Application of the polaron-transport theory to $\sigma(\omega)$ in $Tl_2Ba_2Ca_{1-x}Gd_xCu_2O_8$, $YBa_2Cu_3O_{7-\delta}$, and $La_{2-x}Sr_xCuO_4$

D. Mihailović,* C. M. Foster, K. Voss, and A. J. Heeger
Institute for Polymers and Organic Solids and Department of Physics, University of California, Santa Barbara, Santa Barbara, California 93106
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We analyze the frequency-dependent photoinduced infrared conductivity, $\sigma_P(\omega)$, obtained from photoinduced absorption measurements of the insulators $Tl_2Ba_2Ca_{0.98}Gd_{0.02}Cu_2O_8$, $YBa_2Cu_3O_{6.3}$, and La_2CuO_4 in terms of $\sigma_{PT}(\omega)$ calculated from nonadiabatic polaron-transport theory. The calculated $\sigma_{PT}(\omega)$ is in good agreement with the experimental $\sigma_P(\omega)$ in the midinfrared. We also compare $\sigma_P(\omega)$ with the infrared conductivity, $\sigma(\omega)$, of the high-T_c superconductors $Tl_2Ba_2CaCu_2O_8$, $YBa_2Cu_3O_7$, and $La_{1.85}Sr_{0.15}CuO_4$. The similar spectral shape and systematic trends in both $\sigma_P(\omega)$ and $\sigma(\omega)$ indicate that the carriers in the concentrated (metallic) regime retain much of the character of the carriers in the dilute (photoexcited) regime. Together, these results imply that in the superconducting cuprates and in their "parent" insulators, the carriers are polarons dressed with a phonon polarization cloud.

I. INTRODUCTION

An extensive body of infrared reflectance data has established that the normal-state frequency-dependent conductivity $\sigma(\omega)$ of the high-temperature superconductors, $Tl_2Ba_2CaCu_2O_8$, $YBa_2Cu_3O_7$, and $La_{1.85}Sr_{0.15}CuO_4$ consists of a narrow, temperature-dependent, Drude-like peak centered at zero frequency, and a broad, temperature-independent excitation in the midinfrared, the so-called "mid-ir" feature.[1–5] The systematic appearance of the mid-ir feature in $\sigma(\omega)$ and the direct correlation of its intensity with doping has been established experimentally for many perovskites (including $Ba_{1-x}Pb_xBiO_3$, $La_{2-x}Sr_xNiO_4$, $SrTiO_3$, and $BaTiO_3$).[6–9] In the cuprate perovskites, similar correlations exist between the mid-ir peak and high-temperature superconductivity at moderate doping levels (less than ≈ 0.5 holes/Cu).[1] Further doping into a metallic phase reduces the intensity of the mid-ir feature[10] as well as T_c.[11,12] This close correspondence of the mid-ir intensity with T_c in cuprate perovskites suggests that the origin of $\sigma(\omega)$ in the mid-ir warrants further investigation, particularly in relation to the mechanism for high-temperature superconductivity.

The mid-ir feature in high-T_c superconductors has been interpreted by many groups. In the case of $YBa_2Cu_3O_{7-\delta}$, it has been attributed to a direct electronic transition distinct from the Drude-like free carrier contribution at low frequency,[2] to a contribution from free holes, but with an ω-dependent scattering rate arising from either a polaron shakeoff of dressed carriers (which are renormalized by strong interactions with phonons, spin waves, etc.),[2,4] or to an intrinsic ω dependence of the scattering mechanism.[3,13]

In systems which have large electronic bandwidth, carrier transport can be accurately described by adiabatic processes in which the crystal lattice is assumed to be static with respect to carrier motion (i.e., the Drude model, Fermi liquid theory, etc.). In narrower band systems, carrier motion becomes nonadiabatic. When the sound velocity becomes comparable to the Fermi velocity, as has been shown to be the case in the cuprates,[14] carrier motion is intimately linked to lattice vibrations. In doped titanates, for example, $BaTiO_3$ and $SrTiO_3$, the origin of mid-ir features in $\sigma(\omega)$ and their relationship to transport mechanisms has been a subject of study for many years.[9,15–20] Both adiabatic and nonadiabatic mechanisms have been proposed for modeling the transport properties. The application of polaron transport theory (PTT), in which carriers move nonadiabatically with respect to the lattice, eventually proved successful as a model of carrier transport in these materials.[9,15–20]

