Evidence for *a*-*b*-Plane Coupling to Longitudinal *c*-Axis Phonons in High-T_c Superconductors

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We show that the strong absorption structure observed in the a-b-plane optical conductivity of the high-T_c superconductors is correlated with c-axis longitudinal optical (LO_c) phonons. This suggests a resolution to the long-standing controversy over the origin of these features. The interaction with LO_c phonons is forbidden when the incident wave vector **q** is normal to the c axis which leads to the surprising result, confirmed with experiments, that the optical properties along a or b are different when measured on the a-b-plane face and a face containing the c axis.

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The unconventional response of the high-T_c superconductors to electromagnetic radiation has been well established [1]. A prominent manifestation of the non-Drude behavior is the "knee" that develops in the a-b-plane reflectance of $YBa_2Cu_3O_{7-\delta}$ near 400 cm^{-1} at low temperatures leading to a "notch"-like absorption feature in the optical conductivity [2,3]. This structure has been the focus of much discussion. It has been variously attributed to the superconducting energy gap [4] and to phonons [5].

It has become clear that $YBa_2Cu_3O_{7-\delta}$ is not the only cuprate superconductor to exhibit such structure. $Bi_2Sr_2CaCu_2O_8$ [6], $Tl_2Ba_2Ca_{n-1}Cu_nO_{2n+4}$ [7,8], $Pb_2Sr_2(Y/Ca)Cu_3O_8$ [9], and $Nd_{2-x}Ce_xCuO_{4-\delta}$ [10] all show strong low-frequency absorption features in the non-Drude component of the a-b-plane optical conductivity. In each case the structure appears as a minimum in $\sigma_1(\omega)$. There are, however, significant material to material differences in the number, magnitude, width, and position of the minima. For example, whereas $YBa_2Cu_3O_{7-\delta}$ is dominated by one broad feature near 420 cm^{-1} [Fig. 2(b)], $Tl_2Ba_2CaCu_2O_8$ shows two deep minima at 350 and 600 cm^{-1} [Fig. 3(b)], while $Pb_2Sr_2(Y/Ca)Cu_3O_8$ exhibits two closely spaced sharper minima at 525 and 585 cm^{-1} and a third broader feature centered near 435 cm^{-1} [Fig. 2(a)].

The relative independence of the position of this structure to both temperature [3] and level of doping [2] suggests that it is unrelated to the superconducting energy gap and that phonons are involved. It has been shown that a sharp level, interacting with an electronic continuum, results in a notchlike minimum in the spectrum of the continuum (an antiresonance) if the external field does not interact with the sharp transition [11,12]. This phenomenon, discussed by Fano for atoms [11] and Rice for phonons and an electronic continuum [12], is common in quasi-one-dimensional organic conductors, where charge [13] or spin density [14] waves break symmetry and couple the low-lying charge-transfer continuum to totally symmetric phonon modes not normally ir active [12]. In high-T_c superconductors, the interaction of the phonons is with bound (midinfrared) and not Drude-like carriers because the antiresonant structure remains when the superconducting carriers condense into a zero-frequency delta function.

In what follows we demonstrate that this structure is caused by the interaction of c-axis, longitudinal optical (LO_c) phonons with the carriers responsible for the midinfrared absorption. We do this by first showing that there is a remarkable similarity between the structure in the a-b-plane conductivity [measured, as is customary, with the propagation vector **q** along the c direction, **q**$\parallel$**c**, Fig. 1(a) inset, called parallel geometry henceforth] and the c-axis-polarized loss function $\mathrm{Im}(-1/\epsilon^c)$. Second we

