New features in the microwave response of YB$_2$Cu$_3$O$_{6.95}$ crystals: Evidence for a multi-component order parameter

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New features are reported in precision measurements of the complex microwave conductivity of high quality YB$_2$Cu$_3$O$_{6.95}$ crystals grown in BaZrO$_3$ crucibles. A third peak in the normal conductivity, $\sigma_1(T)$, at around 80 K, and enhanced pair conductivity $\sigma_2(T)$ below $\sim 65$ K are observed. The data are inconsistent with a single order parameter, and instead are indicative of multi-component superconductivity. Overall, these results point to the presence of multiple pairing interactions in YB$_2$Cu$_3$O$_{6.95}$ and also provide a natural explanation to account for the low temperature 35 K conductivity peak observed in all YB$_2$Cu$_3$O$_{6.95}$ crystals.

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The mechanism for superconductivity and the nature of the superconducting state in the cuprates continue to be prime issues that are being debated [1]. Microwave measurements have yielded important information on the nature of the pairing, the quasiparticle density of states and scattering in the cuprate superconductors [2,3]. Surface impedance of YBCO single crystals in the past have consistently shown two features: A linear penetration depth ($\lambda(T) \propto T$) over a limited low $T$ range, and the presence of a bump in the surface resistance $R_s(T)$ which results in a peak in the normal conductivity $\sigma_1(T)$ at $\sim 35$ K well below the superconducting $T_c \sim 93$ K [4]. The former behavior has been attributed to $d$-wave order parameter symmetry or in general, the presence of nodes in the gap. A rapidly decreasing quasiparticle scattering rate below $T_c$ has been proposed to account for the latter feature [5]. There is a general consensus that the experimental results can be explained in the framework of a single $d$-wave order parameter with inclusion of effects due to strong coupling, scattering with strong temperature dependence, and fluctuations [6].

In this paper, we present new results on the microwave response of high quality YB$_2$Cu$_3$O$_{6.95}$ single crystals grown by a new method which avoids crucible corrosion. We measure the temperature dependent surface impedance $Z_s = R_s + iX_s$ and penetration depth $\lambda$ ($= X_s/\mu_0\omega$) from which we extract the complex conductivity $\sigma_s = \sigma_1 - i\sigma_2$. The two features mentioned above, $\lambda(T) \propto T$ at low $T$ and the 35 K peak in $\sigma_1$ are still present in these crystals. However in addition, a new peak in $\sigma_1(T)$ at $\sim 80$ K and a distinct increase in $\sigma_2(T)$ below $\sim 65$ K are observed. A single $d$-wave order parameter is insufficient to describe the new data, and instead we show that an analysis in terms of two-component superconductivity is necessary.

The single crystals (typically 1.3 $\times$ 1.3 $\times$ 0.1 mm$^3$ in size) used in this work were of very high quality grown from BaZrO$_3$ crucibles (BZO) [8]. This growth method leads to crystals with extremely clean surfaces and exceptional purity exceeding 99.995% [9] in contrast to crystals grown in other crucibles which have final reported purities of 99.5 – 99.95% [10,11]. As shown in this paper, this difference in purity appears to play a significant role in the microwave properties. Standard oxygen annealing procedures were followed to obtain optimally doped crystals with oxygen stoichiometry around $O_{6.95}$ [12]. The crystals have $T_c = 93.4$ K, and very sharp transitions in $R_s(T)$ and SQUID magnetic susceptibility measurements. Crystals grown by this method also have unique physical properties due to their exceptional purity, as revealed in other experiments [13,14]. Unlike the case in earlier crystals, the vortex lattice was imaged for the first time in YBCO with a low temperature STM [13]. Specific heat measurements indicated extremely sharp jumps at $T_c$ [14].

Three crystals of optimally doped YB$_2$Cu$_3$O$_{6.95}$ grown in BZO crucibles (hereafter called YBCO/BZO) were measured. In addition, measurements on a YB$_2$Cu$_3$O$_{6.95}$ crystal (of comparable dimensions) grown in the commonly used yttria-stabilized zirconia (YSZ) crucible (hereafter called YBCO/YSZ) are also presented for comparison.

The high precision microwave measurements were carried out in a 10 GHz Nb cavity using a “hot finger” technique [15]. This method has been extensively validated for precision measurements of the surface impedance in cuprate [16] and other superconductors. All measurements reported in this paper were carried out with the microwave field $H_{rf}$ || c so that currents flow predominantly in the ab planes. Any influence due to $c$-axis properties [4] or finite size effects are minimal as the results obtained on three samples with slightly different dimensions and varying edge geometries were identical.

