Measurement of the $ab$ Plane Anisotropy of Microwave Surface Impedance of Untwinned YBa$_2$Cu$_3$O$_{6.95}$ Single Crystals

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The temperature dependence of the surface resistance $R_s$ and the penetration depth $\lambda$ has been measured in both $a$ and $b$ directions in YBa$_2$Cu$_3$O$_{6.95}$. The overall temperature dependences are similar and show a linear behavior at low $T$. This argues against the chains playing a direct role in the unconventional behavior of the surface impedance. The residual $R_s$ of these twin-free crystals is very small, implying a residual conductivity of the order of the minimum value predicted by Lee [Phys. Rev. Lett. 71, 1887 (1993)] for a $d$-wave superconductor.

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Microwave surface impedance measurements have recently been playing an important role in exploring the nature of the superconducting state in high-$T_c$ superconductors. The imaginary part of the surface impedance, the surface reactance, is directly related to a magnetic penetration depth $\lambda$, which provides a measure of the superfluid density. Its temperature dependence $\Delta \lambda(T)$ reflects the quasiparticle density of states available for thermal excitations and therefore probes the gap structure of the superconducting state. High quality Y-Ba-Cu-O (YBCO) single crystals have a linear $\Delta \lambda(T)$ below 25 K [1] that strongly suggests a pairing state with nodes in the gap. Zinc doped crystals [2,3] and many thin films [4,5] have a $T^2$ temperature dependence, a sign of gapless behavior induced by pair-breaking defects. The real part of surface impedance, the surface resistance $R_s$, provides information about the real part of the conductivity $\sigma_{11}$, which is sensitive to the scattering rate of the thermally excited quasiparticles as well as their density of states. The $R_s$ of high quality YBCO single crystals has a broad peak below $T_c$ that we have attributed to a rapid drop in the quasiparticle scattering rate [6,7]. Addition of Ni and Zn impurities limits the rapid drop in the quasiparticle scattering rate, and we have observed that this results in a suppression of the broad peak in $R_s$ [8–10], leaving a lower, monotonically decreasing loss that is similar to that observed in thin films [11,12]. Clearly, impurities and defects play a large role in the behavior of $\lambda(T)$ and $R_s(T)$ in this material.

The presence of the CuO chain layers in YBCO raises serious questions about the origin of the abundant low energy excitations seen in $R_s(T)$ and $\Delta \lambda(T)$. First, the orthorhombic distortion associated with the presence of chains gives rise to twinning in most crystals. Since all microwave measurements up to now have been performed on twinned samples, there is the possibility that weak links at the twin boundaries seriously affect the surface impedance [13]. The second concern is that the chains might play a key role in the qualitative behavior of $\Delta \lambda(T)$ and $R_s(T)$ [12,14]. With these points in mind and the additional motivation of the already well-known anisotropy of resistivity [15] and thermal conductivity [16], we have undertaken microwave measurements of twin-free crystals.

Cavity perturbation techniques were used to obtain the real and imaginary parts of the surface impedance of the crystals. The imaginary part, which gives $\Delta \lambda(T)$, was measured in a split-ring type resonator described elsewhere [17]. The surface resistance was measured by placing the sample at the center of a cylindrical Pb:Sn plated superconducting cavity, which resonates at 34.8 GHz (TE$_{01}$ mode). The surface resistance of a sample can be obtained through

