Measurement of the *ab* Plane Anisotropy of Microwave Surface Impedance of Untwinned $YBa_2Cu_3O_{6.95}$ Single Crystals

Kuan Zhang,[1] D. A. Bonn,[1] S. Kamal,[1] Ruixing Liang,[1] D. J. Baar,[1] W. N. Hardy,[1] D. Basov,[2] and T. Timusk[2]
[1]*Department of Physics, University of British Columbia, Vancouver, British Columbia, Canada V6T 1Z1*
[2]*Department of Physics and Astronomy, McMaster University, Hamilton, Ontario, Canada L8S 4M1*
(Received 4 April 1994)

The temperature dependence of the surface resistance R_s and the penetration depth λ has been measured in both a and b directions in $Yba_2Cu_3O_{6.95}$. 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.

PACS numbers: 74.25.Fy, 74.25.Nf, 74.72.Bk

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 the magnetic penetration depth λ, 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 σ_1, 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_{011} mode). The surface resistance of a sample can be obtained through

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

where Q_s and Q_u 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 40×10^6 which corresponds to $R_s = 35\ \mu\Omega$ for Pb:Sn at this temperature. This is a small correction

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relative to the $\pm 50\ \mu\Omega$ 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 $J_s^2 a[bR_{sb} + cR_{sc}]$, where J_s 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} ($i = a, b$, 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 σ_1 in the c direction being compensated by a larger c-axis penetration depth ($R_s \propto \lambda^3\sigma_1$ in these materials). Since the samples in these studies have a and $b \gg c$, in the measurement configuration described above one has $bR_{sb} \gg cR_{sc}$. 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_s of samples 1 and 2 on a logarithmic scale. For sample 1, when scaled to 10 GHz (using $R_s \propto \omega^2$, where ω is the microwave frequency), R_{sb}

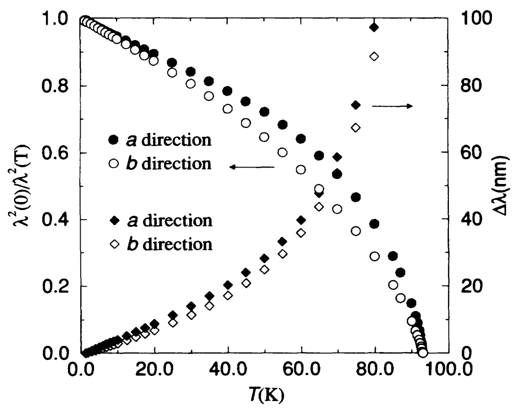

FIG. 1. The change of penetration depth from 1.3 K, $\Delta\lambda(T) = \lambda(T) - \lambda(1.3\ K)$ and $\lambda^2(0)/\lambda^2(T)$ of sample 1 in the a and b directions, using $\lambda_a(0) = 1600$ Å and $\lambda_b(0) = 1030$ Å from infrared data on sample 2.

at 77 K corresponds to 125 $\mu\Omega$ and the average of R_{sa} and R_{sb} corresponds to 140 $\mu\Omega$. 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_s 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_s, 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

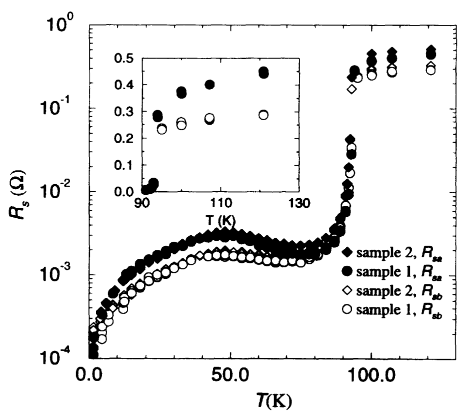

FIG. 2. R_s curves of two untwinned YBCO single crystals, samples 1 and 2 on a logarithmic scale. R_s of sample 2 matches that of sample 1 at low temperatures but is higher in the normal state, due to contamination by R_{sc}. The error is $\pm 50\mu\Omega$ below T_c and ± 5 mΩ above T_c. R_{sa} and R_{sb} of sample 1 in the normal state are displayed in the inset.