In nonadiabatic PTT, carriers interact with both optical and acoustic phonons; the acoustic phonons determine the temperature-dependent contribution to $\sigma(\omega)$ near zero frequency[16] and the optical phonons give rise to a broad temperature-independent "shakeoff" peak in $\sigma(\omega)$ in the midinfrared for temperatures $T < T_D$, where T_D is the Debye temperature.[15,19,20] In addition to successfully describing the shape of the mid-ir peak in $\sigma(\omega)$ upon doping,[17,18] the model also correctly predicted the dc transport properties[16,18] of the doped titanates.

In this paper, we apply PTT as a model of the frequency-dependent transport process experimentally observed as the mid-ir features in $\sigma(\omega)$ in the high-T_c cuprates, restricting ourselves to carrier interactions with optical phonons.[15,17] The calculated $\sigma_{PT}(\omega)$ is fit to the photoinduced infrared conductivity, $\sigma_P(\omega)$, obtained from photoinduced absorption (PIA) measurements of the insulating precursor materials,[21–24] where the carrier concentration is in the dilute limit and carrier-carrier interactions are thus assumed to be negligible. These spectra are compared to the infrared conductivity $\sigma(\omega)$ obtained from Kramers-Kronig analysis of reflectivity measurements in the doped, metallic (superconducting) materials. The similar spectral shape and consistent trend toward lower energy of the mid-ir peak in both $\sigma_p(\omega)$ and $\sigma(\omega)$ indicate that carriers in the concentrated (metallic) regime retain much of the character of carriers in the dilute (photoexcited) regime. We conclude that the mid-ir absorption in $Tl_2Ba_2CaCu_2O_8$, $YBa_2Cu_3O_{7-\delta}$, and La_2CuO_4 can be assigned to nonadiabatic polaron (or possibly bipolaron) transport and suggest that the differences between $\sigma_P(\omega)$ and $\sigma(\omega)$ can be attributed to the onset of carrier-carrier interactions in the concentrated regime. Since we cannot make any experimental distinction between polaron or bipolaron transport, we refer to the carriers simply as polarons.

II. CALCULATION OF $\sigma(\omega)$ AND COMPARISON WITH EXPERIMENTAL RESULTS

The theoretical treatment of polaron transport is based on Holstein's molecular crystal Hamiltonian;[25] $\sigma_{PT}(\omega)$ is calculated[15,16,19] in both the low-temperature ($T\rightarrow 0$) and high-temperature limit ($k_BT>\omega_D/2$), where ω_D is the Debye frequency and k_B is Boltzmann's constant. The $\sigma_{PT}(\omega)$ spectrum reflects the shakeoff of the optical phonon cloud (the localized distortion) from the polaron as a result of nonadiabatic transport from site to site, with the peak[19] in the spectrum corresponding approximately to $2E_b$, where E_b is the polaron binding energy. Whereas the original calculation was done by Reik[15,17] in the limit of strong coupling, [i.e., with $(2E_b/k_BT)(a/r)^3\gg 1$, where k_B is Boltzmann's constant, a is the lattice constant, and r is the polaron radius], the theory has been extended by Emin[19] to the weak-coupling regime to include large polarons where the range of the polaron extends beyond one unit cell.

In PTT, the polarons are considered as noninteracting particles[15–20] in the dilute limit. We can probe the transport of such isolated carriers by measurements of $\sigma_P(\omega)$, and we can probe the transport of carriers in the concentrated regime from measurements of $\sigma(\omega)$. However, we expect departures from the theoretically calculated $\sigma_{PT}(\omega)$ in a system with a high density of carriers exhibiting collective phenomena (e.g., superconductivity), as is the case in the high-T_c cuprates where $\sigma(\omega)$ is the response of the carriers at a density in the range of 0.1–0.5 per unit cell.

