is necessary then to minimize $R$, if antennas are to be miniaturized. This supplies the motivation to use superconducting materials. In a later paper, Wu, et al.\textsuperscript{167} reported that a tunable YBCO small magnetic dipole antenna yielded 5 dB more radiated field strength at 77 K than an identical copper antenna at 77 K.

Stripline resonators are used in microwave circuits as bandpass filters, frequency stabilizers and other applications. Because the $Q$ of a resonator is inversely proportional to the surface resistance of the conductor material, narrower bands and better frequency stabilization is acquired.

![Figure VIII-1 Stripline resonator in the meander line form.](image-url)
with superconducting materials. In addition, device miniaturization is facilitated by $R_s$ reduction. If the meander line geometry illustrated in Figure VIII-1 is used then the necessary line separation is reduced if the conductor separation can be reduced. By reducing the conductor separation, the resonator volume, and hence the $Q$, is reduced. But, if the conductor has very low loss, then a greater degree of miniaturization can be tolerated\textsuperscript{168}.

Mossavati, et al.\textsuperscript{169} made a stripline resonator from a YBCO thick film (23 mm long and 1.3 mm wide) deposited onto a 0.9 mm thick zirconia substrate. The backside of the substrate was completely coated with YBCO and served as the ground plane. The films were deposited in the same manner (and in the same laboratory at ICI) as sample YBCO#4. The resonator had a $Q$ of 1000 at 12 GHz and 20 K and a $Q$ of ~800 at 77 K. The $Q$'s were low but were attributed to dielectric losses. Higher $Q$, miniaturized resonators are achieved by epitaxially depositing the HTSC film onto low loss substrates\textsuperscript{170}.

Thick film microstrip resonators have certain advantages over thin film devices. For low frequency applications, <1 GHz, large area films are needed. In the future, large area flat epitaxial films may be expected, but presently high quality films are limited to two to three inch diameter. Thick films, on the other hand, can be deposited on any size or shape surface. Also at issue is power handling ability. A device made from a 300 nm thin film of YBCO loses its ability to support a resonance at lower power levels than a thick melt processed film.
A potential application of HTSC resonators is found in cellular communication. Illinois Superconductor Corp. (ISC) recently received an Advanced Technology Program project award to construct a receiver for a cellular telephone base station using HTSC resonators\textsuperscript{171}. The higher Q resonators will increase the total number of channels available in the overcrowded cellular band, improve reception and stabilize the frequencies. The devices will be made from thick YBCO films on stainless steel substrates with silver buffer layers. They will be operated between 200 MHz and 2 GHz.

The plausibility of making low $R_s$ YBCO films on stainless steel substrates was demonstrated in a collaboration between ISC and the author. The surface resistance of these films at 1 GHz and 77 K was more than an order of magnitude lower than copper at 1 GHz and 77 K. The $R_s$ measurements were performed at 17.5 GHz. Using the result of the previous chapter that the $R_s$ is nearly quadratic below this frequency at 77 K, and the fact

![Surface resistance at 1 GHz of a YBCO film deposited onto a stainless steel substrate. $T_c=92$ K.](image)

Figure VIII-2: Surface resistance at 1 GHz of a YBCO film deposited onto a stainless steel substrate. $T_c=92$ K.
that the films were well oriented with large grains, the $R_\theta$ was scaled quadratically to 1 GHz and is shown along with copper in Figure VIII-2.

Advanced devices for frequency control stand to benefit greatly from HTSC materials. An active hydrogen maser uses the 1.420405751769 GHz hyperfine transition frequency of hydrogen as a frequency standard. A high Q resonator tuned to this frequency is used to couple power into the maser. The difficulty is that a 1.42 GHz cylindrical cavity resonator is too enormous to place onto satellites. Miniaturization was accomplished in a compact resonator design which incorporates a loop-gap structure illustrated in Figure VIII-3. The resonator structure is an open cylinder which has been split in half along the longitudinal axis. The loop-gap mode is an LC oscillation where the loop inductance is the L and the gap capacitance is the C.

