Infrared Evidence for Gap Anisotropy in $YBa_2Cu_3O_7$

R. T. Collins, Z. Schlesinger, F. Holtzberg, and C. Feild
IBM T. J. Watson Research Center, Yorktown Heights, New York 10598
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Infrared reflectivity measurements of crystalline $YBa_2Cu_3O_7$ ($T_c \simeq 92$ K) are performed with the electric field both parallel and perpendicular to the Cu-O planes. We find evidence for an unusually large in-plane energy gap ($\sim 8kT_c$), while a much smaller energy scale ($\sim 3kT_c$) is obtained with the field perpendicular to the planes, suggesting substantial gap anisotropy. Temperature-dependent measurements near T_c demonstrate a clear relationship between the development of the $\sim 8kT_c$ feature and the superconducting transition.

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In the copper-oxide class of high-temperature superconductors[1,2] there is substantial anisotropy between the in-plane (a-b plane) and out-of-plane (c axis) transport properties. The platelike nature of the crystals and preferred growth directions of the films have made studies with the electric field perpendicular to the planes extremely difficult; consequently, very little is known about the dynamics along the c-axis direction, although they play a major role in some theories. Even within the a-b plane, controversies exist regarding the interpretation of infrared data, which have been cited as evidence for a very large energy gap ($2\Delta_{a\text{-}b} \simeq 8kT_c$),[3] a BCS size gap,[4] and no gap at all.[5] To address these issues, a more complete picture of the infrared response along each of the principal crystal axes is required, particularly since impurity scattering tends to average, to some degree, all directions into any particular measurement.

In this Letter we report the infrared response of $YBa_2Cu_3O_7$ with the incident electric field both parallel and perpendicular to the high-conductivity Cu-O planes. Evidence for a relatively small energy gap, $2\Delta_c \simeq 3kT_c$, associated with the direction perpendicular to the planes is presented. In contrast, the temperature dependence of the a-b-plane reflectivity near T_c provides clear evidence for a much larger energy scale, $2\Delta_{a\text{-}b} \simeq 8kT_c$, associated with the in-plane dynamics, as originally reported.[3]

Experiments were performed on $YBa_2Cu_3O_7$ crystals which typically exhibited a sharp (~ 0.3 K wide) superconducting transition at about 92 K, as determined by ac susceptibility. The crystals have a platelike geometry with a typical thickness of about 0.05 mm in the c-axis direction, and lateral (a-b plane) dimensions of order 1 to 3 mm. a-b-plane reflectivities were measured on individual crystals; however, to obtain sufficient area to measure the reflectivity with the incident electric field along the c axis a stack of about twelve crystals was used. The stack was potted in epoxy and polished to obtain a flat surface; the fill fraction of crystal within the epoxy was roughly 50%. We have previously found that polishing does not severely degrade the infrared properties of a-b-plane crystal surfaces relative to as-grown surfaces. Even smaller effects are expected with the field along the c axis because of the larger penetration depth. Reflectivities were measured at near normal incidence using a scanning interferometer, with the sample mounted in close proximity to an evaporated gold reference mirror which was aligned parallel to the sample surface. A manipulator allowed either the sample or reference to be positioned in the infrared beam. We tested our technique by studying both brass-Au and Au-Au reflectivity ratios; in each case a precision of better than 1% was obtained.

In Fig. 1(a) the measured reflectivity of the a-b-plane face of a twinned $YBa_2Cu_3O_7$ crystal is shown as a function of frequency for temperatures from 45 to 150 K, along with reflectivity ratios [Fig. 1(b)]. At each temperature the conductivity is obtained from a Kramers-Kronig transformation of the reflectivity data in Fig. 1(a), along with additional data extending to 30000 cm^{-1} and a high-frequency termination as described previously.[6] In Fig. 1(c) we show the real part of the conductivity at 45 and at 105 K, respectively, divided by that at 150 K. Between about 500 and 800 cm^{-1} the ratio of the reflectivity in the superconducting state to that at 150 K, shown by the solid line in Fig. 1(b), drops sharply, while the corresponding conductivity ratio exhibits a rapid increase, indicating a rather sudden onset of absorption at roughly 500 cm^{-1} ($\simeq 8kT_c$) in the superconducting state.