$$R_s = A \left( \frac{1}{Q_s} - \frac{1}{Q_i} \right),$$

where $Q_s$ and $Q_i$ are the quality factors of the resonator with and without the sample, respectively. The coefficient $A$ is a function of the sample position in the cavity and is obtained from a measurement with the sample in the normal state, where $R_s$ is readily calculated from the dc resistivity. We found the variation of $A$ to be within 2% for the sample in a range about 0.75 mm along the axis of the cylindrical cavity, which ensures that the motion due to the thermal expansion of the sample probe during a temperature ramp is not a problem. Nevertheless, the sample motion precludes accurate measurement of $\Delta \lambda(T)$ in this geometry at 35 GHz. To correct any systematic error caused by the rearrangement of the field patterns due to the introduction of the sample, a Pb:Sn sample cut to the size of the crystal was also measured. Taking the difference between $\Delta (1/Q)$ for the YBCO and $\Delta (1/Q)$ for the Pb:Sn reference removes the systematic error and yields the difference between the $R_s$ of YBCO and Pb:Sn. The final correction then is to add in the $R_s$ of the Pb:Sn alloy. This $R_s$ can be determined from the $Q_0$ of the Pb:Sn cavity itself. At 1.3 K, the $Q_0$ of a freshly plated cavity is about $40 \times 10^6$ which corresponds to $R_s = 35 \mu \Omega$ for Pb:Sn at this temperature. This is a small correction
relative to the ±50 μΩ scatter in the data. For measurement of the surface resistance in the $b$ direction, as an example, the sample is oriented with the $a$ axis parallel to the $H$ field of the TE$_{011}$ mode. In this case the current runs along the $b$ direction in the $ab$ plane and along the $c$ direction in the $ac$ plane. The total loss is then proportional to $J_x^2[\alpha R_{sb} + c R_{sc}]$, where $J_x$ is the surface current density and $a$, $b$, and $c$ are dimensions of the sample in the $a$, $b$, and $c$ directions, respectively. $R_{si}$ ($i = a, b, \text{or } c$) is the surface resistance with current running along the $i$ direction. We estimated $R_{sc}$ in the superconducting state by analyzing the difference in the loss due to current running in the $ab$ plane in one measurement and running along both the $a$ or $b$ and $c$ directions in another measurement. $R_{sc}$ obtained by this method is of the same order as that in the $a$ or $b$ direction in the superconducting state. This results from a much smaller $\sigma_1$ in the $c$ direction being compensated by a larger $c$-axis penetration depth ($R_c \propto A^3 \sigma_1$ in these materials). Since the samples in these studies have $a$ and $b \gg c$, in the measurement configuration described above one has $b R_{sb} \gg c R_{sc}$. The total loss is thus proportional to $R_{sb}$.

The crystals were grown by a flux method discussed in Ref. [18]. The critical temperature of these crystals is about 93.4 K, and the transition width in magnetization and specific heat measurements is less than 0.25 K. The surface impedance measurements reported here were made on two untwinned samples. Sample 1 is naturally untwinned and has a size $1.2 \times 1.2 \times 0.01$ mm$^3$. Sample 2 was mechanically detwinned and has a size $1.1 \times 1.0 \times 0.035$ mm$^3$. The absence of twins was verified with a polarizing optical microscope. The ratio of the length to the thickness of sample 1 exceeds 100, which ensures that the contamination by $R_{sc}$ can be ignored for the measurements in the superconducting state.

Figure 1 shows $\Delta \lambda$ and $\lambda^2(0)/\lambda^2(T)$ for the $a$ and $b$ directions in sample 1 from 1.3 to 100 K. Below 15 K, the $\Delta \lambda$'s are quite linear in $T$; the slope for $\Delta \lambda_a$ is about 4.7 Å/K and that for $\Delta \lambda_b$ is about 3.6 Å/K. The average is 4.2 Å/K, consistent with the value of 4.3 Å/K reported earlier for twinned crystals [1] (for twinned crystals the observed $\Delta \lambda$ is a simple arithmetic average of the two values). In order to plot $\lambda^2(0)/\lambda^2(T)$, we have used $\lambda(0)$ data obtained on one of our untwinned crystals by infrared techniques [19], with $\lambda_a(0) = 1600$ Å and $\lambda_b(0) = 1030$ Å. The general features of the superfluid fraction, $n_s(T)/n_s(0) = \lambda^2(0)/\lambda^2(T)$, are the same, despite a striking anisotropy in $\lambda(0)$. Quantitatively, however, $n_s(T)/n_s(0)$ is not the same in the two directions. This requires that the distribution of the low-lying states be anisotropic with respect to the two directions, which is not unexpected given the orthorhombic symmetry of the crystal.

