NORMAL STATE INFRARED PROPERTIES OF HIGH-T_c SUPERCONDUCTORS

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Abstract

Infrared studies of ab-plane oriented $YBa_2Cu_3O_{7-\delta}$ films produced by laser ablation have been carried out at temperatures between 20 and 300 K. A strong, temperature-independent absorption is observed in the mid-infrared 150–6000 cm^{-1} energy range, with a temperature-dependent Drude contribution in the far infrared (below 400 cm^{-1}). The Drude plasma frequency is nearly independent of temperature while the relaxation rate depends linearly on temperature, reaching $1/\tau \approx 80$ cm^{-1} (0.01 eV) at 100 K. It is argued that the mid-infrared absorption is a direct particle-hole excitation rather than a Holstein emission process.

In this paper we discuss the infrared properties above T_c of epitaxially-grown laser-deposited films of $YBa_2Cu_3O_{7-\delta}$. There have been at least four interpretations of these properties.[1] Early studies used a Drude model with a constant, very short, relaxation time.[2,3,4] Because this simple model fails to account for the temperature dependence of the infrared properties,[5] recent free-carrier models assume either a strongly frequency-dependent relaxation rate and effective mass[6,7] or parallel conduction by several types of conduction electrons having differing concentrations and relaxation rates.[8] A fourth point of wiew, which we take, attributes the midinfrared absorption to a direct electronic excitation.[9,10,11,12,13]

Our films were made by pulsed eximer laser (248 nm, 30 ns) deposition[14] from a composite $YBa_2Cu_3O_{7-\delta}$target onto 700–750° C $SrTiO_3$ substrates. An oxygen background pressure of 200 mTorr was maintained during the deposition; subsequently, the films were superconducting with $T_c \approx 91$ K. The highly oriented films have the c axis normal to the surface. The normal-state resistivity is linear, with an extrapolated zero-temperature intercept of 0 ± 2 $\mu\Omega$-cm. The superconducting transition is sharp: $\Delta T_c \approx 0.2$ K. Reflectance measurements were made with a Bruker interferometric spectrometer over 50–5000 cm^{-1} (0.0062–0.62 eV) and with a grating monochromator over 1000–32,000 cm^{-1} (0.12–4 eV). A continuous flow cryostat was used to cool the sample to temperatures down to 20 K. The data were corrected for the known reflectance of the Al reference. Because of the high quality of the surfaces there was no need to coat the samples with a metal to estimate the scattering losses. The frequency-dependent conductivity $\sigma_1(\omega)$ and real dielectric function $\epsilon_1(\omega)$ were estimated from the reflectance using Kramers-Kronig analysis.[15]

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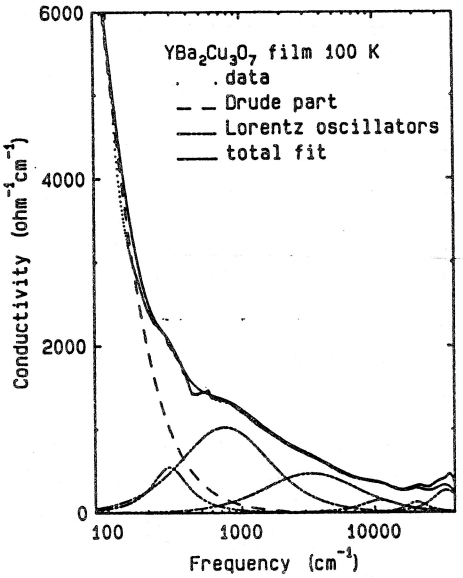

Fig. 1. Conductivity (as points) shown on a logarithmic frequency scale. The $\sigma_1(\omega)$ from fits to a sum of Drude (D) and Lorentzian (L) terms is shown, as well as the individual contributions to the sum. The Drude parameters are (in cm^{-1}): $\omega_{pD} = 8200$ and $1/\tau = 80$. The Lorentzian ω_{el}, ω_{pl}, γ_l are: L_1—290, 3000, 250; L_2—740, 9800, 1550; L_3—3300, 14 500, 7500; L_4—11 200, 9300, 9500; L_5—21 000, 8600, 9500; L_6—34 500, 17 000, 18 000.

