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Comparison of the electrodynamic properties of sintered $YBa_2Cu_3O_{7-y}$ and $La_{1.85}Sr_{0.15}CuO_{4-y}$

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The reflectivity in the superconducting and normal states of $YBa_2Cu_3O_{7-y}$ has been reproduced with the effective-medium theory plus an anisotropic dielectric function and the Mattis-Bardeen conductivity with a gap value $2\Delta/k_BT_c=2.5$ to 2.7. We find that the lowest ir-active phonon polarized in the high-conductivity plane produces structure in the superconducting gap, the lowest-frequency phonon perpendicular to the plane is overdamped, and the electronic susceptibility is more isotropic for $YBa_2Cu_3O_{7-y}$ than for $La_{1.85}Sr_{0.15}CuO_{4-y}$.

The anisotropic reflectivity data[1] on single-crystal La_2NiO_4 has been used to successfully model the far-infrared (FIR) behavior[2] of sintered La_2NiO_4 and $La_{1.85}Sr_{0.15}CuO_{4-y}$ through the effective-medium approximation[3] (EMA). A number of single-crystal properties of $La_{1.85}Sr_{0.15}CuO_{4-y}$ have been deduced from this investigation: The phonons are quite similar both in position and strength to those of La_2NiO_4, and thus the electrodynamic properties of $La_{1.85}Sr_{0.15}CuO_{4-y}$ are also highly anisotropic. In contrast to La_2NiO_4, a large infrared (ir) dielectric constant ε_∞ occurs along the **c** axis in $La_{1.85}Sr_{0.15}CuO_{4-y}$ due to a strong electronic mode at 0.5 eV. For frequencies below 150 cm^{-1}, the two-dimensional (2D) conductivity in the normal state can be described in terms of a frequency-dependent scattering rate. The insights realized from this study have encouraged us to measure and analyze the electrodynamic properties of sintered $YBa_2Cu_3O_{7-y}$ to the same degree in order to compare the "deduced" 2D response of the two high-T_c compounds.

In this paper we show that the two high-T_c compounds have similar electronic transition strengths and frequency-dependent relaxation times but that the strongest ir active phonon mode is overdamped for $YBa_2Cu_3O_{7-y}$ and not for $La_{1.85}Sr_{0.15}CuO_{4-y}$. The yttrium compound has the additional novel feature that the lowest-frequency in-plane ir active lattice mode is below the superconductivity gap energy 2Δ.

The measured FIR reflectivities for the superconducting (10 K) and normal state[4] (100 K) of sintered $YBa_2Cu_3O_{7-y}$ are shown in Fig. 1(a). The measurements are made at a resolution of 6.5 cm^{-1} below 300 cm^{-1} and 13 cm^{-1} for higher frequencies. The reflectivity changes between the superconducting and normal state shown in Fig. 1(a) are consistent with those reported by others.[5-7] The calculated curves shown in Figs. 1(b) and 1(c) will be considered later.

The solid curves in Figs. 2(a) and 2(b) show the FIR parameters obtained for the composite material at a temperature of 100 K, from a Kramers-Kronig analysis as previously reported.[4] These data represent the starting point of our current investigation. The fundamental assumption in what follows is that the electrical anisotropy of the two high-T_c materials is nearly the same, i.e., in both cases the high conductivity is in the plane perpendicular to the **c** direction of the crystallite. Recent dc conductivity measurements[8,9] on single crystals of $La_{1.85}Sr_{0.15}CuO_{4-y}$ and $YBa_2Cu_3O_{7-y}$ support this hypothesis.

As a first step to describe FIR phonon data in Fig. 2 we introduce a dielectric function[10] with enough modes to account for all of the peaks observed in the FIR. For the doublet centered at 300 cm^{-1}, one phonon mode is as-

