Fano effect in the a-b plane of $Nd_{1.96}Ce_{0.04}CuO_{4+y}$: Evidence of phonon interaction with a polaronic background

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(Received 18 July 1997; revised manuscript received 11 September 1997)

Reflectance measurements in different $Nd_{1.96}Ce_{0.04}CuO_{4+y}$ samples with properly selected carrier concentrations provide firm evidence of Fano antiresonances for the four transverse optical E_u phonons in the a-b plane. An analysis of the Fano line shapes and of the dependence on temperature of the renormalized phonon frequencies allows us to determine the origin of the electronic continuum interacting with the phonons. Unlike in doped semiconductors, where the continuum is the free-carrier absorption band, here the continuum is provided by a polaron band at ~ 0.1 eV, present in most parent compounds of high-T_c superconductors. In the most doped sample, the polaron band softens as T decreases, thus indicating a delocalization of the polaronic carriers at low temperature. [S0163-1829(98)08501-4]

In 1961, Fano quantitatively accounted for the so-called Fano antiresonance, i.e., the asymmetric line shape of the $2s2p^1P$ resonance of He observed in electron inelastic scattering experiments.[1] In that work, Fano revised an earlier qualitative interpretation[2] of the spectra of He and other rare gases. Fano pointed out that such effects are expected whenever a set of discrete states is mixed with a continuous spectrum. Thereafter, ''Fano profiles'' have been observed in a number of spectra, including those where phonon discrete states interact with a continuum background due to free carriers. In high-critical-temperature superconductors (HCTS's), phonon Fano profiles have been detected with the electric field of the radiation polarized along the c axis, either in[3–6] $YBa_2Cu_3O_{7-\delta}$ or in[7] $Bi_2Sr_2CaCu_2O_8$. Moreover, in the $400-600\ cm^{-1}$ region of the reflectivity spectra of $Pb_2Sr_2LCu_3O_8$ ($L=Y$, Dy, Eu, Nd, and Pr), phonon line shapes have been reported to become asymmetric for increasing doping.[8] These results have been explained in terms of an interaction of Raman-active modes along the c axis, made infrared active by some symmetry-breaking potential, and an electronic continuum which develops with doping in the midinfrared. As far as we know, no evidence of Fano line shapes has been reported for the infrared-active phonons of the a-b plane. Here, indeed, the optical phonons are shielded by the carriers which form below T_c a fluid of superconducting pairs.

In order to look for Fano resonances in the a-b plane of a cuprate, we have selected a strongly doped, nonmetallic system, where the existence of a considerable electron-phonon coupling is ensured by previous observations of polaronic effects in the a-b plane reflectivity. It is the case of $Nd_{2-x}Ce_xCuO_{4-y}$ (NCCO), where an electronic continuum detected at $\sim 1000\ cm^{-1}$ has been explained in terms of a polaron band d, whose strength increases with either x see (Ref. 9) or y (see Ref. 10). The polaronic origin of the d band has been firmly established in Nd_2CuO_{4-y} (NCO).[10] Here it exhibits a fine structure, well explained in terms of an electron-phonon interaction. This structure has been shown to be related to some infrared-active vibrations (IRAV's), observed in the far-infrared spectra of samples where carriers are either photoinduced or chemically induced.[11–15] These IRAV's add to the extended phonons of the undisturbed stoichiometric crystal, and appear in the far infrared as new peaks or sidebands of the phonons.[16,17] An infrared band at $\sim 1000\ cm^{-1}$ similar to that found in e-doped NCO has been observed also in h-doped, oxygen-enriched La_2CuO_{4+y} (LCO), where both its line shape and temperature dependence are consistent with the absorption from a polaronic impurity state.[18] Such infrared evidence is confirmed by a broad spectrum of experimental techniques, ranging from extended x-ray-absorption fine structure,[19] to electron diffraction,[20] and to neutron scattering[21] and points toward the existence of polaron superstructures in superconducting cuprates as well as in nonsuperconducting perovskites.

