Superconducting Energy Gap and Normal-State Reflectivity of Single Crystal Y-Ba-Cu-O

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We report the first measurements of the normal-state reflectivity and superconducting energy gap of $Y_1Ba_2Cu_3O_{7-x}$ single crystals. With the electric field in the metallic, *a-b* plane of our platelike crystals we observe a Drude normal-state reflectivity, and an enhancement of the reflectivity at temperatures below T_c (90 K) which follows the Mattis-Bardeen form. The superconducting energy gap obtained from these measurements is found to be temperature dependent, increasing to a value of $2\Delta/kT_c \simeq 8$ below $\simeq 50$ K. Within the Eliashberg-BCS framework this unusually large gap value suggests that $Y_1Ba_2Cu_3O_{7-x}$ is a very strongly coupled superconductor.

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The recent discovery[1] of high-temperature superconductivity in layered oxide compounds[1-3] is naturally followed by considerable effort to understand the mechanism of the superconductivity. Within the BCS-Eliashberg framework high T_c's can arise either from the exchange of bosons of high energy or by strong coupling to lower-energy modes. Theoretical models[4] ranging from weak-coupling excitonic theories to strong-coupling phonon and magnetic-exchange mechanisms, some well outside the BCS theory, have been considered as explanations for the high T_c's. In most theories the superconductivity is primarily associated with the high-conductivity (*a-b*) plane[4] and thus an experimental knowledge of the properties within this plane is of central importance.

Typically measurements of the specific-heat discontinuity at T_c or of the superconducting energy gap have been used to distinguish between strong and weak coupling.[5] Measured specific-heat discontinuities consistent with weak coupling have been reported by a number of groups[6-9]; however, Carbotte[10] has shown that, within the Eliashberg formalism, these results are consistent with either weak or very strong coupling, since the specific-heat jump at T_c is a nonmonotonic function of coupling strength. On the other hand, the reduced energy gap, $2\Delta/kT_c$, increases monotonically with increasing coupling strength and thus its measurement allows an unambiguous distinction between strong- and weak-coupling models.[10]

In this Letter we report the first infrared measurement of single-crystal $Y_1Ba_2Cu_3O_{7-x}$ ($T_c \simeq 90$ K). With the incident electric field in the metallic, *a-b* plane of our platelike single crystals, we observe a primarily Drude normal-state reflectivity. Below T_c we observe a reflectivity enhancement which follows the Mattis-Bardeen form,[11] and from which we infer a temperature-dependent energy gap which reaches a value of $2\Delta/kT_c \simeq 8$ below $T \simeq 50$ K. This result suggests that the high T_c of $Y_1Ba_2Cu_3O_{7-x}$ is associated with very strong-coupling superconductivity.

Growth of the single crystals is described in detail elsewhere.[12] The crystals were annealed in flowing oxygen (1 atm) at 420 °C for 30 h. Superconducting transition temperatures of annealed crystals as measured by an inductive technique were typically 92 K with a width of 0.2 K. Twinning characterized by the {110} twin planes and the $\langle 1\bar{1}0 \rangle$ directions was observed in the crystals. A mosaic of individual platelike crystals was prepared to obtain a large area ($\simeq 2\times 2$ mm^2) for the infrared-reflectivity measurements. Reflectivity measurements on individual crystals were also made and equivalent results were obtained, but with a lower signal-to-noise ratio. The infrared- and optical-reflectivity measurements were made with a scanning interferometer (50–800 cm^{-1}), a Perkin-Elmer grating spectrometer (200–4000 cm^{-1}), and a Carey grating spectrometer (4000–40000 cm^{-1}). The infrared radiation was incident at near normal incidence to the crystal surfaces (and hence the *a-b* plane) and was nominally unpolarized.