A copper loop-gap resonator was found to have a Q of 11,500 at 77 K. A Q of at least 14,000 is needed to support maser oscillations. An identical resonator made out of silver electrodes electrophoretically coated with YBCO had a Q of 31,000 at 77K. With such a large Q it is necessary to be able to fine tune the resonator. In a collaboration between the author and Physical Sciences, Inc. in Alexandria Va., theoretical calculations were performed using MAFIA code to determine the extent of tuning achieved by rotating a 1 mm thick sapphire slide into the loop-gap. It was found that a tuning sensitivity smaller than 100 KHz could be achieved by rotating the slide 1°. With finer rotation control and a thinner slide 1
KHz tuning is possible. Work is presently underway to produce a compact hydrogen maser with superconducting electrodes for eventual use on board the Global Positioning System satellites.

Finally mention should be made of work leading toward the goal of HTSC accelerator cavities. Superconducting niobium cavities are used in particle beam accelerators to reduce the necessary level of accelerating power. With the same application in mind for TBCCO, Arendt, et al.\textsuperscript{174} (Los Alamos) constructed a

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure_viii_3}
\caption{Figure VIII-3 MAFIA calculation of the electric field in the loop-gap mode with a rotatable sapphire tuner.}
\end{figure}
clamshell shaped cavity out of Consil 995 and magnetron sputter deposited a 6 µm thick TBCCO film onto the interior. The choice of a clamshell shape was based on the need to study the problems associated with HTSC sputter deposition onto curved surfaces. The cavity had an unloaded Q of 10^5 at 20 K and 6.6x10^4 at 77 K and 10 GHz. This was comparable to an identical copper resonator. Nb cavities at this frequency and 2 K can exhibit Q's larger than 10^8.

From this preliminary result the Los Alamos group was encouraged to pursue further studies yet unpublished. Attempts will also be made by this group to deposit a YBCO film onto a silver clamshell cavity by electrophoresis. Use of HTSC in particle accelerators is presently a long way off. Issues such as field emission and long term material stability have yet to be addressed.

**Experiments in Static Magnetic Shielding**

The hydrogen maser relies upon a very accurate control of the DC magnetic field which induces the hyperfine transition. For this reason the maser needs to be shielded from external magnetic fields. Squid applications and biomagnetic measurements also require a magnetically clean environment. Conventional mu-metal magnetic shielding is bulky and, for the maser, at least four layers of shielding material are needed. The need for the multiple layers lies in the low residual field which penetrates the mu-metal. A superconducting thick film serves
as a perfect shield to low fields. The scheme to be employed for low magnetic field shielding is to have a closed HTSC thick film cylinder enclosed within a closed mu-metal cylinder.

Magnetic shielding experiments were performed in a collaboration involving the author, Physical Sciences Inc. and ICI Advanced Materials, Runcorn, England. Granular HTSC thick films will shield magnetic fields below a critical field referred to in these experiments as the penetration field, $H_p$. It is the field at which fluxons begin to penetrate the intergranular medium and is related to $H_{c1}$. A fluxon must penetrate first at the edge of the film and move in small steps though the intergranular medium. Until a significant number of grain boundaries have broken down to flux penetration a fluxon cannot migrate into the film. Thus, it is expected that $H_p$ is larger than the smallest values of $H_{c1}$ in the sample.

In these experiments a 3% yttria stabilized zirconia(YSZ) cylinder and two
flat YSZ end pieces were coated with YBCO by the screenprinting technique and then melt processed. Screen printing is described by Topfer. In brief, the YBCO is ground to a fine powder and mixed into an organic solvent to form an ink. The ink is applied to the substrate by a high pressure squeegee. The substrate is then heated to ~1050°C for two minutes to induce a partial melt of the YBCO. On cooling, large 0.5 mm to 1 mm grains are formed.

The cylinder, shown in Figure VIII-4, was 5 cm in diameter and 5 cm long. It was placed on the cold head of the closed cycle refrigerator. The cold head was then placed inside a large solenoid which was oriented to provide a magnetic field along the symmetry axis of the HTSC cylinder. The solenoid in turn was enclosed by four layers of mu-metal.

A cryogenic Hall probe with 10 mGauss sensitivity was placed in various locations inside the HTSC cylinder. The solenoid field was ramped from zero to 20 Gauss, from 20 Gauss to -20 Gauss, and from -20 Gauss to 0 Gauss. After arriving at 0 Gauss the field was ramped to whatever

![Figure VIII-5](image_url)  
**Figure VIII-5** Hysteresis loops for the internal field inside the all HTS cavity with the hall probe positioned in the center of the cavity.
value brought the internal field back to 0 Gauss. Thus complete hysteresis loops shown in Figure VIII-5 were generated. The hysteresis is due to flux trapping in the grain boundaries in accordance to the Bean critical state model\textsuperscript{178}.