In the following it will be shown that asymmetric line shapes are found in $Nd_{1.96}Ce_{0.04}CuO_{4-y}$ for the four optically active E_u phonons in the a-b plane. These asymmetric line shapes will be explained in detail in terms of a Fano interaction between the phonons and an electronic background. Contrarily to what is normally found in doped semiconductors, the electronic continuum is not given by the weak Drude term eventually present in these semiconducting compounds, but by *the polaron d band peaked at* $\approx 1000\ cm^{-1}$. This conclusion is based on an analysis of the Fano line shapes and of the temperature dependence of the phonon peak energies in two different, properly selected doped samples. In fact, in order to discriminate the effect of the charges added to the a-b plane from possible effects due to the chemical doping, two single crystals with the same Ce doping ($x=0.04$) but different oxygen concentration y, and therefore different carrier concentration, have been measured, MN10 and MN29. The former is as grown, the latter is enriched in oxygen and almost compensated.

The experimental apparatus has been described in detail elsewhere.[10] Data have been collected from 20 (MN29) or 70 (MN10) through $18\,000\ cm^{-1}$ by use of a Michelson interferometer. The real part of the optical conductivity $\sigma(\omega)$ has been obtained from Kramers-Kronig transformations. The reflectivity data have been extrapolated to zero frequency either by using a Hagens-Rubens formula or by a Drude term obtained by fitting a Drude-Lorentz model for the complex

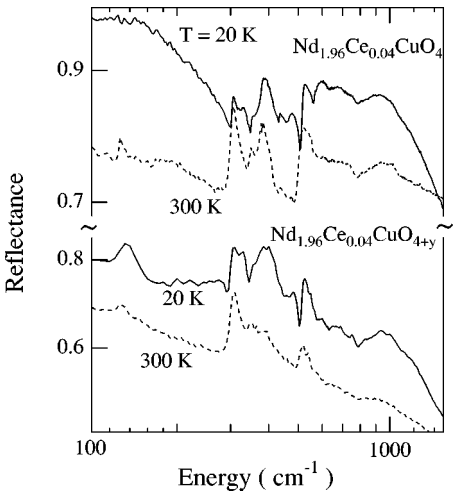

FIG. 1. Far- and midinfrared reflectance of samples MN10 and MN29, from top to bottom, at 20 and 300 K.

dielectric function $\tilde{\epsilon}(\omega)$ to the $R(\omega)$ data.[10] The transformed $\sigma(\omega)$, for $\omega \geq 70$ (20) cm^{-1}, has been found to be independent of the approach used. For $\omega \geq 18\,000\ cm^{-1}$, $R(\omega)$ has been extrapolated up to $\omega = 320\,000\ cm^{-1}$ by using the values at 300 K reported by Tajima *et al.*[22] and, at higher frequencies, by extrapolating $R(\omega)$ with a ω^{-4} law.

The low-energy reflectance $R(\omega)$ of samples MN10 and MN29 is reported in Fig. 1 for $T = 20$ K and $T = 300$ K. $R(\omega)$ is strongly T dependent in both samples and lower in MN29 than in MN10, consistently with the lower number of extra charges determined in the former crystal, as will be shown in the following. In both samples, the room temperature reflectivity is dominated by the four E_u phonons and by a broad but clear peak at $\lesssim 1000\ cm^{-1}$, the d band. In the low-T spectra, the phonon lines show a broadening, instead of a narrowing, due to the insurgence of new satellite bands. This effect, observed also in NCO doped by oxygen vacancies[10] as well as in LCO doped by excess oxygen,[23] has been already ascribed,[10] as mentioned above, to the insurgence of IRAV modes due to self-trapped charges.

