Microwave resistance of ceramic YBa₂Cu₃O_{6.9}

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The temperature dependence of the microwave resistance R was determined at 17.6 GHz using a resonator made entirely of ceramic YBa₂Cu₃O_{6.9}. The dependence R(T) is well described by an empirical expression below the superconducting transition temperature T_c . The strong dependence R(T) observed at $T \ll T_c$ was in agreement with measurements of the depth of penetration. The microwave resistance of ceramic disks prepared in different ways and subjected to different subsequent treatments were compared at liquid helium temperatures. The disks with a higher density had lower residual resistances. The residual resistance was proportional to the square of the frequency.

One of the experimental methods for the investigation of high-temperature superconductors involves determination of the microwave response. In spite of the large number of papers on this topic, ¹⁻⁵ this response is not yet fully understood. In particular, the importance of microinhomogeneities of the ceramic structure has not yet been fully established and it is not known how the impedance of the superconductor with the highest transition temperature varies below this temperature. Moreover, electrodynamic properties are known to provide an important key for understanding the nature of superconductivity.

We determined the temperature dependence of the microwave response of ceramic $YBa_2Cu_3O_{6.9}$. Such a determination should be carried out in a wide range of temperatures. It is usually difficult to identify accurately the contribution of a sample against the background of the changes due to the material from which the resonator is made. We therefore used a resonator made entirely of ceramic $YBa_2Cu_3O_{6.9}$. The advantage of this approach (which affords approximately the same accuracy in the normal and superconducting ranges of temperature) is in our opinion worth the trouble encountered in making a ceramic resonator.

MATERIAL AND MEASUREMENT METHOD

Our initial components used in synthesizing the ceramic material were powdered copper, barium, and yttrium oxides, which were carefully mixed in the required ratio and heat-treated in a stream of oxygen for several hours employing the usual technology.⁶ The resultant ceramic of the YBa- $_2$ Cu₃O_{6.9} composition was ground to a powder with a particle size less than 50 μ m. This powder was then compacted by hydrostatic pressures of 2–5 kbar to form cans or tubes with walls of thickness 5 mm, as well as disks of approximately the same thickness. This was followed by heat treatment in an oxygen stream. Depending on the powder grain size and on the pressure used in the compacting stage, our samples had different densities ranging from 5.0 to 5.9 g/cm². The internal surfaces of some of the resonators were ground and polished in the usual manner.

We also tried other methods for fabricating cylindrical resonators operating at frequencies 9 < f < 19 GHz. We now consider in detail one of these resonators characterized by a natural frequency of f = 17.6 GHz. This was a circular cylinder with the internal diameter 22.8 mm and of height 20.8 mm, consisting of two parts: a cylinder with a base 1 and a disk 2 closing its end (Fig. 1). The two parts were clamped together by an external spring. The quality of the electrical contact between the two parts was umimportant because the TE₀₁₁ resonator oscillation mode was not associated with currents crossing the line of contact between the two parts. The TM₁₁₁ mode, degenerate with TE_{011} , was suppressed by insulating the disk 2 from the cylinder 1 with a thin Mylar film. An electromagnetic wave traveled along a waveguide of 11×5.5 mm cross section and reached the resonator. The excitation of the resonator and the coupling to an external detector were performed by two separate loops 4 introduced through apertures in a side wall of the resonator. The planes of these loops were perpendicular to the lines of force of the magnetic field of the TE_{011} mode shown dashed in Fig. 1. The depth of insertion of the loops was selected to ensure that at any temperature the resonator was coupled weakly to the input and output channels. The detector therefore recorded a signal proportional to the square Q^2 of the intrinsic Q factor of the resonator. The Q(T) curve was calibrated using the absolute value of Q known at several points from the width of the resonance curve. The temperature of the resonator was measured with a germanium thermometer 5.

The main microwave characteristic of a material is its surface impedance. We determined the real part of the impedance in the form of the surface resistance R. This resistance governed the power P absorbed per unit surface area

$$P = \frac{1}{2}RH_{\sim}^{2}$$

 (H_{\sim}) is the amplitude of an hf magnetic field on the surface). Integration over the whole internal surface of the resonator gave the relationship between R and the Q factor. In the case of our resonator the relationship was

$$R(\Omega) = 780/Q$$

We assumed that the radiative losses could be ignored.

The losses observed for different ceramics at $T \approx 2$ K were compared using end disks 2 prepared and treated in different ways; these disks were pressed against the niobium cylinder 1. Under these conditions the absorption in the disk was the main source of losses experienced by the resonator.



FIG. 1. Schematic diagram showing a resonator with coupling loops.

RESULTS

Figure 2 shows the experimental dependence R(T) obtained for a resonator made of a ceramic of density 5.1 g/ cm³. Two important features of this dependence should be mentioned.