The conclusion of this short report on magnetic shielding is that static magnetic fields below 2 Gauss are shielded by HTSC thin films at 77 K. Current work involves depositing YBCO films onto two large, six inch disks of silver and sandwiching them together with the film on the inside\textsuperscript{179}. This makes a metal-superconductor-metal structure which protects the environmentally sensitive YBCO. The sandwich is then drawn into a two inch diameter, four inch deep cup in the same manner as a soda can is drawn from sheet metal\textsuperscript{180}. 
Chapter IX
Conclusion

This dissertation described an experiment which was designed to probe the magnetic field dependence of the surface impedance of granular high temperature superconductors. Sections were found within concerning the Fabry-Perot resonator and static magnetic shielding.

Chapter IV reviewed the technique of surface impedance measurement with cavity resonators. Emphasis was placed upon the Fabry-Perot resonator. It was demonstrated that the geometry factor calculated from scalar theory is 5% to 10% lower than that calculated numerically from the more accurate vector theory. Calculations were also performed to verify that negligible mode mixing losses could be achieved while maintaining reasonable coupling strength. Many details of cavity techniques were intentionally included in Chapter IV so that it could remain as an operator's manual for the cavity resonator systems developed in this work.

In Chapter V both the surface resistance and the surface reactance were seen to be strongly dependent upon static magnetic fields greater than 1 Oe. Furthermore, it was seen that the surface reactance had a double valued dependence upon the field. These facts were argued in Chapter VI to be evidence of grain boundary dominated losses. In order to physically model the grain boundary losses, the weakly coupled grain model was introduced in Chapter VI. In using this
model it was argued that the grain boundaries are resistively shunted Josephson junctions. From the field dependent kinetic inductivity of carriers crossing a Josephson junction in the voltage state, the surface impedance was shown within the model to be magnetic field dependent.

In Chapter VII the measured surface resistance was compared to the model by performing a two step mapping procedure of the data onto the normalized model curve. From this mapping procedure the zero field model surface reactance was determined along with the single model parameter \( x = \frac{2\lambda^2}{\delta^2} \), where \( \lambda \) is the superconducting penetration depth due to the intrinsic granular penetration and the Josephson penetration, and \( \delta \) is the skin depth. From the model parameter, \( x \), the frequency dependence of the surface resistance was predicted and shown to be in good agreement with Fabry-Perot measurements.

Two conclusions were drawn from this experiment. First, the surface impedance of granular HTSC was shown to be dominated by the weak Josephson coupling between the grains. With this mechanism dominating the losses the surface impedance depends upon magnetic field, temperature and frequency in a predictable manner. Second, the surface impedance was shown to saturate at high field because the grain boundaries are shunted by superconducting microshorts which are magnetic field independent. In low magnetic fields the grain boundary impedance was comparable to the microshort impedance. But as the field was increased the grain boundary kinetic inductivity increased linearly. Eventually the
kinetic inductivity of the microshorts shunted the grain boundaries entirely.

Future experimentation should involve large grained thin films. These experiments tested the grain boundary model on samples which were selected specifically for their granularity. With less granular samples the grain boundaries will not necessarily dominate the losses. In such a case the intrinsic granular kinetic inductance will need to be included along with flux flow. Surface impedance studies of thin films in high static magnetic fields will provide the necessary data to test the stripline model discussed at the end of Chapter VI.
Appendix 1

Relationship Between Reflection Coefficient and Coupling Q

Transmission line theory is used to relate the coupling Q to the reflection coefficient of the resonator. The resonator is coupled by a transmission line. This could be either a coaxial cable or a wave guide. The resonator/transmission line network is shown in Figure App1-1. The transmission line is connected to a microwave source with output resistance, $R_s$. The transmission line itself has a characteristic resistance, $R_c=(l/c)\sqrt{\frac{\omega}{L}}$, where $l$ is the per unit length inductance and $c$ is the per unit length capacitance of the transmission line. $R_c$, which is real, is used to represent the transmission line in the equivalent circuit of Figure App1-2.