The low-energy part of the optical conductivity, obtained as described above, is reported in Fig. 2 for the same samples and temperatures of Fig. 1. The room temperature phonon lines of both samples (in particular the bending and stretching Cu-O modes at ~ 300 and $\sim 515\ cm^{-1}$, respectively) deviate appreciably from the ordinary Lorentzian shapes. They display a strong asymmetry, with a dip on the low-energy side which indicates an interaction with a continuum at higher energy.[1] The closest band with such features is the d band. In order to verify if actually *the E_u phonons interact with the polaronic background*, the optical conductivity from 70 (20) to 18 000 cm^{-1} has been fitted in terms of a complex dielectric function given by

$$\tilde{\epsilon}(\omega) = (\tilde{\epsilon}_D + \tilde{\epsilon}_F + \tilde{\epsilon}_{\mathrm{LM}} + \tilde{\epsilon}_d) + (\tilde{\epsilon}_{\mathrm{MIR}} + \tilde{\epsilon}_{\mathrm{CT}} + \epsilon_\infty). \quad (1)$$

Here, the first set of parentheses includes the low-frequency terms of interest for the present study: $\tilde{\epsilon}_D$ is a (weak) Drude-like term, $\tilde{\epsilon}_F$ is the contribution of the Fano-like phonons, $\tilde{\epsilon}_{\mathrm{LM}}$ that of the local modes, and $\tilde{\epsilon}_d$ that of the polaron band. In the second set of parentheses of Eq. (1),

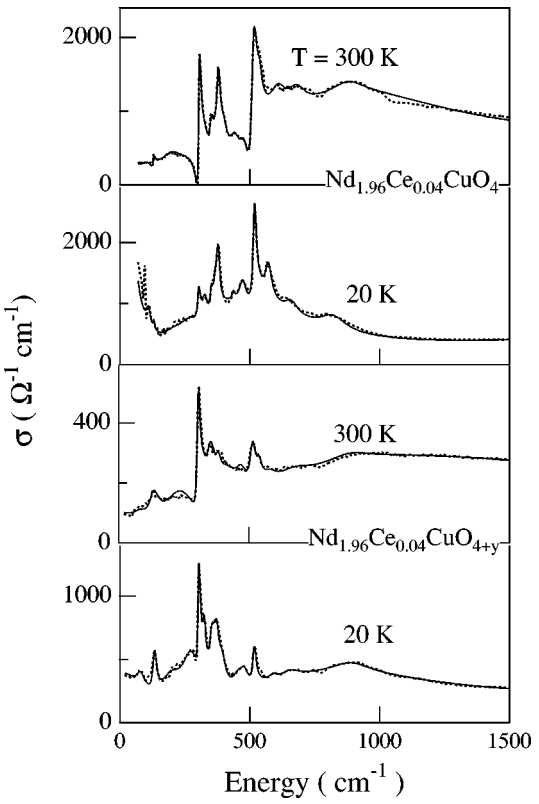

FIG. 2. Far- and midinfrared optical conductivity of samples MN10 and MN29, from top to bottom, at 20 and 300 K.

$\tilde{\epsilon}_{\mathrm{MIR}}$, $\tilde{\epsilon}_{\mathrm{CT}}$, and ϵ_∞ are the standard contributions from the midinfrared band, the charge-transfer band, and the higher-energy oscillators, respectively (see Ref. 9 for further details). Each of these contributions, $\tilde{\epsilon}_*$, but for $\tilde{\epsilon}_F$, is given by a sum of j Lorentzians,

$$\tilde{\epsilon}_*(\omega) = \sum_j \frac{S_j^2}{(\omega_j^2 - \omega^2) - i\omega\Gamma_j}, \quad (2)$$

with strengths, peak frequencies, and damping factors given by S_j, ω_j, and γ_j, respectively.

Let us now focus on the Fano-like-phonon line shapes seen in Fig. 2. The overall contribution to the dielectric function $\tilde{\epsilon}_F$ of an oscillator interacting with an electronic continuum has been reported in an analytical form by Davis and Feldkamp.[24] This form has been never used before to fit the far-infrared optical conductivity of either HCTS's or other ionic compounds, at least to our knowledge. Usually, Fano line shapes are fitted, instead, after an arbitrary subtraction of the electronic background. Davis and Feldkamp extend the Fano original approach to the case of an interaction between an arbitrary number of discrete states and continua. They get, for $\tilde{\epsilon}_F = \epsilon_{1F} + i\epsilon_{2F} = \Sigma_k \tilde{\epsilon}_{F,k}$,

$$\epsilon_{1F,k}(\omega) = R_k \gamma_k \left(\frac{(q_k^2 - 1)(\tilde{\omega}_k - \omega) + 2q_k\gamma_k}{\gamma_k^2 + (\omega - \tilde{\omega}_k)^2} \right) \quad (3)$$

and

$$\epsilon_{2F,k}(\omega) = R_k \left(\frac{[q_k\gamma_k + (\omega - \tilde{\omega}_k)]^2}{\gamma_k^2 + (\omega - \tilde{\omega}_k)^2} - 1 \right). \quad (4)$$

TABLE I. Parameters of the Fano fit for the E_u phonons and the electronic continuum of crystal MN10 (MN29) at different T. Units are cm^{-1}, except for adimensional q and R.

