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Dynamic properties of a high-T_c superconductor: Direct evidence for non-BCS behavior

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Measurements of the temperature dependence of the infrared conductivity of the CuO_2 planes in $YBa_2Cu_3O_7$ in the superconducting state are reported. Remarkably, we find that the temperature dependence of $\sigma_1(\omega,T)$ is independent of ω for $\omega \lesssim 500$ cm^{-1}, and that it is the same as that of the nuclear Korringa product, $1/T_1T$. These observations indicate that for the best-characterized high-temperature superconductor, $YBa_2Cu_3O_7$, the phenomenology of dynamic properties is fundamentally different from that of conventional superconductors.

In conventional superconductors measurements of dynamic properties played a central role in establishing the validity of the Bardeen, Cooper, and Schrieffer (BCS) theory.[1] For example, infrared measurements provided the first spectroscopic evidence of an energy gap,[2] and the observed enhancement of the nuclear relaxation rate[3] below T_c provided evidence for the BCS coherence factors.[4] In the high-T_c cuprate superconductors, one expects that an understanding of the dynamic properties would be of similarly fundamental interest. Although a number of these properties have been very reliably and reproducibly measured, some confusion exists as to whether they lead to a consistent picture of the dynamics in the superconducting state. In this paper we examine the temperature dependence of the ac conductivity, and demonstrate a remarkable relationship between the infrared conductivity and the nuclear Korringa product, $1/T_1T$. We also show consistency between the temperature dependence of the infrared conductivity and that of the penetration depth. These properties exhibit a phenomenology which is fundamentally different from that of conventional superconductors. We concentrate on data from fully oxygenated $YBa_2Cu_3O_7$, since it is for this material that the strongest evidence for electronic homogeneity exists,[5] and the widest range of the properties have been measured.

In Fig. 1 we show the infrared conductivity of the CuO_2 planes in $YBa_2Cu_3O_7$ ($T_c \simeq 93$ K) for temperatures between 30 and 120 K. These conductivities were obtained from reflectivity measurements of single-domain crystals, with the infrared electric-field polarized perpendicular to the CuO chains ($\mathbf{E} \parallel \hat{\mathbf{a}}$), as previously described.[6] Below 50 K the conductivity is essentially independent of temperature, exhibiting a gaplike absorption threshold at $\simeq 500$ cm^{-1} in agreement with earlier observations.[6–9] Above 50 K the conductivity in the gap region grows, causing this feature to fill-in and gradually disappear. Here we concentrate on the temperature dependence of $\sigma_1(\omega,T)$ in the 50–100 K range, and its relationship to other fundamental properties.

In Fig. 1, we find no evidence for a decrease in the energy of the conductivity threshold as T approaches T_c. In fact, the temperature dependence of the conductivity appears to be independent of frequency throughout the range of our measurements. This is illustrated in Fig. 2 where the conductivities at 450 and 225 cm^{-1} (with the latter scaled by 0.58) are shown as a function of temperature. In Fig. 2 we also show the nuclear Korringa product $1/T_1T$ for the planar oxygen in $YBa_2Cu_3O_7$, from the ^{17}O NMR data of Hammel *et al.*[10] Remarkably, we find that the temperature dependence of $1/T_1T$ below T_c is essentially the same as that of the infrared conductivity. These observations are potentially quite significant in that they suggest a universal temperature dependence in the superconducting state extending from NMR to infrared frequencies, as discussed below.

