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Surface Impedance Studies of YBCO

D.A. Bonn[a], S. Kamal[a], A. Bonakdarpour[a], Ruixing Liang[a], W.N. Hardy[a], C.C. Homes[b], D.N. Basov[b], and T. Timusk[b]

[a] Dept. of Physics and Astronomy, Univ. of British Columbia,
6224 Agricultural Rd., Vancouver, B.C. V6T 1Z1, Canada

[b] Dept. of Physics and Astronomy, McMaster Univ.,
1080 Main St. W., Hamilton, ON L8S 4M1, Canada

This paper summarizes recent measurements of the penetration depth and surface resistance of YBCO, with particular emphasis on the asymptotic behaviour at low temperatures and its dependence on hole doping and cation substitution. A complete set of measurements of the penetration depth tensor indicates that $\lambda(T)$ has a predominantly linear temperature dependence in both the a and b directions at low temperatures, but the c-axis penetration depth is much flatter, without a clear linear term. Ni and Ca substitution are found to have little effect on the linear behaviour of $\lambda(T)$, but Zn impurities alter the behaviour; from T linear to T^2 and then to a non-monotonic temperature dependence at a Zn concentration of 1%. The surface resistance in the ab-plane also seems to vary linearly with temperature at low temperatures, except when the sample is doped with Zn. Substitution of Zn for Cu leads to upturns in the surface resistance below 4 K that are seen at 3.8 GHz, but not at 35 GHz, suggesting impurity effects at a very low energy scale.

1. INTRODUCTION

The microwave surface impedance has played an important role in sorting out the nature of the high T_c superconducting state. It has provided crucial information on both the temperature dependence of the superfluid density and on the conductivity of the thermally excited quasiparticles in the superconducting state. Information on the real part of the conductivity and the penetration depth is obtained from the real and imaginary parts of the surface impedance $Z_s = R_s + iX_s$, where the real part is the surface resistance, and the imaginary part is the surface reactance. In the superconducting state the conductivity can be expressed as $\sigma(\omega,T) = \sigma_1(\omega,T) - i/\mu_0\omega\lambda^2(T)$ where the imaginary term is the inductive response of the superfluid and the real term is governed by absorption processses [1]. In the limit of local electrodynamics the surface impedance can be calculated from this using

$$Z_s(\omega) = \left(\frac{i\mu_o\omega}{\sigma_1 - i\sigma_2}\right)^{1/2} \qquad (1)$$

and except very close to T_c, this leads to two simple expressions:

$$R_s = \frac{\mu_o^2}{2}\omega^2\lambda^3(T)\sigma_1(\omega,T) \qquad (2)$$

and

$$X_s(T) = \mu_o\omega\lambda(T). \qquad (3)$$

So, the surface reactance gives a direct experimental measure of the penetration depth, and thus the superfluid density. The surface resistance is somewhat more complicated since it is essentially a measurement of the absorption processes occuring within the depth that the microwave fields penetrate, and thus depends on both $\sigma_1(\omega,T)$ and $\lambda(T)$. After discussing the experimental techniques used to measure these two quantities, they will each be discussed turn.

2. EXPERIMENTAL

All of the crystals studied here were grown by a flux method in yttria stabilized zirconia crucibles [2]. Corrosion of these crucibles leads to impurity levels in the samples of order 0.1 %. Crystals used for the microwave measurements were between 10 and 50 μm thick and were cleaved into rectangular shapes,

typically 1-2 mm^2 in area. The samples were mechanically detwinned and subsequently reannealed in order to set the oxygen doping level.