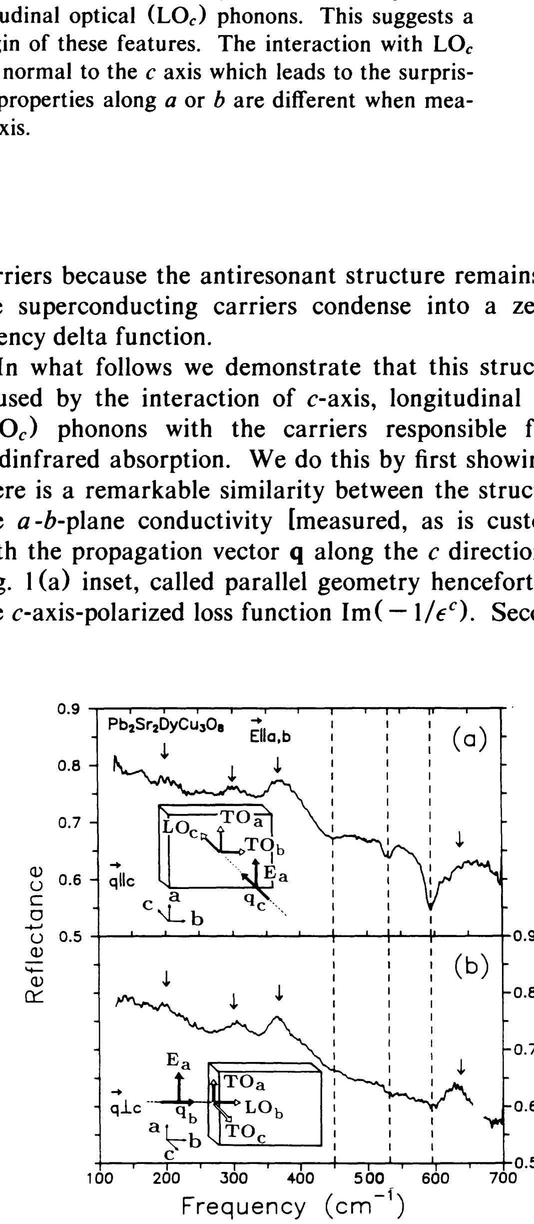

FIG. 1. **E**$\parallel(a,b)$ reflectance of $Pb_2Sr_2DyCu_3O_8$ at room temperature for (a) **q**$\parallel$**c** (parallel geometry) and (b) **q**$\perp$**c** (perpendicular geometry). Direct coupling to unscreened TO_a and TO_b modes is indicated by the small arrows in both spectra. Antiresonances due to strong electron-phonon coupling to longitudinal c-axis modes in the parallel geometry are indicated by the vertical dashed lines. This coupling is forbidden in perpendicular geometry, as can be seen by comparing the insets to (a) and (b), which show the polarization vectors of phonons allowed by momentum conservation. To first order the electron-coupled modes are absent in the spectrum (b).

show that on symmetry grounds, this effect will not be observed if the a-b conductivity is measured in a geometry where light propagates along the a or b axis [$\mathbf{q} \perp \mathbf{c}$, Fig. 1(b) inset, called perpendicular geometry]. Because flux-grown crystals have small faces normal to the planes, perpendicular geometry is not generally used to study the in-plane conductivity. We finally show that the a-b conductivity is different in perpendicular geometry: There are no antiresonances. Previously it was shown that the notchlike minimum observed in the a-b-plane optical conductivity of $YBa_2Cu_3O_{7-\delta}$ could be modeled (using Lorentzian oscillators) by coupling a phonon to an electronic continuum [5]. Here we use experimental data to show which specific phonons are responsible, and the unexpected consequences thereof.

Figure 1(a) shows the a-b-plane infrared reflectance of $Pb_2Sr_2DyCu_3O_8$, a poorly metallic member of the $Pb_2Sr_2RCu_3O_8$ system, where R is a rare earth. Measured in parallel geometry, this system shows clearly an evolution of electron-coupled phonon absorption with doping [9]. The doping-induced modes appear as antiresonances; their positions are indicated by the vertical dashed lines. Figure 1(b) shows the a-b reflectance measured in perpendicular geometry. While the regular transverse optical (TO) phonons (denoted by arrows) have changed little between the two geometries, the antiresonances are absent in the geometry where $\mathbf{q}$ is perpendicular to $\mathbf{c}$. We will show that this surprising result is a direct consequence of the interaction of the continuum with LO_c modes.

The Kramers-Kronig-derived optical conductivity in the geometry where the antiresonances can be seen is shown in the upper curve of Fig. 2(a). Although exhibiting metallic, linearly decreasing resistivity and a T_c of 75 K, $Pb_2Sr_2DyCu_3O_8$ has a low dc conductivity and thus, in contrast to other high-T_c superconductors like $YBa_2Cu_3O_{7-\delta}$ and $Bi_2Sr_2CaCu_2O_8$, the non-Drude component dominates the optical properties. Consequently the symmetry-allowed normal optical phonons are not well screened and appear as *peaks* in the optical conductivity (e.g., near 640, 355, 300, and 200 cm^{-1}). The electron-coupled modes, on the other hand, absent in the undoped parent compound [9], appear as *minima* at 435, 525, and 585 cm^{-1}.