The first feature of the R(T) curve was the temperature dependence well below the transition point. In the case of classical superconductors the surface resistance below $T_c/2$, found after subtracting the residual resistance R_0 , is exponentially small⁷

$$R(\omega, T) - R_0 \propto \exp(-\Delta/T).$$
(1)

The curve plotted in Fig. 2 cannot be described by the above expression. However, the experimental results are fitted well by the following empirical expression down to temperatures $t \equiv T/T_c \leq 0.96$:

$$\frac{R(T)-R_0}{R(T_c)} = \frac{R_o}{R(T_c)} = a\varphi^{\nu},$$
(2)

where R_s is the surface resistance of an ideal superconductor; a = 0.15; $\varphi = t^4(1 - t^4)^{-1}$ is a variable occurring in the two-liquid Gorter-Casimir model; $R_0 = R(2 \text{ K}) = 0.014 \Omega$ (Fig. 3).

The second feature of the curve in Fig. 2 was a very high value of the residual losses, compared with classical superconductors. According to this figure, at liquid helium temperatures the resistance R_0 was only 18 times less than R in the normal state just before the transition. It was established in Ref. 5 that R_0 of superconducting YBa₂Cu₃O_{7-x} ceramics varied quadratically with the frequency: $R_0 \propto \omega^2$. We carried out measurements using two similar niobium resonators



FIG. 2. Temperature dependence of the resistance of a ceramic resonator characterized by $\rho = 5.1$ g/cm³ and f = 17.6 GHz.

at two frequencies 9 and 18 GHz and obtained, in agreement with Ref. 5, values of R_0 differing by a factor of 4.

The value of R_0 depended also on the density of the ceramic and on the state of its surface. This was established in experiments carried out using niobium resonators. The use of a ceramic with a higher density (5.9 g/cm³) reduced R_0 to 6 m Ω and polishing of the surface of this dense ceramic reduced R_0 still further to 4 m Ω . The relationship $R_0 \propto \omega^2$ enabled us to compare these values with the data taken from other investigations and reduced to the frequency 18 GHz: 25 m Ω from Ref. 2, 1.5 Ω from Ref. 4, and 31 m Ω from Ref. 5.

Many microwave absorption experiments have revealed the influence of a weak magnetic field. We took measurements to correct for the field of the Earth to within 1-2% and compared the residual resistance when a sample was cooled in the field of the Earth and in the absence of this field. We found no significant reduction in the Q factor.

However, it should be pointed out that the high value of R_0 was definitely not associated with nonlinear effects. Estimates obtained indicated that the maximum amplitude of the hf field on the surface of the ceramic at the highest power reaching the input waveguide channel was only 0.1 Oe. A reduction in the power by a factor of 1000 did not alter R_0 .

The dc resistivity ρ was determined for one of the disks. To within the limits of the experimental error the room-temperature resistance R was related to ρ by the usual expression applicable in the case of the normal skin effect:

 $R = (\omega \mu_0 \rho/2)^{\frac{1}{2}}$.

DISCUSSION

The mechanism of strong microwave absorption at low temperatures far from T_c is undoubtedly related to the inhomogeneity of the samples and particularly to the presence of normally conducting regions near the surface. The role of such regions had been analyzed earlier⁸ in studies of the residual resistance of lead and niobium. We know that if there are surface cavities filled with a conducting substance, their contribution to the surface resistance can be estimated by assuming that each cavity is a waveguide with a termination, which then makes it possible to estimate the electric field in the cavity. In an idealized model when all the cavities are the same, an estimate of the residual resistance gives



FIG. 3. Comparison of the experimental dependence $R_x/R(T_c)$ with the function (2).

$$R_{0} = \omega^{2} \mu_{0}^{2} s^{3} x / 4\pi^{3} \rho, \qquad (3)$$

where ρ is the resistivity of the substance filling the cavities; s^3 is the characteristic volume of a cavity; x is the part of a surface occupied by a normal conductor. If $\rho \approx 1000 \mu \Omega$.cm, $s \approx 10 \mu$ m, and $x \approx 1$, we obtain $R_0 \approx 10^{-2} \Omega$. Equation (3) yields a quadratic frequency dependence of the residual resistance, which—as demonstrated above—is supported by the experimental results. Equation (3) accounts also for the reduction in R_0 with increasing sample density, because a denser ceramic should contain smaller pores.

The electromagnetic response of ceramic superconductor grains with sizes on the order of penetration depth should be described by average effective parameters that allow for the presence of grain boundaries. The "inductive" part of the conductivity created by superconducting electrons may be balanced by the "capacitative" conductivity due to insulating layers between the grains. The temperature dependence of the microwave power absorption may be very different for homogeneous and ceramic superconductors.

In view of the absence of theoretical calculations of the electromagnetic response of such a ceramic considered as a granulated system, we attempt to understand our temperature dependence using the current ideas on the electrodynamics of homogeneous superconductors.

It is now generally accepted that the depth of penetration of an electromagnetic field into Y-Ba-Cu-O ceramics in the superconducting state is 10^3-10^4 Å, and the coherence length ξ and the mean free path *l* are 5-50 Å. Since $\lambda \ge \xi l$, an analysis of the experimental dependence R(T) should be made using a local coupling between the current and field.

