Evidence for Rapid Suppression of Quasiparticle Scattering below T_c in $YBa_2Cu_3O_{7-\delta}$

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The microwave surface resistance at 2.95 GHz of a very-high-quality crystal of $YBa_2Cu_3O_7$ exhibits unusual nonmonotonic temperature dependence. After a sharp drop below T_c by a factor of 5000 the loss rises to a peak at 35 K and then falls at lower temperature. The peak is due to a rapid decrease in the scattering of thermally activated quasiparticles below T_c and this suppression of scattering suggests that the excitations responsible for the large resistivity of $YBa_2Cu_3O_7$ are gapped below T_c. A technological implication is that disorder may lower the microwave loss.

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Microwave loss studies of $YBa_2Cu_3O_7$ continue to be of interest for both physical and technological reasons. At a fundamental level the frequency and temperature dependence of the surface resistance (R_s) of a superconductor provides information on the coherence length, mean free path, London penetration depth, and energy gap [1]. Microwave studies of $YBa_2Cu_3O_7$ are technologically important because the potential use of high-T_c superconductors in high-frequency electronics exploits their low microwave loss. Measurements of R_s in high-T_c superconductors are currently centered on studies of thin films of $YBa_2Cu_3O_7$, in part because thin films are the form in which they are being incorporated into devices [2]. There are rather few measurements of R_s for high-quality crystals of $YBa_2Cu_3O_7$ because there are few microwave techniques sensitive enough to measure very low loss in very small samples. Wu *et al.* measured the microwave loss of a 0.5-mm^2 crystal of $YBa_2Cu_3O_7$ using cavity perturbation in a superconducting Pb resonator [3]. However, the small size of good $YBa_2Cu_3O_7$ crystals limits the sensitivity of cavity perturbation in large cylindrical resonators operating in the 1–10-GHz range. Even with a resonator with a quality factor (Q) greater than 10^7 [4] Wu *et al.* could only measure losses down to the 400-$\mu\Omega/\square$ level at 10 GHz.

A cavity perturbation technique using superconducting split-ring resonators has recently been developed that is capable of measuring losses of a few $\mu\Omega/\square$ in the 1.7–5-GHz range, even for samples smaller than 1 mm^2 [5]. The chief advantage of split-ring resonators in this frequency range is that they are more compact than conventional cylindrical resonators and this greatly improves sensitivity to small samples. Since the measurement of R_s by cavity perturbation depends on measuring the change in a resonator's Q when a sample is inserted, the key to measurement of small, low-loss samples is the measurement of small changes in large Q's. Reference [5] gives detailed procedures for measuring Q's in the 10^6 range to $\pm 0.2\%$ which allows measurement of losses as small as a few $\mu\Omega/\square$. The results presented below were obtained with a 2.95-GHz resonator that has been used in an ongoing program aimed at identifying the sources of microwave loss in $YBa_2Cu_3O_7$ crystals. The absolute value of R_s has been determined by assuming that the loss at 93 K is what one would expect for a metal with a dc resistivity of 95 $\mu\Omega$ cm but this choice has no effect on the qualitative features discussed below.

Figure 1 displays the temperature dependence of R_s of a crystal that has the lowest loss that we have yet measured. The crystal was grown using a $BaCuO_2$:CuO flux [6]. The growth technique uses high-purity powders of Y_2O_3, $BaCO_3$, and CuO (99.999%) which are reacted to obtain $BaCuO_2$ and $YBa_2Cu_3O_7$. These powders are mixed with CuO to form a flux with the composition $YBa_9Cu_{39}O_x$. A gold sheet is folded in half to an angle of 10° with 1–2 g of flux placed close to the crease and is

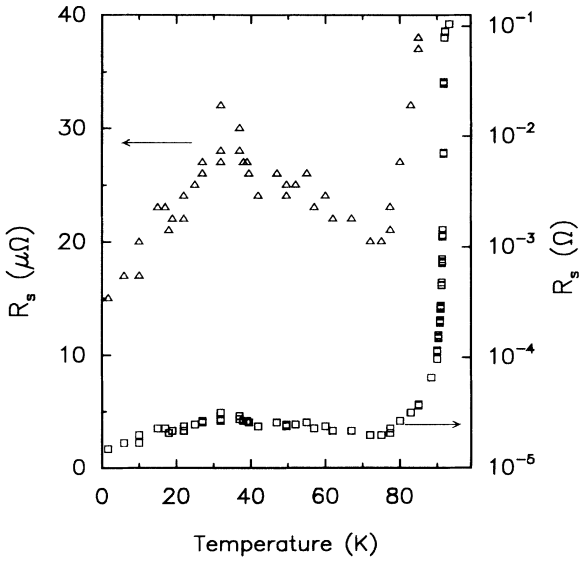

FIG. 1. The surface resistance at 2.95 GHz of a 0.6×0.6-mm^2 crystal of $YBa_2Cu_3O_7$. Below T_c the surface resistance drops much more quickly than BCS theory; nearly 4 orders of magnitude by 80 K. The unusual rise in the loss from 77 to 35 K is interpreted to be an indication that the scattering time of the thermally activated quasiparticles increases rapidly with decreasing temperature.