In the PIA technique, $\sigma_P(\omega)$ is derived from $(-\Delta T/T)$ of the insulating precursor compounds of the same materials, where T is the transmission, and ΔT is the change in transmission upon photoexcitation with weak laser light ($\cong$40 mW/cm^2). In the insulator limit, where $\epsilon_1\gg\epsilon_2$, the photoinduced infrared conductivity $\sigma_P(\omega)$ is related to $-\Delta T/T$ by

$$\sigma_P(\omega)=(nc/4\pi d)(-\Delta T/T)\ ,$$

where $n=\sqrt{\epsilon_1}$ is the refractive index, d is the absorption length, and c is the speed of light. The response of the system in the dilute limit, $\sigma_P(\omega)$, should correspond closely to the idealized theoretical model. Thus we compare $\sigma_P(\omega)$ with the predictions of PTT. We fit $\sigma_{PT}(\omega)$ to the mid-ir $\sigma_P(\omega)$ in $Tl_2Ba_2Ca_{0.98}Gd_{0.02}Cu_2O_8$,[23] $YBa_2Cu_3O_{6.3}$,[22] and La_2CuO_4.[21] This approach is justified because of the observation of infrared active vibrational features in the PIA (which imply the existence of localized lattice distortions) and because the experimentally determined effective masses in the cuprates[21–24] (see Table I) are similar to the titanates ($3<m^*/m_e<30$) (Refs. 9 and 26) and thus suggest polaron formation.

The frequency-dependent conductivity $\sigma_{PT}(\omega,T\rightarrow 0)$ in the low-temperature limit is given by[15,17]

$$\sigma_{PT}(\omega,T\rightarrow 0)=\frac{\sqrt{2\pi}t^2e^2N}{\hbar^3\omega_0^2}e^{-\eta}\left[\frac{\omega_0}{\omega}\right]^{(\omega/\omega_0+2/3)}e^{(\omega/\omega_0)}\eta^{\omega/\omega_0}\ , \quad (1)$$

where η is the number of phonons in the polaron polarization cloud, ω_0 is an averaged phonon frequency, t is the electronic resonance overlap integral, $\hbar$ is Planck's constant, and N is the number density of carriers of charge e; η and ω_0 are parameters describing the electron-phonon interaction and are related to the electron-phonon coupling constant $\alpha_j(\mathbf{q})$ through the relations

$$\omega_0=\frac{\sum_{j,q}\alpha_j(\mathbf{q})\sin^2(\frac{1}{2}q)\omega_j^2(\mathbf{q})}{\sum_{j,q}\alpha_j(\mathbf{q})\sin^2(\frac{1}{2}q)\omega_j(\mathbf{q})} \quad (2)$$

and

$$\eta=\frac{1}{\omega_0}\sum_{j,q}2\alpha_j(\mathbf{q})\sin^2(\tfrac{1}{2}q)\omega_j(\mathbf{q}) \quad (3)$$

with the sum over the wave vector $\mathbf{q}$ of the jth optical phonon branch. The characteristic frequency ω_0 represents an averaged phonon frequency involved in the carrier hopping, weighted by the electron-phonon interaction $\alpha_j(\mathbf{q})$. If the phonon dispersion is not too large, formula (3) can be approximated by the more familiar form,[17]

$$\eta\approx\sum_{j,q}2\alpha_j(\mathbf{q})\sin^2(\tfrac{1}{2}q)\ , \quad (4)$$

which represents a weighted average of $\alpha_j(\mathbf{q})$ over the Brillouin zone. The two parameters η and ω_0 essentially determine the shape of the spectrum, and the prefactor is a constant proportional to the square of t.