Recently there has been some controversy over whether this onset of absorption at $\sim 8kT_c$ is related to the occurrence of superconductivity or merely a continuation of the normal-state temperature dependence.[4,5] To address this point a-b-plane reflectivity ratios in the vicinity of T_c are shown in Fig. 2(a). It is apparent that the reflectivity below 500 cm^{-1} increases much more rapidly below T_c than above T_c, and that in the vicinity of 800 cm^{-1} the derivative of the reflectivity with respect to temperature changes sign at about T_c. The drop in the reflectivity ratio between 400 and 800 cm^{-1}, defined as η in Fig. 2(a), is directly related to the onset of absorption at $\sim 8kT_c$. In Fig. 2(b) we plot η as a function of

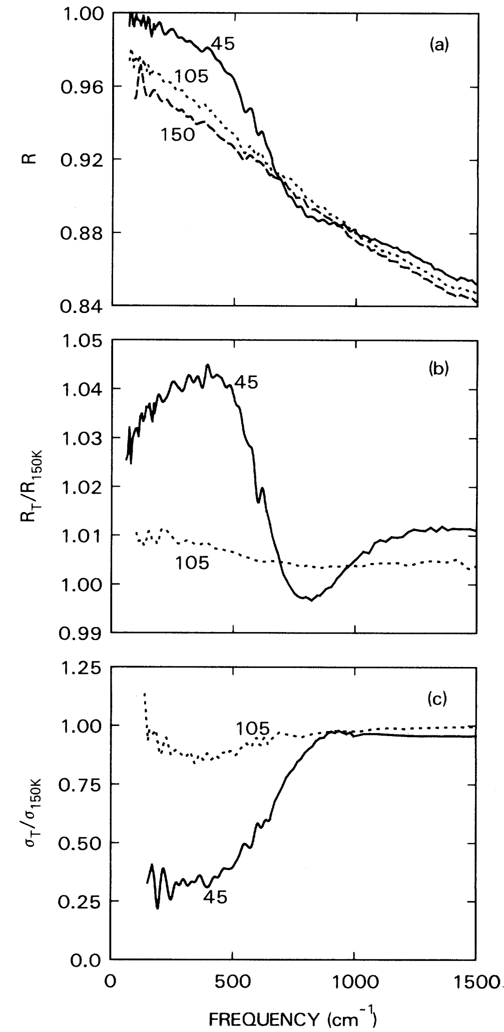

FIG. 1. (a) The a-b-plane reflectivity of a $YBa_2Cu_3O_7$ crystal at $T=45$, 105, and 150 K. (b) The a-b-plane reflectivity at 45 and at 105 K, divided by that at 150 K. (c) The real part of the a-b-plane conductivity at 45 and at 105 K divided by that at 150 K.

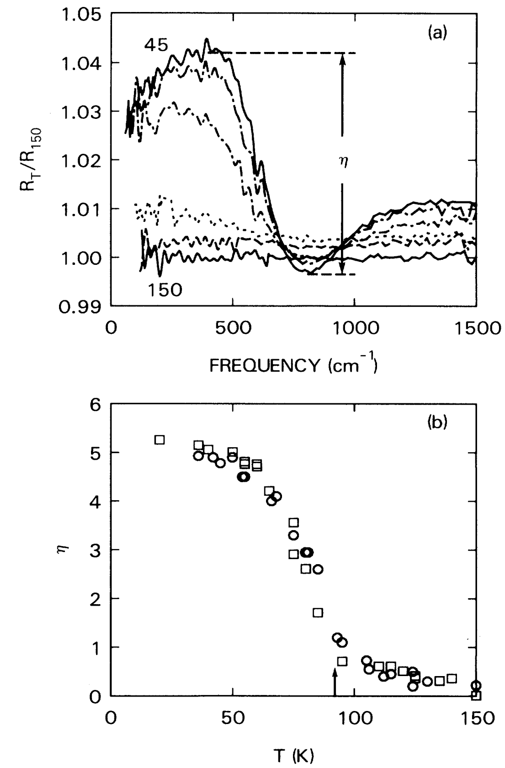

FIG. 2. (a) a-b-plane reflectivity ratios in the vicinity of T_c. R_T/R_{150} is the reflectivity at a temperature T divided by that at $T=150$ K, where $T=150$, 125, 105, 80, 65, and 45 K. (b) The drop in the reflectivity ratio between 400 and 800 cm^{-1} (η) is plotted as a function of temperature for the sample used in (a) (circles) and a second sample (squares) illustrating the relationship between the $\sim 8kT_c$ reflectivity enhancement and T_c (indicated by the arrow).

temperature, demonstrating the relationship between the $\sim 8kT_c$ feature and the transition to superconductivity. The possible persistence of this feature into the normal state is of potentially great interest. We do observe a small depression in the conductivity ratio for $T=105$ K in Fig. 1(c) which seems reminiscent of the much larger depression in the 45-K ratio, but we caution against placing much significance on it at this point. Investigation of the presence of a pseudogap or self-energy structure in the normal state near $\sim 8kT_c$ remains an interesting area for further research.

