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Electrodynamic response of $Y_1Ba_2Cu_3O_y$ and $La_{1.85}Sr_{0.15}CuO_{4-\delta}$ in the superconducting state

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Measurements of the temperature dependence of the surface resistance R_s at 9.85 GHz and the screening length δ_s at 1 MHz for the superconducting states of $Y_1Ba_2Cu_3O_y$ and $La_{1.85}Sr_{0.15}CuO_4$ show strong correlations, suggesting that the superconducting state is the same in the two materials. While the overall temperature dependence of R_s is reasonably well described by the Bardeen-Cooper-Schrieffer (BCS) theory in the very dirty limit, additional features are seen in R_s and also in δ_s which suggest detailed deviations from the BCS *s*-wave state.

A fundamental question concerning the recently discovered oxide superconductors[1] is what is the nature of the superconducting state, and further, is it the same in different families of new materials? Studies of the electromagnetic response of superconductors can yield important information[2] on the nature of the gap, densities of states, quasiparticle excitations, and the pairing, and also establish correlations between different superconducting materials.

We have studied[3] the superconducting state in $Y_1Ba_2Cu_3O_y$ and $La_{1.85}Sr_{0.15}CuO_{4-\delta}$, via experiments on the microwave surface resistance R_s at 9.85 GHz and the rf screening length δ_s (which is related to the penetration depth λ), at 1 MHz. The dependence of the normalized surface resistance R_s/R_n on reduced temperature $t \equiv T/T_c$ is similar for the two materials, and furthermore new features are found in R_s/R_n in the form of local minima near $t \approx 0.96$ and 0.9. Such features do not arise in the BCS *s*-wave theory of the electromagnetic response of superconductors. However, the underlying temperature dependence of R_s/R_n can be approximated by a simple extension of the BCS theory, by appropriate choice of one adjustable parameter. The local minima also appear in the temperature derivative of $\delta_s(t)$, at the same values of t as in the microwave data; however, $\delta_s(t)$ cannot be fitted to the BCS prediction for λ with any reasonable choice of parameters. Taken together, our results provide strong evidence that the superconduction state is the same in $Y_1Ba_2Cu_3O_y$ and $La_{1.85}Sr_{0.15}CuO_{4-\delta}$, and that while it has important similarities with the BCS *s*-wave state, it differs from the latter in detail.

$Y_1Ba_2Cu_3O_y$ (hereafter referred to as Y-Ba-Cu-O) and $La_{1.85}Sr_{0.15}CuO_4$ (La-Sr-Cu-O) were prepared using now standard techniques.[1] The Y-Ba-Cu-O samples showed onsets of decreasing resistance at 94 K and zero resistance by 92.5 K. Powder x-ray patterns are in excellent agreement with the published results,[4] and show no lines corresponding to any other phase. The La-Sr-Cu-O samples had onset temperatures ranging from 39.5 to 42 K and had 2-K transition widths.

Microwave measurements were carried out[5] using a sapphire-loaded copper cavity resonant in the TE_{011} mode. Figure 1 displays the measured superconducting surface resistance at 9.85 GHz, plotted as $r = R_s(T)/(R_n)$ vs t for Y-Ba-Cu-O and La-Sr-Cu-O, where $R_n \equiv R_s(T_c)$. At low temperatures the accuracy of the measurement degrades as R_s improves in comparison with the copper contribution R_{Cu}, which limits the precision of the measurement. For $t > 0.75$ the relative accuracy of r is about 0.02, so that for the temperature range of Fig. 1, the error bars are invisible.

The most striking features of the data in Fig. 1 are the minimum near $t = 0.9$ and possibly another near $t = 0.96$. It is important to note that these features occur at the same value of *reduced* temperature for both oxide superconductors. This clearly precludes accidental material effects such as mixtures of additional phases since the T_c's differ in absolute magnitude by a factor greater than 2.