The $T$ dependence of $\lambda(T)$ of YBCO/BZO and
YBCO/YSZ crystals are shown in Fig.1. The data for the new high purity YBCO/BZO crystals clearly reveal a new high feature - a bump or plateau in the vicinity of 60 – 70K, which has not been reported in any previous λ(T) data for YBa2Cu3O7−δ crystals. A plateau in λ(T) similar to that observed here has been reported in films and was attributed there to a 2-gap behavior.2

The surface resistance Rs(T) is also displayed in Fig.1 as a function of T/Tc. At low temperatures all samples showed the non-monotonic behavior in Rs that is well-known and the low temperature values are comparable to that seen previously.3 However at intermediate temperatures, a new feature is seen - a shoulder in Rs in the YBCO/ZYO sample which is completely absent in the YBCO/YSZ sample. We emphasize that the data for YBCO/YSZ is representative of that reported in literature.4-7

These new features are best studied in terms of the complex conductivity σs = σ1 − iσ2 = iμω/(Rs + iXs)2. The pair conductivity σ2 vs. T is shown in Fig.2 and is a measure of the superfluid density ns(T), since σ2 = ns(T)e2/mω = 1/[μωμωμωλ2](T). Although only changes Δλ(T) = λ(T) − λ(4.2K) and correspondingly ΔXs = μωμωΔλ are measured in the experiment, the absolute value of Xs and hence λ are obtained by assuming the normal state Rn = Xn at and above Tc. This procedure yields a consistent value of λ(0) ≈ 1000Å for all the YBCO/ZYO crystals and a value of λ(0) ≈ 1400Å for the YBCO/YSZ crystal. The σ2(T) data indicates an additional onset of pair conductivity below around 60 – 70K, never observed in previous crystals reported to date as is evident from the comparison with the plot for the YBCO/YSZ crystal. Also the pair conductivity σ2(T) rises to a higher value in the BZO grown crystals compared with the YSZ grown crystals. All the YBCO/ZYO crystals showed this feature in the data. While the accuracy of Δλ ∼ 1Å, the uncertainty in λ(0) is much larger, perhaps of order 100 – 300Å. Nevertheless, the results are consistent with a lower λ(0) and hence enhanced pair conductivity in the YBCO/ZYO crystals compared with the YBCO/YSZ crystals.

The resultant plot of the normal conductivity σ1(T) is shown in Fig.3. The low temperature peak at around 35K (labeled A) is observed in several previous measurements on crystals.2,8,9 Note that very near Tc a sharp peak (labeled C) is also present (see insets to Fig.3) in all YBCO/ZYO and YBCO/YSZ samples and which is often attributed to fluctuations.10 The sharpness of these peaks in the YBCO/ZYO crystals attests to the high quality of these crystals. The most striking feature is a new (third) conductivity peak (labeled B) clearly visible in the YBCO/ZYO data centered at approximately 80K (∼ 0.9Tc), which has not been reported previously in microwave measurements.

It is clear that the YBCO/ZYO crystals reveal two new and important features in the data - (i) the additional enhancement of pair conductivity σ2 with an onset around 60 – 70K, indicative of enhanced pairing below this temperature, and (ii) the new third normal conductivity peak at around 80K (∼ 0.9Tc) in σ1(T). The former was observed in all the YBCO/BZO crystals, while the 80K peak in σ1 is more sensitive to sample details, particularly the normal state scattering, and was lower in one of the samples which had the highest Rs.(Details of results on the other YBa2Cu3O6.95 and also on specially oxygenated YBa2Cu3O7−δ crystals will be published elsewhere).

The data of Fig.1 for the new YBCO/BZO samples is difficult to describe using a single order parameter below Tc. This is evident from the curve for λ−2(T) obtained from a weak-coupling d-wave calculation shown in Fig.4. In the “hydrodynamic” limit, σ1s,d(T) = nqp(T)e2τs(T)/m. A single peak in σ1(T) can arise from a combination of increasing τ and decreasing nqp as T is lowered. Except for detailed dependences on T, this peak should occur for an s- or d-wave superconductor and even in a two-fluid model. (The BCS coherence peak for an s-wave superconductor is masked by this effect). In order to describe the conductivity data in earlier YBa2Cu3O7−δ crystals8-13 in the framework of a d-wave model, one needs a dramatic drop in the scattering rate below Tc. However any model using a single gap or order parameter (s- or d-wave) will only lead to a single conductivity peak, in disagreement with the present data.

