Infrared-active phonons and the superconducting gap of T_c-reduced double-chain $YBa_2Cu_4O_8$ superconductors

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We report a far-infrared reflectivity study in the temperature range 10–300 K of the double-chain $YBa_2Cu_4O_8$ superconductor with Zn and Sr substituting for Cu and Ba, respectively. Superconductivity-induced self-energies of the transverse optical $B_{1u}(z)$ phonon were used to estimate the position of the superconducting gap. Evidence for a shift of the gap to lower energies upon decreasing the superconducting transition temperature T_c was found for Zn- as well as for Pr-doped materials. A comparison of the linewidth changes of the plane-oxygen phonon (at $\sim 300\ cm^{-1}$) with the results of a strong-coupling calculation yields the gap-to-T_c ratio $2\Delta_0/kT_c = (8.2 \pm 1.0)$ for $YBa_2(Cu_{3.96}Zn_{0.04})O_8$ which is close to $2\Delta_0/kT_c = (6.6 \pm 0.2)$ reported for $(Pr_{0.37}Y_{0.63})Ba_2Cu_4O_8$. For Sr in place of Ba in $Y(Ba_{2-z}Sr_z)Cu_4O_8$, for which T_c remains essentially the same within a broad range of compositions ($0 \leq z \leq 0.6$), no change of the temperature behavior of the phonon and thus no variation of the position of the superconducting gap was observed. These experimental findings confirm that the picture of superconductivity-induced changes in the phonon parameters holds in the double-chain compound, similarly to the single-chain material. We also report the effects of these dopants on the phonon anomaly which has been attributed to a spin gap.

I. INTRODUCTION

Optical phonons and their interactions with other elementary excitations in high-temperature superconductors were investigated intensively after the discovery of these remarkable materials. The aim of these investigations is twofold: (i) to identify the excitations responsible for the pairing mechanism and (ii) to determine the size of the superconducting gap. As far as optical measurements are concerned, the behavior of the Raman-active B_{1g}-like phonon is probably one of the best studied features of $RBa_2Cu_3O_{7-\delta}$. This mode was shown to be strongly affected by the transition to the superconducting state: It exhibits large changes of the phonon linewidth and frequency (up to 10 cm^{-1}; see, e.g., Ref. 1 for references) and also a remarkable increase of its intensity.[2] Changes of the phonon parameters are believed to be a consequence of the interaction of the phonon in question with a continuum of states of the same (B_{1g}) symmetry which redistributes upon entering the superconducting state. The relatively large magnitude of the effects implies either the closeness in energy of the phonon to the superconducting gap or a large coupling strength. A quantitative analysis of these effects within a strong electron-phonon coupling approach was shown to yield a rather accurate estimate of the value of the superconducting gap $2\Delta_0/kT_c = (4.95 \pm 0.10)$ in $RBa_2Cu_3O_7$ and an electron-phonon coupling constant $\lambda = 0.01$–0.02, in agreement with predictions from calculations in the local-density approximation.[3] The z-polarized infrared- (IR-) active transverse optical phonons (B_{1u} symmetry) were also shown to exhibit superconductivity-induced self-energy effects in cuprate superconductors.[4–6] Interestingly, the position of the gap and even the electron-phonon coupling constants of the IR modes obtained for $RBa_2Cu_3O_7$ are quite similar to those obtained by Raman-scattering experiments despite the different symmetry of the respective phonons and, hence, the possible interaction channels: For the odd IR phonons interband transitions have to be invoked in an electron-phonon interaction, while for the even Raman phonons intraband transitions suffice.[5]

