Far-Infrared Conductivity of the High- T_c Superconductor YBa₂Cu₃O₇

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We present reflectance data for the high-temperature superconductor $YBa_2Cu_3O_7$, and the Kramers-Kronig-transformed conductivity from 50 to 900 cm⁻¹. There is a rich phonon structure with peaks at 151, 191, 279, 310, 548, and 609 cm⁻¹. The continuous background is Drude type at high temperature but below the superconducting transition there is a region of suppressed conductivity consistent with a superconducting gap with the weak-coupling BCS value. Also associated with superconductivity are changes to the low-lying phonons and a plasma-type edge at 60 cm⁻¹.

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High-temperature superconductivity, first reported by Bednorz and Müller¹ in the Ba-La-Cu-O system, has been observed with a transition temperature of 90 K in $YBa_2Cu_3O_7$ ² It is important to find out if these novel materials get their unusual superconducting properties from an extremely strong interaction within the normal electron-phonon mechanism or if an entirely new excitation has to be invoked. One signature of a very strong electron-phonon interaction is the amount by which the energy gap to transition temperature ratio $2\Delta/kT_c$ exceeds the BCS weak-coupling limit of 3.53. In this work we report on reflectance measurements from 50 to 950 cm⁻¹ above and below the transition temperature in polycrystalline YBa₂Cu₃O₇. With our complete data set we are able to perform accurate Kramers-Kronig analysis to obtain the optical conductivity and the dielectric constant from 50 to 900 cm $^{-1}$.

The YBa₂Cu₃O₇ sample was prepared by firing Y₂O₃, BaCO₃, and CuO in a two-step procedure. First, the well-mixed powders were fired at 900 °C in a platinum crucible for 8 h, the green-black material was reground, pressed into pellets, and refired in 1 atm of O₂ at 925 °C for 10 h, cooled to 500 °C in 3 h and then removed from the furnace. A thermogravimetric analysis showed that the oxygen loss was consistent with the formula YBa₂Cu₃O_{6.85}. The lattice constants, measured with neutrons and with x rays at room temperature, are a = 3.8282 Å, b = 3.8897 Å, and c = 11.6944 Å. A complete neutron-diffraction structure analysis of the sample will be published elsewhere.³

Magnetic susceptibility measurements show that the sample has an onset of diamagnetism at 93 K. The superconducting fraction is close to unity as determined by the magnitude of the magnetic susceptibility. Fourprobe resistance measurements give a 2-K-wide transition at 89 K, and the magnitude of resistivity just above the transition temperature is $1000 \ \mu\Omega$ cm, rising to 2000 $\mu \Omega$ cm at room temperature.

The far-infrared reflectance was measured with single-reflection geometry near normal incidence. At the end of the experiment the sample was coated with a highly reflecting metal film, lead or aluminum for the low- and high-frequency regions, respectively. The metal-coated sample allows one to determine the effects of geometrical differences between the reference and the sample. This method allows absolute determination of the reflectance of the sample to a precision of better than 0.5%.

Figure 1 shows the reflectance of YBa₂Cu₃O₇ above the transition temperature at 105 K and at 55 K well below it. While the overall reflectance is superficially similar to other perovskites,⁴ there are unique temperature-dependent effects in this exotic material. One of the most striking features is the plasmon-type edge at 60 cm⁻¹ at low temperatures that disappears completely at high temperature. This feature has also been seen in the related material La_{1.85}Sr_{0.15}CuO₄. It is



FIG. 1. Reflectance of $YBa_2Cu_3O_7$ in the normal state (solid curve) and superconducting state (dashed curve).

closely associated with the superconductivity in that compound but is not the actual energy gap.⁵

The directly measured reflectance is difficult to interpret on the basis of elementary absorption processes since it involves both the real and imaginary part of the dielectric response. Kramers-Kronig analysis of the reflectance data yields the optical conductivity, which is a direct measure of microscopic energy-absorbing processes. Figure 2 shows the conductivity of $YBa_2Cu_3O_7$ for the two temperatures in the range from 50 to 900 cm⁻¹, obtained this way from reflectance measurements up to 4000 cm⁻¹.

The overall spectrum has a background of constant conductivity of the order of 250 $(\Omega \text{ cm})^{-1}$. Below the superconducting transition temperature, there is a marked depression of this level by about 100 $(\Omega \text{ cm})^{-1}$ extending to the neighborhood of 200 cm⁻¹. Above this frequency, the difference between normal and superconducting conductivities diminishes and there is a crossover at about 250 cm⁻¹ where the conductivity in the normal state is lower than in the superconducting one. These observations are consistent with a BCS electromagnetic response⁶ with an energy gap of about 200 cm⁻¹. It should be added, however, that in the BCS theory the conductivity in the gap region goes to zero; whereas in our curve, below 190 cm⁻¹, there remains 60% of the normal-metal conductivity.

Superimposed on the continuous Drude-type conductivity are three sets of sharp double peaks that we identify with phonon structure. At 105 K the pairs are at 151 and 191 cm⁻¹, at 279 and 310 cm⁻¹, and at 548 and 609 cm⁻¹. The lowest doublet is strongly temperature dependent. In particular, the peak at 191 cm⁻¹ narrows from a width of 14 cm⁻¹ in the normal state to a width of 8 cm⁻¹ in the superconducting state. Both of the



FIG. 2. The frequency-dependent conductivity of YBa_2Cu_3 -O₇ in the normal state (solid curve) and superconducting state (dashed curve). We associate the region of depressed conductivity below 200 cm⁻¹ with the superconducting energy gap. The sharp peaks are due to phonons, the peaks at 279 and 312 cm⁻¹ seem to be coupled to the electrons that participate in the superconducting transition.

peaks in the second doublet shift down in energy by 4 $\rm cm^{-1}$ upon entering the superconducting state. The two highest peaks, on the other hand, show no discernible temperature dependence when the material goes through the superconducting transition.

We will attempt to interpret our results in terms of ordinary BCS theory, assuming the material to have a gap of the order of 200 cm⁻¹ with a coupling to phonons. It should be noted, however, that we are presenting data on polycrystalline samples that are very likely quite anisotropic. Some of the structure must arise from grains that are oriented with the c axis, the nonsuperconducting direction, parallel to the electric vector of the incident light. These grains would present a set of phonon lines that would show little change on entering the superconducting phase. In fact, all of the lines except the pair around 300 cm⁻¹ fall into this inert category. Only the peaks at 279 and 310 cm⁻¹ seem to be directly coupled to the electron system. The lines at 151 and 191 cm⁻¹ seem to be coupled in a different way; they sharpen and gain intensity upon entering the superconducting state but do not shift in energy.

There seems to be no evidence of structure that could be associated with Holstein phonon emission seen in strong-coupling superconductors such as lead.⁷⁻⁹ If the 312-cm⁻¹ phonon, for example, is particularly strongly coupled to the electronic system, one would expect to see some structure at a frequency of $2\Delta + \omega$ or at 512 cm⁻¹. There appears to be considerable temperature-dependent absorption in this frequency range, but it is not possible to identify it as a characteristic Holstein structure.

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