Motivated by the temperature dependences discussed above, we find that we can fit the conductivity to the form

$$\sigma_1(\omega,T) = f(T)\sigma_{1s}(\omega) + [1-f(T)]\sigma_{1n}(\omega) , \quad (1)$$

where $\sigma_{1s}(\omega)$ is the conductivity at 30 K and $\sigma_{1n}(\omega)$ is

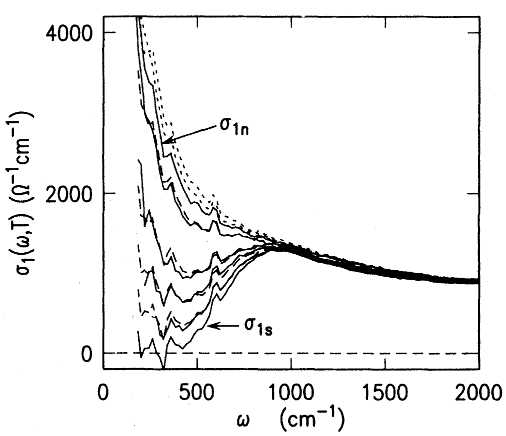

FIG. 1. The real part of the infrared conductivity of the CuO_2 planes in a single domain $YBa_2Cu_3O_7$ crystal is shown for T=30, 60, 70, 80, 90, and 100 K (solid lines), and for 110 and 120 K (dotted lines). The dashed curves are fits, based on Eq. (1), using weighted averages of the 30 and 100 K conductivity spectra, which are labeled as σ_{1s} and σ_{1n}, respectively.

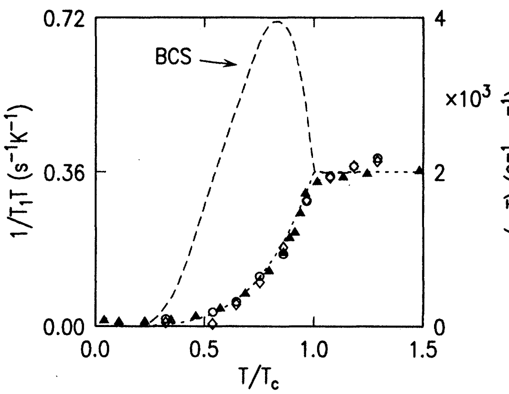

FIG. 2. The nuclear Korringa product (triangles) and the infrared conductivity at $\omega = 450$ (circles) and 225 cm^{-1} (diamonds) (scaled by 0.58) are shown as a function of reduced temperature, T/T_c. The data refer to the planar oxygen (Ref. 10) and to the conductivity of the CuO_2 planes in $YBa_2Cu_3O_7$. The dashed curve shows the behavior expected for $1/T_1T$ (and for the microwave conductivity) in the BCS theory, with the broad Hebel-Slichter peak (Ref. 4). The dotted curve through the data is proportional to $(T/T_c)^4$.

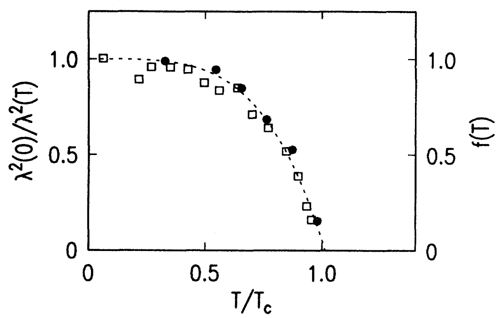

FIG. 3. A comparison of $\lambda^2(0)/\lambda^2(T)$, as determined from magnetic measurements of the penetration depth (Ref. 17) (open squares) and from the infrared conductivity of Fig. 1, via Eq. (2) (circles), is shown. The dotted curve through the data is $1-(T/T_c)^4$.

the conductivity at 100 K ($T_c = 93$ K). The fits are shown as dashed lines in Fig. 1. Since $\sigma_{1s}(\omega)$ is essentially zero below 500 cm^{-1}, this functional form implies a simple temperature dependence $1-f(T)$ independent of frequency up to ~ 500 cm^{-1}, as discussed above.