An important, more recent experimental finding is that the phonon self-energies in $YBa_2Cu_3O_{7-\delta}$ are sensitive to the presence of impurities in the sample even within the concentration range where they have only a minor influence on the superconducting transition temperature T_c.[7–11] To explain this effect the idea of impurity-

induced averaging of an anisotropic superconducting gap was put forward.[8] A further point is that the self-energies of the Raman-active phonons in $YBa_2Cu_3O_{7-\delta}$ were shown to depend strongly on oxygen content and ordering.[12–15] The superconductivity-induced variation of the B_{1g} phonon frequency and damping almost disappears for an oxygen deficiency $\delta \approx 0.15$, while T_c has decreased by only a few degrees from the maximal value ($T_c \sim 93$ K at $\delta \approx 0.05$).[14] Whether this effect is the consequence of the variation of the free-carrier (holes) concentration within the CuO_2 planes or is associated with the disorder of the chain-related states around the Fermi surface in oxygen-deficient materials remains controversial.[15,16]

We report below a study of the infrared phonon self-energy effects for the double-chain counterpart of $YBa_2Cu_3O_{7-\delta}$, namely, $YBa_2Cu_4O_8$. A remarkable property of the 124 material, the crystallographic unit cell of which consists of two face-centered $YBa_2Cu_3O_7$ blocks,[17–19] is the fixed oxygen stoichiometry. One can thus exclude effects of the variation of the chain-oxygen content on the properties of the double-chain materials and, in particular, the phonon self-energy effects. Early investigations showed that the superconductivity-induced changes of the phonon frequency and linewidths of the z-polarized plane-oxygen mode with frequency $\sim$300 cm^{-1} are as strong for $YBa_2Cu_4O_8$ as for $YBa_2Cu_3O_7$;[20–24] their analysis yielded also the position of the superconducting gap of $YBa_2Cu_4O_8$, which seems to be slightly larger than that of $YBa_2Cu_3O_7$.[20,21,23] The aim of the present study is to perform a systematic investigation of the influence of the metal-superconductor transition on the infrared-active phonons and on the position of the superconducting gap of $YBa_2Cu_4O_8$ materials with transition temperatures T_c covering a range between 8 and 82 K; the variation of T_c was achieved by substituting elements at different crystallographic sites: Zn for Cu, Pr for Y, and Sr for Ba, respectively.

II. EXPERIMENTAL DETAILS

A series of ceramic samples $(Pr_xY_{1-x})Ba_2Cu_4O_8$, and $YBa_2(Cu_{4-y}Zn_y)O_8$, and $Y(Ba_{2-z}Sr_z)Cu_4O_8$ with nominal composition $0 \le x \le 0.8$, $0 \le y \le 0.15$, and $0 \le z \le 0.6$ was synthesized at an oxygen pressure of 1 atm by a polymerized complex method. Details of the sample preparation and characterization are given in Ref. 25. The superconducting transition temperature was determined by a conventional four-probe resistivity technique and magnetic susceptibility measurements. T_c was found to decrease continuously with increasing either Zn or Pr content from 82 to 46 K (for $y = 0.05$) or 8 K (for $x = 0.8$). In the case of substitution of Sr for Ba T_c was unaffected by the doping. The phase purity and lattice parameters were checked by x-ray diffraction and Raman spectroscopy. The impurity level was below 2% for all samples studied. The oxygen content was determined by iodometric titration and found to be 7.95(4) in all samples. The results of a detailed study of the microscopic structure of some of the samples are given in Ref. 25.

The far-infrared reflectivity measurements at near normal incidence were carried out on a Bruker-113v Fourier-transform interferometer in the spectral range 40–650 cm^{-1}. The samples were mounted on a cold finger of a He cryostat and the temperature was measured by a thermocouple placed close to the sample. A gold mirror was used as a reference.