We now turn our attention to measurements in which the incident electric field is oriented perpendicular to the conducting planes, i.e., along the c axis. The c-axis reflectivities in the normal (100 K) and superconducting (45 K) states are presented in Fig. 3(a). The spectra have been adjusted to account for the open space between individual crystals, as described above, in this edge-on geometry. The ratio of the superconducting to normal reflectivity is given in Fig. 3(b). The real part of the conductivity, shown in Fig. 3(c), is obtained from a Kramers-Kronig transform of the reflectivity using additional higher-frequency data[7] scaled to match our spectra. The normal-state c-axis conductivity in Fig. 3(c) extrapolates to a dc value near 400 $(\Omega\,\mathrm{cm})^{-1}$. A comparison with the extrapolated dc conductivity of about 16000 $(\Omega\,\mathrm{cm})^{-1}$ for the a-b plane yields a conductivity anisotropy estimate at 100 K of about 40:1 which is in the range of the lowest estimates from dc transport studies.[8] As a consequence of this low c-axis conductivity the c-axis phonons are not well screened, and the reflectivity enhancement below T_c is large because the normal-state reflectivity is rather low. The changes in reflectivity at the superconducting transition are similar to those observed in ceramic samples,[9] which is not completely unexpected since the large change in reflectivity

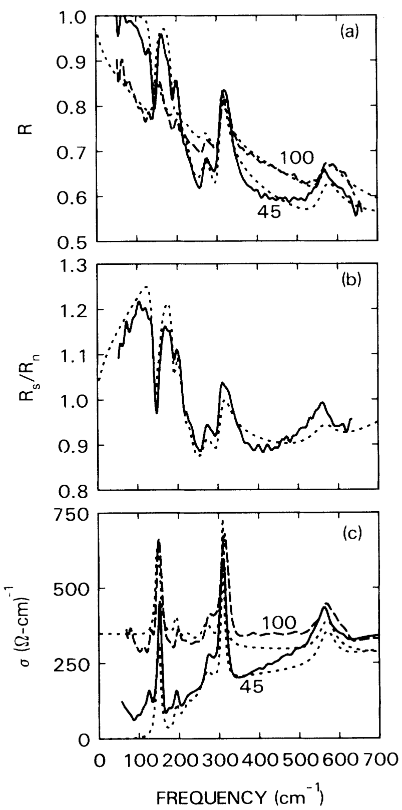

FIG. 3. (a) The reflectivity in the superconducting ($T=45$ K) and normal ($T=100$ K) states for the incident electric field along the c-axis direction. (b) The c-axis reflectivity in the superconducting state divided by that in the normal state. (c) The real part of the conductivity in the superconducting ($T=45$ K) and normal ($T=100$ K) states. The dotted lines are calculated fits to the data including both phonon and electronic contributions to $\sigma(\omega)$.

associated with the c axis is likely to dominate the a-b-plane contribution in an unoriented sample as we have previously proposed.[10]

The rapid drop in the reflectivity ratio [Fig. 3(b)] at about 200 cm^{-1} suggests the possibility of a c-axis energy gap in this vicinity. The dotted curves in Fig. 3 are an approximate fit to our spectra, calculated using a superconducting energy gap of 170 cm^{-1}, with normal-state conductivity of 350 $(\Omega\,\text{cm})^{-1}$, and phonons (Lorentz oscillators) at 155, 195, 220, 270, 310, and 570 cm^{-1}. The frequency of the lowest (155 cm^{-1}) phonon in Fig. 3(c) is slightly higher (~ 2 cm^{-1}) in the superconducting state than in the normal state, whereas the other phonons all shift in the opposite direction. In addition, the response of the lowest-frequency phonon changes from dispersive to absorptive in the superconducting state [Fig. 3(a)], while the higher-energy phonons are dispersive in both spectra.[9] Both observations are consistent with the existence of an energy gap at a frequency between the 155 and 195 cm^{-1} phonons. Bonn *et al.*[11] have suggested the possibility of an energy gap in this range from observed changes in the damping of these phonons in their study of ceramic samples. Based on the phonon behavior, the frequency range in which R_s/R_n drops most substantially, and the calculated fits in Fig. 3, we estimate a superconducting frequency scale for the c axis of about 170 cm^{-1} $\simeq 3kT_c$, which is much lower than that obtained for the a-b-plane response ($\sim 8kT_c$). Our estimate of $\simeq 3kT_c$ is slightly larger than the lower bound of $\simeq 2.1kT_c$ which Noh, Kaplan, and Sievers[12] have recently placed on the c-axis gap of $La_{2-x}Sr_xCuO_4$ from a study of far-infrared sphere resonances.