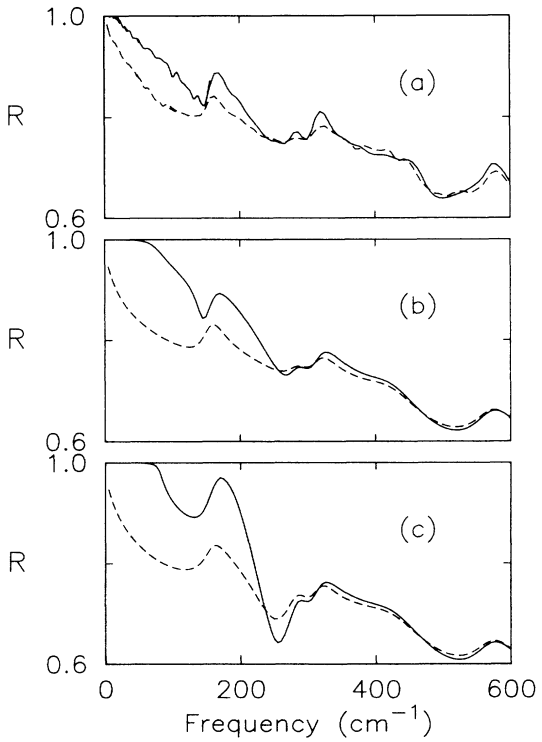

FIG. 1. Normal incidence reflectivity in the superconducting (solid curves) and normal (dashed curves) states of sintered $YBa_2Cu_3O_{7-y}$ vs frequency. (a) Measured reflectivity. (b) Curves calculated with parameters in Table I (assuming the lowest-frequency phonon is polarized in the **a-b** plane) using EMA with $L_c=0.7$ and $2\Delta=170$ cm^{-1}. (c) Same as (b) except assuming phonon polarized along the **c** direction.

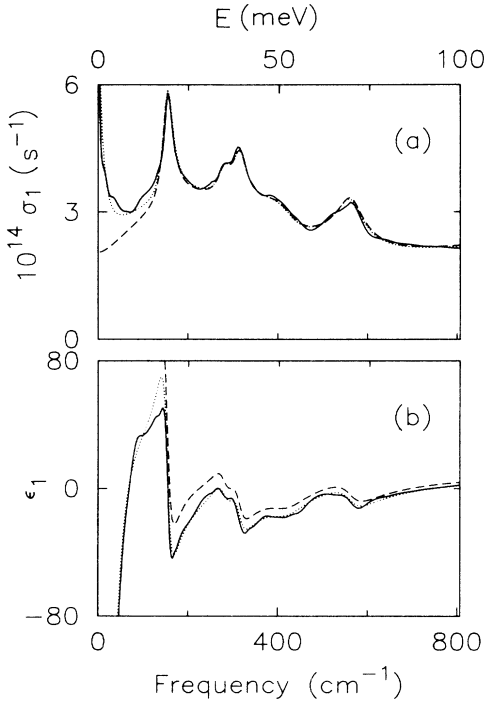

FIG. 2. Effective optical parameters vs frequency for sintered $YBa_2Cu_3O_{7-y}$. (a) Real part of the conductivity. (b) Real part of the dielectric function. Results obtained from a Kramers-Kronig analysis of the reflectivity (solid curve) compared with the EMA calculations with $L_c=0.7$ (dashed curve) and also to a related model with a frequency-dependent scattering rate for conduction electrons (dotted curve).

signed to the **a** and one to the **b** directions consistent with the orthohombic crystal symmetry.[11] The particular crystallographic mode assignment is given in Table I. Since each mode introduces three parameters in the dielectric function it is understood that, although the normal-state data can be fit, the modes cannot be uniquely determined. In addition, the static contribution from the anisotropic bound electronic modes $\sigma_{1el}(\omega)$ is characterized by $\varepsilon_{\infty,a}$, since

$$\varepsilon_{\infty,a} \approx 1+8\int_0^{\infty} \frac{\sigma_{1el}(\omega)}{\omega^2}d\omega\,, \tag{1}$$

where a identifies the crystal direction.

Figure 1(a) indicates that the reflectivity shapes produced by the phonon modes display different signatures in the superconducting and normal states. Below it is shown that if the free-carrier conductivity is 2D-like then the symmetry of the lowest-frequency phonon mode which reproduces the two reflectivity signatures can be found.