III. REFLECTIVITY DATA ANALYSIS

In order to obtain precise information on the temperature behavior of the z-polarized TO phonon frequencies, linewidths, and intensities we performed Kramers-Kronig transforms of the reflectivity. The question arises as to how accurately the phonon parameters obtained without an effective-medium analysis are and how are they affected by the strong optical anisotropy of the superconductor, in particular by the high average reflectivity associated with the CuO_2 planes. All data discussed below refer to *transverse* optical phonons with an ionic displacement along the c axis of the material under investigation. By symmetry, these excitations cannot interact directly with the *longitudinal* plasma oscillations of the free carriers within the CuO_2 planes. A change of the average reflectivity level upon, e.g., sample cooling may, however, influence the phonon parameters obtained in the way described. To estimate the error due to changes in reflectivity, we present below the results of a calculation performed with a simple phenomenological model. The normal-incidence reflectivity of a semi-infinite bulk materials is

$$R(\omega) = \left| \frac{\sqrt{\tilde{\epsilon}(\omega)} - 1}{\sqrt{\tilde{\epsilon}(\omega)} + 1} \right|^2 . \tag{1}$$

We can simulate the complex dielectric function $\tilde{\epsilon}(\omega)$ by a Drude function and a series of uncoupled Lorentzian oscillators representing the phonons:

$$\tilde{\epsilon}(\omega) = \epsilon_\infty - \frac{\epsilon_\infty \omega_p^2}{\omega^2 + i\omega\Gamma} + \sum_n \frac{S_n \omega_{\mathrm{TO},n}^2}{\omega_{\mathrm{TO},n}^2 - \omega^2 - i\omega\gamma_{\mathrm{TO},n}} . \tag{2}$$

Here ϵ_∞ is the high-frequency dielectric constant; $\omega_{\mathrm{TO},n}$, $\gamma_{\mathrm{TO},n}$, and S_n are the TO phonon frequency, damping, and the oscillator strength, respectively; ω_p and Γ are the screened plasma frequency and carrier scattering rate.

First, we calculated two sets of reflectivity spectra from Eqs. (1) and (2) for two sets of free-carrier parameters approximately resembling the reflectivity values observed experimentally: ω_p =1500 cm^{-1} [curve 1 in Fig. 1(a)] and 3000 cm^{-1} (curve 3); Γ = 800 cm^{-1} for both curves; the contribution of three phonons at 150, 300, and 600 cm^{-1} was also included (see captions of Fig. 1 for more details). The frequency dependence of the imaginary part of the dielectric function obtained by the Kramers-Kronig transform of these simulated reflectivity spectra is shown