The dashed curves in Fig. 1 show the calculated $\sigma_{PT}(\omega)$ [Eq. (1)] using the parameters η and ω_0 listed in Table I. The fits to $\sigma_P(\omega)$ for $Tl_2Ba_2CaCu_2O_8$ and $YBa_2Cu_3O_{7-\delta}$ [Figs. 1(a) and 1(b)] are excellent over the entire frequency range. The value of $\omega_0=200$ cm^{-1} is

TABLE I. Effective masses and parameters describing the electron-phonon interaction.

	m^*/m_e	η	ω_0 (cm^{-1})
$La_{2-x}Sr_xCuO_4$	23	10	450
$YBa_2Cu_3O_7$	13	7	200
$Tl_2Ba_2CaCu_2O_8$	11	5.6	200

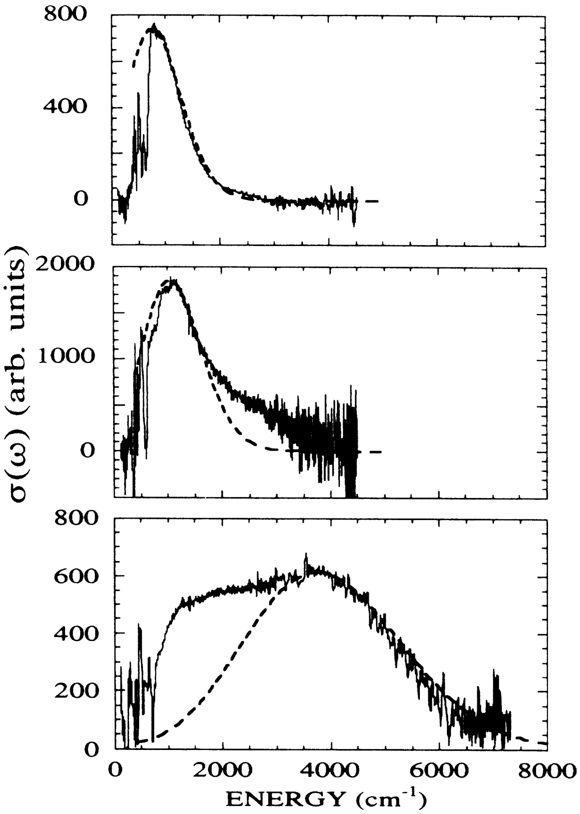

FIG. 1. The photoinduced infrared conductivity $\sigma_P(\omega)$ (solid lines) in the insulator precursors for $Tl_2Ba_2CaCu_2O_8$ (top), $YBa_2Cu_3O_7$ (middle), and $La_{2-x}Sr_xCuO_3$ (bottom) compared with fits to polaron-transport theory $\sigma_{PT}(\omega)$ as calculated using Eq. (1) (dashed lines).

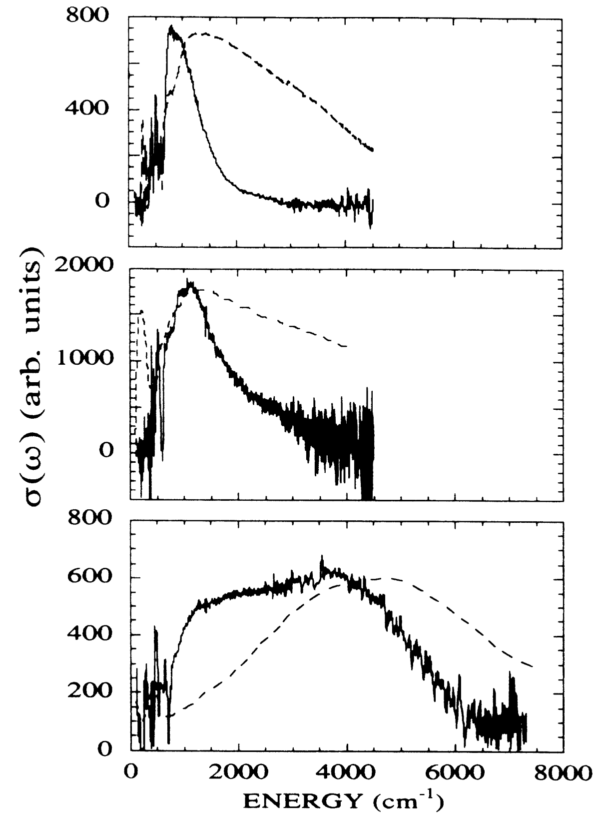

FIG. 2. The infrared conductivity $\sigma(\omega)$ (dashed lines) for $Tl_2Ba_2CaCu_2O_8$ (top), $YBa_2Cu_3O_7$ (middle), and $La_{2-x}Sr_xCuO_4$ (bottom) compared with the photoinduced infrared conductivity $\sigma_P(\omega)$ (solid lines) in their respective insulator precursors.