Since these crystals are too small to perform sensitive cavity perturbation measurements of the surface impedance in the 1-5 GHz range with cylindrical resonators, superconducting split-ring resonators are used in a variety of measurement geometries [3, 4]. For measurements of the penetration depth, the sample is mounted on a sapphire plate and held fixed in the center of a loop-gap resonator, with the microwave magnetic field lying in the ab-plane of the thin crystal. Measurements of shifts in the resonant frequency then provide a direct measure of changes in the penetration depth as the sample temperature is varied. This geometry avoids systematic errors associated with sample motion and large demagnetization factors, allowing for precision measurements of the temperature dependence of the penetration depth relative to its value at a base temperature, usually 1.3 K ($\Delta\lambda(T) = \lambda(T) - \lambda(1.3K)$). The absolute value of $\lambda(0)$ can not usually be determined with this technique, so we instead use far infrared measurements to determine the absolute value of the penetration depth at low temperatures [5, 6]. One complication of this technique is that it mixes in a contribution from the temperature dependence of the c-axis penetration depth. For instance, with the field lying in the b direction in a thin platelet the frequency shift is given by $\Delta f = K[a\Delta\lambda_a + c\Delta\lambda_c]$ where λ_a is the penetration depth for currents running in the a direction, λ_c is the penetration depth for currents running in the c direction, and a and c are the crystal's dimensions in the a and c directions, respectively. Although the contribution from λ_c is small for a thin platelet, it is not negligible because λ_c is typically much larger than λ_a. This complication can, however, be exploited to measure λ_c. Cleaving the plate into several thin needles multiplies up the contribution from the c-axis, so that measurements of the frequency shift before and after cleaving allow λ_c and λ_a to be separated.

Measurements of the surface resistance have been performed using cavity perturbation of a split-ring resonator at 3.8 GHz in which the surface resistance is determined from the change in 1/Q of the resonator when the crystal is inserted into it. The usual measurement geometry involves microwave magnetic fields applied perpendicular to the ab-plane, so the measurements are a combination of a and b axis surface resistance, without contamination from the c direction. Even with the high sensitivity of a superconducting split ring resonator, the microwave surface resistance is so low at this frequency that the Q of the resonator typically only changes by 1 to 10 % when the sample is inserted. Measuring these low losses requires repeated insertion and removal of the sample from the cavity at each measurement temperature. Careful attention must be paid to reducing microphonics because slight vibration of the sample inside a high Q resonator leads to rapid modulation of the resonant frequency that can spoil the Q measurement. Also, at the lowest surface resistance levels, nonperturbative effects must be taken into account. When a sample is inserted into the split ring the slight rearrangement of the microwave fields can lead to small changes in the microwave losses in the cavity walls. This effect is accounted for by measuring identically sized samples of Pb, which has a low and well known microwave surface resistance at low temperature and frequency.

3. PENETRATION DEPTH

Fig. 1 shows the measured values of $\Delta\lambda(T) = \lambda(T) - \lambda(1.3K)$ for all three crystal axes and at three different oxygen doping levels. Both $\Delta\lambda_a$ and $\Delta\lambda_b$ have a nearly linear temperature dependence at low temperatures for all three of the doping levels that we have tried. We have found that there is some sample to sample variation in the degree of curvature below 4 K, a problem which might be attributed to the residual impurity level in these samples. The variation from sample to sample does suggest that the curvature at low temperatures is not intrinsic behaviour, but higher purity samples will have to be measured to make certain that it is simply an impurity problem. The very large change in the magnitude of $\Delta\lambda_c$ as a function of doping level reflects the large change in the absolute value of the c-axis penetration depth. Far infrared measurements indicate that $\lambda_c(0)$ increases from 1.1 μm to 6.5 μm upon deoxygenating from $O_{6.95}$ to $O_{6.60}$.

The difference between the behaviour of the penetration depth within the ab plane and along the c-axis is more apparent in the plots of $\lambda^2(0)/\lambda^2(T)$ shown in Fig. 2. $\lambda^2(0)/\lambda^2(T)$ is constructed from the microwave measurements of $\Delta\lambda(T)$ coupled with the far infrared measuremnts of $\lambda(0)$ and provides a relatively direct measure of the superfluid fraction. For all of the oxygen dopings that we have studied; underdoped, optimally doped, and slightly overdoped, the superfluid fraction decreases linearly with increasing temperature in both the a and b directions. When the temperature scale is normalized