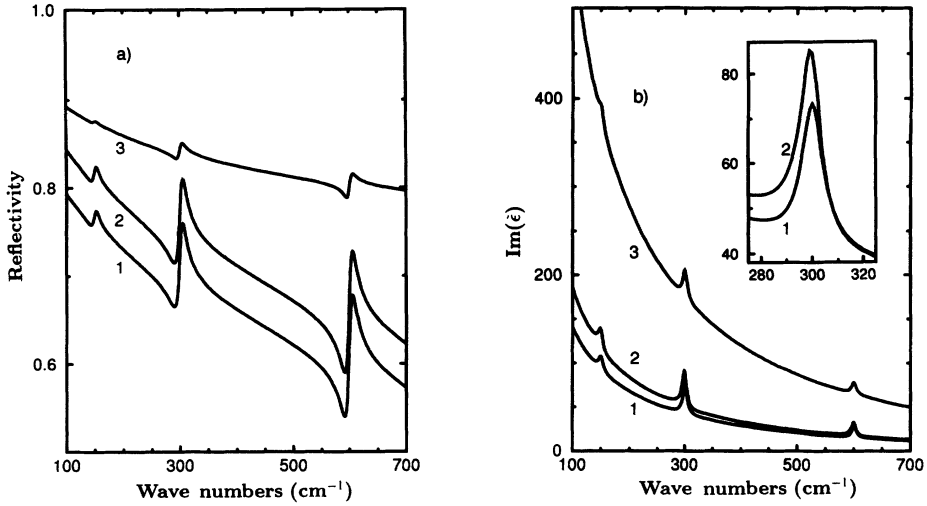

FIG. 1. (a) Model reflectivity spectrum obtained according to Eqs. (1) and (2) for three Lorentz oscillators with parameters (ω_{TO}, γ_{TO}, S, respectively) 150 cm^{-1}, 10 cm^{-1}, 1; 300 cm^{-1}, 10 cm^{-1}, 1; 600 cm^{-1}, 10 cm^{-1}, 0.2; $\epsilon_\infty = 5$. The other parameters are Γ=800 cm^{-1}; ω_p = 1500 cm^{-1} (curves 1 and 2) and 3000 cm^{-1} (curve 3). Curve 2, same as curve 1 with a constant additional reflectivity background of 0.05. (b) Imaginary part of dielectric function obtained by a Kramers-Kronig analysis of the reflectivities in (a). Note the variation of the line intensities, and thus of the oscillator strengths, for curves 1 and 2 (the latter is shifted down by 5 in the inset) upon the changes in reflectivity.

in Fig. 1(b). The phonon frequencies and the linewidths extracted from curves 1 and 3 in Fig. 1(b) by a fit to a Lorentzian line shape differ by not more than 0.2 cm^{-1} with respect those used initially for the calculation of the reflectivity according to Eqs. (1) and (2). The phonon oscillator strength also remains unaffected to within a few percent. In principle, this is an expected result for TO phonons which are uncoupled to the carriers. The situation is, of course, different for the longitudinal phonons, which can couple to the charge waves of longitudinal nature producing mixed plasmon-phonon excitations; the spectral function of the latter is described by the loss function Im$[-1/\tilde{\epsilon}(\omega)]$.

The second step consists of a simulated increase of the calculated [curve 1, Fig. 1(a)] reflectivity level by, say, 5% (curve 2). The corresponding frequency dependence of Im$[\tilde{\epsilon}(\omega)]$, obtained by a Kramers-Kronig analysis of this curve, is given by curve 2 in Fig. 1(b). A fit of the phonons to Lorentzian line shapes shows that the TO-phonon frequencies and linewidths remain weakly affected (by less than 1 cm^{-1}) by the artificial change in reflectivity; more apparent is, however, the increase in phonon oscillator strengths [see inset to Fig. 1(b)]. This means that one has to consider with care the data on the phonon oscillator strengths obtained by a Kramers-Kronig analysis in anisotropic media without an effective-medium analysis. An increase of the average reflectivity level upon sample cooling is probably one of the main reasons for the apparent increase of the IR phonon oscillator strengths observed experimentally for $YBa_2Cu_3O_7$ ceramics.[5,26] We point out in conclusion that the phonon frequencies and linewidths obtained by an analysis of Im$[\tilde{\epsilon}(\omega)]$ are only weakly dependent on the absolute reflectivity.

IV. RESULTS

The two upper curves in Fig. 2 are the reflectivity spectra of the undoped double-chain $YBa_2Cu_4O_8$ ceramic in the normal and the superconducting states (after Ref. 21). As for $YBa_2Cu_3O_{7-\delta}$,[26] the structures in the spectra of $YBa_2Cu_4O_8$ are known to be due to z-polarized optical phonons and the broad background is associated with the response of the highly conducting CuO_2 planes.[20,21,27] The seven phonon modes observed in the spectra have already been assigned[21,24,27–29] to the particular $B_{1u}(z)$ eigenmodes of $YBa_2Cu_4O_8$. As far as the transition to the superconducting state is concerned, the background reflectivity of $YBa_2Cu_4O_8$ does not change drastically upon lowering the temperature below T_c; the strongest relative changes occur around 200 and 500 cm^{-1}, where it increases by about 5%. One can already from the untreated reflectivity data of Fig. 2 see that the structure at about 300 cm^{-1}, which corresponds to the in-phase vibrations of the oxygen ions within the CuO_2 planes, becomes narrower and shifts considerably to the lower frequencies.