In Fig. 1(a) the normal-state *a-b*–plane reflectivity of the single crystals is shown. A Kramers-Kronig transform is used to obtain $\sigma(\omega)$ [Fig. 1(b)] which is primarily Drude-type, although there may be some frequency-dependent scattering as suggested previously.[13] A Drude fit to the reflectivity is included in Fig. 1(a) and the corresponding Drude conductivity is plotted in Fig. 1(b). The parameters used in the fit are $\omega_p \simeq 25000$ cm^{-1} and $\tau^1 \simeq 7500$ cm^{-1}. With $\sigma = ne^2\tau/m$, these imply a dc resistivity of $\simeq 700$ $\mu\Omega\cdot$cm which is comparable to typical room-temperature measured values on both polycrystalline and single-crystal samples.[14] (Exact agreement is not expected since, in the presence of strong inelastic scattering, the optical scattering rate will tend to be somewhat larger than the dc value, especially at low temperatures.[15]) Arbitrarily choosing a band mass of unity, one would obtain a carrier density of 7×10^{21} cm^{-3}, which is within the range established by Hall measurement.[16] In contrast to several previous studies of polycrystalline material,[17-19] however, we do not find the low-frequency optical conductivity to be dominated

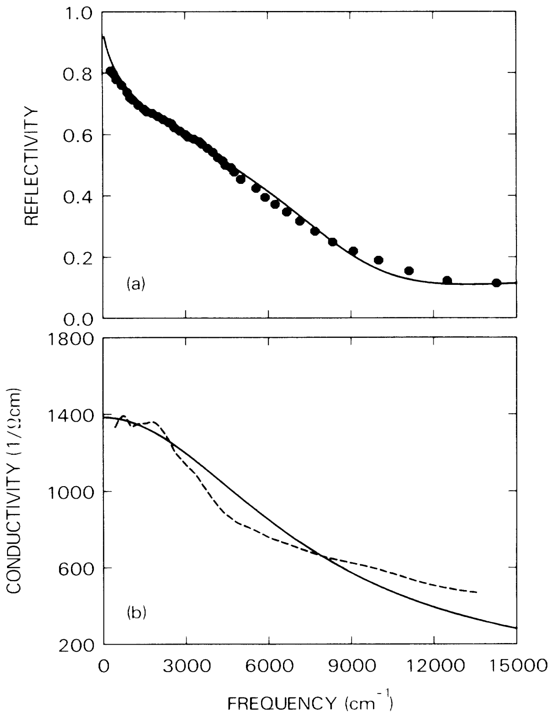

FIG. 1. (a) Measured room-temperature *a*-*b*–plane reflectivity of single crystal $Y_1Ba_2Cu_3O_{7-x}$ and a Drude fit to the data (solid line). (b) Optical conductivity obtained by a Kramers-Kronig transform (dashed line) and the Drude conductivity (solid line) from the fit in (a). (Drude parameters are given in the text.)

by a mode at $\simeq 0.5$ eV. Such a mode has figured prominently in pictures of excitonic superconductivity.[20]

As the temperature of the crystals is reduced below T_c, we expect to see an enhancement in the low-frequency reflectivity due to the opening of the superconducting gap. In Fig. 2 ratios of the reflectivity in the superconducting state to that in the normal state ($\simeq 90$ K) are shown for several temperatures below T_c. As the temperature is reduced below T_c (90 K), the reflectivity increases over a broad spectral range below about 700 cm^{-1}. No significant change in reflectivity is observed above T_c (e.g., from 90 to 120 K) or below about 50 K, demonstrating that the reflectivity increase we observe is indeed correlated with the superconducting transition. Because our sample was not large enough to fill the beam completely, some of the radiation reaching the detector was reflected from the copper sample mount. The vertical scale of the reflectivity ratios is therefore a lower bound on the actual size of the reflectivity enhancement, which could be as much as a factor of 2 or 3 larger than shown in Fig. 2. Even with this correction our reflectivity ratios indicate that the reflectivity in the superconducting state is less than 100%, which may be due to inhomogeneity at the surface. (The possibility also exists that this is an intrinsic result, associated for example with nonzero angular momentum pairing[4] and hence regions of the *a*-*b* plane in which small or vanishing gaps occur.) (See note added.)