consistent with calculated values of $\alpha_j(\mathbf{q})$ obtained recently from a linearized-augmented-plane-wave calculation[27] for $YBa_2Cu_3O_{7-\delta}$ as well as with experimental evidence for strong coupling of the low-frequency modes from infrared spectroscopy.[28] In addition to the polaron shakeoff, we observe structure from photoinduced localized phonon modes in all three systems; for $Tl_2Ba_2CaCu_2O_8$ and $YBa_2Cu_3O_{7-\delta}$, they[21–24] distort the low-frequency edge of the peak in $\sigma_P(\omega)$. These vibrational modes are not described by Eqs. (1)–(3) (which are valid only above $\omega > \omega_D/2$) and will therefore be discussed elsewhere. In La_2CuO_4, the fit is good for $\omega > 3000$ cm^{-1}. The deviations at lower frequency possibly arise from the fact that the La_2CuO_4 PIA experiments[21] were carried out before highest quality materials were available.

In agreement with polaron theory, the values of η and ω_0 obtained from the theoretical fits for the three systems scale with the polaron effective masses obtained from the PIA data[24] (see Table I) reflecting a trend in $\alpha_j(\mathbf{q})$. In addition, as η and ω_0 (and the effective mass) decrease, T_c of the system increases. We have previously shown[24] that T_c is inversely proportional to m^* for the cuprates. This behavior is consistent with theories of bipolaronic superconductivity which predict $k_B T_c \cong \hbar^2 n^{2/3} m^*$ (Ref. 29), where n is the number of bipolarons of effective mass m^*.

In Fig. 2, we compare the frequency-dependent conductivity $\sigma(\omega)$, obtained from reflectivity measurements of $Tl_2Ba_2CaCu_2O_8$ thin films[4] ($T_c = 100$ K), $YBa_2Cu_3O_7$ single crystals[5] ($T_c = 90$ K), and $La_{2-x}Sr_xCuO_4$ ceramic samples[30] ($T_c = 35$ K), with the photoinduced infrared conductivity $\sigma_P(\omega)$. The similar spectral shape and the consistent trend of the mid-ir peak toward lower energy in both $\sigma_P(\omega)$ and $\sigma(\omega)$ indicate that carriers in the concentrated (metallic) regime retain much of the character of carriers in the dilute (photoexcited) regime.

III. DISCUSSION

A number of qualitative observations follow from the comparison of $\sigma(\omega)$ and $\sigma_P(\omega)$:

(i) In all perovskites exhibiting superconductivity, the peak energy in both $\sigma(\omega)$ and $\sigma_P(\omega)$ shifts toward lower energy as T_c of the material increases; from $\cong 1.2$ eV (12 000 cm^{-1}) in $Ba_{1-x}Pb_xBiO_3$ (Refs. 6 and 31) to $\simeq 0.5$ eV (4000 cm^{-1}) in $La_{2-x}Sr_xCuO_4$ (Refs. 1, 21, and 30) to 0.13 eV (1300 cm^{-1}) in $YBa_2Cu_3O_7$ (Refs. 1–3, 5, and 22) and to 0.09 eV (950 cm^{-1}) in $Tl_2Ba_2CaCu_2O_8$ (Refs. 4 and

23) (see Table II).

(ii) The concurrent shift in energy of both $\sigma(\omega)$ and $\sigma_P(\omega)$ from system to system implies that both are intrinsic features of the frequency-dependent conductivity and that the origin of the mid-ir feature is the same in both the dilute and concentrated limits.

(iii) The primary difference between $\sigma_P(\omega)$ (the dilute limit) and $\sigma(\omega)$ (the concentrated limit) is the broadening of the mid-ir peak which we expect to occur as the carrier concentration increases to a level at which the exclusion principle and polaron-polaron interactions become important.

