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Observation of a Narrow Superconducting Transition at 6Ghz in Crystals of $YBa_2Cu_3O_7$

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ABSTRACT

We report measurements of the temperature dependence of the surface resistivity of high quality $YBa_2Cu_3O_7$ crystals at $5.95Ghz$. A narrow transition with 89K onset is observed. Resistivity in the normal state is found to be $R_{normal} = 126 \times 10^{-3}\Omega$. We establish an upper limit for the resistivity at $77K$ of $1.0 \times 10^{-3}\Omega$ and at $2K$ of $1.5 \times 10^{-5}\Omega$. The dissipation in four crystals with a total area of about $20mm^2$ is measured to be nearly independent of peak microwave magnetic field over the range of 0.5 gauss to 93 gauss in sharp contrast to the strong field dependence observed in bulk sintered samples.

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Dissipation of microwaves in superconductors is due to the interaction of the oscillating fields with the unpaired electrons. The interaction occurs in a surface layer characterized by the penetration depth λ. The power dissipated is proportional to the number density of unpaired electrons within this surface layer. At zero temperature, the pair binding energy 2Δ corresponds to a frequency of $7 \times 10^{11} Hz$ in lead and niobium. Our measurements are at a frequency of about $6 \times 10^{9} Hz$ and therefore do not involve direct transitions.

The microwave surface resistance R is defined so that

$$P_{loss} = \frac{1}{2} R \int H^2 da$$

where H is the magnetic field tangential to the surface and P_{loss} is the dissipated power.[1]See e. g., J. D. Jackson, Classical Electrodynamics, 2nd edition, Wiley, New York (1975), pp. 338-339. To date, RF measurements for frequencies above $1Ghz$ on bulk ceramic disks with an exposed surface area of about $1cm^2$ indicate a broad superconducting transition and resistivities within an order of magnitude of copper, even at $2K$.[2] [3] [4] Furthermore, the microwave losses in such samples are observed to increase rapidly with the magnitude of the RF magnetic fields. Similar results are obtained for comparable area thin films on $Zr0_2$ and MgO substrates.[4]

In order to determine whether the observed behavior is an intrinsinc property of the compound or due to complications associated with fabrication, we have attempted to characterize the microwave properties of crystals.

The crystal platelets of $YBa_2Cu_3O_7$ were grown from copper-rich eutectic melts contained in ZrO_2 crucibles as described previously .[5] The crystals were annealed in oxygen for more than three weeks. T_c's of similarly grown crystals were measured to be $89K$ using a dc susceptibility (Meissner effect) measurement.

We have measured the temperature and field dependence of the microwave resistivity of the $YBa_2Cu_3O_7$ crystals in a superconducting niobium cavity resonating in the TE_{011} mode at $5.95Ghz$. A sketch of the cavity appears as Fig. 1.

The niobium cavity is operated between $4K$ and $2K$ so that it is superconducting with a high Q of about 10^8. The high T_c samples are placed near the outer edge of a $3.8cm$ diameter sapphire disk thermally anchored to a sapphire rod introduced into the cavity through an axial cutoff tube. The temperature of the sample is thus independent of that of the cavity, and can be heated from the bath temperature to room temperature. The apparatus is described in detail elsewhere.[6] D.L.Rubin, J.Gruschus, J.Kirchgessner, D.Moffat, H.Padamsee, J.Sears, Q.S.Shu, S.Tholen, E.Wilkins, R.Burhman, S.Russek, and T.W.Noh, Third Workshop on RF Superconductivity, Argonne, Illinois, September 14-18, (1987),(to be published). Surface magnetic fields of the TE_{011} mode vanish at the axis of the cavity so placing the crystals off axis yields better sensitivity. Good thermal contact between crystals and sapphire is provided by a small dab of Apiezon N grease. Similarly good conductivity between sapphire rod and the disk is established with the grease. The temperature dependence of the dissipation in the assembly without high T_c crystals ultimately limits the sensitivity of the technique. If no sample is present on the sapphire disk, the cavity Q is about 1.5×10^8 at $2K$. Dissipation in the sapphire increases with temperature and when the temperature of the sapphire is near $100K$ the cavity Q is about half that measured when the sapphire disk is at liquid helium temperature.