The dashed curve of Fig. 2(a) shows the experimentally obtained dielectric loss function, $\mathrm{Im}(-1/\epsilon)$ of $Pb_2Sr_2DyCu_3O_8$ with the electric field $\mathbf{E}$ polarized along the c axis. A striking similarity between the position, width, and relative strength of the peaks of the c-axis loss function and the minima in the real part of the a-b-plane optical conductivity $\sigma_1^{a\text{-}b}$ is evident. The peaks in the dielectric loss function yield the positions of longitudinal optical phonon modes. This correlation suggests that these features are caused by an interaction (henceforth referred to as "coupling") of the a-b-plane electronic continuum with LO_c phonons. For comparison, the contribution of the phonons to the c-axis dielectric loss function of $YBa_2Cu_3O_{7-\delta}$ at 100 K determined from recent measurements of Homes *et al.* [15] by subtracting the Drude component is shown in Fig. 2(b) along with the a-b conductivity of Kamáras *et al.* [3]. The notch in $\sigma_1^{a\text{-}b}$ coincides with a broad peak in $\mathrm{Im}(-1/\epsilon^c)$. Additional weaker features in $\sigma_1^{a\text{-}b}$ can be associated with smaller loss-function peaks.

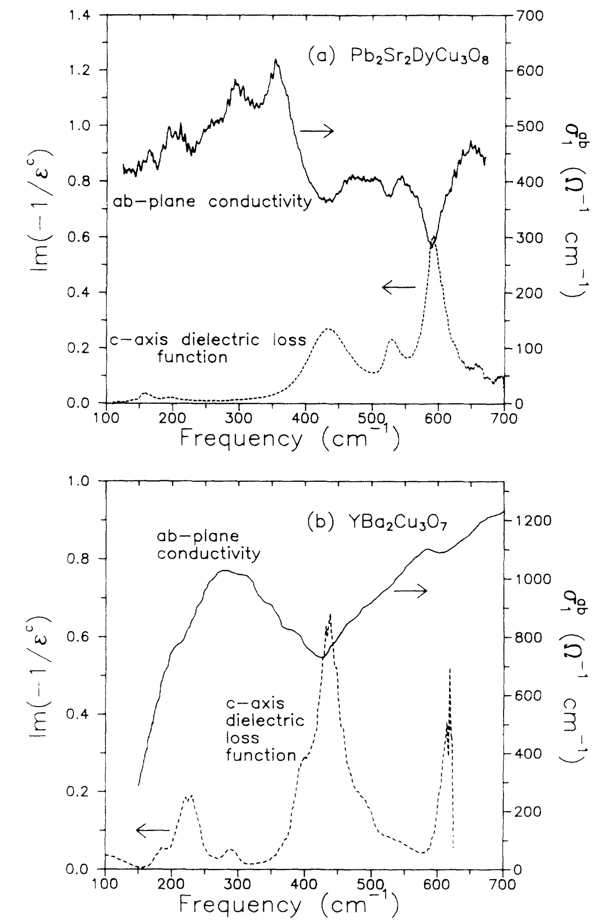

FIG. 2. Comparison of the a-b-plane midinfrared component of the optical conductivity of (a) $Pb_2Sr_2DyCu_3O_8$ and (b) $YBa_2Cu_3O_{7-\delta}$ with the dielectric loss function of the c-axis phonons. Note the striking correspondence of the position, width, and relative strength of the minima in $\sigma_1^{a\text{-}b}$ and the peaks in $\mathrm{Im}(-1/\epsilon^c)$. $\sigma_1^{a\text{-}b}$ of $YBa_2Cu_3O_{7-\delta}$ is after Kamáras *et al.* (Ref. [3]), while $\mathrm{Im}(-1/\epsilon^c)$ is determined after Homes *et al.* (Ref. [15]).

In Fig. 3 further evidence that association of antiresonances and c-axis LO phonons is common to the cuprates is derived from similar comparisons between the non-Drude component of the a-b-plane optical conductivity and the dielectric loss function of the c-axis phonons for $Nd_{2-x}Ce_xCuO_{4-\delta}$, $Tl_2Ba_2CaCu_2O_8$, and $Bi_2Sr_2CaCu_2O_8$ [16]. In each case minima in $\sigma_1^{a\text{-}b}$ correspond to peaks in $\mathrm{Im}(-1/\epsilon^c)$, providing evidence for a universal interaction of c-axis phonons with the a-b-plane midinfrared continuum in the cuprate superconductors.