The resonator has an impedance, $Z_L=R+j\omega L+1/(j\omega C)$, which is modelled in Figure App1-2 as an LCR oscillator. The reflection coefficient at the resonator is
\[ \bar{\Gamma} = \frac{Z_L - R_C}{Z_L + R_C}. \] (142)

At resonance, \( \omega = (LC)^{\frac{1}{2}} \), the reflection coefficient is real, and

\[ \Gamma_{\text{res}} = \frac{R - R_C}{R + R_C} = 1 - \frac{R_C}{R} = \frac{1 - \beta}{1 + \beta}, \] (143)

where \( \beta \) is the ratio of the transmission line characteristic resistance to the resonator effective resistance.

The \( Q \) of an isolated LCR oscillator is \( Q_0 = \frac{L \omega}{R} \). In reality the oscillator is connected to external circuitry and the actual \( Q \) is descriptive of the entire network shown in Figure App1-2.

The measured, or loaded, \( Q \) is

\[ \frac{1}{Q_L} = \frac{1}{Q_o} + \frac{1}{Q_c} = \frac{R}{\omega L} + \frac{R_C}{\omega L} \] (144)

where it is assumed that \( R_C \) is matched to \( R_s \) (and thus invisible). The term...
external $Q$ is often used instead of coupling $Q$ to emphasize that it refers to everything external to the resonator. (If $R_C$ is not much larger than $R_s$ then the analysis is simply altered by replacing $R_C$ everywhere with $R_C + R_s$.) The loaded $Q$ is now written as

$$Q_L = \frac{Q_o}{\frac{R_C}{1+\frac{R}{R}}}$$  \hspace{1cm} (145)$$

where $\beta$ is determined from the measured reflection coefficient. If there are two couplers then further analysis leads to

$$Q_L = \frac{Q_o}{1+\beta_1+\beta_2}$$  \hspace{1cm} (146)$$

where $\beta_1$ and $\beta_2$ refer to the first and second coupler respectively.
Appendix 2

Fortran Code FITTER to map the surface impedance data onto the weakly coupled grain model curve

c......Program "FITTER" to fit Rs vs. Xs data to the Weakly Coupled Grain Model

Dimension R(500), X(500), Rc(150), Rs(150), dXs(150), Xso(150)
Dimension y1(150), Hfield(150)

Real newsum,q,w,mu
Logical repeat,ww,minim

The user inputs the frequency and the length of the data file. The data file wcgmfit contains three columns. Column 1 is the magnetic field, Hfield. Column 2 is the surface resistance, Rs. Column 3 is the change in surface reactance from zero field, dXs.

Print*,'Enter the frequency'
Read*,freq
pi=3.141592654
mu=12.56637e-7
Print*, 'how many pairs?'
Read*,n
Open(unit=10, file='wcgmfit.in')
DO 10, j = 1,n
read(10,*) Hfield(j), Rs(j), dXs(j)
dXs(j)= 2*pi*freq*dXs(j)*mu*10
print*, 'dXs= ',dXs(j)
10 Continue
Close(10)

c......The user enters a guess for Rc. Experience indicates this guess should be about 180 mohms. The zero field surface reactance, Xso, needed to plot the (dXs,Rs) point onto the model (Xs/Rc, Rs/Rc) curve is then determined. Ideally Xso should be the same for each pair. The standard deviation in Xso is calculated over all of the pairs. Then Rc is
In the entire process, the user inputted $R_c$ minus 75 mohms up to the user inputted $R_c$ plus 75 mohms.

```fortran
Print *, 'Enter a guess for $R_c$ in mohms'
Read*, $R_c(1)$
j = 1

Print*, 'Please enter the max acceptable std. dev. for Xsoffset'
Print*, '(suggest something < 2.0)'
Read*, std

ww = .TRUE.
repeat = .FALSE.

Create array $R(k) = R_s/R_c$
20 Do 30, k = 1, n
   $R(k) = R_s(k)/R_c(j)$
30 continue

Determine $X_s/R_c$ for each $R_s/R_c$. The file lookup.dta contains four columns. Column 1 is the model parameter, $\omega_1/\rho_1$. The second column is the derivative $dR_s/dX_s$ for the given $y$. The third column is $R_s/R_c$ and the fourth is $X_s/R_c$ calculated from the weakly coupled grain model.

i = 1
Open(unit=9, file='c:lookup.dta', status='old')
Read(9, *) $y(1), dRdX(1), RsRc(1), XsRc(1)$

Do 40 k = 1, n
   i = i + 1
   IF(i .GT. 2000) THEN
      Print*, 'Out of range $R_c = ', R_c(j)
      goto 62
   END IF
```