Figure 2 shows $R_x$ of samples 1 and 2 on a logarithmic scale. For sample 1, when scaled to 10 GHz (using $R_x \propto \omega^2$, where $\omega$ is the microwave frequency), $R_{sb}$ at 77 K corresponds to 125 μΩ and the average of $R_{sa}$ and $R_{sb}$ corresponds to 140 μΩ. These values are substantially lower than all reported surface resistance values for YBCO material at 77 K, as far as we know. We note that in the normal state $R_x$ of sample 2 (the thicker one) is slightly higher due to contamination by $R_{sc}$. In the superconducting state, it also has a higher residual $R_x$, perhaps indicating a slightly lower quality. For these reasons, the rest of the analysis will concentrate on sample 1. In the inset we show the surface resistance of sample 1 plotted to emphasize the normal state surface resistance. It can be seen that $R_{sa}$ is about 1.5 to 1.6 times as large as $R_{sb}$ at 121 K. This corresponds to a ratio of 2.4 for the resistivities in the $a$ and $b$ directions, which

![FIG. 1. The change of penetration depth from 1.3 K, $\Delta \lambda(T) = \lambda(T) - \lambda(1.3 \text{ K})$ and $\lambda^2(0)/\lambda^2(T)$ of sample 1 in the $a$ and $b$ directions, using $\lambda_a(0) = 1600$ Å and $\lambda_b(0) = 1030$ Å from infrared data on sample 2.](image)

![FIG. 2. $R_x$ curves of two untwinned YBCO single crystals, samples 1 and 2 on a logarithmic scale. $R_x$ of sample 2 matches that of sample 1 at low temperatures but is higher in the normal state, due to contamination by $R_{sc}$. The error is ±50 μΩ below $T_c$ and ±5 mΩ above $T_c$. $R_{sa}$ and $R_{sb}$ of sample 1 in the normal state are displayed in the inset.](image)
agrees with the highest anisotropies reported so far [15,20] for the dc resistivities.

In Fig. 3, $R_s$ is shown on a linear scale in order to highlight the low temperature behavior. $R_{sa}$ is seen to have a very prominent broad peak at about 48 K, whereas the peak for $R_{sb}$ is much smaller. Below 35 K, both $R_{sa}$ and $R_{sb}$ are very linear, with the $R_{sa}$ curve about 1.8 times steeper than that of $R_{sb}$. At 1.5 K $R_{sa}$ and $R_{sb}$ are about 100 $\pm$ 50 and 60 $\pm$ 50 $\mu\Omega$, respectively. The $R_s$'s are reaching our resolution limit, but even taking this into account these are the lowest values ever reported for YBCO [11,12]. For comparison, the typical residual $R_s$ of twinned crystals is about 300 $\mu\Omega$ at 34.8 GHz.

For the complex conductivity, except where the temperature is very close to $T_c$, the imaginary part is much larger than the real part. In this case the full expression for the surface resistance simplifies to

$$R_s = \mu \sigma^2 \omega^2 \lambda^2 / 2.$$  

Assuming the penetration depth at 34.8 GHz to be the same as the dc value, the real part of the conductivity in both directions at 34.8 GHz was derived using equation (2). (This assumption will introduce a slight distortion in $\sigma$ in the region to 30 K, due to $\omega \tau$ approaching unity [3]. The maximum error is $\pm 15\%$.)