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 ± 50 and 60 ± 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

$$R_s = \mu_0^2 \sigma_1 \omega^2 \lambda^3 / 2 \,. \tag{2}$$

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 σ_1 in the region 10 to 30 K, due to $\omega\tau$ approaching unity [3]. The maximum error is $\leq 15\%$.) The result is displayed in Fig. 4. It is interesting to note that the large anisotropy in $\lambda(T)$ has caused $R_{sb} < R_{sa}$, in spite of the fact that $\sigma_{1b} > \sigma_{1a}$. At temperatures just below T_c, σ_{1b} is about 2.4 times as large as σ_{1a}, similar to the normal state conductivity anisotropy. Both σ_{1a} and σ_{1b} 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, σ_{1b} remains about a factor of 2 larger than σ_{1a}. 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.

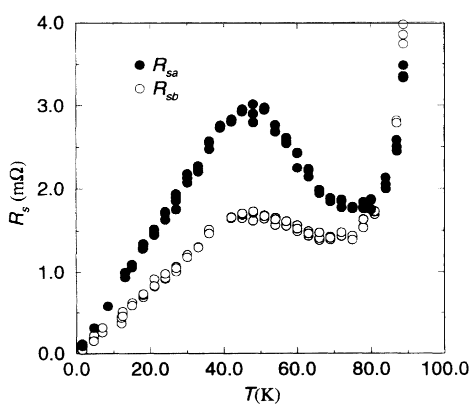

FIG. 3. The surface resistance of sample 1 in the superconducting state. The error is ± 50 $\mu\Omega$ as indicated by the scatter in the data. R_{sa} is 80% larger than R_{sb} at the low temperatures and has a more prominent peak. The residual R_{sa} and R_{sb} at 0 K are much less than that of twinned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

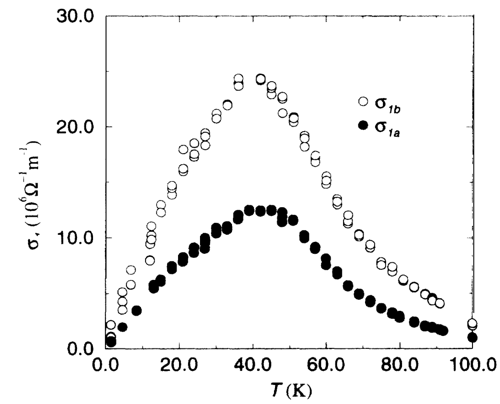

FIG. 4. The real part of conductivity at 34.8 GHz. Note that it is the large anisotropy in λ that causes $R_{sb} < R_{sa}$, in spite of the fact that σ_{1b} is larger than σ_{1a}.

Although these experiments on twin free crystals were initially aimed at studying the ab plane anisotropy of $YBa_2Cu_3O_{6.95}$, 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 $\sigma_{00} = ne^2/m\pi\Delta(0)$ that is independent of the scattering rate [21,22]. If one takes $\hbar/\tau(T_c) \approx 2k_BT_c$ and $\Delta(0) \approx 2k_BT_c$, then $\sigma_{00} \approx 0.3\sigma_{\mathrm{dc}}(T_c)$. If we fit our low temperature conductivities to a straight line we obtain residual conductivities of $\sigma_{1a}(T \to 0) \approx (0.45 \pm 0.15) \times 10^6\ \Omega^{-1}\,\mathrm{m}^{-1} \approx 0.45 \pm 0.15\sigma_{1a,\mathrm{dc}}$ and $\sigma_{1b}(T \to 0) \approx (0.7 \pm 0.2) \times 10^6\ \Omega^{-1}\,\mathrm{m}^{-1} \approx (0.35 \pm 0.10)\sigma_{1b,\mathrm{dc}}$. The residual conductivity is near our resolution limit and close to the predicted σ_{00} for a d-wave superconductor.

A number of issues are addressed by the observed anisotropies of $\lambda^2(0)/\lambda^2(T)$ 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 σ_1, which has been attributed to a rapid rise in the quasiparticle lifetime τ 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^*. Since $\lambda^{-2}(0) \propto n/m^*$ for a superconductor in the clean limit, the anisotropy of $\lambda(0)$ implies $(n/m^*)_b/(n/m^*)_a \approx 2.4$ for $YBa_2Cu_3O_{6.95}$. Such a large anisotropy explains most of the difference in the magnitudes of σ_b and σ_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 ρ_{dc} 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.

We would like to thank D.C. Morgan and J. Gan for helpful discussions. This work was supported by the Natural Sciences and Engineering Research Council of Canada and the Canadian Institute for Advanced Research.

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