The reflectivity spectra of the ion-substituted materials are shown in Fig. 2. Their evolution upon cooling is very similar to those of undoped $YBa_2Cu_4O_8$. This fact, according to the results of Sec. III, suggests a weak influence of the background reflectivity on the TO-phonon frequencies and linewidths discussed below. Note only that at low temperatures the structure around 300 cm^{-1} exhibits pronounced broadening for the x=0.4 sample and, instead, narrows for Sr-doped material (z=0.4)

The temperature dependence of the linewidth and the frequency of the plane-oxygen mode for

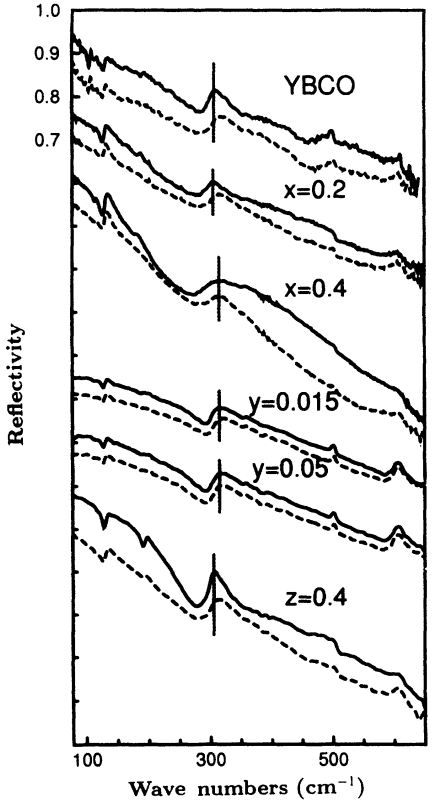

FIG. 2. Far-infrared reflectivity spectra of undoped $YBa_2Cu_4O_8$ (two upper curves) and $(Pr_xY_{1-x})Ba_2Cu_4O_8$ (x=0.2 and 0.4), $YBa_2(Cu_{4-y}Zn_y)O_8$ (y=0.015 and 0.050), and $Y(Ba_{2-z}Sr_z)Cu_4O_8$ (z=0.4) substituted ceramics in the superconducting state at $T \sim 10$ K (solid line) and the normal state just above T_c (dashed line). The superconducting transition temperatures are 82, 74, 63, 70, 46, and 82 K (from top to bottom). The reflectivity scale refers to the undoped and x=0.2 samples; offsets for the other samples are (from top to the bottom) −0.2, −0.8, −0.95, −1.0. For each sample the reflectivity in the normal state has been shifted vertically by −0.05. Vertical lines mark the low-temperature position of the local maximum in reflectivity around 300 cm^{-1}.

$YBa_2(Cu_{4-y}Zn_y)O_8$, obtained by a Kramers-Kronig transform of the reflectivity, are displayed in Fig. 3. Variation with composition of the temperature behavior of both frequency and linewidth is evident. The other solid-solution system studied is $Y(Ba_{2-z}Sr_z)Cu_4O_8$. Sr^{2+} is known not to affect the superconducting transition temperature of the double-chain $RBa_2Cu_4O_8$ compounds, but it reduces the size of the unit cell because of the fact that the ionic radius of Sr^{2+} is smaller than that of Ba^{2+} (see, e.g., Ref. 30). The temperature dependence of the linewidth and the frequency of the plane-oxygen phonon are shown in Fig. 4 for three compositions. Neither the linewidth nor the frequency are significantly affected by the disorder.