Fig. 1 shows $\sigma_1(\omega)$ at 100 K over a wide frequency range. Also shown is a fit to a model dielectric function which consists of a Drude part and several Lorentzian contributions. The Drude part represents the contribution of free carriers while the Lorentzians are meant to represent excitations of bound electrons, interband transitions, etc. The total dielectric function is

$$\epsilon(\omega) = -\frac{\omega_{pD}^2}{\omega^2 + i\omega/\tau} + \sum_{l=1,6} \frac{\omega_{pl}^2}{\omega_{el}^2 - \omega^2 - i\omega\gamma_l} + \epsilon_\infty \qquad (1)$$

where ω_{pD} and $1/\tau$ are the plasma frequency and relaxation rate of the Drude carriers, while ω_{el}, ω_{pl}, and γ_l are the center frequency, strength, and width of the l^{th} Lorentzian contribution. The fit to Eq. 1 is shown in Fig. 1 along with the contributions of the individual terms.

To fit the broadband mid-infrared absorption requires terms at 290, 740, and 3300 cm^{-1}, although the wings of the first two Lorentzian terms are broader than the structure in $\sigma_1(\omega)$, indicating that these are not particularly good model dielectric functions. A better fit can be obtained with the aid of an electron-phonon coupling model recently proposed.[16] The mid-infrared bands have high oscillator strengths and contribute most of the bound electron contribution to the static dielectric constant, which we estimate to be $\epsilon_{1b}(0) \approx 300$. Other electronic excitations[17] occur at 11 200, 21 000, and 34 500 cm^{-1} (1.4, 2.6, and 4.3 eV).

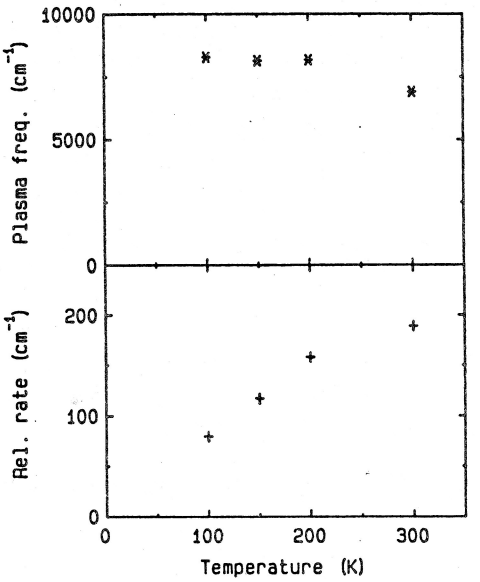

Fig. 2. Temperature dependence of the Drude plasma frequency and relaxation rate.

Fig. 2 shows the temperature dependence of the low-frequency Drude terms. The Drude plasma frequency is $\omega_{pD} \approx 8200$ cm^{-1}, nearly T-independent. The scattering rate is $1/\tau \approx 80$ cm^{-1} at 100 K and is essentially T-linear (like the resistivitiy) with zero intercept. The 100 K value corresponds to $\tau = 6.6 \times 10^{-14}$ sec and (if $v_F = 3 \times 10^7$ cm/sec) a mean free path of $\ell = 200$ Å. From $\omega_p \equiv \sqrt{4\pi ne^2/m^*}$, we can estimate $n/m^* \approx 0.7$–0.8×10^{21} free electrons/cm^3. Assuming one hole per $YBa_2Cu_3O_7$ formula unit this would imply $m^* \approx 6$–8 free-electron masses.

We turn now to the issue of whether the mid-infrared absorption in these materials is due to a strongly frequency-dependent scattering.[6,7] Frequency-dependent scattering arises from a Holstein[18] process, in which an electron absorbs a photon of energy $\hbar\omega$, emits some excitation at $\hbar\Omega$, and scatters. Energy conservation requires $\hbar\omega \geq \hbar\Omega$. By assuming a narrow band of excitations in the 300–500 cm^{-1} range strongly coupled ($\lambda \approx 3$–9) to the conduction electrons, Thomas *et al.*[6] and Collins *et al.*[7] were able to describe the strongly temperature dependent far-infrared behavior and the temperature-independent midinfrared absorption within a Holstein picture. No direct midinfrared absorption was included in these models.

There are two problems with this strong coupling picture. First, there would be a pronounced nonlinear dc resistance as $k_BT \rightarrow \hbar\Omega$, in disagreement with experiment where a highly linear resistivity with zero intercept is measured.[19] Second, *if there is a superconducting gap* then the Holstein process requires that the photon energy provide both the gap energy 2Δ and the energy of the emitted excitation $\hbar\Omega$. Therefore, the Holstein structure in $\sigma_1(\omega)$ is shifted by 2Δ to higher frequencies below T_c. No such shift is observed.

The T-linear resistivity also rules out several parallel conduction channels.[8] If there were temperature independent conduction in parallel with temperature-dependent conduction, the resistivity would saturate at high temperatures.

We conclude that the midinfrared absorption is a direct electronic excitation. Earlier we had suggested a (Frenkel) exciton,[10] but other possibilities obviously exist.[20]

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