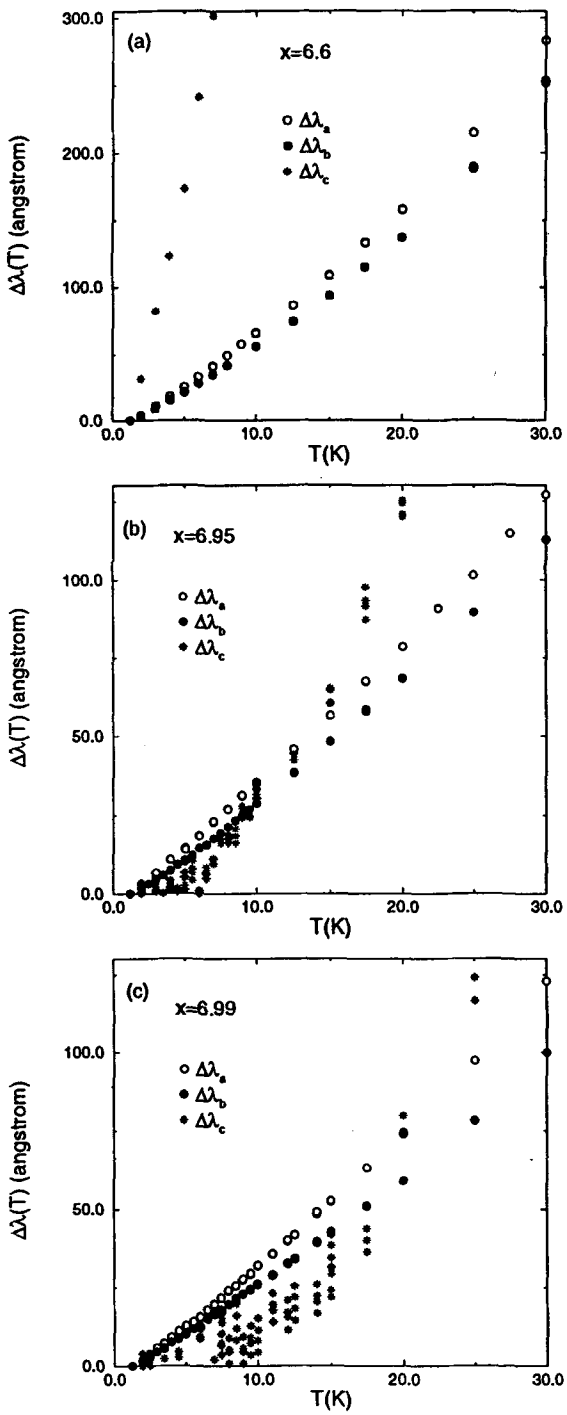

Figure 1: $\Delta\lambda(T) = \lambda(T) - \lambda(1.3K)$ versus T for the a, b, and c directions for three different oxygen doping levels in $YBa_2Cu_3O_x$.