We have carried out a comparison of our experimental results with numerical computations based upon the BCS

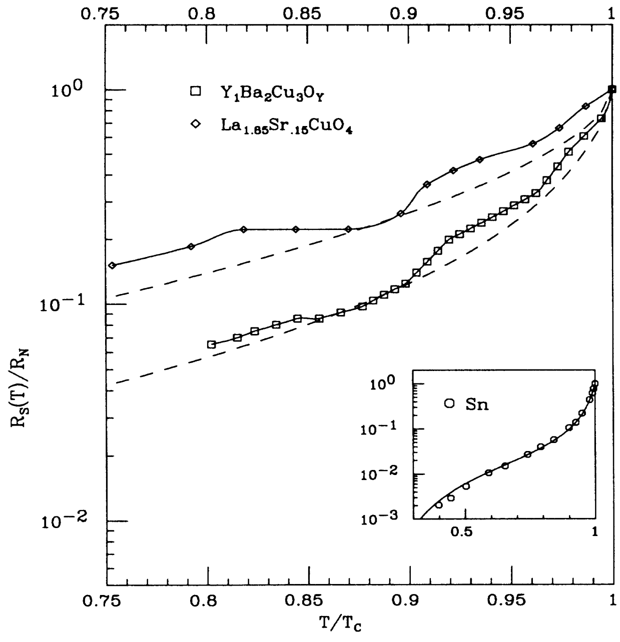

FIG. 1. Surface resistance ratio R_s/R_n for $Y_1Ba_2Cu_3O_y$ and $La_{1.85}Sr_{0.15}CuO_4$. The dashed lines are calculations using the BCS theory in the dirty limit approximation. The inset shows similar results for Sn, along with a comparison (solid line) to the BCS theory in the same approximation.

s-wave theory, using a "dirty" limit approximation. For a bulk superconductor in the pure local limit, it is easy to show[6] that $Z_s/R_n = \sqrt{2i}\,[\sigma_s(\omega,T)/\sigma_n]^{-1/2}$, where $R_n = (2\pi\omega/c\sigma_n)^{1/2}$ and σ_n are the normal-state surface resistance and conductivity, respectively. Also $\sigma_s(\omega,T) \equiv \sigma_1 - i\sigma_2$ is the local limit Mattis-Bardeen[7] complex conductivity. For finite mean free path, one has in general to consider the detailed wave-vector or mean-free-path dependence[7-9] of σ_s, and this entails lengthy numerical computation. For a dirty superconductor, considerable simplification can be achieved by recognizing that the principal effect of a short mean free path is to greatly reduce the superelectron screening, so that

$$\sigma_{2,\mathrm{eff}}/\sigma_n = (\lambda_L/\lambda_{\mathrm{eff}})^2(\sigma_2/\sigma_n) ,$$

while the "normal" electron response represented by (σ_1/σ_n) is unchanged. The surface resistance of an impure superconductor may then be computed from

$$R_s/R_n = \mathrm{Re}\{2i/[\sigma_1/\sigma_n - i(\lambda_{\mathrm{eff}}/\lambda_L)^{-2}(\sigma_2/\sigma_n)]\}^{1/2} . \qquad (1)$$

In carrying out the numerical computations, the Mattis-Bardeen expressions for the conductivities and the BCS forms for the coherence factors and density of states are used. The temperature dependence of R_s/R_n arises solely from $\Delta(t)$, for which we used the approximation $\Delta(t) = \Delta(0)\,[\cos(\pi t^2/2)]^{1/2}$. For $\Delta(0)$, the weak-coupling result $\Delta(0) = 1.74 k_B T_c$ was used. One parameter of the calculations is the measured ratio $\hbar\omega/k_B T_c$. There is only one adjustable parameter, $(\lambda_{\mathrm{eff}}/\lambda_L)^2$.

The validity of both the experimental technique and the analysis is demonstrated in the inset of Fig. 1, where we show data for $R_s(T)/R_n$ at 9.85 GHz for the conventional BCS superconductor Sn. (The data were taken in the same experimental configuration as used for the high-T_c materials.) The solid curve in the inset represents the analysis above, using the measured ratio $\hbar\omega/T_c = 0.125$ ($T_c = 3.78$ K), and choosing $(\lambda_{\mathrm{eff}}/\lambda_L)^2 = 2.5$.

