Temperature Dependence of Electrodynamic Properties of YBa$_2$Cu$_3$O$_y$ Crystals

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(Received 8 June 1989)

The temperature dependences of the radio-frequency penetration depth $\lambda_{ab}$, microwave surface resistance $R_s$, and the lower critical field $H_{c1}$ were measured for YBa$_2$Cu$_3$O$_y$ crystals. Pair-breaking effects of magnetic fields on $\lambda_{ab}$, represented by a parameter $k$ $=$ $\partial \lambda_{ab}/\partial (T^2)$, were also measured. The temperature dependences of all these parameters $\lambda_{ab}$, $H_{c1}$, $k$, and $R_s$ are in good agreement with the Bardeen-Cooper-Schrieffer temperature dependences for an $s$-wave superconductor, and suggest a mean-field behavior of the gap parameter.

PACS numbers: 74.30.Gn, 74.20.Fg, 74.70.Vy

Since the discovery$^1$ of the high-$T_c$ oxide superconductors, two outstanding questions continue to be debated. One concerns the microscopic mechanism leading to superconductivity, and the other the nature of the superconducting state. Conclusions regarding the latter require experiments which will enable building up a consistent phenomenological picture. In particular, experiments which probe the electrodynamic response of the superconductors at high frequency can yield information regarding the gap, the density of states, and the nature of the pairing. The results should then be explained by a successful theory of superconductivity for these materials, as happened for the "low-$T_c$" superconductors.$^2$

In this paper, we report the results and analysis of a comprehensive set of electrodynamic measurements of high-quality YBa$_2$Cu$_3$O$_y$ crystals. Measurements were carried out on the temperature dependence of the penetration depth at 6 MHz and the surface resistance at 10 GHz. The data for these parameters are in good agreement with detailed calculations based upon a Bardeen-Cooper-Schrieffer (BCS) theory. A new type of experiment, in which the effect of small magnetic fields on $\lambda$ was studied, yielded quadratic dependences of $\lambda$ on the magnetic field, i.e., $\lambda_{ab}(T,H) = \lambda_{ab}(T,0) + k(T)H^2$. The quadratic coefficient $k$, which varies over 5 orders of magnitude with temperature, is in good agreement with a Ginzburg-Landau (GL) theory, using BCS temperature dependences. In the same experiment, the lower critical field $H_{c1}(\perp c$ axis) is observed as a sharp deviation from the quadratic behavior, and is a new method of determining this parameter. The complete temperature dependence of $H_{c1}$ is in good agreement with a BCS calculation. These results provide strong evidence for an $s$-wave superconducting state, and a mean-field behavior of the gap parameter in YBa$_2$Cu$_3$O$_y$.

The experiments and the analysis further yield a complete set of electrodynamic parameters for YBa$_2$Cu$_3$O$_y$. The 6-MHz experiments directly yield $k(0) = 1 \times 10^{-3}$ Å/G$^2$, $H_{c1,\perp}(0) = 250$ G, and from fits with the BCS theory, $\lambda_{ab}(0) = 1400$ Å, $\Delta(0)/kT_c = 2.15$. The microwave results are consistent with these parameters.

Our experiments may be viewed in a general framework as testing the fundamental constitutive current-vector-potential relation: $J = -Q_1 A - Q_2 A^3$. For the zero-dc-field experiments, $A = A_f$, and the only linear term is relevant, with the kernel $Q_1 \propto 1/\lambda^2$ for the penetration-depth experiment, and $Q_1$ complex for microwave absorption. For the field dependence of $\lambda$, $A = \delta A_{dc} + A_{hf}$, but only the response $J_f = -Q_2 \delta A_{dc}A_{hf}$ is measured, with $Q_2 \propto k$. The kernel $Q_1$ is determined by the nature of the superconducting ground state, and $Q_2$ is described by pair-breaking effects of the magnetic field, and these can in principle be obtained from microscopic theories.