V. DISCUSSION

The temperature behavior of the IR-active phonons of the undoped and Pr-substituted double-chain $YBa_2Cu_4O_8$ superconductor has been analyzed in earlier studies.[21–24,31] We summarize here only the important results regarding temperature effects near T_c. A pronounced narrowing just below T_c of the plane-oxygen mode of undoped $YBa_2Cu_4O_8$ at $\sim$ 300 cm^{-1} and a broadening of the chain-oxygen mode[29] at $\sim$ 500 cm^{-1} yielded boundaries[21,32] for the position of the superconducting gap to which $B_{1u}(z)$ phonons couple: 310 cm^{-1} $\leq 2\Delta_0 \leq$ 497 cm^{-1}. Investigations of Pr-substituted double-chain $(Pr_xY_{1-x})Ba_2Cu_4O_8$ superconductors provided more precise information on the size of the gap. A narrowing upon cooling of the mode at 300 cm^{-1} for all samples with $x \leq 0.35$ signals the presence of a gap at higher energies. For higher Pr content ($x \geq 0.4$), however, a broadening of the TO mode is observed which was interpreted as a downshift of the superconducting gap across the phonon energy, creating in this way a new channel for phonon decay.[31] The crossover of the gap and the TO-phonon energies at $\sim$ 300 cm^{-1} occurs within a narrow range of Pr concentrations $0.35 \leq x \leq 0.40$ which

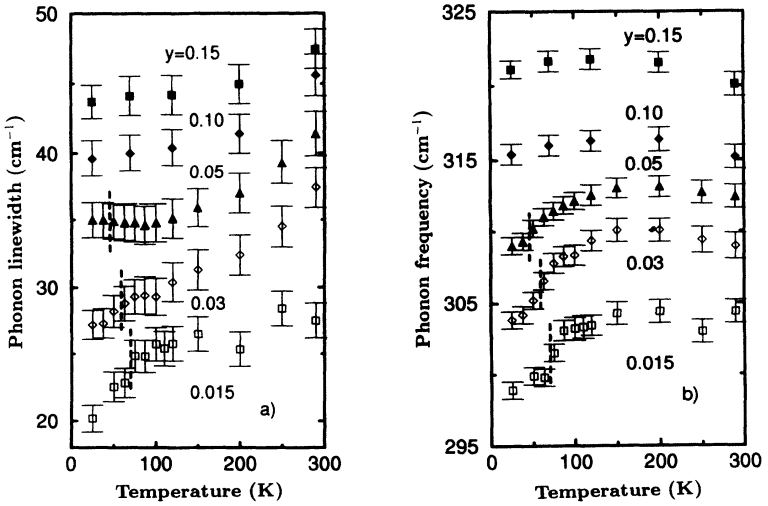

FIG. 3. Temperature dependence of the full linewidth at the half-maximum (a) and the frequency (b) of the plane-oxygen phonon of $YBa_2(Cu_{4-y}Zn_y)O_8$ superconductors. The curves from top to bottom are offset vertically by (a) 19, 13, 7, 3, 0 cm^{-1} and (b) 15, 10, 5, 2, 0 cm^{-1}. The dashed vertical lines mark the T_c for each sample.

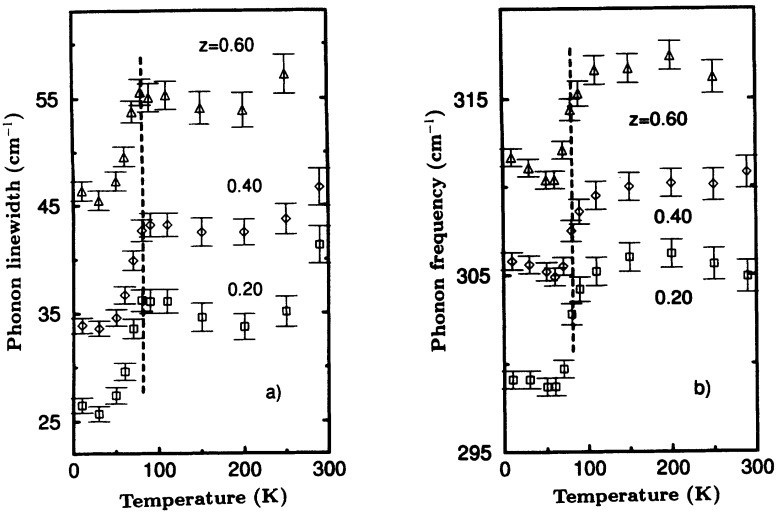

FIG. 4. The same as in Fig. 3 for $Y(Ba_{2-z}Sr_z)Cu_4O_8$. The offsets are (a) 15, 10, 0 cm^{-1} and (b) 10, 5, 0 cm^{-1} from top to bottom.