If the composite crystallites are spheroids with the same symmetry as the conductivity tensor then, as shown in Ref. 3, a single parameter, the depolarization factor L_c along the **c** direction of the crystallite, specifies the EMA since a fixed fill fraction of $f=\frac{2}{3}$ applies for a 2D metallic component. The effective conductivity σ_e is obtained from[3]

$$\frac{2}{3}\frac{\sigma_{ab}-\sigma_e}{(1+L_c)\sigma_e+(1-L_c)\sigma_{ab}} + \frac{1}{3}\frac{\sigma_c-\sigma_e}{2[(1-L_c)\sigma_e+L_c\sigma_c]}=0\,. \tag{2}$$

TABLE I. Classical oscillator parameters used to fit the sintered $YBa_2Cu_3O_{7-y}$ FIR reflectivity data. ε_∞ represents the static dielectric constant contribution produced by the electronic modes.

	ω_{TO} (cm^{-1})	ω_{LO} (cm^{-1})	γ (cm^{-1})	ε_∞	Ω_p (eV)	Γ (eV)
(a)	155	225	26			
	281	500	40			
(b)	155	225	26			
	315	545	40			
$L_c=\frac{1}{3}$				8	2.6	1.9
$2\Delta=156\ cm^{-1}$						
(c)	200	490	270			
	425	490	80			
	565	880	55			
				3.5		
(a)	155	235	26			
	281	560	35			
(b)	155	235	26			
	315	631	35			
$L_c=0.7$				8.0	2.6	1.2
$2\Delta=170\ cm^{-1}$						
(c)	200	450	270			
	415	475	90			
	565	710	55			
				3.5		

This EMA equation should be composed of three terms, one for each component of the orthorhombic dielectric function, but we have chosen to simplify the problem by approximating the conductivity in the **a-b** plane by the average of the conductivities in the **a** and **b** directions. Since these conductivities differ only by the splitting of the small doublet centered at 300 cm^{-1}, this approximation is reasonable. With the lowest-frequency phonon **c** or **a-b** plane directed, the single-crystal dielectric function is combined with Eq. (2) to fit the data shown in Fig. 2. The results for the **a-b** plane phonon case are represented by the dashed curves and these parameters are tabulated in Table I.

Inspection of the phonon parameters listed in Table I shows that the lowest-frequency **c**-directed phonon has an anomalously large linewidth. We have been able to reproduce the same response with an alternate model which replaces this anharmonic phonon with a **c**-directed Drude term plus a more harmonic phonon; however, in addition to requiring two more parameters this model does not provide enough anisotropy in the dc conductivity to be consistent with the data.[8,9]

As has been noted earlier the Drude plus Lorentz oscillator dielectric function can be fitted to the data down into the FIR spectral region, but for the lowest frequencies a frequency-dependent relaxation time is required[3,4] to complete the dynamical picture. The phenomenological scattering rate process described by Eqs. (3) and (4) in Ref. 3 is characterized by three parameters: the dc scattering rate Γ_{dc}, the strength of the effect λ_0, and the saturation frequency $1/\alpha$. These fitted values are $\Gamma_{dc}=2500\ cm^{-1}$, $\lambda_0=100$, and $1/\alpha=75\ cm^{-1}$, with all other parameters in Table I unchanged. The resulting fit appears in Fig. 2 as the dotted curves. Because this frequency-dependent rate has not yet been incorporated into the superconducting state response this contribution cannot be included in the comparison of the superconductivity-to-normal-state properties which follows.

With the normal-state Drude-Lorentz dielectric function determined, the Mattis-Bardeen equations[12] are used to model the 2D superconductivity in the **a-b** plane. The new parameter is the electromagnetic energy gap 2Δ with all other parameters in the dielectric function model fixed. We find that the data can be fit either by spherical or plate-shaped grains but *only* if the lowest-frequency phonon is in the **a-b** plane. Figure 1(b) shows the fit for platelike grains ($L_c=0.7$) with $2\Delta/k_BT_c=2.7$. An equally good fit for spherical grains can be obtained with $2\Delta/k_BT_c=2.5$ (not shown). The best fit when the lowest-frequency phonon is **c** directed [shown in Fig. 1(c)] does not agree with the data; the normal-super change in reflectivity is much larger and has a different frequency dependence than that found for the **a-b** plane case shown in Fig. 1(b). We conclude that the lowest-frequency phonon is polarized in the high-conductivity plane direction.