Details of the fabrication and the characteristics of the crystals are discussed elsewhere.$^3$ The 6-MHz measurements of $\lambda_{ab}$ were carried out with resonant coils,$^4$ using an ultrastable tunnel-diode oscillator. Changes $\delta \lambda$ in the resonant frequency $\nu$ of the oscillator are related to changes $\delta \lambda_{ab}$ by $\delta \lambda_{ab} = -G \delta \nu / \nu$, where $G$ is a geometric factor. At zero external field, the temperature dependence $\delta \lambda_{ab}(T)$ was measured. (By use of a Mumetal shield, the residual magnetic field was $\approx 10^{-2}$ G.) The background temperature dependence of the coil$^5$ was carefully accounted for in the analysis of $\delta \lambda_{ab}(T)$, and presently limits the low-temperature resolution to $\delta \lambda_{ab}(T) \approx 200$ Å, and the temperature range to above $T \approx 55$ K. In what follows, we discuss the results for $\delta \lambda_{ab}(T) = \lambda(T) - \lambda(T = 55$ K). In a separate experiment using the same configuration, small dc magnetic fields were applied at fixed temperature, and the changes $\delta \lambda_{ab}(H)$ were also measured at various temperatures. In this latter experiment, the highly stable oscillator (1 Hz in $6 \times 10^6$ Hz) enables a resolution $\delta \lambda_{ab}(H) \approx 20$ Å, over the entire temperature range 4.2 K to $T_c$.

The temperature dependence of $\delta \lambda_{ab}(T)$ is shown in Fig. 1, and the data are compared to the same quantity using the local-limit BCS result$^6$

\[ \lambda^{-2}(T) = \lambda^{-2}(0) \left[ 1 - 2 \int_{\Delta}^{\infty} (-\partial \lambda/\partial E)[E/(E^2 - \Delta^2)^{1/2}]dE \right], \]

\[ \lambda_{ab}(0) = 1400 \text{ Å}, \quad \Delta(0)/kT_c = 2.15. \]
FIG. 1. Temperature dependence of $\delta_{ab}(T)$ for a YBa$_2$Cu$_3$O$_y$ crystal ($T_c = 86.2$ K) at 6 MHz. The lines represent BCS calculations: solid, $\lambda_{ab}(0) = 1400$ Å, $\Delta(0)/kT_c = 2.15$; dashed, $\lambda_{ab}(0) = 1000$ Å, $\Delta(0)/kT_c = 1.76$.

where $f$ is the Fermi function. The BCS temperature dependence$^7$ for $\Delta(T)$ was used. We have used a semilogarithmic plot to emphasize the range of measurement of $\delta_{ab}$, rather than the more usual linear plot$^8$-10 The scatter in the data indicates the resolution limit due to the background subtraction. The transition temperature $T_c$ ($= 86.2$ K) was determined from the peak in $d\lambda/dT$, and coincides with the midpoint of the inductive transition. The weak-coupling$^{[7]}$ with $\Delta(0)/kT_c = 1.76$] temperature dependence for $\lambda$ agrees with the data near $T_c$, with $\lambda_{ab}(0) = 1000$ Å, but deviates from the data at lower temperature (dashed line). If, however, $\Delta(0)/kT_c$ is treated as an additional fitting parameter, as is commonly done for conventional superconductors, it is possible to fit the data over the entire temperature range. Best fits were obtained for $\lambda_{ab}(0) = 1400 \pm 100$ Å and $\Delta(0)/kT_c = 2.15 \pm 0.1$, as shown in Fig. 1 (solid line).

We next discuss the results of a novel experiment in which we study the effects of small dc magnetic fields on the rf penetration depth. Typical results for $\delta_{ab}(H)$ are shown in Fig. 2, at $T = 77$ K. Similar results were observed over the entire temperature range 4.2 to 85.9 K ($T_c = 86.2$ K). Two distinct regimes are observed: At low fields, the data can be clearly represented as $\delta_{ab}(T,H) = k(T)H^2$, where $k(T)$ is an experimentally determined quadratic coefficient. A distinct change of slope is observed at a characteristic field which we identify as $H_{c1}$, the lower critical field. For $H > H_{c1}$, $\lambda$ increases with a dependence weaker than quadratic, with evidence of saturation at high fields. The high-field behavior due to flux is the subject of a separate study; in this paper we have focused on the electrodynamics in the Meissner state and discuss the complete temperature dependences of both $k(T)$ and $H_{c1}(T)$.