then rested on zirconia supports. A typical growth involves heating at 200 °C/h to 970 °C in air, holding at 970 °C for 1–3 h, cooling at 30 °C/h to 800 °C, and then turning the furnace off. $YBa_2Cu_3O_7$ crystals are found along the crease in the gold and are subsequently annealed in purified oxygen at 450 °C for 48 h. This growth technique produces free-standing crystals with flux-free surfaces. Although the short growth time produces small crystals (a 0.4-mm^2 sample was used for the data in Fig. 1), they are easy to oxygenate and have only a small amount of time to pick up impurities. Magnetic susceptibility measurements indicate that the crystals grown by this technique are consistently of high quality. With a field of 10 Oe applied parallel to the c axis the transition width was 4 K (10% to 90%) with an onset at 91.5 K.

A surprising feature of the loss depicted in Fig. 1 is the broad peak centered near 35 K which is clearly resolved because the uncertainty in the measurements is only ± 3 $\mu\Omega$ in this low-loss regime. The peak has been observed in two samples grown by the technique outlined above. In order to further verify whether or not this feature is intrinsic, measurements have been made on larger crystals produced by flux growth in zirconia crucibles. Crystals grown by this technique have 92.5-K critical temperatures and the transition width measured by magnetic susceptibility (100-Oe field parallel to c axis) is only 1.8 K. The specific heat of these crystals exhibits a jump below T_c that is only 0.6 K wide which indicates a high degree of homogeneity. During the course of these measurements it was found that spots of flux on the surface were the dominant cause of losses greater than those depicted in Fig. 1. A 1×2-mm^2 crystal with a 50-μm spot of flux on the surface had a loss that was 50% larger than the best crystal. However, once a temperature-independent residual loss was subtracted, this large crystal exhibited the same broad peak at 35 K in the surface resistance. In addition to being grown differently, the latter crystal was twin-free over more than 90% of its surface, whereas the crystal whose loss is shown in Fig. 1 was heavily twinned with a spacing between twin boundaries of the order of 1 μm. Although this comparison does not rule out twin boundaries as a source of residual loss, it does indicate that twinning is not the source of the nonmonotonic temperature dependence of R_s.

The loss at 77.3 K is 22 $\mu\Omega/\Box$ which is equivalent to 250 $\mu\Omega/\Box$ at 10 GHz if R_s varies as the square of the frequency. This loss at liquid-nitrogen temperature is comparable to the lowest losses measured in thin films at 10 GHz and 77 K [7]. A particularly low loss of 2 mΩ at 82 K has been measured in a thin film at 59 GHz [8] but this is too high in frequency to confidently make comparisons to 3-GHz data by assuming the loss scales as frequency squared. The residual loss at 1.7 K is 15 $\mu\Omega/\Box$ which is comparable to the calorimetric measurement of 15 $\mu\Omega/\Box$ made at 3 K and 5.95 GHz by Rubin *et al.* [9].

The peak in R_s is understandable within existing theories of superconductivity if the scattering time of the thermally excited quasiparticles increases rapidly with decreasing temperature. Calculations of the surface resistance based on the theory of Bardeen, Cooper, and Schrieffer (BCS) indicate that in the clean limit R_s increases with increasing scattering time [1]. In a BCS-like calculation of surface resistance in a layered superconductor the loss was also found to increase with increasing scattering time [10]. This unusual result can be seen in a simple two-fluid model. At frequencies much less than the gap frequency the electrodynamics of the two-fluid model can be expressed as a frequency- and temperature-dependent complex conductivity with the simple form

$$\sigma(\omega,T)=\sigma_n(T)+ic^2/4\pi\omega\lambda(T)^2, \tag{1}$$

where the imaginary part models the purely inductive response of the superfluid with a temperature-dependent London penetration depth $\lambda(T)$, and $\sigma_n(T)$ is the predominantly real conductivity of the normal fluid. With this conductivity and local electrodynamics one obtains a microwave loss that is given by

$$R_s=(8\pi^2/c^4)\omega^2\lambda^3(T)\sigma_n(T). \tag{2}$$

If one ignores coherence factors, σ_n can be modeled by the low-frequency limit ($\omega\tau\ll 1$) of the Drude free-electron model, $\sigma_n(T)=\tau(T)n_n(T)e^2/m^*$, where $n_n(T)/m^*$ is the ratio of the normal-fluid density to the effective mass, e is the electron's charge, and $\tau(T)$ is the scattering time of the normal fluid. Substitution of this normal-fluid conductivity into Eq. (2) yields a surface resistance that increases with increasing scattering time and that can exhibit nonmonotonic temperature dependence because of competition between two temperature-dependent quantities, $n_n(T)$ and $\tau(T)$.