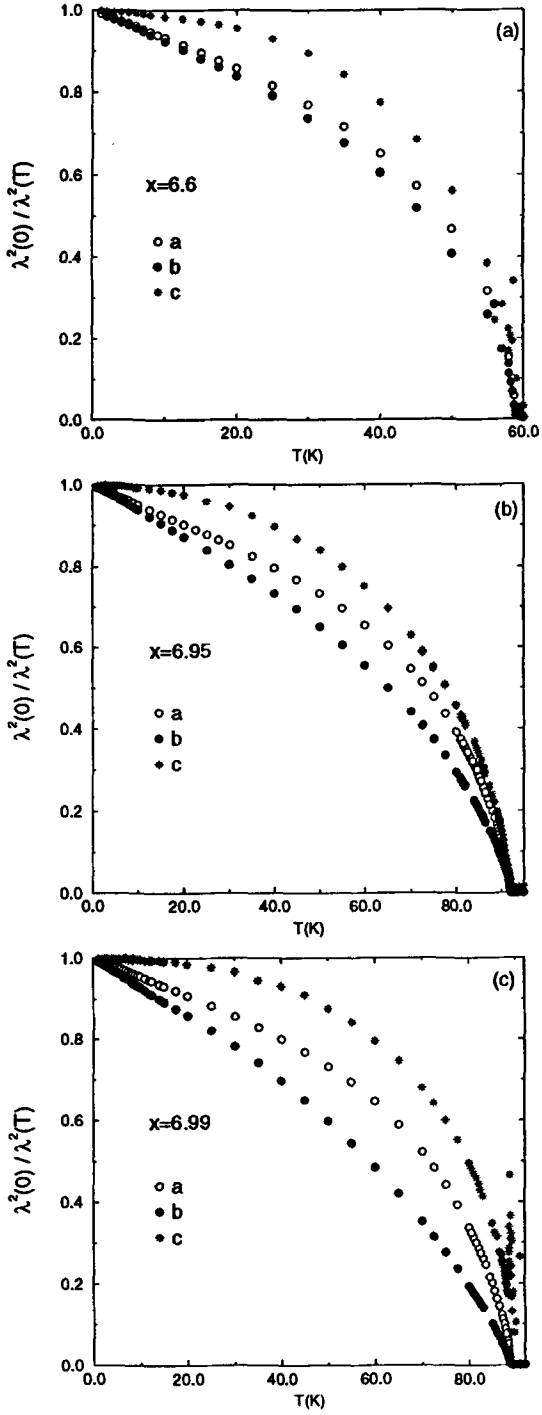

Figure 2: The superfluid fraction, $\lambda^2(0)/\lambda^2(T)$, extracted from the data of Fig. 1 along with infrared determinations of $\lambda(0)$.

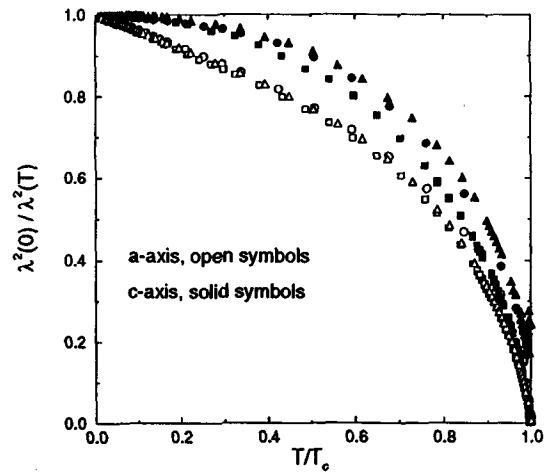

Figure 3: The superfluid fraction versus reduced temperature for three different doping levels of $YBa_2Cu_3O_x$; x=6.60 (circles), x=6.95 (squares), x=6.99 (triangles). In both the a and c directions, the temperature dependence of the superfluid fraction is largely independent of hole doping level in this range.

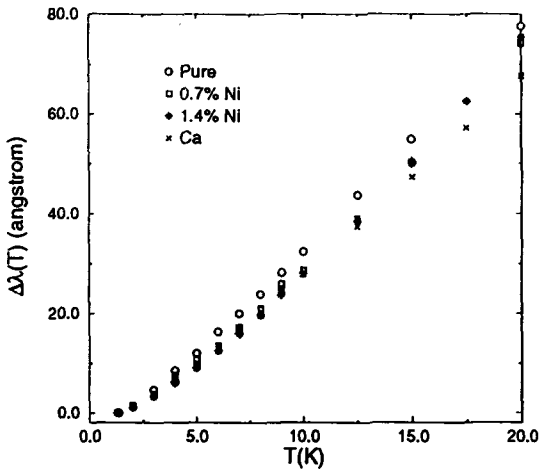

Figure 4: Ni substitution at the 0.75 % and 1.4 % level and Ca substitution at the 2 % level do not significantly affect the temperature dependence of the penetration depth.

by T_c the overall temperature dependence in the a direction (currents perpendicular to the CuO chains) is remarkably independent of doping, as is shown in Fig. 3. In the b direction the superfluid fraction also varies linearly with temperature at low temperatures, but its slope evolves as a function of doping, being quite close to the a-axis curve in $O_{6.60}$, and somewhat steeper at higher hole doping levels.

The c-axis superfluid fraction is quite different, with a very flat temperature dependence at low temperatures and no clear sign of linear temperature dependence. It is perhaps even more surprising that the overall behaviour of the c-axis penetration depth shown in Fig. 3 does not seem to change much with the oxygen doping level, even though $\lambda_c(0)$ changes by a factor of 6 through this doping range [5].