Equation (1) also implies a specific prediction for the temperature dependence of the penetration depth. This can be tested via the Glover-Ferrel-Tinkham sum rule,[11,12] which relates the area missing from the superconducting conductivity (relative to the normal-state conductivity) to the penetration depth,

$$1/\lambda^2(T) = (8/c^2)\int_{0^+}^{\infty}[\sigma_{1n}(\omega)-\sigma_1(\omega,T)]d\omega$$
$$= f(T)/\lambda^2(0)\,, \qquad (2)$$

where $\lambda(0)$ is the penetration depth at low temperature, and the last equality is based on using Eq. (1) for $\sigma_1(\omega,T)$. Previously, we have shown[6,7] that from the infrared conductivity one obtains $\lambda(0) \simeq 150$ nm, in good agreement with values inferred from more direct measurements of penetration depth.[13,14] We now find that, with the ansatz of Eq. (1), there is also good agreement between the temperature dependence of the infrared conductivity and that of the penetration depth. This is demonstrated in Fig. 3, where $\lambda^2(0)/\lambda^2(T)$ (from Ref. 13) and $f(T)$ (from the infrared data of Fig. 1) are shown. The observation of very acceptable agreement increases our confidence in the intrinsic nature of both the penetration depth and infrared data, as well as in the low-frequency extrapolation implied by Eq. (1).

If one associates $f(T)$ in Eq. (1) with the fractional density of a superconducting condensate and $1-f(T)$ with a normal component, Eq. (1) has a simple interpretation in terms of a two-fluid model. In this picture, the superconducting fraction has a temperature-independent energy gap ($\simeq 500$ cm^{-1}), and the superfluid density $f(T)$ has a temperature dependence which is consistent with the Gorter-Casimir form $(T/T_c)^4$, shown as dotted lines in Figs. 2 and 3. (This fit is not unique; e.g., exponents between about 3 and 5 are also plausible.) The possibility of fitting microwave data to a two-fluid picture has been considered by Drabeck et al.[15] While this is a convenient way to describe the data, it is not clear what it refers to at a microscopic level. A real-space two-fluid picture (phase separation) seems inconsistent with experimental and theoretical considerations; for example, it would imply two coexisting NMR lines due to the difference in the normal and superconducting state Knight shifts, which is not seen. A momentum-space two-fluid picture (in analogy with ^{4}He or Bose condensation) is intriguing, although key questions are unanswered, e.g., what is the nature of the excitation spectrum responsible for the emergence of a normal fraction for $T>0$. The possibility that the temperature-dependent normal fraction is associated with vortex pairs could also be considered. For the present, we prefer to view Eq. (1) as a phenomenological description of the superconducting dynamics which can be compared to various theories of high-temperature superconductivity.

In order to place the data presented here in perspective, it is useful to review the phenomenology of conventional (BCS) superconductivity. In the BCS theory $1/T_1T$ exhibits the well-known Hebel-Slichter enhancement below T_c, which arises from the coherence factors of the nuclear hyperfine interaction,[4] and the fact that the energy gap becomes small (compared to T) as T approaches T_c. The observation of this striking effect[3] played a central role in establishing the validity of the BCS theory.[4] The conductivity at finite frequency obeys the same coherence factors as $1/T_1T$, and at the NMR frequency $\sigma_{1s}(\omega,T)$ therefore follows the same temperature dependence as $1/T_1T$.[4] At higher frequency, however, $\sigma_{1s}(\omega,T)$ is not enhanced below T_c. (When the excitation frequency is greater than the nominal width of the BCS density of states peak, enhancement does not occur.[4]) These two regimes of qualitatively distinct temperature dependence are shown in the BCS calculations[16] of Fig. 4, where the enhancement of $\sigma_{1s}(\omega,T)$ at low frequency and the absence of enhancement at higher frequency are shown explicitly in