The surface impedance of a homogeneous superconductor with a smooth surface is described by the following expression valid at temperatures $T < T_c$:

$$Z = R + iX = (i\omega\mu_0/\sigma)^{\frac{1}{2}} = R_n(2i\sigma_n/\sigma)^{\frac{1}{2}},$$

where $\sigma = \sigma_1 - i\sigma_2$ is the complex conductivity of a superconductor in an hf field; $R_n = (\omega \mu_0 / 2\sigma_n)^{1/2}$ and $\sigma_n = nl^2 \tau / m$ are, respectively, the surface resistance and the conductivity in the normal state. According to the two-liquid model, if $\omega \tau \ll 1$, then

$$\left(\frac{R_s}{R_n}\right)^2 = \frac{\beta}{\alpha^{1/2}} (1 - (1 - \alpha^{-1})^{1/2}).$$
(4)

Here, $\beta = n/n_n$, $\alpha = 1 + (n_s/n_n\omega\tau) = (1 + n_n\omega^2\tau^2/n_s)^2$, n_n and n_s are the densities of normal and superconducting electrons at $T < T_c$, and $n_n + n_s = n$. If $n_n/n_s \leq 1$, then $\alpha \ge 1$ and

$$\frac{R_s}{R_n} = \frac{(\omega\tau)^{\frac{\gamma_h}{2}}}{2^{\frac{\gamma_h}{2}}} \left(1 + \frac{n_n}{n_s}\right)^{\frac{\gamma_h}{2}} \frac{n_n}{n_s}.$$
(5)

If in Eq. (4) or (5) we substitute the following relationship from microscopic theory:

$$\frac{n_n}{n} = \left[\frac{2\pi\Delta(T)}{kT}\right]^{\frac{1}{2}} \exp\left(-\frac{\Delta}{kT}\right),\tag{6}$$

valid in the range $T < T_c / 2$, we obtain the exponential dependence of Eq. (1). If instead of Eq. (6), we use the Gorter-Casimir relationship

$$\frac{n_n}{n_s} = \varphi(t) = \frac{t^4}{1 - t^4}, \quad t = \frac{T}{T_c},$$

it follows from Eq. (5) that

$$R_{s}(t) \propto \varphi(\varphi+1)^{\frac{1}{2}} = t^{4}(1-t^{4})^{-\frac{3}{2}}.$$
(7)

Since the experimental result of Eq. (8) does not agree with Eq. (1) or Eq. (7), we have to compare results with other experimental data.

A strong dependence R(T) at low temperatures was also reported in Refs. 2–5. In particular, the temperature dependence of the resistance obtained in Ref. 5 for ceramic samples was reasonably described by Eq. (2). Unfortunately, quantitative comparisons were limited by the fact that the results reported in Refs. 2–5 gave the R(T) dependence and these were presented simply in the form of graphs.

Moreover, Eqs. (4) and (5) allow us to carry out a comparison with the measured values of the penetration depth $\lambda(T)$. According to the two-liquid model, we have

$$\frac{\lambda(T)}{\lambda(0)} = \left(1 + \frac{n_n}{n_s}\right)^{\frac{1}{2}},\tag{8}$$

i.e., the temperature dependence $\lambda(T)$ includes the same ratio n_n/n_s as Eq. (5). This allows us to use the experimental dependence of Eq. (2) to plot the corresponding dependence $\lambda(T)/\lambda(0)$, which, however, includes an unknown parameter $\omega \tau$ [compare Eqs. (5) and (8)]. In particular, if $t \leq 1$ (and, consequently, $n_n/n_s \leq 1$), it follows from Eqs. (2), (5), and (8) that

$$at^2 = \frac{(\omega\tau)^{\frac{\eta_2}{2}}}{2^{\frac{\eta_2}{2}}} \frac{n_n}{n_o}$$

and

$$\frac{\Delta\lambda(t)}{\lambda(0)} = \frac{\lambda(t) - \lambda(0)}{\lambda(0)} = \frac{a}{2^{\frac{1}{2}}(\omega\tau)^{\frac{n}{2}}}t^{2}.$$
(9)

The dependence of the coefficient in front of t^2 on ω is only apparent. Since $R_n \propto \omega^{1/2}$ and $R_s \propto \omega^2$ (Ref. 5), the coefficient *a* should be described by $a \propto \omega^{3/2}$.

Quadratic $\Delta\lambda(t)$ dependence has been observed in a number of experiments on a ceramic⁹ and on single crystals in the (a, b) plane.¹⁰ As is known, a similar dependence $\Delta\lambda \propto t^2$ is observed for heavy-fermion systems.¹¹⁻¹²

A comparison of the coefficient of t^2 with that in Eq. (9) yields an estimate $\omega \tau \approx 0.3$.

We conclude by noting that the experimental results were analyzed on the assumption that the contribution of the defects to the resistance described by R_0 is independent of t. However, at low values of t ($t \le 0.3$) the resistances R_0 and $R_s(t)$ are comparable. One could determine the reliability of the $R_s(t)$ dependence at these temperatures by ensuring that R_0 is at least an order of magnitude less.

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