The effects of various cation substitutions on the temperature dependence of the penetration depth have been extensively studied, particularly Ni and Zn which substitute for Cu in the YBCO structure. Fig. 4 shows that Ni impurities, which can occupy both the plane and chain Cu sites, have very little effect on the temperature dependence of the penetration depth in the ab-plane. The variation seen in Fig. 4 is within the variation seen from sample to sample in our nominally pure crystals. This result contrasts interestingly with studies that indicate that somewhat higher levels of Ni impurities have a very large effect on the magnitude of the penetration depth [7]. Ca impurities, which occupy the Y site lying between the CuO_2 bilayers, also seem to have very little effect on the temperature dependence of the penetration depth. Also, if one regards oxygen vacancies in the CuO chains as being a defect, then the oxygen doping results shown in Fig. 1 indicate that oxygen vacancies in the chains do not strongly affect the temperature dependence of the penetration depth in the ab plane.

Zn substitution for Cu in the CuO_2 planes is the only defect that we have found so far that does have a strong effect on the temperature dependence of the penetration depth. Early studies of the effect of Zn indicated that the penetration depth at low temperatures changed from linear in T to T^2 with a substitution of only 0.3 % [8, 9] and this change in the power law has been attributed to the effects of a unitary scatterer in a d-wave superconducting state [10]. Within this model of a dirty d-wave superconductor, it has been suggested that the difference between Ni and Zn substitution might result from Zn acting as a unitary scatterer, but Ni acting as a weaker, Born scatterer. The T^2 behaviour that is also seen in many thin films might then also be attributed to some strongly scattering defect, but the identity of this defect, or defects, has not been determined.

Fig. 5 shows this evolution towards quadratic temperature dependence upon going from a nomi-

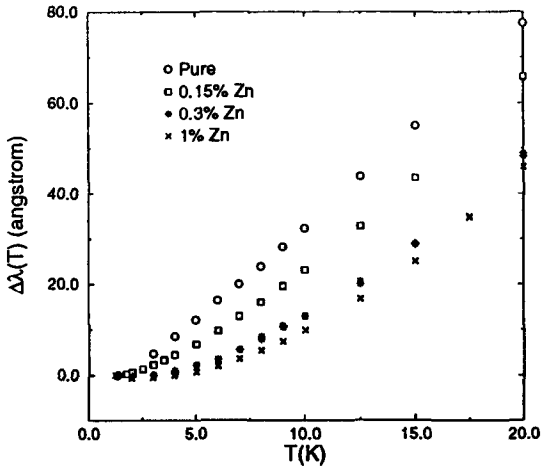

Figure 5: Zn substitution for Cu changes the linear temperature dependence of the penetration depth to a quadratic one and then to non-monotonic temperature depedence at higher impurity concentrations.

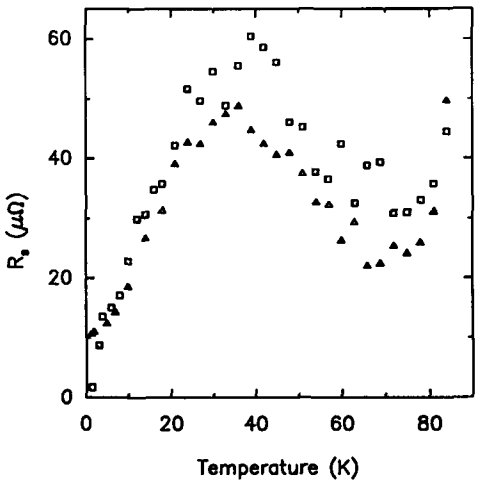

Figure 6: The surface resistance of two different samples of $YBa_2Cu_3O_{6.95}$ at 3.8 GHz. At low temperatures and frequencies the surface resistance varies roughly linearly with temperature, but with considerable sample variation in the curvature and residual loss at low temperatures.

nally pure $YBa_2Cu_3O_{6.95}$ crystal, through 0.15 % Zn substituted, to a crystal with 0.31 % Zn substitution. More recent measurements of samples with 1 % substitution of Zn for Cu complicate this picture. At this higher impurity level the penetration depth develops a weak, non-monotonic temperature dependence; $\Delta\lambda(T)$ starts to turn up again as temperature is decreased below 3 K. If the behaviour of $\Delta\lambda(T)$ is interpreted as a measure of normal fluid density, then this result indicates that at the lowest temperatures measured, the superfluid density starts to decrease slightly with decreasing temperature.