The dashed curves in Fig. 1 represent the calculations of Eq. (1), using the experimental values $\hbar\omega/k_B T_c = 5.06\times 10^{-3}$ for Y-Ba-Cu-O and 1.2×10^{-2} for La-Sr-Cu-O, and the choices $(\lambda_{\mathrm{eff}}/\lambda_L)^2 = 65$ for Y-Ba-Cu-O and 43 for La-Sr-Cu-O. At temperatures $t < 0.8$, Eq. (1) reduces to $R_s/R_n \propto (\lambda_{\mathrm{eff}}/\lambda_L)^3(\sigma_1/\sigma_n)(\sigma_2/\sigma_n)^{-3/2}$, and also $\sigma_2/\sigma_n \sim (k_B T_c/\hbar\omega)(\pi\Delta(0)/k_B T_c)$. This provides an understanding of the effect of the ratio $(\lambda_{\mathrm{eff}}/\lambda_L)$. Since T_c is so high, $\hbar\omega/k_B T_c$ is very small, and hence if these compounds behaved like pure elemental superconductors, where $(\lambda_{\mathrm{eff}}/\lambda_L) \sim 1$, then R_s/R_n would be two orders of magnitude smaller than is observed. The same argument holds for roles played by the gap ratio. If $\Delta(0)/k_B T_c$ is much smaller than the BCS value as has been reported for La-Sr-Cu-O (Ref. 10) and some two-dimensional superconductors,[11] then the best fit would be found using a smaller value of $(\lambda_{\mathrm{eff}}/\lambda_L)$ than we have reported. It may be noted that at low temperatures $R_s/R_n \propto \exp[-\Delta(0)/k_B T_c]$, so that lower temperature ($t < 0.5$) results are required to obtain an independent value of the gap ratio $\Delta(0)/k_B T_c$.

This analysis yields two important conclusions. The overall temperature dependence is reasonably well described by the BCS s-wave theory, using the BCS temperature dependences and the weak coupling value 1.74 for the gap ratio. However, it is also clear from Fig. 1 that the data deviate from the BCS s-wave theory as regards the features near $0.9T_c$ and $0.96T_c$. We note that the analysis presented above captures the essentials of the BCS theory. To our knowledge, detailed computations using the full Mattis-Bardeen mean-free-path dependence[7] or the equivalent kernel of Ref. 9 do not yield the features observed in the data.[12]

The values of $(\lambda_{\mathrm{eff}}/\lambda_L)$ deduced from the measurements may be used to deduce other parameters of the superconducting state. Using for the electron density $n = 10^{21}$/c.c. and hence $\lambda_L = 52$ nm gives $\lambda_{\mathrm{eff}} = 419$ nm for Y-Ba-Cu-O and 340 nm for La-Sr-Cu-O. For La-Sr-Cu-O this result agrees with the value (330 nm) deduced from magnetization data,[13] while for Y-Ba-Cu-O it is larger (by a factor 4) than reported.[1]

The screening length δ_s was determined using a minor variant of the Schawlow and Devlin technique[14] in which the resonant frequency of a coil containing the sample is measured as a function of T. The conversion from frequency shift to absolute values of δ_s was found by calibrating with Cu disks. In principle, δ_s is related to the penetration depth by $\delta_s = \lambda \tanh(d/2\lambda)$, assuming an exponential penetration law. In practice, factors associated with sample defects such as voids, filamentary networks, and granularity may greatly complicate the relationship, as will anisotropy in polycrystalline samples.

Figure 2 shows $\delta_s(T)$ for one Y-Ba-Cu-O and one La-Sr-Cu-O sample, normalized to the asymptotic value, $d/2$, and plotted against the reduced temperature t (d is about 1 mm in both cases). The two curves are in overall agree-

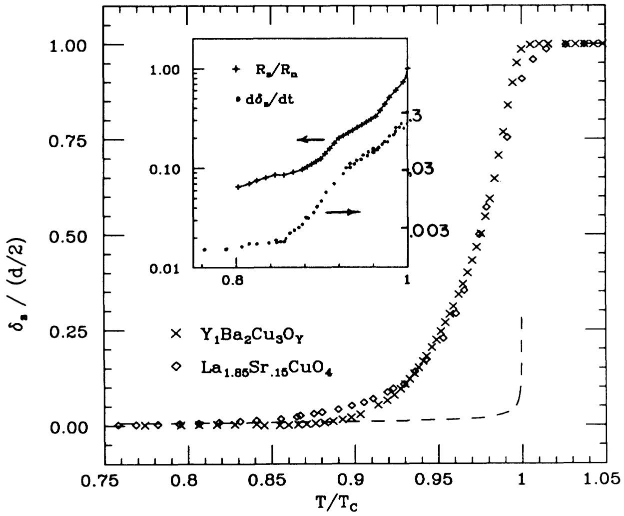

FIG. 2. Screening length δ_s at 1 MHz vs reduced temperature t for Y-Ba-Cu-O and La-Sr-Cu-O. δ_s is normalized to the asymptotic normal-state value $d/2$, where d($\sim$1 mm) is the sample thickness. The dashed line is the BCS prediction for the penetration depth λ, using $\lambda_0 = 0.5$ μm. The inset shows the temperature derivative $d\delta_s/dt$ (in cm) and also R_s/R_n for Y-Ba-Cu-O.