The result is displayed in Fig. 4. It is interesting to note that the large anisotropy in $\Delta(T)$ has caused $R_{sb} < R_{sa}$, in spite of the fact that $\sigma_{tb} \approx \sigma_{ta}$. At temperatures just below $T_c$, $\sigma_{tb}$ is about 2.4 times as large as $\sigma_{ta}$, similar to the normal state conductivity anisotropy. Both $\sigma_{ta}$ and $\sigma_{tb}$ rise 6 to 7 times from $T_c$ to the peak values around 42 K, showing the rapid drop of quasiparticle scattering rates in both directions. Throughout the entire temperature range, $\sigma_{tb}$ remains about a factor of 2 larger than $\sigma_{ta}$. At low temperatures, both curves are quite linear up to 15 K and start to bend over slightly around 15 K. The extrapolated zero temperature conductivities are very small, less, in fact, than the scatter in our experimental points.

Although these experiments on twin free crystals were initially aimed at studying the $ab$ plane anisotropy of YBa$_2$Cu$_3$O$_{6.95}$, they have also clearly shown that the qualitative features of the surface impedance are not caused by twin boundaries. However, it is clear that much of the residual conductivity previously observed in twinned crystals was caused by the twin boundaries. It has been pointed out that the conductivity of a $d$-wave superconductor approaches a limit $\sigma_{00} = n e^2/m\Delta(0)$ that is independent of the scattering rate [21,22]. If one takes $\hbar/\tau(T_c) \approx 2k_B T_c$ and $\Delta(0) \approx 2k_B T_c$, then $\sigma_{00} \approx 0.3 \sigma_{dc}(T_c)$. If we fit our low temperature conductivities to a straight line we obtain residual conductivities of $\sigma_{ta}(T \rightarrow 0) \approx (0.45 \pm 0.15) \times 10^6 \Omega^{-1} m^{-1} \approx 0.45 \pm 0.15 \sigma_{ta,dc}$ and $\sigma_{tb}(T \rightarrow 0) \approx (0.7 \pm 0.2) \times 10^6 \Omega^{-1} m^{-1} \approx (0.35 \pm 0.10) \sigma_{tb,dc}$. The residual conductivity is near our resolution limit and close to the predicted $\sigma_{00}$ for a $d$-wave superconductor.

A number of issues are addressed by the observed anisotropies of $\Delta^2(0)/\lambda^2(T)$ and $\sigma_1(T)$. First, both the penetration depth and the conductivity at low temperatures are nearly linear with $T$ in both directions, which indicates that the chains are not likely to be the sole source of the low-lying states responsible for the linear $T$ dependence. Also, the broad peak in $\sigma_1$, which has been attributed to a rapid rise in the quasiparticle lifetime $\tau$ below $T_c$, is very similar in the two directions. All of the qualitative features of the electrodynamics are the same in the $a$ and $b$ directions, it is the magnitude that differs. In fact, the differences in magnitude can largely be subsumed under an anisotropy in $n/m^*$.

![FIG. 3. The surface resistance of sample 1 in the superconducting state. The error is $\pm 50 \mu\Omega$ as indicated by the scatter in the data. $R_{sa}$ is 80% larger than $R_{sb}$ at the low temperatures and has a more prominent peak. The residual $R_{sa}$ and $R_{sb}$ at 0 K are much less than that of twinned samples.](image)

![FIG. 4. The real part of conductivity at 34.8 GHz. Note that it is the large anisotropy in $\lambda$ that causes $R_{sb} < R_{sa}$, in spite of the fact that $\sigma_{tb} > \sigma_{ta}$.](image)
a large anisotropy explains most of the difference in the magnitudes of $\sigma_b$ and $\sigma_a$ below $T_c$ and is also consistent with the overall size of the dc resistivity anisotropy above $T_c$. Somehow the presence of the chains leads to a large anisotropy in $n/m^*$ without affecting any of the qualitative features below $T_c$. One transport feature that does not fit trivially into this scenario is the observation of a nonlinear temperature dependence in $\rho_b$, in the chain direction [20]. Resolving the apparently simple behavior below $T_c$ with details of the $ab$ anisotropy above $T_c$ may be addressed by further experiments on samples with different levels of chain vacancies.

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