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EVIDENCE FOR STRONG BOUND-ELECTRON–PHONON INTERACTION AT 52 meV IN $YBa_2Cu_3O_7$

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Recent infrared and neutron scattering data provide evidence for strong electron–phonon coupling in the 52 meV/420 cm^{-1} range in $YBa_2Cu_3O_7$. From a simple model of two coupled oscillators it is shown that a linear electron–phonon coupling can give the observed antiresonance structure in the frequency-dependent conductivity. We argue that the coupling is to the *bound* carriers responsible for the midinfrared absorption rather than to the superconducting free carriers.

1. Introduction

Recent inelastic neutron scattering measurements of Reichard et al. [1] on $YBa_2Cu_3O_7$ show a remarkable broad branch in the 55 meV/440 cm^{-1} region, near the zone boundary in the $\zeta00/0\zeta0$ direction. Reichard et al. interpret this as evidence for strong electron–phonon coupling. We show that this band has been also seen by several groups in the optical conductivity as an antiresonance at 52 meV/420 cm^{-1} superimposed on a continuous electronic absorption [2–5]. It also appears as a peak in the ratio of reflectances at different temperatures [6,7]. We calculate the frequency-dependent conductivity using a simple model of a linear interaction between a (bare) phonon at 433 cm^{-1} and an electronic continuum. The model accounts for the position, shape, temperature dependence and doping level dependence of the optical conductivity in the 150–2000 cm^{-1} region in $YBa_2Cu_3O_7$. We show that the electrons which couple strongly to the oscillator at 433 cm^{-1} are not the free carriers but bound electrons responsible for the midinfrared band. Indeed, there is no evidence that the free carrier–phonon coupling is strong. Together these new results point to the complexity of the low-lying states in the new superconductors.

Many measurements [6–11] have shown that the infrared response of $YBa_2Cu_3O_7$ is characterized by two contributions: a low-frequency free carrier or Drude absorption and a strong electronic midinfrared band. The free carrier response is conventional: the infrared data are consistent with Drude behavior with a scattering rate that varies linearly with temperature [5] above T_c and a temperature-independent plasma frequency of about 1.2 eV, a value in excellent agreement with other experiments [12,13]. In the superconducting state all the oscillator strength of the Drude absorption condenses to a delta function centered at zero frequency, as expected for a clean limit superconductor [5,11].

The midinfrared band in contrast has a complex structure [14]. Absent in the undoped nonsuperconducting $YBa_2Cu_3O_6$, the band is, at low doping levels, centered at 5000 cm^{-1} (0.6 eV). As T_c approaches 90 K, the oscillator strength grows at lower frequencies. Independent of doping level there is a sharp minimum in the optical conductivity at 420 cm^{-1}. Above T_c the low-frequency portion of the midinfrared band overlaps the Drude absorption from the free carriers. However, in the superconducting state when the free carriers have condensed, the conductivity is zero (to within experimental error) below 19 meV/150 cm^{-1}.

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Figure 1 shows the midinfrared band [5] at several temperatures in a high-quality laser-ablated film with a T_c of 90 K. These curves were calculated by subtracting from the frequency-dependent conductivity (determined by Kramers–Kronig analysis of the reflectance) the Drude contribution. The Drude parameters were estimated by least squares fitting to the low-frequency reflectance. The two characteristic features of the midinfrared conductivity are evident: the onset or gap-like feature at 150 cm^{-1} and a dip or antiresonance at 420 cm^{-1}. Whether the gap at 150 cm^{-1} is associated with superconductivity is controversial. The structure at 420 cm^{-1} is definitely not the superconducting gap: it is present in the normal state and its position is independent of doping level, T_c and temperature.