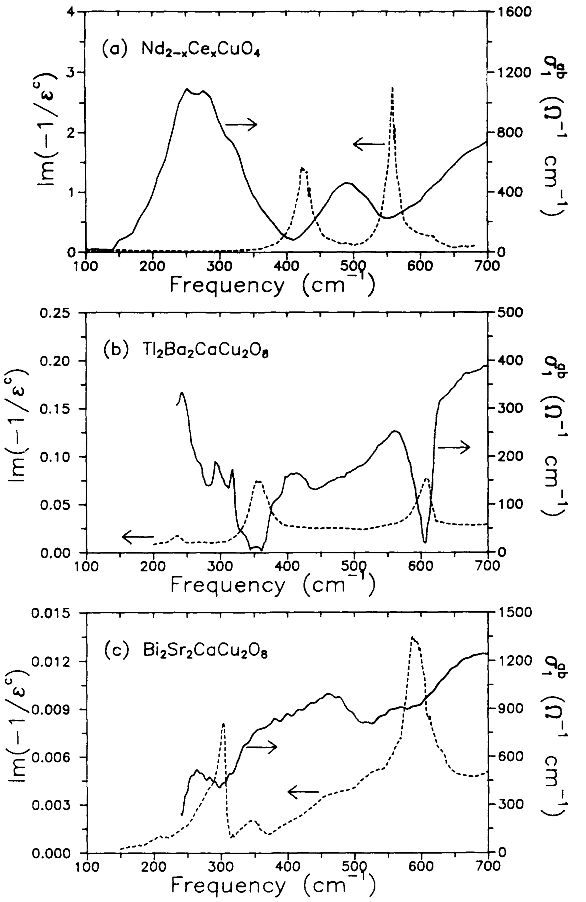

FIG. 3. Further evidence for a universal coupling of the a-b-plane optical conductivity to c-axis phonons is derived from the comparison of $\sigma_1^{a\text{-}b}$ with $\mathrm{Im}(-1/\epsilon^c)$ for (a) $Nd_{2-x}Ce_xCuO_{4-\delta}$, (b) $Tl_2Ba_2CaCu_2O_8$, and (c) $Bi_2Sr_2CaCu_2O_8$. The $\sigma_1^{a\text{-}b}$ curves shown in (a), (b), and (c), respectively, are after Hughes *et al.* (Ref. [10]), Foster *et al.* (Ref. [7]), and Reedyk *et al.* (Ref. [6]), while the $\mathrm{Im}(-1/\epsilon^c)$ curves were obtained via Kramers-Kronig analysis of the reflectance measured by Tajima *et al.*, Zetterer *et al.*, and Kamáras *et al.* (Ref. [16]), respectively.

The hypothesis that the coupling is to c-axis longitudinal modes explains the difference between the spectra in parallel and perpendicular geometry in Fig. 1. In parallel geometry, Fig. 1(a), the $\mathbf{q}$ of the incident light is in the c direction and can excite LO phonons propagating in the c direction (positions indicated by vertical dashed lines). In perpendicular geometry, Fig. 1(b), there is no incident momentum component in the c direction and the excitation of LO_c phonons is forbidden.

TO_a phonons can be excited directly by the electric field along a (peaks denoted by arrows) in both geometries. Momentum selection rules allow the antiresonant coupling to these modes as well as to (provided an appropriate dipole moment exists) TO_b and LO_c in parallel geometry and TO_c and LO_b in perpendicular geometry. Since in the usual parallel geometry there is little evidence for an electron-coupled interaction with the allowed a-b-plane TO phonons, the coupling to these modes must be small. The results of the perpendicular measurement of Fig. 1(b) lead to the conclusion that coupling to TO_c and LO_b is also weak. Since we do not distinguish between a and b in pseudotetragonal $Pb_2Sr_2DyCu_3O_8$ (there are no chains as in $YBa_2Cu_3O_{7-\delta}$), we can conclude that a strong electron-phonon interaction only couples the a-b-plane carriers responsible for the midinfrared absorption and longitudinal c-axis phonons.