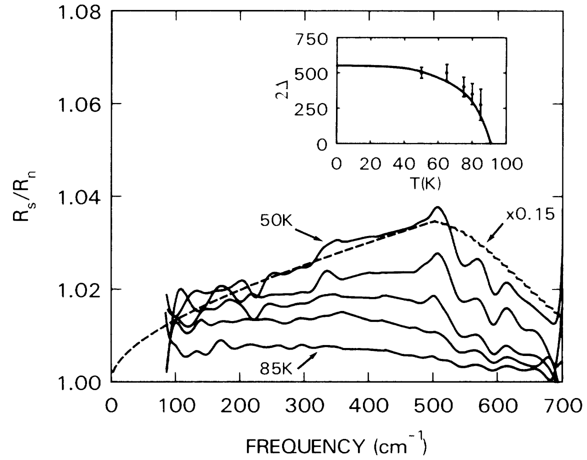

FIG. 2. The measured ratios of the reflectivity in the superconducting state to the reflectivity in the normal state ($T=90$ K) for temperatures of 50, 65, 75, 80, and 85 K. The dashed line indicates a Mattis-Bardeen fit to the 50-K spectrum with an energy gap of 500 cm^{-1}. Inset: The temperature dependence of the peak in the ratios along with a BCS temperature dependence for the superconducting gap (solid line).

Figure 2 also shows a calculated reflectivity ratio, R_s/R_n, where we assume a $T=0$ Mattis-Bardeen conductivity[11] with an energy gap of $2\Delta = 500$ cm^{-1} for the superconductor, and a Drude normal state. This ratio rises to a broad maximum at 2Δ and then drops gradually back to unity (the form of the curve above 2Δ depends on the skin-effect regime, for which we use the dirty limit[21]). A comparison of this calculated reflectivity ratio to the measured spectrum for $T \simeq 50$ K shows that the data follow the Mattis-Bardeen form, which, along with the direct correlation with T_c, establishes the primary basis for the identification of the reflectivity enhancement with the superconducting energy gap. Specifically, on the basis of Mattis-Bardeen theory, the energy gap is identified with the maximum in R_s/R_n, which occurs at $\simeq 500$ cm^{-1} for $T \lesssim 50$ K and, for example, at $\simeq 350$ cm^{-1} at $T=80$ K. (The sharper peak at 500 cm^{-1} is of instrumental origin and should not be confused with the broad maximum which is the signature of the energy gap.) We are thus observing, for the first time, an infrared-reflectivity enhancement which provides a direct and accurate measure of the energy gap 2Δ. Superconducting energy gaps thus obtained are plotted as a function of temperature in the inset to Fig. 2. A BCS temperature dependence is also shown; however, the substantial error bars near T_c preclude the possibility of making detailed conclusions regarding the temperature dependence of the gap.

These results are in marked contrast to observations made on polycrystalline samples[22-34] where strong contributions from the c-axis orientation, which is not particularly metallic, very much complicate the data analysis.[28] For example, in polycrystalline $La_{2-x}Sr_xCuO_4$ the gap measurement is obscured by a very low-energy plasmalike edge,[27] which is primarily a property of the c-axis response,[28] and in which effective-medium effects may be important.[29] Values of $2\Delta/kT_c$ ranging from 1.5 to 4.0 have been reported.[22-29] Similar measurements[30-34] of polycrystalline $Y_1Ba_2Cu_3O_{7-x}$ exhibit very strong, possibly c-axis related, phonon structure, which makes an estimation of 2Δ from that data difficult. Values of $2\Delta/kT_c$ ranging from about 2.5 to 4.5 have been inferred.[30-34] Results for $Y_1Ba_2Cu_3O_{7-x}$ films[32] seem to lie somewhere between the earlier polycrystalline results and the present single-crystal data, in terms of both the shape of the reflectivity ratio and the estimated size of the gap ($2\Delta \simeq 5kT_c$) inferred from the data. These films are primarily, but not completely, aligned with the c axis perpendicular to the surface. One may therefore presume that the discrepancy between the film results[32] and our a-b–plane single-crystal data is due to a small amount of c-axis data in the film spectra, especially since the low reflectivity of the c axis tends to be preferentially represented in reflectivity ratios.[28]