$\sigma_n(T)$ can be extracted from $R_s(T)$ by using Eq. (1) and assuming some form for $\lambda(T)$ that is a good representation of established experimental values. Figure 2 depicts the $\sigma_n(T)$ that one obtains by using

$$\lambda(t)=\lambda_0[1-t^{3-t}]^{-1/2}, \tag{3}$$

where t is the reduced temperature T/T_c, and λ_0 is the zero-temperature limit of the penetration depth. This form is a close approximation to BCS theory [11] and fits the mean-field behavior of $\lambda(T)$ that is observed near T_c in magnetization measurements [12]. The real part of the conductivity depicted in Fig. 2 has been generated using a T_c of 91.8 K and a λ_0 of 1250 Å; choices that give a $\sigma_n(T)$ which smoothly approaches the normal-state conductivity of $YBa_2Cu_3O_7$ at T_c. An alternative $\lambda(T)$ that is used to fit some muon-spin-relaxation measurements is the Gorter-Casimir form, where the exponent $3-t$ in Eq. (3) is replaced by 4 [13] and λ_0 used in Eq. (3) is replaced by $\sqrt{2}\times 1250$ Å $=1770$ Å in order to recover the same mean-field behavior near T_c. Although the behavior near T_c is nearly identical, the values of $\sigma_n(T)$ below 85 K are up to a factor of 2 smaller than those obtained

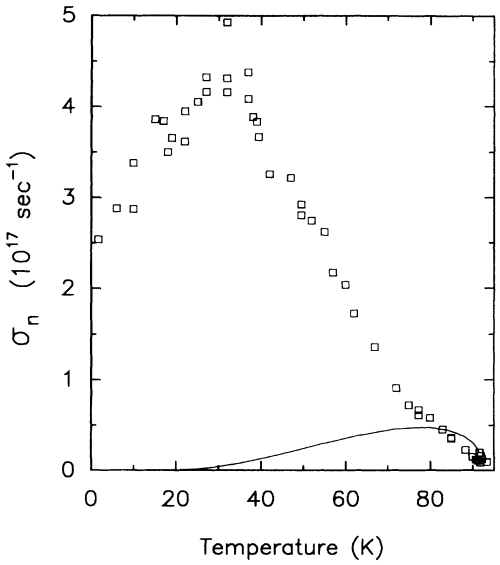

FIG. 2. The real part of the conductivity extracted from the surface resistance of $YBa_2Cu_3O_7$ (squares). The BCS conductivity (solid curve) is calculated using a T_c of 91.8 K, a gap ratio of 3.52, a coherence length of 20 Å, a $\lambda(T=0, 1/\tau=0)$ of 1008 Å, and a mean free path of 25.5 Å. The observed initial rise near T_c is not nearly as abrupt as the onset of the coherence peak in BCS theory.

with Eq. (3).

Along with the measured values of $\sigma_n(T)$, Fig. 2 includes BCS conductivity calculated for a set of parameters appropriate for $YBa_2Cu_3O_7$. The $\lambda(T=0, 1/\tau=0)=1008$ Å is a choice of clean-limit penetration depth that is consistent with the choices of λ_0 used in the two versions of the two-fluid model. It is tempting to associate the peak in $\sigma_n(T)$ below T_c with coherence effects, but in detail it is quite different from a BCS coherence peak. Initially $\sigma_n(T)$ in $YBa_2Cu_3O_7$ rises with a shallower slope than the BCS result. This gives rise to a surface resistance that falls more sharply at T_c than BCS surface resistance; the fact that the observed drop in R_s is *sharper* means that the behavior of $\sigma_n(T)$ cannot be attributed to a transition broadened by inhomogeneity. The relatively slow rise in $\sigma_n(T)$ suggests that coherence effects in $YBa_2Cu_3O_7$ are less apparent than those predicted by BCS theory. On the other hand, the measured $\sigma_n(T)$ for $YBa_2Cu_3O_7$ eventually rises much farther than the BCS result before falling again below 35 K. The nonzero low-temperature limit of σ_n is sample dependent and probably not intrinsic.