The total power dissipated in the cavity is due to the sum of the losses in the niobium cavity walls, sapphire crystal holder, and in the high T_c sample:

$$P_{diss} = \frac{1}{2} R_{Nb} \int\limits_{Nb} H_{Nb}^2 da + \frac{1}{2} R_c \int\limits_{c} H_c^2 da + P_{holder}. \tag{1}$$

Here R_{Nb} and R_c are the microwave resistivities of the niobium walls and crystals respectively. $\int_{Nb} H_{Nb}^2 da$ and $\int_c H_c^2 da$ are the integrals of the magnetic fields over the surfaces of the niobium and crystals. The unloaded Q_0 of the cavity relates power and stored energy according to $P_{diss} = \omega U/Q_0$. (The loaded Q includes the effects of the input and output couplers.) Then (1) can be solved for $1/Q_0$ as follows

$$\frac{1}{Q_0} = R_{Nb}G_{Nb} + R_cG_c + \frac{1}{Q_{holder}(T)}, \tag{2}$$

where $G_{Nb} = \frac{1}{2}\int_{Nb} H_{Nb}^2 da/\omega U$ and $G_c = \frac{1}{2}\int_c H_c^2 da/\omega U$. G_{Nb} and G_c are factors determined strictly by the cavity geometry and sample placement and relate stored energy to surface field. $Q_{holder}(T)$ represents the effect on the cavity Q due to the temperature dependent losses in the sapphire holder which have been measured separately. Sample resistivity R_c is thus simply related to the unloaded Q_0. We determine Q_0 by measuring the ratio of the power incident at one coupler to the power transmitted from the second coupler.

We extract the Q_0 as follows: The cavity Q_0 is related to the ratio of incident power and stored energy per cycle according to :[7]

$$\omega U = \frac{4\beta_1}{(1+\beta_1+\beta_2)} P_1 Q_0, \tag{3}$$

where $\beta_i = Q_0/Q_i$. Q_0 is the unloaded Q, Q_1 is the external Q associated with the loop that couples power (P_1) into the cavity and Q_2 with the loop used to measure the output power (P_2) which is proportional to the stored energy. Since $\omega U = P_2 Q_2$ and by design $\beta_2 \ll 1$ for $Q_0 \lesssim 10^9$ we can write

$$Q_0(T) = \frac{Q_1}{2\left(\frac{Q_1}{Q_2}\frac{P_1(T)}{P_2(T)}\right)^{\frac{1}{2}} - 1}. \tag{4}$$

The measure of P_1 and P_2 as a function of temperature with a Hewlett Packard 8569B spectrum analyzer yields the temperature dependence of the Q_0 with a relative precision of better than 0.5% for $1/Q_0 \sim 10^{-8}$. Q_1 and Q_2 are determined by measuring the loaded Q and the coupling parameters β_1 and β_2 at a fixed temperature. The loaded Q is determined by a measurement of the decay time of the stored energy, and the coupling parameters from a measure of standing wave ratios. Then the unloaded Q_0 is given by[7]

$$Q_0 = Q_{loaded}(1 + \beta_1 + \beta_2).$$

Q_1 and Q_2 are related to Q_0 and the coupling parameters as noted above. Note that Q_1 and Q_2 are characteristic of the couplers and as such are independent of Q_0. In particular $Q_0(T)/\beta_i(T) = Q_i$ and Q_i is temperature independent.

An alternative technique for determining the microwave dissipation in the crystals is by a direct measure of the induced temperature rise. The RF power dissipated in the crystals leads to an elevated temperature with respect to the bath. When the same equilibrium temperature is attained by application of heater power then

$$P_{heater} = \frac{1}{2}\int_c R_c H_c^2 da + P_{holder}.$$

To minimize the power dissipated in the holder the sapphire disk is replaced by a $1.27cm$ diameter niobium disk and the experiment restricted to temperatures below $4.5K$. Without the crystals present we measure first the temperature rise as a function of transmitted microwave power and then as a function of heater power. Below $3.5K$ the temperature rise is proportional to microwave power. We conclude that the dissipation is temperature independent and thus dominated by losses other than in the niobium disk. At higher temperatures the dissipation begins to grow much more rapidly with temperature and at $4.5K$ the losses are consistent with the BCS resistivity.[8]J. P. Turneaure, Proceedings of the Eighth International Conference on High Energy Accelerators, ed. M. H. Blewitt amd N. Vogt-Nilsen (CERN Scientific Information Service, Geneva, Switzerland, 1971, pp. 52-54. The temperature independent background limits the sensitivity of the technique.