corresponds to 63 K $\leq T_c \leq$66 K and yields[31] a superconducting gap-to-T_c ratio $2\Delta_0/kT_c = (6.6 \pm 0.2)$.

A question of interest is whether the observed superconducting gap scaling with T_c is a general property of substituted $YBa_2Cu_4O_8$ or just inherent to the substitution by Pr ions in particular. To address this issue we investigated the reflectivity spectra and analyzed the phonon behavior of Zn-substituted $YBa_2Cu_4O_8$. Zn^{2+} is known[33] to occupy the plane Cu(2) sites in the lattice (at least for low doping levels). In spite of having the same valency state as Cu, Zn seems to act as an effective magnetic pair-breaking center because of the filled $3d$ electronic shell (see, e.g., Ref. 34): Zn substitution produces the strongest suppression of superconductivity in both $YBa_2Cu_3O_7$ and $YBa_2Cu_4O_8$.[33]

As can be seen from Fig. 3(a), at low Zn content (y=0.015, T_c=70 K) there is a pronounced narrowing of the phonon line just below T_c, like in the case of undoped $YBa_2Cu_4O_8$ and Pr-doped material with $T_c \geq$ 66 K [see Fig. 2(a) of Ref. 31]. This narrowing is suppressed for y=0.03 (T_c=59 K). For higher Zn contents a slight broadening of this phonon is observed, instead of a narrowing [see curve for y=0.05 (T_c=46 K) in Fig. 3(a)] indicating a shift of the gap to lower energies. The situation seems to be qualitatively similar, but less pronounced, to what is known for the Pr-doped double-chain material.[31] The crossover of the superconducting gap and the phonon occurs within the range $0.03 \leq y \leq 0.05$ which corresponds to 46 K $\leq T_c \leq$ 59 K and yields $2\Delta_0/kT_c = (8.2 \pm 1.0)$. This value is reasonably close to that obtained for $(Pr_xY_{1-x})Ba_2Cu_4O_8$. The temperature dependence of the TO-phonon frequency is also similar to that discussed for $(Pr_xY_{1-x})Ba_2Cu_4O_8$: A softening of the mode is observed for doping levels where the gap is situated above or slightly below the phonon energy [$y \leq 0.05$, Fig. 3(b)]. It is important to mention that a possible decrease of the electron-phonon coupling constant for T_c-reduced materials, as is known[35] to be the case for $YBa_2Cu_3O_{7-\delta}$, cannot explain the observed effects: It cannot account for the change in *sign* of the self-energy effect, e.g., a phonon narrowing instead of a broadening. Summarizing, we have thus established that for Zn and Pr concentrations differing by almost one order of magnitude the phonon self-energies are affected similarly by changes in T_c.