It is unlikely that the large peak in $\sigma_n(T)$ can be attributed to temperature dependence of the normal-fluid density. Coherence effects in BCS theory cause a peak in $\sigma_n(T)$ below T_c which one can think of as a rise in normal-fluid density just below T_c followed by an exponential decrease at lower temperatures. However, the related coherence peak in the nuclear spin-lattice relaxation rate is not observed in NMR measurements [14], so an alternate explanation of the peak in $\sigma_n(T)$ is needed. Rather than being a manifestation of coherence it seems likely that the peak is due to two competing temperature dependences; a normal-fluid density that decreases with decreasing temperature and a scattering time that increases. These two temperature dependences cannot be disentangled without a model for the temperature dependence of the normal-fluid density. However, for temperatures not too far below T_c a crude estimate of $1/\tau(T)$ is provided by the quantity $ne^2/m^*\sigma_n(T)$ (see Fig. 3), where n/m^* is the ratio of the total density of charge carriers to their effective mass. To be internally consistent n/m^* is fixed by the low-temperature penetration depth, $n/m^* = c^2/4\pi e^2\lambda_0^2 = 2\times10^{48}$ cm^{-3}g^{-1}. In the absence of coherence effects $ne^2/m^*\sigma_n(T)$ provides an upper limit on $1/\tau(T)$ since one expects the normal-fluid density $n_n(T)$ to fall below n as the temperature decreases.

Even though neglect of the temperature dependence of the normal-fluid density and coherence effects leads to substantial uncertainty in identifying $ne^2/m^*\sigma_n(T)$ with the quasiparticle scattering rate, the drop in this quantity below T_c (see Fig. 3) is so large and rapid that it is clear that $1/\tau(T)$ falls much faster than linearly with temperature. This rapid decrease in the scattering indicates that the strong inelastic scattering process responsible for the linear temperature dependence of the dc resistivity of $YBa_2Cu_3O_7$ is suppressed below T_c. This behavior below T_c would be expected if an energy gap develops in the spectrum of excitations responsible for the scattering above T_c. By positing just such a gapping of the excitation spectrum, marginal-Fermi-liquid theory [15] is able to model experimental results at much higher frequency,

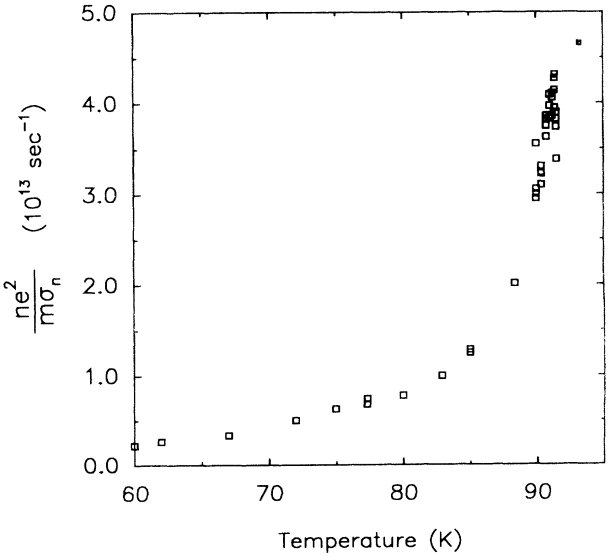

FIG. 3. The quantity $ne^2/m\sigma_n(T)$ provides an estimate of the quasiparticle scattering rate in $YBa_2Cu_3O_7$ for temperatures close to T_c. The open squares are extracted from the microwave data and the solid square at 95 K is derived from the normal-state resistivity.

particularly time domain terahertz spectroscopy [16]. Recent calculations of the scattering within the framework of marginal-Fermi-liquid theory are qualitatively in agreement with the measurements presented here [17]. Whatever the microscopic explanation, a rapidly suppressed scattering rate is a major feature of the behavior of $YBa_2Cu_3O_7$ at the superconducting transition and should be taken into account in any interpretation of the changes in physical properties below T_c.

A technological implication of this result is that the loss below T_c in a pure sample of $YBa_2Cu_3O_7$ is limited by the rapid decrease in $1/\tau(T)$. If one could introduce sufficient disorder to limit how far $1/\tau(T)$ drops, the low-temperature loss might be decreased. In fact, the lowest measured R_s in a thin film in this frequency range falls monotonically with temperature to a lower loss than the crystal studied here [7]. This film had a residual dc resistivity of 22 $\mu\Omega$ cm, about 25% of the resistivity observed above T_c in $YBa_2Cu_3O_7$, whereas, the residual resistivity of our crystals is very close to zero. This suggests that the film contains enough defects to keep $1/\tau(T)$ from falling below 25% of its value at T_c. Such a limit would give rise to a low-temperature $1/\tau(T)$ that does not fall below the value for the clean single crystal. This would eliminate the peak in R_s and reduce the low-temperature loss, as is observed in the film. Thus it seems that a certain amount of disorder may actually improve the low-temperature surface resistance of a superconducting film of $YBa_2Cu_3O_7$.

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