Instead it is necessary to consider a two-component system to understand the new data. The new feature in the σ2(T) data suggests that there is additional pairing of carriers below approximately 60 – 70K. The essential features of the data can be well described by a simple model of two superconducting components with Tc,A = 65K and Tc,B = 93K. We have calculated the total conductivity σ = σ1 − iσ2 = (σ1,A + σ1,B) − i(σ2,A + σ2,B). For ease of calculation we used s-wave order parameters for both the A and B components to calculate the conductivities using Mattis-Bardeen expressions. In the case of σ2, calculations assuming s and d which is closer to the realistic case are also presented. Gap values of ΔA(0)kTc,A = 1.5 and ΔB(0)/kTc,B = 2.8 were used. For σ1, temperature dependent scattering times τA,B = τ0A,B/(1 + (T/Tλ,A,B)4), with τ0A/τ0B = 2, Tλ,A = 37K, Tλ,B = 83K were assumed. The calculations reproduce the essential features of the data , viz. the onset of pairing around 60K observed in σ2(T) and the 2 peaks in σ1(T), as shown in Fig.4. Although we have used two decoupled components in the above model for illustrative purposes, a small attractive coupling between the two components is probably essential. It is easy to show in the framework of Ginzburg-Landau theory15-17 that such coupling does not change the results much, and the essential features of the data are retained in more elaborate models.

The microwave measurements do not directly yield the
symmetry of the order parameter in the $A$ and $B$ components, except as can be inferred from the temperature dependence of the data. The fact that \( \lambda(T) \) is linear at low temperatures would indicate that pairing in component $A$ could be d-wave like, exhibiting nodes in the gap. Several theories have suggested the strong possibility of a mixed \( s + d \) state in the presence of orthorhombic distortion as is the case in \( YBCO \), and detailed BCS calculations, such as those in ref. \[21\] of coupled \( s-d \) mixtures do yield penetration depth (\( \lambda^{-2}(T) \) vs. \( T \)) curves similar to the present \( \sigma_2(T) \) data.

It is tempting to assign the two superconducting components $A$ and $B$ with the associated condensates residing on \( Cu-O \) planes and chains respectively in the \( YBCO \) system. However such an interpretation might be inappropriate given the facts that the crystals are twinned and microwaves probe a length scale spanning several hundred unit cells. Instead the two sets of condensates may originate from different types of pairing associated with different regions of the Fermi surface. Such a scenario based on band structure taking into account the chain-plane coupling in \( YBCO \) and eventually leading to two types of pairing interactions has been proposed \[21\].

Our results resolve an important issue regarding the origin of the conductivity peak at 35K. While in previous cases, it was necessary to invoke a precipitous drop in the scattering rate to account for its location, it is natural from Figs. 2 and 3 to associate the low \( T \) (35K) peak with the \( A \)- (\( T_{cA} \approx 60K \)) component and not the \( B \) (\( T_{cB} \approx 93K \)) component. The location at 35K (\( \sim 0.5T_{cA} \)) is now not unreasonable for a conductivity peak associated with onset of pairing at \( T_{cA} \). This implies that the type-\( A \) condensate is present in the \( YBCO/YSZ \) samples also but has a weaker, almost gapless, temperature dependence. We therefore believe that the same mechanisms were operating in earlier samples also but are clearly distinguished in the new \( YBCO/BZO \) crystals.

A natural suspicion that arises is whether the data in the new \( YBCO/BZO \) crystals arise from inadequate oxygen annealing leading to macroscopic chemical phase separation. It is important to note that the annealing procedures for the \( YBCO/BZO \) crystals were exactly the same as for the \( YBCO/YSZ \) crystals \[12\]. An estimate suggests that the data cannot be explained by macroscopic segregation of an \( O_{6.5} \) 60K phase and an ideal \( O_7 \) 90K phase. For \( O_{6.5} \) one would require \( \sim 6 - 10\% \) of the 60K phase which cannot however account for the relative weights of the two components in our \( \sigma_2(T) \) data. Instead, our data points to a physical mechanism like interlayer coupling rather than to chemical segregation for our results.

In conclusion, measurements of the microwave properties of ultra-pure \( YBa_2Cu_3O_{7-\delta} \) crystals reveal new features suggesting the presence of two superconducting components in this compound. Our work provides a possible explanation for the 35K microwave conductivity peak, and yields new insights into the pairing mechanism in the high temperature superconductors.

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FIG. 1. $\lambda(T)$ vs. $T$ for YBCO/BZO (curve 1) and YBCO/YSZ (curve 2) crystals. Data from ref.2 is also shown for comparison (curve 3). Lower inset shows the low T part. Top inset shows $R_s(T)$ vs. $T/T_c$ of YBCO/BZO and YBCO/YSZ.

FIG. 2. Pair conductivity $\sigma_2(T)$ of YBCO/BZO (top) and YBCO/YSZ (bottom).

FIG. 3. $\sigma_1(T)$ of YBCO/BZO (top) and YBCO/YSZ (bottom). The sharp peaks C present in both cases are shown in insets. Note the appearance of a new peak B in YBCO/BZO.

FIG. 4. Calculations of $\sigma_1(T)$ and $\sigma_2(T)$ using the two component model assuming two $s$--wave order parameters (long dashed lines). For $\sigma_2(T)$, the case for a more realistic situation of an $s + d$ symmetry is also shown (solid line). The short dashed line represents the calculations for a single weak-coupling $d$--wave order parameter.