	20 K	100 K	200 K	300 K
$\tilde{\omega}_1$	130 (133)	130	130	128 (132)
R_1	1 (1.2)	2	2	4.5 (0.6)
γ_1	3 (8)	4	4	4 (20)
q_1	8 (10)	8	8	1.5 (10)
$\tilde{\omega}_2$	303 (303)	303.5	303	304.5 (304)
R_2	1.9 (3.9)	1.7	3.7	9 (2.7)
γ_2	6 (5)	4	4.5	5 (6.5)
q_2	5 (3.1)	7	3.8	1.8 (3.1)
$\tilde{\omega}_3$	351 (352)	350	349.5	350 (351)
R_3	2.3 (0.68)	2.7	2.3	2.8 (0.48)
γ_3	5 (10)	5	5	7 (18)
q_3	3 (10)	3	3	2.4 (10)
$\tilde{\omega}_4$	517 (515)	514	512	511 (514)
R_4	2.9 (0.83)	1.7	2	6 (0.55)
γ_4	8 (7)	7	8	11 (12)
q_4	4.5 (7)	6.5	6	2 (7)
ω_d	480 (850)	500	570	900 (1100)
S_d	5400 (4800)	5800	6900	8000 (5400)
γ_d	500 (1100)	650	800	1100 (1900)

Different phonons are indicated by the index k, while R_k are scale parameters which include the transition rates to the excited states and q_k are the Fano-Breit-Wigner parameters, an inverse measure of the electron-phonon interaction.[1] $\tilde{\omega}_k$ is the renormalized frequency of the kth phonon. The dependence of R_k, q_k, and γ_k on energy is assumed to be smooth and is therefore neglected in the model of Ref. 24.

The fits obtained in this way are excellent in both samples at all temperatures, as shown by the close match in Fig. 2 between the theory (solid lines) and the data (solid dots) in the case of 300 K and 20 K. It has here to be pointed out that most of the contributions to $\tilde{\epsilon}(\omega)$, in particular those due to the interacting extended phonons which are relevant in the present context, are well distinguished in the spectra where they appear as nonoverlapping peaks. Therefore, although the total number of parameters entering the fits is high, the uncertainty relative to the four parameters needed to reproduce each extended phonon interacting with the electronic continuum is small. The values of these parameters are given in Table I for both samples MN10 and MN29, together with those of the polaron band. For all E_u modes of sample MN10, q is roughly constant for $T \leqslant 200$ K, while it decreases by a factor of 3–4 at room temperature. These values are similar, but for the sign, to those reported in $YBa_2Cu_3O_{7-\delta}$ for the Raman mode at 312 cm^{-1} interacting with an electronic background. Therein, q is equal to -4.3 at room temperature[25] and ranges from -6 to -2 for temperatures varying between 4 K and 300 K.[6] In that case, however, Raman-active c-axis phonons were claimed to interact with a Drude continuum at zero frequency, hence the negative sign for q.

It is now also possible to estimate for each sample the effective carrier concentration below a frequency ω, which is given by

$$n_{\mathrm{eff}}(\omega) = \frac{2m^*V}{\pi e^2} \int_0^{\omega} \sigma(\omega')\, d\omega', \tag{5}$$

where m^* is the effective mass and V is the volume of the unit cell. If one puts, in Eq. (5), $\omega = 12\,000$ cm^{-1} (the charge-transfer energy gap), one finds that in the compensated sample MN29 the effective charge is lower than the n_{eff} of MN10 (by about 40%).

