Temperature Dependence of Electrodynamic Properties of $YBa_2Cu_3O_y$ Crystals

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The temperature dependences of the radio-frequency penetration depth λ_{ab}, microwave surface resistance R_s, and the lower critical field H_{c1} were measured for $YBa_2Cu_3O_y$ crystals. Pair-breaking effects of magnetic fields on λ_{ab}, represented by a parameter $k \equiv d\lambda_{ab}/d(H^2)$, were also measured. The temperature dependences of all these parameters λ_{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.

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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 $YBa_2Cu_3O_y$ 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 λ was studied, yielded quadratic dependences of λ 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 $YBa_2Cu_3O_y$.

The experiments and the analysis further yield a complete set of electrodynamic parameters for $YBa_2Cu_3O_y$. The 6-MHz experiments directly yield $k(0) = 1\times10^{-3}$ 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: $J = -Q_1A - Q_2A^3$. For the zero-dc-field experiments, $A = A_{\text{rf}}$, and only the 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 λ, $A = A_{\text{dc}} + A_{\text{rf}}$, but only the response $J_{\text{rf}} = -Q_2A_{\text{dc}}^2A_{\text{rf}}$ 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 λ_{ab} were carried out with resonant coils,[4] using an ultrastable tunnel-diode oscillator. Changes δf in the resonant frequency f of the oscillator are related to changes $\delta\lambda_{ab}$ by $\delta\lambda_{ab} = -G\delta f/f$, 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 $\leq 10^{-2}$ 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 $\delta\lambda_{ab}(T) \sim 200$ Å, and the temperature range to above $T = 55$ K. In what follows, we discuss the results for $\delta\lambda_{ab}(T) \equiv \lambda(T) - \lambda(T = 55\text{ K})$. 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 6×10^6 Hz) enables a resolution $\delta\lambda_{ab}(H) \sim 20$ 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]

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

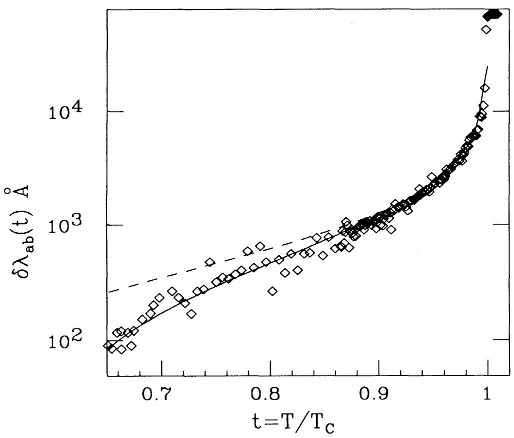

FIG. 1. Temperature dependence of $\delta\lambda_{ab}(T)$ for a $YBa_2Cu_3O_y$ crystal ($T_c=86.2$ K) at 6 MHz. The lines represent BCS calculations: solid, $\lambda_{ab}(0)=1400$ Å, $\Delta(0)/kT_c=2.15$; dashed, $\lambda_{ab}(0)=1000$ Å, $\Delta(0)/kT_c=1.76$.

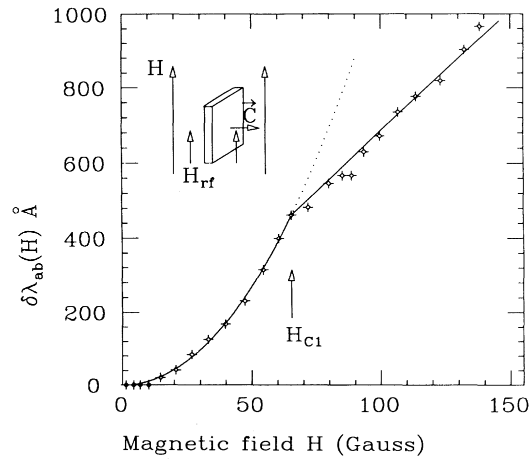

FIG. 2. Change in 6-MHz penetration depth $\delta\lambda_{ab}(H)$ for the crystal of Fig. 1 with external dc magnetic field H at $T=77$ K. Similar data were obtained between 4.2 and 85.9 K. Note that quadratic behavior at low fields, and the sharp change in slope at $H=H_{c1}$. Inset: field configuration.

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 $\delta\lambda$, 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 [with $\Delta(0)/kT_c=1.76$] temperature dependence for λ 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 $\lambda_{ab}(0)=1400\pm100$ Å and $\Delta(0)/kT_c=2.15\pm0.1$, 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 $\delta\lambda_{ab}(H)$ 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 $\delta\lambda_{ab}(T,H)=k(T)H^2$, 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 $H>H_{c1}$, λ 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 $\delta\lambda_{ab}$ 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 λ_{ab}, should vary as H^2 to first order.[11] The magnitude of such effects can be calculated using a Ginzburg-Landau model.[12] This yields for the quadratic coefficient $k(T)=\frac{3}{4}\lambda_{ab}(T)/H_{c0}^2(T)$, 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 $k(4.2\text{ K})=10^{-3}$ Å/G^2 and $\lambda_{ab}(0)=1400$ Å, we deduce