The scheme is repeated with the crystals in place on the niobium disk and again we establish the correspondence between heater power and transmitted microwave power. The fraction of the power that is dissipated in the crystals as compared to that dissipated in the niobium disk is readily determined. We have as usual that

$$P_{diss} = \tfrac{1}{2} R_c \int\limits_c H_c^2 da + \tfrac{1}{2} R_{disk} \int\limits_{disk} H_{disk}^2 da + P_{background}.$$

If the temperature of the disk is above the transition temperature of niobium but below the transition temperature of the crystals then the dissipation is dominated by the losses in the disk. In particular at $70K$

$$G_{disk} \simeq \frac{1}{Q_0(70K) R_{disk}(70K)}. \tag{5}$$

Then a measure of the Q_0 at $90K$ and knowledge of the resistivity of niobium and the normal conducting resistivity of the crystals yields:

$$G_c = \left(\frac{1}{Q_0(90)} - \frac{R_{disk}(90)}{R_{disk}(70)} \frac{1}{Q_0(70)} \right) \frac{1}{R_c(90)}, \tag{6}$$

where we neglect background losses which is appropriate when crystals and/or niobium disk are normal conducting. The magnetic field (H^2) is given for a particular value of transmitted power P_2 by

$$\tfrac{1}{2} \int\limits_c H_c^2 da = G_c P_2 Q_2. \tag{7}$$

The data for a $7.3mm^2$ crystal are shown in Fig. 2. The Q_0 of the cavity increases by nearly two orders of magnitude as the crystals are cooled from just above the transition temperature to $75K$. Near transition the cavity losses are clearly dominated by the crystal sample. Therefore at high temperatures the last term in (2), representative of the cavity losses, is negligible and the resistivity of the crystals is simply proportional to $1/Q_0$. The Q_0 measured with the high T_c sample at $75K$ is indistinguishable from that of the bare cavity and sapphire assembly. At $75K$ total dissipation in the crystals is below the level of dissipation in the cavity walls and sapphire assembly and we thus quote an upper limit for sample resistivity.

An absolute calibration of crystal resistivity is achieved by replacing the $YBa_2Cu_3O_7$ with a similarly shaped sample of niobium foil. The measurement of Q as a function of sample temperature is repeated and the niobium data appear together with the $YBa_2Cu_3O_7$ data in Fig. 2. The transition at 9K is clear. For the calibration run, the Q_0 of the cavity was slightly higher presumably due to lower indium joint losses which fluctuate slightly on breaking apart and reassembly. The measured Q with the niobium sample at a temperature of 295K coupled with our knowledge of the resistivity of room temperature niobium at $5.95Ghz$ determines the coefficient G_c in (2). Then the surface resistivity of the crystal above transition (100K) is $0.126 \pm 0.025\Omega$. At 70K, the resistivity of the crystals is less than $1 \times 10^{-3}\Omega$.

Dissipation in a collection of crystals with total area of about $20mm^2$ is indicated in Fig. 3. Data from single platelet and multiple platelets are superimposed and the good correspondence suggests all crystals are of comparable quality. To improve the cavity Q further, magnetic shielding was added during the measurement of four platelets to exclude the earth's field. For this measurement, the overall Q changed by a factor of 250 between normal state and $2K$. While the cavity Q above transition does indeed fall by a factor of two as compared to the single platelet measurement, the Q below transition remains dominated by system losses. We thus reduce the upper limit of the losses representative of these crystals to $5 \times 10^{-4}\Omega$ at $70K$.

An improved upper limit to the crystal resistivity was obtained at about $3K$ using the calorimetric technique. The power dissipated in the holder assembly with and without platelets present is determined as a function of surface field and shown in Fig. 4. Apparently the crystals contribute no significant additional dissipation beyond that observed when the crystals are not present. Note that the ratio of the effective areas of crystals to niobium disk is 0.3. For a difference in dissipated power as indicated by Fig. 4 of less than $20\mu watts$ at $3.5Oe$ we find using $\frac{1}{2}R_c \int_c H_c^2 da \leq P_{diss}$ that $R_c(3K) \leq 15\mu\Omega$ at a frequency of $6Ghz$. We have used measured Q_0's at $70K$ and $90K$ in (6) and (7) to compute $\frac{1}{2}\int_c H_c^2 da$. We assume that the fields are uniform over the area of the crystals.

The temperature dependence of the microwave resistivity can be determined according to the BCS theory of superconductivity. The theory can be solved numerically with the aid of computer programs developed by Turneaure and Weissman[9], and by Halbritter.[10] The resistivity depends on the energy gap, the London penetration depth, coherence length, and mean free path of the material. We plot the theoretical resistivity of niobium along with the measured resistivity of the crystals in Fig. 5. Apparently the relative change in resistivity over the reduced temperature range near transition is greater in the $YBa_2Cu_3O_7$ crystals than in niobium. This may be a consequence of the relatively high resistivity of the normal conducting state of the crystals.

Table 1 compares the $77K$ and liquid He temperature resistance values of these crystals with those of bulk, thin films and earlier batches of crystal samples studied at $6Ghz$.[4] Note that the figures for the new crystals are only upper limits. The resistance of the new crystals is at least 5 times lower than that of copper at $77K$.

Table 1. RF Resistance values for bulk, thin films and crystals of $YBa_2Cu_3O_7$ at $6Ghz$.