4. SURFACE RESISTANCE

The penetration depth provides a rather straightforward measure of the superfluid density since the superfluid density is directly responsible for the measured screening of the microwave fields. Surface resistance is somewhat more complicated since it is essentially the absorption of microwave power within the penetration depth of the fields and thus depends both on the screening and the absorption processes going on within that screening depth. However, $\lambda(T)$ mainly influences the temperature dependence of $R_s(T)$ close to T_c where the rapid drop in the screening length leads to a precipitous drop in surface resistance just below T_c. At lower temperatures the behaviour of $R_s(T)$ is largely governed by the temperature dependence of the conductivity $\sigma_1(\omega, T)$. In particular, the asymptotic behaviour of $R_s(T)$ at low temperatures is essentially the asymptotic behaviour of $\sigma_1(\omega, T)$.

Fig. 6 shows the low loss region of the surface resistance in the superconducting state for two different samples of nominally pure $YBa_2Cu_3O_{6.95}$. The broad peak in $R_s(T)$ for the pure sample has been studied in detail at a number of frequencies and in a large number of samples (see ref. [1] and references cited therein). It reflects a large peak in the temperature dependence of $\sigma_1(\omega, T)$ and it has been found that the height of this peak in the temperature dependence diminishes with increasing frequency; it is substantially smaller at 35 GHz [9] and eventually disappears altogether in the THz frequency range [11]. The picture that has emerged from this is that as the temperature falls below T_c the conductivity spectrum $\sigma_1(\omega)$ becomes sharply peaked at low frequency. In the normal state, the width of the conductivity peak is of order $2k_BT$, lying in the far infrared, but by 30 K the width of the peak is roughly 35 GHz - a collapse in the width of almost a hundredfold. This dramatic change has been interpreted as a collapse in the scattering rate of the thermally excited quasiparticles below T_c [12]. Since this suggests that the inelastic scattering that dominates the normal state transport properties is drastically diminished, then the conductivity in the superconducting state should become very sensitive to scattering by impu-

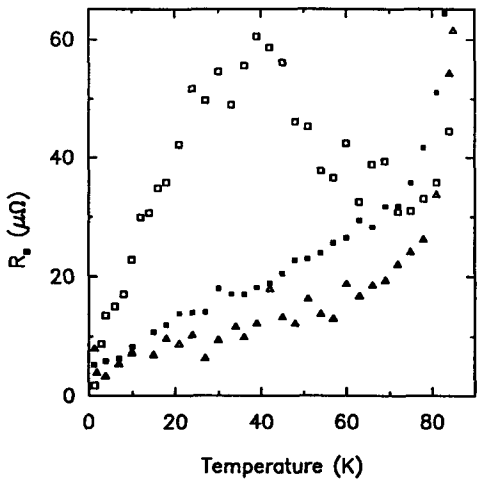

Figure 7: At 0.75 % substitution (triangles) Ni completely suppresses the broad peak in the 3.8 GHz surface resistance and at 1.4 % (solid boxes) the overall surface resistance begin to increase in magnitude, perhaps due to the effect that high Ni concentrations have on the absolute value of λ. For all Ni concentrations studied, the surface resistance varies linearly with temperature at low temperatures.

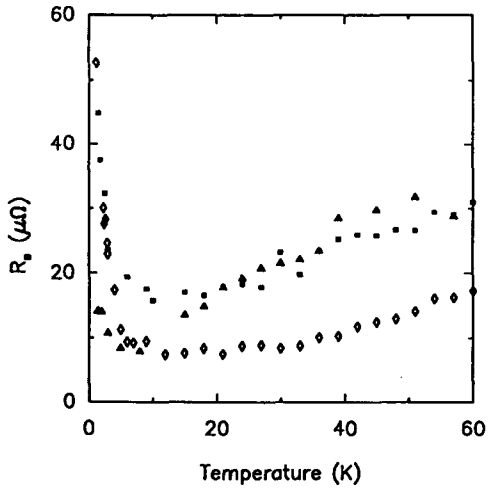

Figure 8: Like Ni impurities, Zn quickly suppresses the peak seen in the surface resistance of pure samples at 3.8 GHz. However, Zn radically changes the temperature dependence at low temperatures, giving rise to an increasingly large upturn in R_s as the impurity concentration goes from 0.15 % (triangles), through 0.31 % (diamonds) to 1 % (solid boxes).

rities. This is clearly illustrated in Fig. 7 where it is shown that 0.75 % substitution of Ni for Cu is sufficient to completely eliminate the broad peak in $R_s(T)$. Conversely, the presence or absence of a peak in $R_s(T)$ can give some measure of the presence of scattering centers in a sample.