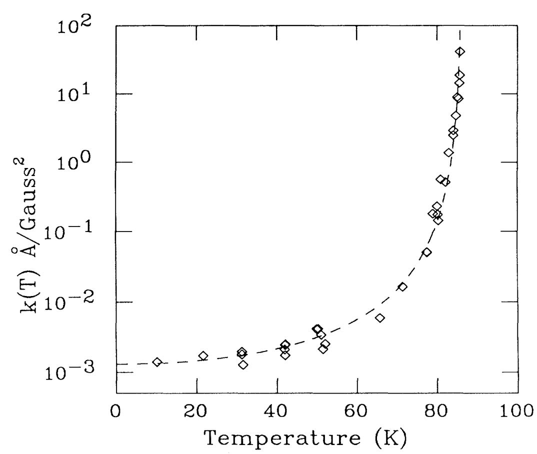

FIG. 3. Temperature dependence of the quadratic coefficient $k(T)$, where $\delta\lambda_{ab}(T,H)=k(T)H^2$. The line represents a Ginzburg-Landau calculation using BCS temperature dependences.

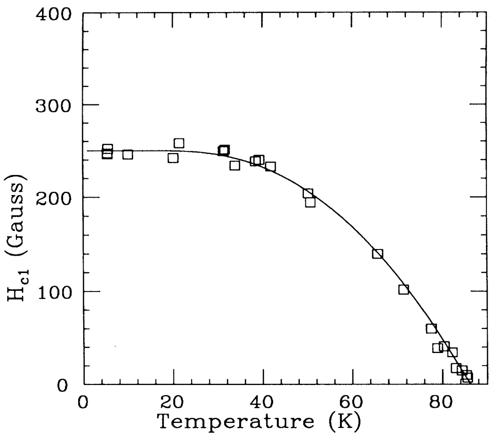

FIG. 4. Temperature dependence of $H_{c1}(\perp\ c$ axis). The line represents a BCS calculation.

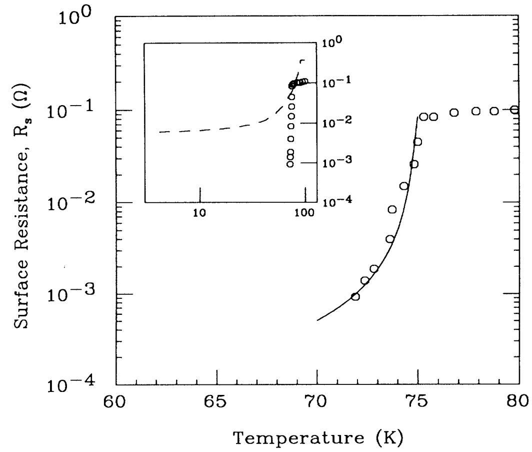

FIG. 5. Temperature dependence of surface resistance R_s at 10 GHz for a larger crystal ($T_c=75$ K). The solid line represents a BCS calculation. Inset: Comparison of the crystal with a typical polycrystal (dashed line)—note the sharp transition of the former.

$H_{c0}(0)=1.02$ kG. This value is comparable to some estimates[13] (1.8 kG) and lower than others[14] (~ 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

$$(1/\Delta)d\Delta/dH^2=-(2/\lambda)d\lambda/dH^2=-1.4\times10^{-6}/\mathrm{G}^2$$

at $T \ll T_c$, similar in magnitude to that observed in Al.[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.[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 (λ is calculated as previously described). From the experimental values $H_{c1}(0)=250$ ± 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[18] the analysis of this κ 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 $\mathrm{mm}^2\times 40$ μm, $T_c=75$ K) crystal is shown in Fig. 5. The inset compares results for the high-quality crystal with our best polycrystal ceramic. The crystal data drop sharply near the transition, and disappear below our sensitivity of ≈ 400 $\mu\Omega$, 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 $Z_s=R_n\{2i/[1+i\delta_n^2/2\lambda(T)^2]\}^{1/2}$. This approximation, which ignores quasiparticles, is only valid near T_c. R_n and δ_n 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, $\delta_n=2.5$ μm (from R_n using classical-skin-depth analysis). Theory and experiment are consistent, and Fig. 5 demonstrates[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 $YBa_2Cu_3O_y$.

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,[21] and vastly different electrodynamic parameters:[3] $\lambda_{ab}(0)\approx 7.5$ μm, $R_s(4.2\ \mathrm{K})=5$ 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_{c1}^* is approximately 3–22 G (i.e., $\ll 250$ G). We have shown elsewhere[4] that the electrodynamics of polycrystalline samples can be well described by a Josephson-junction network, with typical junction area 2 to 4 $\mu\mathrm{m}^2$. Nominal single crystals of moderate quality exhibit electrodynamic parameters[3] of intermediate values, e.g., $\lambda_{ab}(0) \sim 2000$ to 8000 Å, $k(0) \sim 10^{-2}$ to 1 Å/G^2, and $H_{c1}(0) < 250$ G. In contrast, the electrodynamic parameters reported here are the lowest $\lambda_{ab}(0)$, $k(0)$, and $R_s(T)$, 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 $YBa_2Cu_3O_y$, 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 $\delta\lambda_{ab}(T)$ and R_s 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_s and the λ_{ab} experiments is the best achievable to date, substantial improvements in the state of the art are required to address this aspect.

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