Specimen	RF T_C (K)	$R_s(77K)$ $m\Omega$	$R_s(4.2K)$ $m\Omega$
copper	-	5.1	3.8
Pellet #1	87	78	20
Pellet #2	96	10.5	4
Film #1/ZrO_2	86	243	31
Film #2/ZrO_2	90	171	20
Film #3/MgO	$77-87$	200	7
Crystals #1	85	100 [a]	< 0.02 [b]
Crystals #2	85	150 [a]	2 [a]
AT &T Bell Labs Crystals	89	< 1	< 0.015 [c]

[a] Based on an estimate of $R_s(100K) = 200m\Omega$

[b] Separate measurement in an $8.6Ghz$ Nb cavity at $1.5K$

[c] Calorimetric measurement at $6Ghz$ and $3K$

In Fig. 6 is a comparison of the transition in a single piece to that of a larger batch (crystals-2 in Table 1) of crystals reported in reference 4. Finally, in Fig. 7 we compare the temperature dependence of the resistivity of the crystals to that of the best ceramic pellet we have measured.

The Q_0 of the TE_{011} cavity has been measured as a function of the peak RF magnetic field at the crystal surface. The average surface field at the crystals is calibrated against the cavity stored energy above T_c from the knowledge of the microwave resistivity above transition. We see from Fig.2 that if the temperature of the sample is above transition that the cavity losses are dominated by dissipation in the crystals so that $P_{diss} = \frac{1}{2} R_c \int_c H^2 da$. Measurement of the dissipated power then yields the average H over the known area of the crystals. Then changes in the square of the field are proportional to changes in the stored energy and therefore the power monitored from the fixed output coupler.

We observe less than a 10% decrease in cavity Q_0 for fields ranging from 0.5 to 93±5 gauss. For these measurements the crystal temperature is always below about $20K$. Even at the highest field levels the resistivity was less than our measured upper limit at $77K$ for the four platelets of $6 \times 10^{-4}\Omega$. Above the 93 gauss threshold there is an apparent breakdown of the superconducting state. This is in marked contrast to the behavior of a film and pellet studied previously and shown in Fig. 8[4]. Once again the resistance values for the crystals are only upper limits.

The measurements on high quality crystals show that the $6Ghz$ surface resistance of the $YBa_2Cu_3O_7$ compound decreases by at least two orders of magnitude within a few degrees of transition. Calorimetric measurements at liquid helium temperatures yield an upper limit on the surface resistivity nearly four orders of magnitude below the normal state value. The sharp drop in resistivity near T_c resembles the behavior of familiar superconductors, such as niobium.

We have confirmed the previous observation that the resistivity of the crystal $YBa_2Cu_3O_7$ does not have a significant dependence on RF magnetic field and we note that the field dependence is quite strong in the bulk ceramic material. The absence of this effect in crystals supports the interpretation that weak links between grains in polycrystalline material are responsible for field dependence.

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FIGURE CAPTIONS

1. The cylindrical niobium cavity operates in liquid helium and resonates at $5.95Ghz$ in the TE_{011} mode. If no high T_c sample is present, the cavity Q_0 is about 1.5×10^8 at 2K.

2. Surface resistivity of the largest crystal platelet including the effect of losses in the sapphire assembly. Below transition the data represent an upper limit to the surface resistivity. Superimposed are the data for the Nb calibration piece which was 40% larger in area. If no crystals are present the Q_0 of the cavity would correspond to the curve labelled "without crystals".

3. Surface resistivity for a collection of crystals grown by the same procedure. Data from Fig. 2 are superimposed. Note that below about $80K$ losses are dominated by dissipation in the sapphire assembly and the plotted resistivities are upper limits for the crystals.

4. Microwave power dissipated in a collection of 4 crystals placed on top of a $1.27cm$ diameter niobium disk in the $6Ghz$ cavity. Power dissipated without crystals is also shown.

5. The theoretical superconducting microwave resistivity of niobium is computed for $\Delta/kT_c = 1.86$, a zero temperature penetration depth $\lambda_L = 360\mathring{A}$, coherence length $\xi = 640\mathring{A}$, a mean free path $l = 500\mathring{A}$, and $T_c = 9.2K$. Data for the crystal resistivity are also shown. Again note that below about $80K$ losses are dominated by dissipation in the sapphire assembly and the observed temperature dependence is systematic. Comparison with the theory at temperatures below about $80K$ is therefore not meaningful. Resistivities are plotted as a function of reduced temperature T/T_c.

6. Comparison of the transition in high quality crystals to a previous batch of crystals.[4]

7. Comparison of the transition in high quality crystals to the best bulk ceramic pellet measured.

8. Field dependence of $6Ghz$ surface resistance of bulk ceramic, thin film and crystals. The ratio of superconducting to normal state surface resistance is plotted.