ELECTRODYNAMICS OF SUPERCONDUCTING $Y_{1}Ba_{2}Cu_{3}O_{7}$

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The response of superconductors to time-varying electromagnetic fields can yield detailed information on the nature of the superconducting state. Experiments carried out at frequencies in the rf to mm-wave spectral region directly probe the collective response of the condensed electrons and of the quasiparticles, and in conventional superconductors, have been important in determining the gap, the density of states and confirming the nature of the pairing.

The basic parameter that is measured at these frequencies is the complex surface impedance $Z_{S} = R_{S} + iX_{S}$. The surface resistance $R_{S}$ is a measure of the power absorbed, while the surface reactance, which is proportional to the penetration depth $\lambda$, is a measure of the reactive power exchanged between the field and the superconductors. These quantities can be computed, from the fundamental constitutive relation which relates current and vector potential for the superconductor, $\mathbf{J} = -\mathbf{Q} \cdot \mathbf{A}$, along with Maxwell's equation and suitable boundary conditions. Experiments can then be compared with detailed predictions for the kernel $Q$ from microscopic theories.

Significant conclusions concerning the intrinsic electrodynamic response require reliable samples. Recent advances in materials processing have yielded high quality samples and experiments indicate that a limiting intrinsic behaviour is observed in these samples. In this paper we summarize measurements of $R_{S}$ and $\lambda$ carried out at rf and microwave frequencies on $Y_{1}Ba_{2}Cu_{3}O_{7}$ crystals. The results are compared with calculations based on the BCS theory, for which detailed comparison is possible at present. Results for the lower critical field $H_{c1}$, along with temperature dependences of $\lambda(T)$ and $R_{S}(T)$ and the field dependence of $\lambda$, are also discussed. A key feature of the
high $T_c$ superconductors is the importance of granularity, and attempts are described to compare the results on polycrystalline samples to models of the granular response in terms of Josephson junction networks.

In addition to the fundamental interest in the high frequency response, a prime motivation is the potential for device applications. Results on two devices that have been fabricated, a microwave superconducting cavity and a filter are discussed briefly.

Measurements on the high $T_c$ superconductors pose new challenges, ironically because of the high temperatures involved. For instance sensitive measurements of $R_s$ require microwave structures whose background absorption is much less than that of the sample. This is difficult to achieve with normal metals like Copper. At 10 GHz, a method was developed\(^3\) which utilizes a high $Q$ ($\sim 10^7$) superconducting cavity made of Pb- plated Cu or Nb and maintained at 4.2K. The sample mounted on a sapphire rod, was placed at the center of the cavity at a maximum of the microwave magnetic field. The sample temperature could be varied from 4.2K to 200K independent of the cavity temperature, thus taking advantage of the low background absorption in Pb or Nb. $R_s$ is determined from the cavity $Q$ using the relation $R_s = \Gamma [Q^{-1}(T) - Q_{0}^{-1}(T)]$, and changes in the penetration depth are determined from the resonant frequency using $\delta \lambda = - \zeta \delta f$, where $Q_0$ is the background $Q$, and $\Gamma$ and $\zeta$ are geometric factors. This method has one of the highest sensitivities currently available for small samples. The technique also circumvents the background thermal contraction (due to the high temperatures) which can severely limit measurements of $\lambda$. In addition to the superconducting cavity, Cu cavities at 10, 17 and 35 GHz are also available, in which the sample forms the bottom plate of the cavity. These latter methods are more appropriate for larger samples ($\geq 1$ cm. dia) and have the advantage of rapid turnaround, although at the expense of sensitivity.

The radio frequency measurements are carried out with an ultra stable oscillator utilizing a tunnel diode. The sample is placed in a small coil (typically 2 mm. dia, 20 turns) which forms the inductive part of the resonant circuit. In order to achieve high stabilities (1 Hz in $10^7$ Hz), the tunnel diode was maintained at 77K, close (10 inches away) to the sample whose temperature could be varied from 4.2K to well above 100K. Changes in the resonant frequency are directly related to changes in the penetration depth via the relation $\delta \lambda = - G \delta f/f$, where $G$ is a geometric factor. The high stability enables measurements of changes of 20Å in $\lambda$. The temperature dependence of the resonant frequency yields the temperature dependence of $\lambda$.

An additional experiment using the rf coil, which has proved to be very informative, is to study the variation of $\lambda$ with an applied dc magnetic field at fixed temperature. This experiment yields a sharp signature of $H_{c1}$ and has enabled the determination of the complete temperature dependence of $H_{c1}$ in $Y_1Ba_2Cu_3O_y$. The
experiment is particularly sensitive to low field effects, both below and above $H_{c1}$, a region not easily accessible by other methods, but of great interest in the high $T_c$ superconductors, because of the possibility of novel flux states\textsuperscript{4}.