Note that the onset of the frequency softening occurs above T_c for $YBa_2(Cu_{4-y}Zn_y)O_8$ [Fig. 3(b)] similar to $(Pr_xY_{1-x})Ba_2Cu_4O_8$.[31] For both systems, however, this is not the case for the change in the linewidth with T. Similar effects have been observed earlier for either oxygen-deficient or ion-substituted single-chain $YBa_2Cu_3O_{7-\delta}$ and were interpreted in terms of superconducting fluctuations[36] or they were suggested to be a consequence of a modification of the Fermi surface due to the opening of a spin gap.[37] The close similarity of these above-T_c features in oxygen-depleted $YBa_2Cu_3O_{7-\delta}$ and stoichiometric $YBa_2Cu_4O_8$ becomes clear from the fact that despite the different chain structure they are both *underdoped* (see, e.g., Ref. 38). This is apparently a necessary condition for the opening of a spin-related gap already in the normal state.[39] Provided this particular property of the underdoped superconductors is responsible for the observed phonon softening in the normal state,[37,40] one can consistently explain other above-T_c anomalies, i.e., the deviation of the resistivity from linear behavior or the change in slope of the Hall coefficient.[41,42] A question of interest is how this anomaly reacts to the reduction of the superconducting transition temperature. According to the calculations of Nagaosa and Lee,[39] in the underdoped region one expects a shift of the above-T_c feature to higher temperatures upon decreasing the carrier density. This is indeed what is seen experimentally for $YBa_2(Cu_{4-y}Zn_y)O_8$: Upon lowering T_c the onset of the phonon softening seems to shift from about 90 K for $y = 0.015$ up to $\approx$ 140 K for $y = 0.050$ [Fig. 3(b)]. In the case of Pr doping (Fig. 2 of Ref. 31), however, the normal-state anomaly appears to follow T_c. These experimental facts may indicate a different relation between T_c and the in-plane carrier density of the two systems studied, which is not surprising because of different mecha-

nisms of T_c suppression (see, e.g., Ref. 43 and references therein).

As can be seen from Fig. 4 there is no measurable change in the temperature dependence of the frequency and the linewidth of the plane-oxygen mode upon replacement of the Ba^{2+} for Sr^{2+} in $Y(Ba_{2-z}Sr_z)Cu_4O_8$. Since this substitution is not accompanied by a change in T_c, we may conclude that the plane-oxygen phonon is sensitive only to those dopants which affect T_c. These qualitative observations prove that the phonon anomalies reported for the double-chain compound $YBa_2Cu_4O_8$ are indeed superconductivity related. The experimentally observed response of the phonon to the various dopants would be hard to explain otherwise. Similarly, the above-T_c effect in the phonon frequency remains constant in z.

More quantitatively, we have shown that variations near T_c of the frequency and the linewidth of the particular phonon are *all* compatible with strong-coupling calculations. Assuming the existence of an s-wave gap[32] we estimated the value of the gap from the experimentally observed crossover in energy of the gap and the phonon for a particular doping level. The numerical values are $2\Delta_0/kT_c = (8.2 \pm 1.0)$ and (6.6 ± 0.2) for $YBa_2(Cu_{3.96}Zn_{0.04})O_8$ and $(Pr_{0.37}Y_{0.63})Ba_2Cu_4O_8$, respectively. Under the assumption of a constant gap-to-T_c ratio, these values may be taken to represent the B_{1u}-symmetry gap of the undoped double-chain $YBa_2Cu_4O_8$ as well. Interestingly, the values found here are very close to others found in the literature for $YBa_2Cu_4O_8$ (between $6.0kT_c$ and $7.9kT_c$).[23,44] Note also that these results do not rule out the possibility of the existence of an anisotropic (d-wave) gap.[45]

VI. CONCLUSION

In this study we have significantly extended previous reports of the phonon self-energy effects near the superconducting transition temperature in the double-chain compound $YBa_2Cu_4O_8$. We found that doping which is accompanied by a reduction of the superconducting transition temperature T_c (Zn^{2+} in place of Cu^{2+} or Pr^{3+} in place of Y^{3+}) affects the phonon self-energy changes near T_c in a similar manner independently of the substituting ion. On the other hand, a replacement of Ba^{2+} by Sr^{2+} has no effect on T_c over a wide range of concentrations, and does not influence the temperature behavior of the phonon. These facts provide experimental evidence for the coupling of the plane-oxygen phonon in double-chain $YBa_2Cu_4O_8$ to electronic bands near the superconducting part of the Fermi surface.

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