2. The model

To describe the interaction between the phonon at 433 cm^{-1} and the electronic continuum we use a simple model of two coupled oscillators to describe the electronic and lattice degrees of freedom. The electronic absorption is represented by a Lorentzian oscillator centered at frequency ω_e with an oscillator strength ω_{pe} and damping γ_e. The phonon is represented by a harmonic oscillator, with bare frequency ω_0 and damping γ_0. The electrons and phonons are coupled by a linear force constant g. The model for the electronic band is too simple in that the gap at 150 cm^{-1} is not included. Within the model, the complex dielectric function is

$$\epsilon(\omega) = \epsilon_\infty + \epsilon_D + \omega_{pe}^2 \Big/ \left[\omega_e^2 - \frac{g^2}{m^* M \omega_0^2} D(\omega) - \omega^2 - i\omega\gamma_e \right], \qquad (1)$$

where ϵ_∞ is the contribution of higher frequency interband processes, ϵ_D the Drude contribution, m^* the electronic effective mass, M the reduced mass of the phonon normal mode, and

$$D(\omega) = \frac{\omega_0^2}{\omega_0^2 - \omega^2 - i\omega\gamma_0}. \qquad (2)$$

It is convenient to introduce a frequency ω_g that characterizes the electron–phonon coupling by $\omega_g^2 = g/\sqrt{m^* M}$.

Figure 2 shows the conductivity calculated with this model. The bare phonon frequency of 433 cm^{-1} was taken from the 300 K data and the remaining parameters of the model were fit to the midinfrared

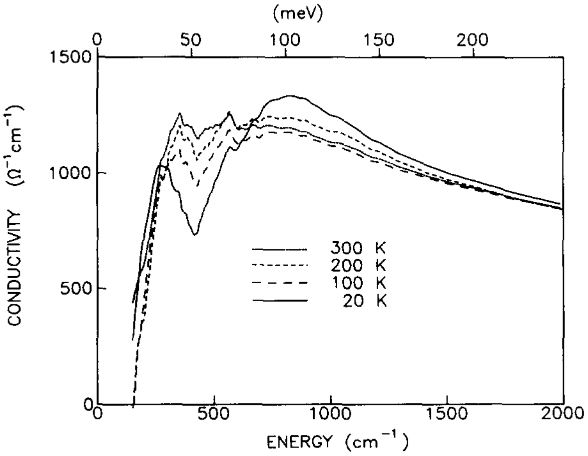

Fig. 1. Midinfrared absorption in an *ab*-plane oriented film of $YBa_2Cu_3O_{7-\delta}$ according to Kamarás et al. [5]. Shown is the difference between the total conductivity and the Drude contribution to the conductivity at four temperatures. We interpret the sharp minimum that appears at 420 cm^{-1} as a result of an interaction between a bare phonon at 433 cm^{-1} and the electronic continuum.

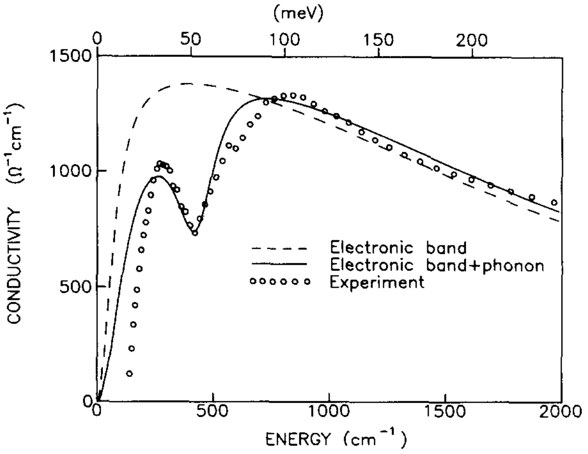

Fig. 2. Calculated conductivity for a model of two coupled oscillators. An electronic band represented as a highly damped harmonic oscillator, shown as the dashed curve is coupled to a phonon at 433 cm^{-1} resulting in the solid curve. The circles show the experimental conductivity at 20 K from ref. [5].

conductivity of Kamarás et al. [5] at 20 K shown in fig. 1. The parameters used are shown in table I. It is evident that the fit at low frequencies is poor. In particular, the 150 cm^{-1} gap is not well described by the simple oscillator model for the midinfrared conductivity.