Recent results have shown that spectacularly sharp microwave reponse can be obtained in well prepared single crystals\textsuperscript{5} and thin films\textsuperscript{6,7}, in contrast to earlier data\textsuperscript{8} on polycrystals. These results have raised the hopes of microwave devices with performance far surpassing those made from conventional materials such as Copper. It is apparent however that sample preparation is crucial to obtaining high quality microwave response. The preparation of the single crystals is discussed in ref.5. Below we summarize several electrodynamic properties of these crystals.

**PENETRATION DEPTH**

The temperature dependence of the penetration depth in the best crystals which possess sharp transitions, are in excellent agreement with calculations based on the BCS theory, down to temperatures of about 0.7 $T_c$. (Higher sensitivities are required for lower temperatures). The actual quantity that is measured is the change in penetration depth referred to the value at a low temperature $T_0$ << $T_c$ i.e. $\delta \lambda(T) \equiv \delta \lambda(T) - \delta \lambda(T_0)$. The data are then compared to the calculations, using the zero temperature penetration depth $\lambda(0)$ as a fitting parameter. This is typical of all electrodynamic measurements, in which $\lambda(0)$ cannot be measured absolutely, unless sample dimensions are known to accuracies comparable to $\lambda(0)$ - however the same restriction does not apply to changes $\delta \lambda(T)$. Near $T_c$ ($T > 0.9 T_c$) the data can be equally well described by a simple two-fluid model or by the BCS theory. As the temperature range is extended to lower temperatures, the characteristic signature of the gap becomes apparent in the BCS model and the two theories begin to deviate. We have compared the data introducing the gap ratio $\Delta/kT_c$ as an additional parameter (see Fig. 1). Best fits to the data are found for $\lambda(0) = 1400\,\text{Å}$ and $\Delta/kT_c = 2.15$, with neither the two fluid model nor a weak coupling value of 1.76 providing good fits\textsuperscript{2}.

The penetration depth measurements are themselves very sensitive tests of sample quality. Using fits to the two-fluid model to extract $\lambda(0)$, we find that $\lambda(0)$ varies from 7.5\,\mu m in polycrystals to the lowest\textsuperscript{9} value 1400 Å in best crystals, with intermediate values of 0.5 to 1\,\mu m in "nominal" crystals.

**FIELD DEPENDENCE OF PENETRATION DEPTH $\lambda(H,T)$**

London first pointed out on thermodynamic grounds that the penetration depth should vary with magnetic field in the Meissner state. Subsequent experiments by Pippard\textsuperscript{10} on the elemental superconductors played an important historical role, since they were the basis of Pippard's concept of a coherence length. Although these variations
are small, we have observed them in the rf coil experiment by applying a dc magnetic field, and the results have yielded important information on the relation of the electrodynamic response to granularity.

For $H < H_{c1}$, we find experimentally that the penetration depth increases quadratically with $H$, i.e. $\delta\lambda(T,H) = k(T) H^2$. This is exactly what one might expect on simple thermodynamic grounds - the magnetic field depresses the order parameter, making all thermodynamic parameters, such as $\lambda$, dependent on the magnetic field. To first order the variation should be quadratic (for a $T$-invariant superconducting state). The coefficient $k(T)$ was determined experimentally for several $Y_1Ba_2Cu_3O_y$ crystalline and polycrystalline samples.

In the best crystals, $k(4.2K) = 10^{-3} \text{Å}/G^2$ and increases strongly with temperature by a factor of $10^5$ at $T_c$. The magnitudes and temperature dependence are in good agreement with a Ginzburg-Landau model for the order parameter depression which yields $k(T) = \frac{3}{4} \lambda(T)/H_{c0}^2(T)$. Using BCS temperature dependences for $\lambda$ and the thermodynamic critical field $H_{c0}$, we find excellent agreement with data over the entire temperature variation of 5 orders of magnitude.

What is remarkable is the extreme sensitivity of the absolute values of $k$ to sample quality and microstructure. For instance, $k(4.2K)$ varies from $10^{-3} \text{Å}/G^2$ in the best crystals to $160 \text{Å}/G^2$ in polycrystalline samples, with intermediate values of $10^{-2}$ to $10^{-1} \text{Å}/G^2$ in nominal crystals. The very large values in the polycrystals are in clear disagreement with the Ginzburg-Landau calculation for the pure bulk superconductor. However, we have shown earlier\textsuperscript{11} that these large values can be understood on the basis of a Josephson junction picture. For a single junction, the Josephson penetration depth $\lambda_j$

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Fig. 1. Temperature dependence of changes in the penetration depth $\delta\lambda(T)$ at 6 MHz with $H_{rf} \perp c$-axis. The solid line represents a BCS calculation.
will vary quadratically with external magnetic field as \( \delta \lambda_j(H) = k_j(T) H^2 \), with \( k_j = \pi^2 / 24 \lambda_j \Sigma^2 / \phi_0^2 \), where \( \Sigma \) is the effective junction area, and \( \phi_0 \) the flux quantum. This relation yields \( \Sigma \sim 4 \mu m^2 \) in the polycrystals. As the samples improve, the sharp decrease in \( k(0) \) reflects the decrease of the junction contribution, until the intrinsic properties dominate in the best crystals.