Beginning with the work of Glover and Tinkham in 1957,[13] infrared estimates of energy gaps in conventional superconductors have been based on either an extrapolated cutoff of σ_{1s}/σ_{1n}, or the frequency of a peak in a measured ratio. We have used these approaches, which emphasize the data above 2Δ, to obtain our estimates for the energy scales associated with the superconducting state of $YBa_2Cu_3O_7$. From our a-b-plane data, a characteristic energy scale of 500 cm^{-1} ($\sim 8kT_c$) is inferred from either the peak in R_s/R_n or the onset of absorption in σ_{1s}/σ_{1n} (Fig. 1). The very rapid rise of σ_{1s}/σ_{1n} above 500 cm^{-1} is suggestive of a sharp density-of-states peak as one expects with an s-wave gap; however, our data also show that there is considerable absorption [i.e., nonzero $\sigma_{1s}(\omega)$] below 500 cm^{-1} in the superconducting state. Because of the reproducibility of this behavior and the high quality of our samples, we believe that a significant fraction of this absorption is intrinsic to $YBa_2Cu_3O_7$; thus it is very important to understand its origin. The most interesting possibility is that it could be reflective of an in-plane gap anisotropy (e.g., due to non-s-wave pairing symmetry), although measurements on both twinned and untwinned samples have failed to show any anisotropy of R_s/R_n within the a-b plane. Another approach, however, is to consider that it may arise from a contribution to $\sigma_1(\omega)$ that does not have a 500-cm^{-1} gap. For example, crude estimates of the chain-related contribution to the conductivity indicate that, within a factor of 2 or so, it may be sufficient to account for the background conductivity. A third possible source of finite absorptivity below 500 cm^{-1} is the anisotropy of the gap and the tendency of impurity scattering to mix, to some degree, contributions from all directions in k space into any infrared conductivity measurement.[14]

Recently Thomas *et al.*[4] have suggested the possibility of an energy gap of conventional magnitude ($\sim 3.5kT_c$), based on their observation of unity reflectivity up to about 120 cm^{-1} in the superconducting state of oxygen-deficient $YBa_2Cu_3O_{7-x}$ crystals ($T_c \simeq 50$–70 K). This approach emphasizes the infrared data below 2Δ, and, in principle, will produce the smallest energy gap in an anisotropic or multiband superconductor, as discussed above. Our present experiments neither refute or support the idea of a low-frequency region with unity reflectivity, since we obtain a reflectivity in excess of 99%, which we cannot definitively distinguish from unity, below about 150 cm^{-1}. Our a-b-plane reflectivity data are reasonably consistent with that of Timusk *et al.*,[5] and with our previously published reflectivity ratios.[3] We note that several other techniques, including nuclear quadrupole resonance[15] and most recently photoemission,[16] have provided evidence for energy gaps of $\sim 8kT_c$ in the Cu-O superconductors. Many tunneling studies also suggest gaps well in excess of the BCS value (see, for example, Ref. 17), although there is some scatter in the reported values, possibly because of the anistropy reported here. Tsai *et al.*[18] have reported a similar in-plane versus out-of-plane anisotropy from their tunneling study of oriented $YBa_2Cu_3O_{7-x}$ films.

Recently we have also studied the infrared properties of the cubic bismuth-oxide superconductors.[19,20] We obtain superconducting-state reflectivity enhancements very much like that shown in Fig. 1(b), and from the peak in the reflectivity ratio, R_s/R_n, estimate an energy gap of conventional magnitude ($\sim 4kT_c$) for both $Ba_{0.6}K_{0.4}BiO_3$ and $BaPb_{0.8}Bi_{0.2}O_3$. The layered cuprates differ from the cubic bismuthates in terms of their highly anisotropic nature and their proximity to antiferromagnetism, either or both of which may be relevant to understanding the very large energy scale associated with the a-b-plane response of superconducting $YBa_2Cu_3O_7$. Our data indicate that this unusually large energy scale, $2\Delta_{a\text{-}b} \simeq 8kT_c$, is not associated with conventional strong coupling, since in that case modifications of the density of states above 2Δ should be clearly evident in both the reflectivity and conductivity ratios shown in Fig. 1.[21]

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