The quadratic dependence of $\delta_{ab}$ on $H$ in the Meissner region ($H < H_{c1}$), which is due to pair-breaking effects on the gap, can be understood on thermodynamic grounds. All thermodynamic parameters, including $\lambda_{ab}$, should vary as $H^2$ to first order. The magnitude of such effects can be calculated using a Ginzburg-Landau model. This yields for the quadratic coefficient $k(T) = \frac{1}{2} \lambda_{ab}(T)/H_{c0}^2(T)$, where $H_{c0}$ is the thermodynamic critical field. The line in Fig. 3 is the temperature dependence for $k(T)$ calculated using BCS numerical results$^7$ for $\lambda_{ab}(T)$ and $H_{c0}(T)$. We note the remarkable agreement over 5 orders-of-magnitude variation of $k(T)$ with temperature. Using the measured value of $k(4.2$ K) $= 10^{-3}$ Å/G$^2$ and $\lambda_{ab}(0) = 1400$ Å, we deduce

FIG. 2. Change in 6-MHz penetration depth $\delta_{ab}(H)$ for the crystal of Fig. 1 with external dc magnetic field $H$ at $T = 77$ K. Similar data were obtained between 4.2 and 85.9 K. Note that quadratic behavior at low fields, and the sharp change in slope at $H = H_{c1}$. Inset: field configuration.

FIG. 3. Temperature dependence of the quadratic coefficient $k(T)$, where $\delta_{ab}(T,H) = k(T)H^2$. The line represents a Ginzburg-Landau calculation using BCS temperature dependences.

1874
$H_{c0}(0) = 1.02$ kG. This value is comparable to some estimates\textsuperscript{13} (1.8 kG) and lower than others\textsuperscript{14} ($\sim 10$ kG). A more detailed theory including density of states and anisotropy would be expected to retain the overall temperature dependence, but could alter the magnitudes, and hence our estimates of $H_{c0}$. What is significant is that the overall temperature dependence agrees with the BCS temperature dependence. We note further that this experiment implies a field-dependent gap given by

$$(1/\Delta) d\Delta/dH^2 = -(2/\Delta) d\Delta/dH^2 = -1.4 \times 10^{-6}/\text{G}^2$$

at $T \ll T_c$, similar in magnitude to that observed in Al.\textsuperscript{15}

The present experiment yields a sharp signature at $H_{c1}$, and is a new method of determining $H_{c1}$, in contrast to magnetization experiments in which $H_{c1}$ is inferred from a gradual change in magnetization.\textsuperscript{16} The complete temperature dependence of $H_{c1}(T)$ is presented in Fig. 4. The line in Fig. 4 is a BCS calculation using $H_{c1}(T) \propto \lambda^{-2}(T)$, which would apply to an isotropic superconductor where $H_{c1} = (\Phi_0/4\pi\lambda^2)\ln(\kappa)$. Again we find good agreement ($\lambda$ is calculated as previously described). From the experimental values $H_{c1}(0) = 250 \pm 5$ G (this agrees with Ref. 17) and $\lambda_{ab}(0) = 1400$ Å, the above relation yields $\kappa = 21$, but it should be noted that anisotropy complicates the analysis of this $\kappa$ value.

The conclusions of the radio-frequency experiments are further validated by microwave measurements of $R_s$ at 10 GHz, using the technique of Ref. 19, with a superconductivity cavity made of Nb. The behavior of $R_s$, with temperature, for a larger (0.5 mm$^2 \times 40$ µm, $T_c = 75$ K) crystal is shown in Fig. 5. The inset compares results for the high-quality crystal with our best polycrystalline ceramic. The crystal data drop sharply near the transition, and disappear below our sensitivity of $\approx 400$ µΩ, in contrast to the polycrystals. Although this method for $R_s$ is currently one of the most sensitive available, the data range is limited due to the small sample size, and the superior sample properties. The sharpness of the transition attests to the high quality of the crystal. The main figure shows a comparison to the BCS theory, using a simple local-limit approximation $Z_s = R_s/[(1 + i\delta_s/2\lambda(T)^2)]^{1/2}$. This approximation, which ignores quasiparticles, is only valid near $T_c$. $R_s$ and $\delta_s$ are the normal-state surface resistance and the classical skin depth, respectively. The line in Fig. 5 was obtained using the experimentally determined parameters: $\lambda_{ab}(0) = 1400$ Å, $\delta_s = 2.5$ µm (from $R_s$ using classical-skin-depth analysis). Theory and experiment are consistent, and Fig. 5 demonstrates\textsuperscript{20} the BCS-type nature of $R_s$.

In this paper, we have considered several electrodynamic experiments, and have shown that in every instance, the BCS theory provides a good description of the results. One must ask to what extent our work constitutes conclusive evidence for an $s$-wave superconducting state in YBa$_2$Cu$_3$O$_y$.