**LOWER CRITICAL FIELD \( H_{c1} \)**

In the above experiment, in which a dc magnetic field is applied in the presence of a probe rf magnetic field, flux entry manifests itself as a sharp deviation from the quadratic dependence of \( \delta \lambda \). The feature at \( H_{c1} \) is much sharper than is typically seen in magnetization measurements, and the present experiment is a new method to measure \( H_{c1} \). To our knowledge these results constitute the first complete determination of the temperature dependence of \( H_{c1} \). The data for the case of \( H \perp c \)-axis are in very good agreement with BCS calculations\(^2\), assuming \( H_{c1} \propto 1/\lambda^2(T) \). At 4.2K the measured value of \( H_{c1}(0) = 250 \) G.

The \( H_{c1} \) values also vary considerably with sample quality, providing a plausible explanation for the wide variation reported in the literature. In polycrystals, typical values are \(< 10 \) G. This is consistent with the Josephson junction model, since for fields \( H_{c1} < \phi_0 / \Sigma \sim 5 \) to \( 10 \) G, flux entry occurs into the junction. The weakest junctions dominate the observed response, and as the junction contribution is minimised with improved sample quality, \( H_{c1} \) appears to saturate at 250 G.

**MICROWAVE SURFACE RESISTANCE \( R_s \)**

The parameter of greatest interest for high frequency applications is the surface resistance \( R_s \). This quantity is typically measured at frequencies \( > 500 \) MHz, where the absorption is appreciable.

In contrast to the polycrystals, \( R_s \) drops extremely rapidly below \( T_c \) in the best crystals. At 10 GHz the drop is nearly 3 orders of magnitude within 4 K below the transition temperature. Comparable results have also been obtained in thin films prepared by laser ablation\(^6,7\).

The temperature dependence near \( T_c \) was compared with calculations based on the BCS theory. Such comparisons can be made at several levels, determined primarily by the number of input parameters that one is comfortable with. A complete calculation was carried out using the numerical program of Halbritter\(^12\), which includes the detailed \( \omega \) and \( q \) dependent kernel. Theory and experiment were consistent with the following parameters: \( \lambda(0) = 1400 \) Å, \( \Delta/kT_c = 2.15 \), mean free path \( l = 70 \) Å and coherence length \( \xi_0 = 31 \) Å, although good fits were also achieved over a region of parameter space surrounding the above values. A wider temperature range is necessary to limit the
uncertainty in these parameters.

It is important to realize that near $T_c$, the $T$ dependence of $R_s$ is dominated by $\lambda(T)$. This is exemplified by the good fit\(^2\) of the data to a simple local limit approximation: $Z_s = R_n \left\{ 2i/[1 + i \delta_n^2/2\lambda^2(T)] \right\}^{1/2}$. Here quasiparticles are ignored, and all parameters are directly determined from experiment, viz. $\lambda(T)$ as described before, and $\delta_n$ and $R_n$ from the measured normal state values. The details of the quasiparticle spectrum become important at lower temperatures. Because of the sensitivity limitations due to sample size and high quality, the characteristic signature of an $s$-wave state, viz. $R_s \propto \exp(-\Delta/kT)$ remains to be verified.

**INTRINSIC VS. GRANULAR RESPONSE**

A close look at the magnitudes of electrodynamic parameters of several samples reveals that the wide variability in these parameters cannot be dismissed as "junk effects", but rather contain an important piece of physics characteristic of the high $T_c$ superconductors. As one goes from polycrystalline samples to the best crystals, the parameters $\lambda(0)$, $k(0)$ and $R_s$ progressively decrease over several orders of magnitudes, while $H_{c1}$ increases. In the best crystals the behaviour of these parameters is consistent with BCS calculations. On the other extreme, some of the properties of the polycrystals, particularly $k(0)$, can be well described by a single Josephson junction analysis\(^{11}\), with appropriate large values of the junction area $\Sigma$. There have been several attempts\(^{13}\) to describe the surface impedance of granular material on the basis of junctions.