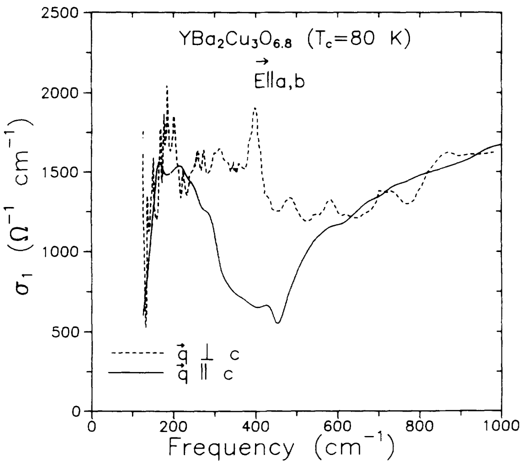

FIG. 4. Comparison of the optical conductivity of $YBa_2Cu_3O_{6.8}$ ($T_c = 80$ K) measured in the usual parallel geometry (solid curve, $T = 10$ K after Orenstein *et al.*; Ref. [17]) and in perpendicular geometry (dashed curve, $T = 10$ K after Bauer; Ref. [18]). Note that the prominent minimum at 420 cm^{-1} is absent in perpendicular geometry.

Finally, in Fig. 4 we return to the optical conductivity of $YBa_2Cu_3O_{7-\delta}$ measured in the usual way on the a-b-plane face (parallel geometry) by Orenstein *et al.* [17] (solid curve) and compare this with the only measurement of the a-b optical properties that we are aware of done on a face containing the c axis ($\mathbf{q} \perp \mathbf{c}$, by Bauer [18]; dashed curve). In this system too, the prominent minimum at 420 cm^{-1} vanishes in the perpendicular geometry.

The results of Figs. 2 and 3 suggest that the coupling is to all higher-frequency modes, i.e., oxygen bond stretching and bending motions, if not to the entire group of c-axis phonons, rather than to specific branches exhibiting a strong doping-induced electron-phonon interaction as was originally proposed [19]. Furthermore, the correlations of the infrared spectra with neutron and tunneling data [20] suggest that the c-axis LO phonons form the structure in all these measurements. A subtle point is whether the coupling is to the c-axis LO modes or more generally to the long-range electric field, described by the dielectric loss function. The fact that no coupling to transverse modes along the c axis is observed and the remarkable similarities displayed, not only in the position but in the relative strength and width, between the peaks in the loss function and the minima in the optical conductivity suggest the latter. In $Pb_2Sr_2DyCu_3O_8$ and $YBa_2Cu_3O_{7-\delta}$, where the similarity is most striking, the results are also most reliable because of their relative independence to subtraction of a dominant, highly frequency-dependent Drude component.

A question that remains in the connection of these features to superconductivity. The resonances are associated with the midinfrared conductivity, and not the Drude carriers that condense. For example, both $La_{2-x}Sr_xNiO_4$ [21] (isostructural to $La_{2-x}Sr_xCuO_4$) and $Pb_2Sr_2RCu_3O_8$ with R=Sm, Tb, which are lightly doped but nonsuperconducting, exhibit such structure. The midinfrared band is, however, a common feature of metallic oxides [22,23], while only the layered structure associated with the high-T_c cuprates seems to yield the electron-coupled modes. It is important to extend these studies to other examples before a clear correspondence can be made between high-T_c superconductivity and the presence of the interaction between the in-plane midinfrared absorption and the LO_c phonons. Thus it seems that while the resonances are a significant feature of the high-T_c superconductors, the LO_c-midinfrared coupling may not contribute to the superconducting pairing.

By surveying the optical properties of several high-T_c cuprate superconductors, we have found that the low-frequency absorption structure observed in the non-Drude component in the a-b-plane optical conductivity can be attributed to normally forbidden c-axis longitudinal optical phonons which are rendered accessible to this experimental configuration by a strong electron-phonon coupling to the midinfrared continuum. This would appear to resolve the long-standing controversy over the origin of these features, and may be related to the presence of superconductivity in these materials. A further consequence of this finding is that the frequency-dependent a-b-plane electronic properties are not decoupled from the c-axis phonons, even in the highly anisotropic Bi- and Tl-based materials, suggesting that in terms of the optical properties, at least, these materials have a three-dimensional aspect.

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