Rice, in discussing a closely related problem in organic conductors, the electron-molecular vibration interaction [15], has introduced the dimensionless constant λ to describe the strength of the coupling given by $\lambda=\omega_g^4/\omega_e^2\omega_0^2$. Our fit, subject to a large uncertainty in the parameter ω_e, gives a value of λ of the order of unity consistent with strong coupling of the bound electrons to this particular branch. Since the frequency ω_g is of the order of ω_e it can be argued that the carriers responsible for the midinfrared conductivity are bound by the coupling to the phonon at 420 cm^{-1}; in other words they are localized by lattice polarization.

The coupled oscillator model explains in a simple way the apparent inertness of the midinfrared mode to changes in temperature and doping level, since phonon frequencies are generally only weakly affected by doping and temperature [16]. Nevertheless, we can see from the experimental curves of Kamarás et al. a softening of the mode in going from room temperature where the antiresonance is at 433 cm^{-1} to 20 K where, as a result of the apparent increase in coupling it shifts to 420 cm^{-1}. The model gives exactly the sample softening as the coupling constant ω_g is allowed to vary from 0 to 400 cm^{-1}.

3. Discussion

We have shown in the previous section that the detailed spectrum of the midinfrared conductivity can be understood in terms of an electronic continuum coupled very strongly to a phonon at 420 cm^{-1}. We now turn to a discussion of the relevance of this model to high-temperature superconductivity.

Table I
Parameters used in two-oscillator fit to the conductivity.

Parameter	Value (cm^{-1})
ω_{pe}	13500
ω_e	400
γ_e	2200
ω_0	433
γ_0	160
ω_g	400

There seems to be little evidence for a general strong interaction between the Drude electrons and phonons in the oxides. The magnitude of the scattering rate obtained by Drude fits [5] in the best sample is on the order of $1.2kT$, which, in the formula $1/\tau=2\pi\lambda kT$ [17] gives $\lambda=0.2$. An estimate based on the lack of curvature and saturation of the DC resistivity [18] gives $\lambda=0.3$. Changes in phonon width on passing through T_c [9,19] are generally less than 1% and point to values of λ that are comparable to estimates for the A15 compounds [20].

It is clear from fig. 1 that the free carriers do not contribute to the structure in $\sigma_1(\omega)$ in the 300–500 cm^{-1} region. As the free-carrier contribution to $\sigma_1(\omega)$ narrows and then condenses to a delta function with decreasing temperature, the 420 cm^{-1} minimum deepens and becomes more prominent in the data. This occurs as the free carrier contribution to $\sigma_1(\omega)$ at these frequencies is decreasing (because $\omega\tau\gg 1$). If the free carriers did contribute, one would expect to see the opposite: a weakening of the structure with decreasing temperature, particularly through the superconducting transition.

Another sort of coupling to free carriers would be via a Holstein [21] process. Such a coupling, if strong, would show u by the appearance of a Holstein sideband at the phonon frequency ω_0 above T_c. Below T_c, in the superconducting state, this sideband would shift to a frequency of 2Δ plus the phonon frequency ω_0 [22]. No such sideband has been seen in any of the low-temperature conductivity spectra of $YBa_2Cu_3O_7$ that have been published. This observation is also in agreement with a picture of a weak interaction between the normal phonons and the free carriers.

The results of Reichard et al. [1] suggest a strong electron–phonon coupling for one phonon branch only. Our results here show that the coupling is mainly to the bound carriers responsible for the midinfrared band and not to the free carriers that undergo the superconducting condensation. Thus it is unlikely that this mode is directly involved in superconducting pairing.

Several issues remain open. The model of two coupled oscillators used here is strictly for the convenience of a closed analytical formula for the dielectric function. It fails to describe the gap-like onset of the midinfrared band at 150 cm^{-1}. The overall nature of this band is also not clear. From the lack of changes associated with the entry to the superconducting state the band lacks the characteristics of a Holstein band. However, an explanation in terms of a simple interband absorption also seems forced since both initial and final states in the relevant bands have to be within 150 cm^{-1} of the Fermi surface and there seems to be little of the temperature dependence one expects of absorption within kT of the Fermi surface.

Further analysis of the neutron scattering results should aid in identifying the location of the 420 cm^{-1} mode in the structure of $YBa_2Cu_3O_7$ and therefore the location of the electrons that produce the midinfrared band.

Acknowledgements

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