177
Read(9,*) y(i),dRdX(i),RsRc(i),XsRc(i)

c......Look for end of file marker.
   IF (y(i).GT.87.5) THEN
      REWIND(9)
   END IF

   IF ((R(k) .LT. RsRc(i)) .AND. (R(k) .GT. RsRc(i-1))) THEN
      goto 45
   ELSE
      goto 35
   END IF

c......Interpolate Xs/Rc linearly
   45   f=(R(k)-RsRc(i-1))/(RsRc(i)-RsRc(i-1))
        X(k)=f*(XsRc(i)-XsRc(i-1))+XsRc(i-1)
        y1(k)=y(i-1)+f*(y(i)-y(i-1))
        i=1
   40   Continue
   43   close(9)

c......Calculate Xsoffset(k)=X(k)*Rc(j)-dXs(k)
   sum=0
   Do 50 k=1,n
      Xso(k)=X(k)*Rc(j)-dXs(k)
      sum=sum+Xso(k)
   50   Continue

c......Calculate standard deviation in Xso, dXso
   q=n
   avg=sum/q

c......Note: newsum is real.
   newsum=0
   Do 60 k=1,n
      newsum=(Xso(k)-avg)*(Xso(k)-avg)+newsum
   60   Continue

   w=newsum/q
   dXso(j)=SQRT(w)
   print*, 'dXso= ', dXso(j)

   IF (ww .eqv. repeat) goto 70
The user entered a maximum acceptable standard deviation, std. If a standard deviation less than this minimum is achieved then the computation ends. Normally the user should enter std=0. In this case after 150 turns of the loop the value of Rc which resulted in the smallest dXso will be determined through a bubble sort.

```
IF (dXso(j) .LT. std) goto 70
62 IF (j .EQ. 150) GOTO 65
j=j+1
zz=j
IF (2*j/2) .EQ. j) THEN
   Rc(j)=Rc(1)-(zz/2)
ELSE
   Rc(j)=Rc(1)+(zz/2)-1
END IF
Print*, 'Begin trial number ', j
Print*, 'Now change Rc to: Rc= ', Rc(j)
goto 20
```

Find minimum Xs,offset

```
65 Do 68 k=1,150
   minim=.TRUE.
   Do 66 11=1,150
      IF (11 .eq. k) GOTO 66
      IF (dXso(k) .GT. dXso(11)) THEN
         minim=.FALSE.
      END IF
   66 Continue
IF (ww .eqv. minim) GOTO 69
68 Continue
69 j=k
Print*, 'min,j,dXso is = ', j,dXso(j)
repeat=.TRUE.
goto 20
```

If the std. dev. in Xs0 is less than the user defined minimum then the program terminates with the following code. Otherwise the minimum Xs0 must be found and re-evaluated before this code can run.
70 PRINT*, 'Xso = ', avg
PRINT*, 'Rc = ', Rc(j)
PRINT*, 'Spread in Xso = ', dXso(j)
PRINT*, 'units in mohms'

C.......Calculate the actual Xs/Rc = (Xs, offset + delta Xs)/Rc.
       DO 75 k = 1, n
         X(k) = (avg + dXs(k))/Rc(j)
75  CONTINUE

C.......Dump to data file.

80 FORMAT (1F10.5, 2X, 1F10.5, 2X, 1F10.5)
OPEN (9, FILE = 'C:RESULT.PRN', STATUS = 'NEW')
WRITE (9, *) 'Rc = ', Rc(j), ' Xso = ', avg, ' dXso = ', dXso(j)
WRITE (9, *) 'Xs/Rc Rs/Rc'
WRITE (9, 80) (yl(k), X(k), R(k), k = 1, n)
CLOSE (9)

90 FORMAT (2X, 1F10.5, 2X, 1F10.5)
OPEN (9, FILE = 'C:DXSO.PRN', STATUS = 'NEW')
WRITE (9, 90) (dXso(k), Rc(k), k = 1, 150)
END
References for Chapters I-III


33. The discussion of Cu-O planes is drawn from G. Burns, reference 4.


References for Chapter IV


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67. Guzin Armagon, College of William and Mary, private communication.


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References for Chapter V


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118. Suzanna Orbach, private communication.


Reference for Chapter VI


135. See, for example, discussion in Reference 9, p.152.


References for Chapter VII


References for Chapter VIII


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Vita

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