The first question which one may ask is the following: To what extent are the present results "intrinsic?" This, of course, bears directly on sample quality. The results presented here are for our best samples, taken from extensive measurements on several polycrystalline and single-crystal samples. The polycrystal samples possess different temperature dependences,\textsuperscript{21} and vastly different electrodynamic parameters: \textsuperscript{3} $\lambda_{ab}(0) = 7.5$ µm, $R_s(4.2$ K) = 5 mΩ, and most importantly, $k(0) = 160$ Å/G$^2$ which is 5 orders of magnitude larger than for the crystals quoted here. Furthermore, the threshold field for flux entry, which we call $H_{c2}$, is approximately 3–22 G (i.e., $\ll 250$ G). We have shown elsewhere\textsuperscript{4} that the electrodynamics of polycrystalline samples can be well described by a Josephson-junction network, with typical junction area 2 to 4 µm$^2$. Nominal single crystals of
moderate quality exhibit electrodynamic parameters\textsuperscript{3} of intermediate values, e.g., $\lambda_{\text{ab}}(0) \sim 2000$ to 8000 Å, $k(0) \sim 10^{-2}$ to 1 Å/G, and $H_{c2}(0) \leq 250$ G. In contrast, the electrodynamic parameters reported here are the lowest $\lambda_{\text{ab}}(0)$, $k(0)$, and $R_\ell(T)$, and the highest $H_{c1}(0)$, and all the temperature-dependent data are characteristic of superior quality samples.

Turning to the implications of the comparison of the temperature dependences, we note that the gap parameter $\Delta(T)$ is the central parameter in the BCS s-wave theory which determines the temperature dependence of the electrodynamic parameters. Our work provides strong evidence that $\Delta(T)$ varies with $T$ according to the BCS prediction. The data further suggest that the BCS description of the quasiparticle spectrum applies to $\text{YBa}_2\text{Cu}_3\text{O}_y$, with a larger gap value characteristic of strong coupling superconductors.

Further evidence for a BCS quasiparticle spectrum would be the low-temperature dependence of the transport parameters. In particular, both $\delta\lambda_{\text{ab}}(T)$ and $R_\ell$ should be $\propto \exp[-\Delta(0)/kT]$ at $T \ll T_c$, for an s-wave superconductor. To our knowledge, such a behavior has not been conclusively demonstrated or disproved in the oxide superconductors. Although the sensitivity of both the $R_\ell$ and the $\lambda_{\text{ab}}$ experiments is the best achievable to date, substantial improvements in the state of the art are required to address this aspect.

This work was supported by NSF Grant No. ECS-8811254. We thank C. A. Shiffman and J. V Jose for useful discussions.

\textsuperscript{5}The thermal contraction of the sample itself is small compared to the $\delta\lambda$ values for the temperature range discussed here because of the small thickness (20 μm).


\textsuperscript{7}B. Muhlschlegel, Z. Phys. 155, 313 (1959).


\textsuperscript{11}Similar quadratic effects on $R_\ell$ and $\lambda_{\text{ab}}$ are well known in "low-$T_c$" superconductors, e.g., A. B. Pippard, Proc. Roy. Soc. London A 203, 210 (1950); S. Sridhar and J. E. Mercereau, Phys. Rev. B 34, 203 (1986).

\textsuperscript{12}Starting from the normalized GL equations, $(1/\kappa^2)d^2\psi/dz^2 = -(1-a^2)\psi + \psi^3$ and $d^2\alpha/dz^2 = \psi^2 a$, one can show in the limit $x \to \infty$ that $\delta\lambda_{\text{ab}}/\lambda_{\text{ab}} = (1+2\cos\theta)H^2/4H^2_c$, to lowest order in $H^2$, where $\theta$ is the angle between $H$ and $H_c$. In the present experiment $\theta = 0$.


\textsuperscript{15}R. Meservey and B. B. Schwartz, in Ref. 2.


\textsuperscript{18}L. Krusin-Elbaum \textit{et al.}, Phys. Rev. B 39, 2936 (1989). These authors present the partial temperature dependence of $H_{c1}$ between 40 K and $T_c$.


\textsuperscript{20}We have also compared the data to a detailed BCS calculation, with the full frequency- and wave-vector-dependent kernel, using the numerical program of J. Halbritter, Z. Phys. 266, 209 (1974). Reasonable fits to the data were obtained for the following parameter choice: $\lambda(0) = 1400$ Å, $\Delta(0)/kT_c = 2.15$, $\xi_0 = 31$ Å, mean free path $l = 70$ Å. This comparison is not discussed in detail because of the larger number of parameters involved.