ment, despite the factor of 2 difference in transition temperatures. At small values of t, δ_s is less than about 5μm in both cases (i.e., within the uncertainties of the measurement), which corresponds to $T \rightarrow 0$ flux exclusion of about 99%. The onset of screening is quite sharp in the Y-Ba-Cu-O compound but appreciably broadened in the La-Sr-Cu-O case. Assuming a probability distribution $P(T_c) \propto \cos^2[\pi(T_c - T_{C0})/\tau]$, we find that τ, the spread of T_c values, is equal to 0.8 K for Y-Ba-Cu-O and 2 K for the La-Sr-Cu-O. These are in good agreement with the estimates based upon the resistivities.

The dependence of δ_s on t clearly does not follow the BCS theory (or the results[15] for heavy-fermion superconductors). The dashed curve in Fig. 2 is the BCS result for the choice $\lambda_0 = 0.5\mu$m, which is well within our margin of error and consistent with estimates of λ, taking into account the mean-free-path enhancement. Obviously δ_s is extremely large compared to the theoretical result once t exceeds about 0.9. In this context it is important to note the distinction between screening currents arising from the perfect conductivity of multiply connected regions and Meissner currents which insure $B = 0$ inside a homogeneous superconductor.[16] The first exaggerates the volume of superconductor, and hence if this form of screening dominated Meissner behavior in our samples, then the correct λ would be larger than the measured δ_s. On the other hand, a sample which consists of isolated superconducting grains in a normal or insulating matrix will appear to have a large penetration depth,[13] and the high δ_s values which we find might be evidence of just such a microstructure. However, both the measured[17] densities (92% to 95% of theoretical values) and optical microscope (1200×) examination argue against this. Surface roughness will certainly lead to exaggerated λ values via an underestimate of the effective sample area. This would be important near $t = 0$, but irrelevant above $t \sim 0.9$, where δ_s is many tens of microns.

A more interesting possibility is that the very high anisotropy of these materials induces an effective granularity even in stoichiometrically perfect samples, if they are polycrystalline. At large values of t, where δ_s is much larger than the grain size d_g, one would expect δ_s to be much larger than λ^*, the value for directions of easy current flow. When λ^* becomes comparable to d_g, screening currents will dip from the surface to sample favorably oriented grains, and the measured screening length should be an average of λ^* and d_g. A crossover temperature t^* can be defined by $\lambda^*(t^*) \sim d_g$ and substituting $t^* \sim 0.95$ (cf. Fig. 2) gives the reasonable value $\lambda_0^* \sim 0.4 d_g$.

The inset to Fig. 2 displays the temperature derivative $d\delta_s/dt$ for the Y-Ba-Cu-O sample, showing that the local minima observed in the 10-GHz R_s data are also contained in the 1-MHz result. Similar features were not seen in $d\delta_s/dt$ for the La-Sr-Cu-O sample, but this is to be expected since the transition width $\tau \approx 0.05 T_c$, which is just about the width of the oscillatory structure. In contrast to this, $\tau < 0.01 T_c$ for the Y-Ba-Cu-O sample, allowing the correlation between the microwave surface resistance and the rf screening length to emerge clearly.

The important conclusion that can be drawn from the R_s and δ_s data is that Y-Ba-Cu-O and La-Sr-Cu-O are similar superconductors, insofar as their electromagnetic response is concerned. The R_s data strongly suggest that the superconducting state is to first order and describable in terms of the BCS s-wave state. However, strong impurity scattering, leading to greatly enhanced $(\lambda_{\text{eff}}/\lambda_L)$ and hence reduced effective superfluid density, is required to fit the data.

Equally important are the oscillatory features in the R_s and $d\delta_s/dT$ data common to both superconductors, which clearly indicate departures from BCS theory. For a homogeneous superconductor, R_s is determined by the temperature dependence of the gap, the coherence factors, the density of states, and the quasiparticle excitations. It is not possible to decide which of these factors needs to be modified to account for these discrepancies without a detailed theory.

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[17] Densities were measured using a standard immersion technique. Values obtained using two-propyl alcohol and benzene agreed to better than 1%, and there were no signs of absorption of fluid. (The density of copper test disks was found to be 8.97 ± 0.01, using the same apparatus.) Theoretical densities were based on the formula $Y_1Ba_2Cu_3O_7$ and $La_{1.85}Sr_{0.15}CuO_4$ and unit cell volumes 1.736×10^{-22} and 1.886×10^{-23} cm^3, respectively.