A realistic network model by Clem\(^{14}\) provides some insight into the electrodynamic response in the intermediate case where both intrinsic and granular contributions are present. The penetration depth of a regular network is calculated to be $\lambda_{\text{eff}}^2 = \lambda_j^2 + \lambda_{\text{int}}^2$. (The inductive impedances of the junction and the superconductor appear in series, leading to a quadrature sum for the penetration depths). Similarly, the field dependent effects represented by $k$ can be written $k_{\text{eff}} = k_j + k_{\text{int}}$, where $k_j$ and $k_{\text{int}}$ are the junction and intrinsic G-L contributions discussed earlier. The observed $H_{c1}$ will be the lower of $H_{c1,j}$ and the intrinsic $H_{c1}$. A detailed model for the surface impedance of networks and quantitative comparison to experiments, remains to be carried out. In any such model however, the junction dissipation can only be included ad hoc, whereas the reactive contributions (such as $k$ and $\lambda$) are determined by geometrical factors, such as junction size.

In polycrystals, the normal state $R_n$ is high due to the large resistivity, and is followed by a rather broad transition below $T_c$ with very high residual values measured at 4.2K. Although the data near $T_c$ can be described in terms of a modified Mattis-Bardeen model\(^9\), the low temperature data do not show\(^{15}\) an $\exp(-\Delta/kT)$ behaviour, even with the residual resistance subtracted. It appears that the temperature dependence of $R_s$ may be almost entirely due to the temperature dependence of $\lambda$, which is very large in the
polycrystals as stated earlier. Whether a junction model will be able to account in detail for the $R_s(T)$ and $\lambda(T)$ in polycrystals remains to be seen.

A characteristic feature of a superconductor is the $f^2$ dependence of $R_s$. This is best demonstrated by measurements on the same sample, as in ref. 15 on a polycrystalline sample. This feature is also present in the junction models, since it arises from the presence of an inductive supercurrent branch.

SUPERCONDUCTING MICROWAVE DEVICES - CAVITIES AND FILTERS

In parallel with the effort aimed at understanding the basic electrodynamic properties of the high $T_c$ superconductors, we have also been involved in the development of superconducting microwave devices fabricated out of these materials. A microwave cavity is a fundamental device with potential applications in particle accelerator structures$^{16}$ and ultra stable clocks$^{17}$. A cylindrical cavity was fabricated$^{18}$ from ceramic $\text{Y}_1\text{Ba}_2\text{Cu}_3\text{O}_y$ The two pieces were formed using a cold isostatic press. Coupling holes were mechanically drilled into the material. The cavity was resonant in the TE$_{011}$ mode at 8 GHz. Starting from a Q of $10^3$ at 100K, the Q improved to $10^4$ at 77K and nearly $10^5$ at 4.2K. The Q(T) data correlated very well with independent measurements of $R_s$ using the technique mentioned earlier. This device, the first of its kind to be fabricated out of the high $T_c$ superconductors, demonstrated that practical devices are feasible, in which the performance limitations were due to the material and not due to extraneous factors such as joint losses, etc. Based on the single crystal results discussed earlier, Q values exceeding $10^6$ at 77K are achievable in principle using high quality material.

Another device of great practical importance in communication systems is a superconducting filter. A typical use of these filters is as a front end to low noise amplifiers. The primary requirement is extremely low insertion loss, so as to avoid added noise to the following amplifier. Although a high Q cavity could be used as a filter, the large number of nearby spurious modes can lead to amplifier saturation. We$^{19}$ have designed an interdigital stripline filter with a center frequency of 7.8 GHz and a passband of about 0.3 GHz. The higher order modes, which are all above 20 GHz, are well separated. The filter was machined out of Ag, and coated with YBCO using a process developed at Argonne. At 4.2K the insertion loss was 2 dB, and improvement by a 13 dB from 100K (see Fig. 2). It appears that the limitation in this case is not due to the material, but rather to a temperature independent loss mechanism, probably due to the joints. Insertion losses of 0.1 dB, limited primarily by the connector mismatch, should be achievable with improvements in design.
CONCLUSIONS

Since the discovery of the high critical temperature superconductors, significant progress has been achieved in understanding the electrodynamic response of these materials. Advances in materials fabrication, leading to high quality samples, has been an important factor. In $Y_1Ba_2Cu_3O_y$ the electrodynamic parameters display temperature dependences which are in good agreement with calculations based on the BCS theory. Since the central parameter common to the calculations is the gap, the results strongly suggest a mean field temperature dependence for the gap parameter, and a value of $\Delta(0)/kT_c = 2.15$. Further confirmation of the quasiparticle spectrum, viz. an $\exp(-\Delta/kT)$ dependence of $\delta\lambda$ and $R_s$ at low temperatures, requires substantially more sensitivity than is currently available.

From a technological view point, the results on carefully prepared single crystals and thin films offer great promise for device applications at microwave frequencies. Orders of magnitude better performance than conventional devices using Cu are achievable in principle at temperatures exceeding 77K, provided high sample quality can be retained in realistic device structures.

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REFERENCES

Fig. 2. Insertion loss vs. temperature of the 7.8 GHz interdigital stripline filter.
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