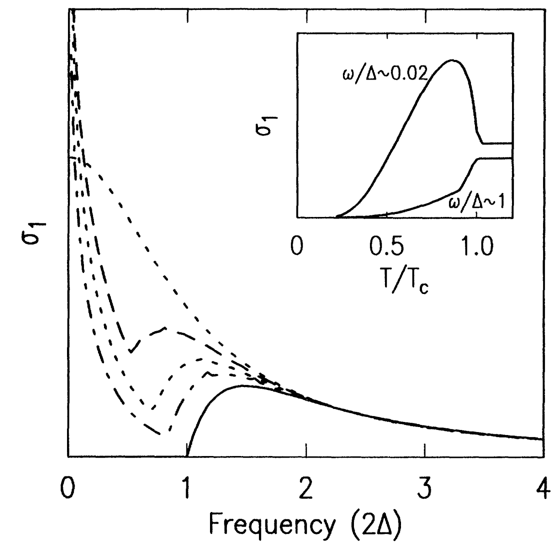

FIG. 4. (a) BCS calculations of the conductivity of a conventional superconductor for $T \simeq 0$, $0.7T_c$, $0.8T_c$, $0.9T_c$, and T_c, show both the temperature dependence of the energy gap, and the enhancement of $\sigma_1(\omega,T)$ at low ω below T_c. In the inset, the calculated temperature dependence of $\sigma_1(\omega)$ is shown for a very low frequency, where enhancement occurs, and a higher frequency ($\omega \sim \Delta$), where $\sigma_1(\omega)$ decreases monotonically below T_c. For these calculations we use the Chang-Scalapino formalism (Ref. 16), with an elastic scattering rate equal to 2Δ, which corresponds to $l/\xi_0 \simeq 1.8$.

the inset.

What is particularly unusual about $YBa_2Cu_3O_7$ is that the temperature dependences of $1/T_1T$, measured at very low ω, and of $\sigma_{1s}(\omega,T)$, at much higher frequency (up to $\sim 500\ cm^{-1}$), are essentially equivalent. Although strictly speaking effects associated with a temperature-dependent transport scattering rate[17] may allow some differences, it is reasonable to think of the temperature dependence of $1/T_1T$ as representative of the essential aspects of the temperature dependence of $\sigma_{1s}(\omega,T)$ at very low frequency. This association makes the essence of our data even clearer: that in $YBa_2Cu_3O_7$ there is a single temperature dependence operative at all relevant frequencies in the superconducting state (i.e., $\omega \lesssim 2\Delta$). This phenomenology represents a fundamental departure from conventional BCS theory, one of the central predictions of which is very different temperature dependence at low and high frequency (see, e.g., Fig. 4).

In the calculations of Fig. 4, one also observes explicitly that the energy gap [the threshold in $\sigma_{1s}(\omega)$] shifts to lower energy as T approaches T_c. In the data of Fig. 1, the conductivity in the gap region ($\omega \lesssim 500\ cm^{-1}$) fills in as T approaches T_c, however, the energy scale, or magnitude of the gap, does not appear to change with temperature. Our ability to fit the data in Fig. 1 with Eq. (1), which includes only one (fixed) energy scale in the superconducting state, supports this observation. This is another aspect of the $YBa_2Cu_3O_7$ data which differs significantly from the conventional expectations.

Since in BCS theory both gap collapse and coherence factors are essential to the occurrence of enhancement of $1/T_1T$, it is natural to consider the possibility that the temperature-independent gap of the infrared data, and the absence of enhancement in the NMR data are directly related. It is not clear whether a previously considered scenario,[17] in which a strong and highly temperature-dependent inelastic scattering influences the dynamics near T_c, is consistent with the phenomenology presented here. A more exotic possibility is that the unconventional temperature dependence of $\sigma_1(\omega,T)$ and $1/T_1$ may be associated with unconventional statistics and altered coherence factors, since (Fermi) statistics play a central role in establishing the usual (BCS) temperature dependence of dynamic properties.[4]

In conclusion, we have examined fundamental dynamic properties, which have been reliably measured in the high-T_c superconductor $YBa_2Cu_3O_7$. We find no inconsistencies in the data, and we demonstrate a remarkable relationship between the infrared conductivity and the nuclear Korringa product, which suggests a fundamentally non-BCS phenomenology for $YBa_2Cu_3O_7$. Ultimately the implications of, and the relationships within, the data cannot be properly understood without a microscopic theory. What the data seem to suggest is that the nature of the superconducting transition in $YBa_2Cu_3O_7$ lies between the physics of the BCS Fermi-surface instability and of Bose condensation.

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