Recently Carbotte[10] explored the Eliashberg equations at arbitrary values of the coupling strength, and obtained an upper limit of $2\Delta/kT_c = 11.5$ for extreme strong coupling. (The lower limit, obtained for BCS weak coupling, is $2\Delta/kT_c = 3.5$.) Within this framework, our measured value of $2\Delta/kT_c \simeq 8$ would imply very strong coupling, well beyond values found in previously studied conventional strong-coupling superconductors, such as Pb ($2\Delta/kT_c = 4.3$) and Hg ($2\Delta/kT_c = 4.6$). In this regime of very strong coupling, Carbotte found that the energy scale associated with $\alpha^2F(\omega)$ is comparable to kT_c. In a quite distinct approach Lee and Read[35] used the temperature dependence of the resistivity to infer strong, low-frequency, inelastic scattering, which they treated as exclusively pair breaking. Invoking this strong pair-breaking mechanism to suppress T_c they obtained very large values for $2\Delta/kT_c$ without a strong-coupling pairing mechanism. Limits to the generality of this approach have been examined by Sachdev, Millis, and Varma.[36]

For $Y_1Ba_2Cu_3O_{7-x}$ the observed absence of an isotope shift[37,38] and the very high T_c have discouraged the application of phonon-related models.[39] We note, however, that our observed gap value is not inconsistent with an electron-phonon coupling strength, λ, in the very strong-coupling regime for which solutions with $T_c = 90$ K and no oxygen isotope effect are found.[38] (Both for lattice stability and to obtain a null isotope effect, these solutions require values of μ^* significantly larger than found in other superconductors.[38]) Although a very strong electron-phonon coupling model has not been definitively ruled out, arguments against it are numerous. For example, Gurvitch and Fiory[40] placed an upper bound on 1.2 on λ for $Y_1Ba_2Cu_3O_{7-x}$, based on the absence of resistivity saturation, and Maletta *et al.*[41] inferred a small electron-phonon coupling in $La_{2-x}Sr_xCuO_4$ from a comparison of susceptibility and specific-heat data.

There is a great deal of interest in pairing via magnetic excitations, either alone or in concert with phonons,[4] since the proximity to magnetism appears to be one of the unique aspects of the oxide superconductors. In many of these models the energy gap is expected to exhibit d-wave or extended s-wave symmetry[4]; however, the details of the size and nature of the gap have not as yet been calculated. In the resonating-valence-band model[42] the procedure for the comparison of theory and infrared data has not yet been established.

In conclusion, in single-crystal samples of superconducting ($T_c \simeq 90$ K) $Y_1Ba_2Cu_3O_{7-x}$ we observe a primarily Drude reflectivity in the a-b plane and a reflectivity enhancement below T_c which follows the Mattis-Bardeen form (peaked at 2Δ). The superconducting energy gap rises to a value of 500 cm^{-1} below $T \simeq 50$ K, which corresponds to $2\Delta/kT_c \simeq 8$. Regardless of the nature of the mode which mediates the pairing (e.g., magnetic fluctuations or phonons) this result indicates that the coupling of the electrons to the intermediate mode is extremely strong.

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Note added.—We have attempted to observe gap anisotropy, which may be associated with p- or d-wave pairing symmetry. Using a mosaic in which the a and b axes are aligned along one of two orthogonal directions in the plane, we find the infrared response to be independent of the polarization of the incident electric field in the a-b plane.

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