The asymptotic low temperature behaviour of the surface resistance has been more problematic, because the losses being measured are very low and because there seems to be a substantial degree of sample variation. Like the penetration depth, the surface resistance seems to be nearly linear below 30 K, with a sample dependent curvature and residual loss at low temperatures. Earlier measurements at 35 GHz also showed a linear temperature dependence, with a much greater signal to noise ratio because of the substantially larger loss at higher frequencies [9, 13]. The measurements at 35 GHz also tended to exhibit somewhat less sample dependence in the curvature and residual loss at low temperatures. However, 35 GHz is not strictly in the low frequency limit, since we have already determined that the conductivity spectrum at low temperatures has a width of order 35 GHz. Thus, measurements at a few GHz are important if one is trying to determine the behaviour in the low frequency, low temperature limit.

Although Fig. 7 indicates that Ni impurities introduce sufficient scattering to suppress the broad peak in the surface resistance, they seem to leave the linear temperature dependence of the surface resistance intact, much as Ni does not seem to affect the temperature dependence of the penetration depth. At 1.4 % Ni substitution, the overall magnitude of the surface resistance is larger than it is at 0.75 %, which may be due to an overall increase in the magnitude of λ, as suggested by the Ni substitution studies of Ulm et al. [7]. A beneficial product of this increase in the overall surface resistance is that the linear temperature dependence is particularly apparent in the sample with 1.4 % Ni substitution.

A study of the effects of Zn substitution on the surface resistance is summarized in Fig. 8. Like Ni, Zn introduces sufficient scattering to suppress the broad peak in the surface resistance. However, Zn substitution has more drastic effects on $R_s(T)$ at low temperatures. Below 4 K, the microwave loss turns upwards, particularly strongly at doping levels of 0.31 and 1 %. This to some extent mirrors the effect that Zn substitution has on the penetration depth. This upturn in the microwave loss is completely absent in the earlier 35 GHz measurements [9], where the surface resistance was found to have a quadratic temperature dependence with no sign of

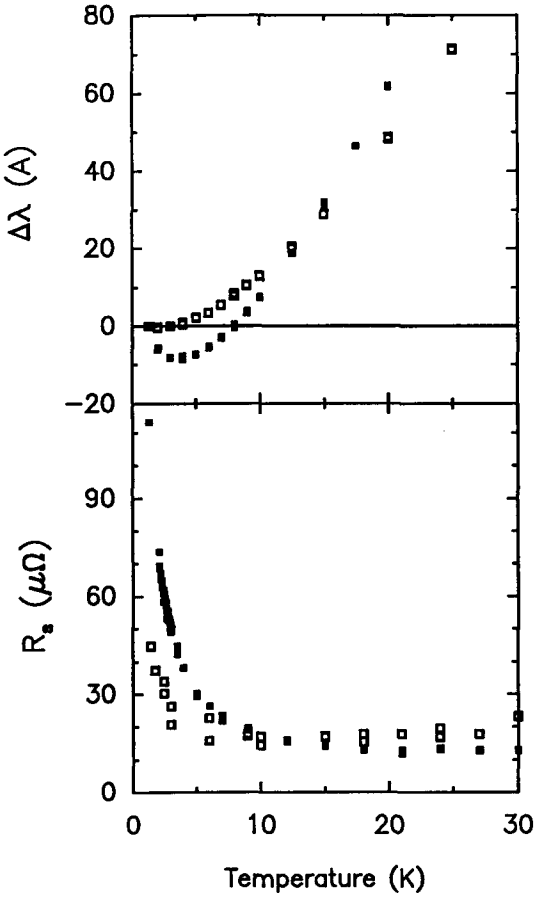

Figure 9: An extreme case of sample variation is shown here for two samples of $YBa_2Cu_3O_{6.95}$ with 0.31 % Zn substitution (open and filled squares are two different samples). The sample with the larger upturn in the 3.8 GHz surface resistance has non-monotonic behaviour in the penetration depth at low temperatures.

non-monotonic behaviour. This indicates that this peculiar behaviour at low temperatures in both the penetration depth and the 3.8 GHz surface resistance involves an absorption process on an energy scale smaller than 1 K. At the present time, there is not sufficient spectroscopic information to say whether or not this feature is peaked at zero frequency or at some finite frequency.