Let us finally discuss the dependence on temperature of the d band in the two samples with different doping, for its relevance to the Fano effect. The midinfrared optical properties of NCCO at room temperature have been already described in detail elsewhere.[9] We can extend here that discussion to the low-temperature measurements of Fig. 2 and to the results of Table I. As a whole, the behavior of the d band for a 0.04 Ce doping follows that already observed in ligthly oxygen-deficient NCO.[10] In the compensated sample MN29 (see Fig. 2 and Table I), the peak energy of the d band is $\omega_d = 1100$ cm^{-1} at 300 K and slightly decreases to 850 cm^{-1} at 20 K. In the as-grown MN10, $\omega_d = 900$ cm^{-1} at 300 K, and drops to 480 cm^{-1} at 20 K. In Table I one can also notice that in sample MN10 the highest-frequency (stretching) mode hardens for decreasing temperature, by an amount much larger than the experimental uncertainty of ± 0.5 cm^{-1}, while the energy of the other phonons remains roughly constant. On the contrary, in sample MN29 no appreciable dependence on T has been detected either for the energies of all the four E_u phonons, *including the stretching mode*, or for their Fano parameters. In MN29, for all phonons $q \approx 10$ but for the bending mode, where $q \sim 3$. The dependence on T reported above for the two samples confirms the conclusion based on the position of the dip in the asymmetrical phonon line shapes, namely, that *the phonons interact via the Fano effect with the polaron d band*. In fact, for decreasing T the d band shifts to low energy by a large amount in sample MN10, only slightly in sample MN29. Moreover, in the former sample the difference between ω_d and the peak energy of the phonon changes drastically for the highest-energy phonon ω_4, which hardens as predicted by the theoretical model.[1,7] Also the dependence on T of the Fano parameter q, inversely related to the strength of the electron-phonon interaction, is consistent with the behavior with T of the polaron band in the two samples. Indeed, $E_p = g^2/\omega^*$, where E_p is the binding polaron energy, g is an average electron-phonon-interaction parameter, and ω^* is an average interacting-phonon frequency. The latter is ~ 210 cm^{-1} in NCO from Ref. 10, in a Lorentzian band model. From $\omega_d = 2E_p$ ($\sim 4E_p$) for a small (large) polaron,[26] one obtains $g = (A\omega^*\omega_d)^{1/2}$, with $A = 0.5 (\sim 0.25)$. In a small polaron approach, therefore, the softening of ω_d in sample MN10 is related to a parallel decrease of g (from 300 cm^{-1} to 220 cm^{-1} for T going from 300 K to 20 K) and an ensuing increase of q in the same temperature range (in a large polaron approach, g changes from 220 cm^{-1} to 160 cm^{-1}). In sample MN29, instead, the energy of the polaron band has a smaller relative change with T while q is roughly constant. If one extrapolates these results to even higher doping (where direct observations of phonon and d bands are prevented by shielding effects), one may infer that the insulating-to-metal transition observed in NCCO for x

$=0.13$ could be due to a collapse of the binding energy of the polaron, which could turn from a partially localized quasiparticle to a fully delocalized carrier.

The strength of the electron-phonon interaction can be quantitatively estimated also from the T dependence of the stretching mode energy in sample MN10, *the only mode which shows a sizable T dependence*. It has to be noticed that this T dependence cannot be ascribed to changes in the lattice parameters since the same stretching mode is roughly T independent in sample MN29. By using the Green's function formalism,[7] one obtains for the renormalized phonon frequencies

$$\tilde{\omega}_k = \omega_k \left(1 - \frac{g_k^2(\omega_d^2 - \omega_k^2)/2}{(\omega_d^2 - \omega_k^2)^2 + (\gamma_d \omega_k)^2} \right), \tag{6}$$