The **a-b** plane single-crystal dielectric response deduced from our composite media study in Fig. 1(b) is shown in Fig. 3. The normal-state results are represented by the dashed curves and the superconducting response by the solid ones. The σ_1 curve for the superconducting state in Fig. 3(a) is particularly noteworthy since $2\Delta=170\ cm^{-1}$,

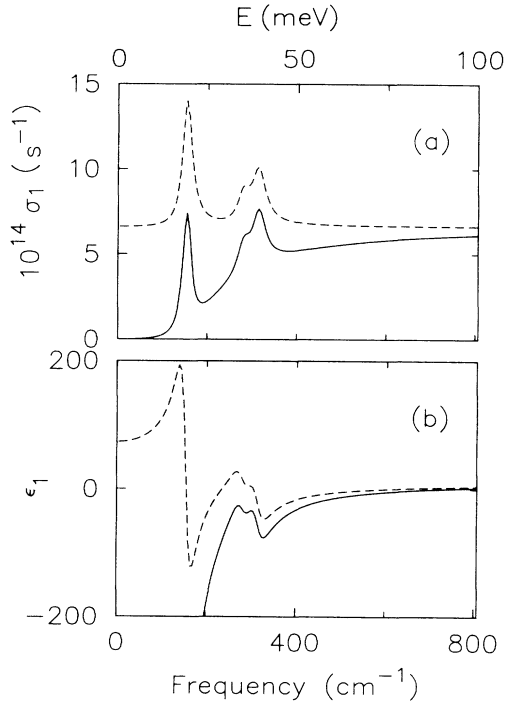

FIG. 3. Deduced **a-b** plane optical parameters vs frequency for single-crystal $YBa_2Cu_3O_{7-y}$. (a) Real part of the conductivity. (b) Real part of the dielectric function. Curves for superconducting (solid) and normal state (dashed) as determined from the fit to the sintered data.

so the strongest ir-active phonon centered at 155 cm^{-1} produces structure in the gap. Hence, in general, we expect single-crystal oxide superconductors to be lossy for frequencies less than 2Δ because of excitation of the lattice degrees of freedom.

Comparing these results with those obtained for $La_{1.85}Sr_{0.15}CuO_{4-y}$ shows that the relative superconducting gap energies are similar with $2\Delta/k_BT_c\approx 2.6$ for both materials and that the strengths of the strongest ir-active phonon modes are comparable to the ir-active mode in MgO and hence not anomalously large as previously suggested.[13] Also, both materials demonstrate frequency-dependent relaxation, with parameters of similar magnitude.

The most apparent difference is the absence of enhanced absorptivity in the superconductivity state above the gap for $YBa_{12}Cu_3O_{7-y}$ either in the published data[5-7] or in the composite media modeling which we have carried out here. We have shown previously[3] with the same composite medium model that the large excess absorptivity in the superconducting state observed for $La_{1.85}Sr_{0.15}CuO_{4-y}$ is due in part to the composite structure, which effectively couples the extremely large value of the dielectric function in the **c** direction to the **a-b** plane through the EMA. Inspection of Table I shows that the single-crystal electronic susceptibility is smaller for $YBa_2Cu_3O_{7-y}$ than it is for $La_{1.85}Sr_{0.15}CuO_{4-y}$; in addition, the superconductivity gap energy falls in a region of

negative dielectric function in the former case, while in the latter it is positive. Modeling shows that for $YBa_2Cu_3O_{7-y}$ the resultant superconducting-normal-state reflectivity difference is only weakly dependent on the shape of the composite grains in contrast with the strong dependence found for $La_{1.85}Sr_{0.15}CuO_{4-y}$.

Another difference is in the strength of the electronic transitions, as monitored by ε_∞ [see Eq. (1)]. The values of ε_∞ for $La_{1.85}Sr_{0.15}CuO_{4-y}$ given in Ref. 3 (Table I) show that the **c**-polarized transition is stronger (even after correcting for the different transition frequency) for the La compound than for the Y one ($\varepsilon_\infty = 3.5$ for the former versus 35 for the latter), while the **a-b** plane transition is stronger for the Y compound ($\varepsilon_\infty = 8$ vs 1).

Finally, the last major difference between the two systems is that the strongest ir-active phonon (**c** directed) for $YBa_2Cu_3O_{7-y}$ is overdamped, while for $La_{1.85}Sr_{0.15}CuO_{4-y}$ it is not.

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