5. DISCUSSION

It has been pointed out throughout this work that there is sample dependent behaviour, particularly below 4 K, in both the penetration depth and the surface resistance. The strong effect that Zn has on the low frequency, low temperature electrodynamics sheds some light on this problem. A particularly severe example of sample dependence is shown in Fig. 9. The two samples used for the surface resistance and penetration depth measurements shown in Fig. 9 were both from the same batch of crystals with 0.31 % Zn substitution and both samples had the same T_c, as observed in the microwave measurements, which indicates that the overall Zn concentration really is the same in both samples. However, the low temperature behaviour is strikingly different. One of the crystals has a much larger upturn in the microwave loss at low temperatures and strong non-monotonic temperature dependence of the penetration depth. The sample with the smaller upturn in the surface resistance has a monotonic penetration depth that varies as T^2. It is not yet clear what the origin of this difference is, but in light of this result it is perhaps not surprising that there is some small variation from sample to sample in our nominally pure crystals.

Despite the caveat about sample dependent curvature below 4 K, all of the results that we have obtained so far in detwinned crystals over a wide range of oxygen doping indicate that the penetration depth has a predominantly linear temperature dependence in both the a and b directions in $YBa_2Cu_3O_x$. In fact, as is shown in Fig. 3, the a axis penetration depth is strikingly insensitive to oxygen content when plotted as a function of reduced temperature. Since the a axis is perpendicular to the CuO chains, this indicates that the penetration depth for the CuO_2 planes has a temperature dependence that is completely insensitive to the large changes in the normal state properties of $YBa_2Cu_3O_x$ that occur when the hole doping is taken from near the optimum value down into the underdoped regime. Whatever factors control the quantitative behaviour of the superfluid density are apparently insensitive to the changes in the degree of c axis coupling and the opening of the normal state pseudogap observed in underdoped samples. It is even more surprising that these changes in the normal state c axis properties are also not accompanied by large changes in the behaviour of the c axis penetration depth. The c axis penetration depth is different from the in-plane penetration depth, but just as for the in-plane properties, its temperature dependence is not strongly affected by the degree of doping.

The linear temperature dependence of the penetration depth has been attributed to a gap function that has lines of nodes [4], suggesting a d-wave superconducting order parameter. Within models of

the effect of impurities on the penetration depth of a d-wave superconductor, the strong effect that Zn substitution has on the temperature dependence of the penetration depth has been attributed to the effects of unitary scattering, with other impurities such as Ni being Born scatterers. This explanation is in accord with other measurements such as NMR studies which indicate that Zn substitution produces a significant density of states at the fermi energy [14]. However, the details of this model of Zn impurities must now also come to terms with the non-monotonic temperature dependence seen at low frequencies in both the penetration depth and surface resistance of samples with relatively high Zn doping.

The ab plane surface resistance of $YBa_2Cu_3O_{6.95}$ also seems to have a nearly linear temperature dependence except when the crystal contains Zn impurities. This temperature dependence, however, is at odds with the present calculations of the microwave conductivity, which predict a quadratic temperature dependence [15]. The surface resistance of those samples (both pure and Ni doped) that exhibit the lowest residual microwave loss does seem to approach the zero temperature limit predicted by P.A. Lee for a d-wave superconductor [16], but this limit has still not been strongly tested because the measurement sensitivity is comparable to the magnitude of the theoretical prediction [1]. The lowest loss samples do typically have the most clearly linear temperature dependence of the surface resistance, so there is no evidence yet of a sample that has surface resistance with quadratic temperature dependence that approaches the zero temperature value expected for a d-wave superconductor. Thus, the low temperature behaviour of the surface resistance persists in being a puzzle that has not been completely solved.

6. ACKNOWLEDGEMENTS

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