where g_k is the coupling constant for the interaction of the kth phonon of energy ω_k with an electronic oscillator with resonance frequency ω_d and damping constant γ_d. It is worth noticing here that the coupling constants g_k do not necessarily coincide with the electron-phonon interaction parameter g, for at least two reasons. First, g is an effective parameter averaged over the different local modes[10] or phonons which build up the polaron band. Second, the strength of the electron-phonon interaction which leads to the formation of a polaron may differ from that of the electron-polaron interaction which results in the Fano resonance. Therefore, in evaluating the renormalized phonon frequencies $\tilde{\omega}_4$ from Eq. (4) we take g_4 as a fitting parameter which, for the sake of simplicity, is assumed to be temperature independent. The bare phonon frequency ω_4 is the second fitting parameter, while the values of ω_d and γ_d entering Eq. (4) are those obtained at each temperature by the Lorentz-Fano fit of the optical conductivity and reported in Table I. The dependence on temperature measured for the stretching-mode energy $\tilde{\omega}_4$ is reported by solid dots in Fig. 3(a), together with the theoretical estimate of $\tilde{\omega}_4$ as obtained for $g_4 \simeq 150\ \mathrm{cm}^{-1}$ and $\omega_4 = 514 \pm 1\ \mathrm{cm}^{-1}$ (solid line in the figure). The agreement between the model and the experiment is excellent. The value for g_4 is on the same order of that estimated for g, thus strengthening the whole model of interaction between carriers and the polaron band. The same model, when applied to the other three modes in sample MN10 or to all the four modes in sample MN29, gives values of the phonon energies which are nearly constant within the experimental uncertainty, in agreement with the experimental results, provided $g_k \lesssim 80\ \mathrm{cm}^{-1}$. As an example, the dependence on temperature of the bending mode $\tilde{\omega}_2$ is reported by open circles in Fig. 3(b). This mode has also a sizable asymmetry in the line shape, as strong as that of the stretching mode. The theoretical estimate of $\tilde{\omega}_2$, as obtained for $g_2 \simeq 70\ \mathrm{cm}^{-1}$ and $\omega_2 = 304 \pm 1\ \mathrm{cm}^{-1}$, is given by a dashed line.

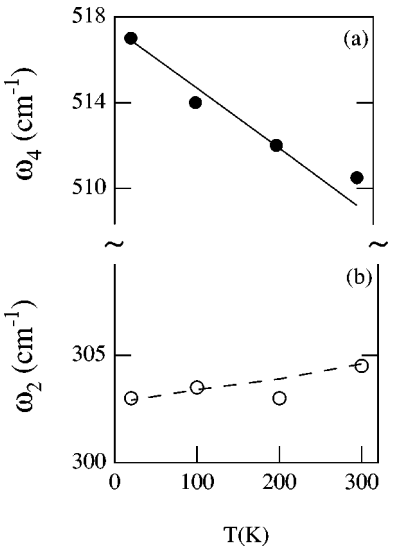

FIG. 3. Frequencies of the stretching, solid dots in part (a), and bending, open circles in part (b), modes in the most doped sample MN10 vs temperature and their behavior as calculated from Eq. (4) (solid and dashed lines). See the text for details.

In summary, a clear signature of a Fano interaction between all the transverse optical E_u phonons and an electronic continuum has been obtained by measuring the reflectance of two $Nd_{1.96}Ce_{0.04}CuO_{4+y}$ samples with different carrier concentrations. A fit has been made of the optical conductivity by a formula which accounts for the whole effects of the electron-phonon interaction, both on the discrete and on the continuum states. In this way it has been shown that those phonons interact with the polaron band at $\sim 1000\ \mathrm{cm}^{-1}$ reported by several authors for the insulating parent compounds of high-T_c superconductors. The dependence on temperature of the renormalized phonon frequencies has been analyzed and the strength of the electron-phonon interaction has been estimated, further supporting the above interaction model. These results point out the role of polarons in the optical properties of the insulating parents compounds of HCTS's. The increase of the Fano parameter for decreasing T observed in the more doped sample and the corresponding large softening of the polaron band indicate that the insulator-to-metal transition in cuprates could be triggered by a collapse to zero frequency of the polaron band. Measurements in more doped samples, as well as in other cuprates, are needed to confirm this intriguing scenario.

We are indebted to W. Sadowski for providing the samples here investigated, and to G. Strinati and M. Grilli for helpful discussions and suggestions. This work has been supported in part by the HC&M program of the European Union under Contract No. 94-0551.

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