The differences between the experimental spectra obtained in the concentrated and the dilute limits (Fig. 2) imply that polaron-transport theory ceases to provide an accurate description of $\sigma(\omega)$ in the concentrated limit and emphasizes the need for generalization of the PTT to the concentrated regime appropriate to the metallic phases of the high-T_c cuprates.

In a given cuprate system, the energy of the mid-ir peak shifts only slightly to higher energy with increased carrier concentration. However, the intensity of the mid-ir peak significantly increases until the maximum T_c is reached at approximately 0.12–0.25 holes/Cu.[1,11,30] A similar dependence of $\sigma(\omega)$ and T_c with doping concentration is seen in the titanates and bismuthates.[6,8,9] Recent data on $La_{2-x}Sr_xCuO_4$ show that upon further doping into the metallic region ($x>0.2$), T_c decreases[10,12] concurrent with a reduction of the intensity of the mid-ir feature. Table II compares the energy ($\approx 2E_b$) of the mid-ir peak and T_c for some cuprate, titanate, and bismuthate superconductors.

In the present analysis, we have not explicitly considered the effect of spin-wave "shakeoff" resulting from coupling of carriers to spin excitations (rather than phonons) as a source of $\sigma(\omega)$ in the mid-ir. The similarity of the line shapes of the $\sigma(\omega)$ spectra and the two-magnon spectra observed in Raman scattering of $Tl_2Ba_2CaCu_2O_8$,[32] $YBa_2Cu_3O_{7-\delta}$,[33] and La_2CuO_4 (Ref. 34) suggest that spin-polaron shakeoff might provide a contribution. A strong spin-lattice interaction has recently been suggested by the temperature dependence of the two-magnon relaxation[35] via coupling to the lattice as inferred from Raman scattering experiments. However, since the electron-phonon interaction arises from the spatial dependence of the transfer integral t, it is expected to be larger than the spin-phonon interaction which originates in the spatial dependence of $\mathcal{J}\cong t^2/4U$, where $\mathcal{J}$ is the magnetic coupling and U is the on-site Coulomb interaction strength.[36] Thus, any contribution to $\sigma(\omega)$ arising from magnon shakeoff should be significantly weaker than the direct contribution to $\sigma(\omega)$ arising from electron-phonon coupling.

TABLE II. Critical temperatures and values of the mid-ir peak position in superconducting perovskites.

	T_c (K)	Mid-ir peak (eV)
$SrTiO_3$	0.3[8]	> 1[8]
$Ba_{1-x}Pb_xBiO_3$	13[6]	1.2[6]
La_2CuO_4	34	0.5
$YBa_2Cu_3O_7$	93	0.13
$Tl_2Ba_2CaCu_2O_8$	110	0.09

We have not discussed in detail the low-frequency, temperature-dependent part of the $\sigma(\omega)$ spectra,[1–5] which has been attributed to free carriers scattering in the weak-coupling limit of a quasimetallic system. In the context of polaron transport theory, these free carriers are dressed polarons with dc transport lifetime (and mean free path) limited by acoustic mode scattering for $k_B T < \hbar\omega_D$ and by a combination of acoustic and optical-mode scattering at higher temperatures.

IV. CONCLUSION

We conclude that the photoinduced infrared conductivity of dilute carriers photoinjected into $Tl_2Ba_2CaCu_2O_8$, $YBa_2Cu_3O_{7-\delta}$, and La_2CuO_4 is well described by nonadiabatic polaron hopping (polaron-transport theory). The similar spectral shape and systematic trends in both $\sigma_P(\omega)$ and $\sigma(\omega)$ indicate that carriers in the concentrated (metallic) regime retain much of the character of carriers in the dilute (photoexcited) regime implying that the charge carriers in the normal state of high-T_c cuprates are polarons or bipolarons. This is in a general sense consistent with the evidence of ferroelectric distortions (doping-induced distortions correlated from cell to cell) in the high T_c cuprates.[37] Furthermore, the absence of a shift[2,4] in the mid-ir feature in $\sigma(\omega)$ at the superconducting transition suggests that if bipolaronic superconductivity is present in the cuprates, bipolarons are formed above T_c.

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*Permanent address: J. Stefan Institute, Jamova 39, 61111, Ljubljana, Yugoslavia.

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