Temperature dependence of the phonon frequencies and linewidths of $YBa_2(Cu_{1-x}M_x)_4O_8$ for M=Ni and Zn

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(Received 30 June 1997)

We report on the temperature dependence of Raman spectra for $YBa_2(Cu_{1-x}Ni_x)_4O_8$ ($x=0-0.02$) and $YBa_2(Cu_{1-x}Zn_x)_4O_8$ ($x=0-0.01$) polycrystals. Below the transition to the superconducting state an anomalous abrupt mode softening and line broadening of the Ba vibrational mode at 100 cm^{-1} are observed in the Raman spectra. The largest shift is observed in the undoped Y1:2:4 samples in which the frequency softens by approximately 5.5 cm^{-1} between T_c and 10 K while the amount of softening is greatly reduced with increasing dopant M concentration. This can be interpreted by considering that under the presence of two gap structure for Y1:2:4 the smaller gap $2\Delta_1$ is near or just above the Ba phonon frequency in undoped samples and its energy decreases with Ni or Zn doping. These phonon anomalies are consistent with the Zeyher-Zwicknagl model assuming the s-wave symmetry of pair function. [S0163-1829(98)05801-9]

I. INTRODUCTION

The superconductivity-induced phonon self-energy effects were found soon after the discovery of $Y_2Ba_4Cu_{6+n}O_{14+n}$ (YBCO) superconductors.[1] Generally, when the temperature decreases below T_c, the superconducting gap opening affects the frequencies and linewidths of the phonons whose frequencies are close to the gap frequency. For example, the frequency of the 340 cm^{-1} Raman mode of $YBa_2Cu_3O_{7-\delta}$ (Y1:2:3), which involves vibrations of the O(2) and O(3) atoms in the CuO_2 planes,[1] softens significantly as the temperature is lowered below T_c.[2] This phonon has also been observed to broaden as the sample is cooled below T_c.[2] These superconductivity-induced changes in the frequency and linewidth of the 340 cm^{-1} phonon appear to provide evidence for the strong electron-phonon interaction in the Y1:2:3 system. The superconductivity-induced phonon self-energy effects have also been theoretically explained within the BCS theory by Klein and Dierker,[3] or within the Eliashberg theory by Zeyher and Zwicknagl (ZZ).[4] Applying the predictions of the ZZ theory, the superconducting gaps were estimated in several polycrystalline $RBa_2Cu_3O_{7-\delta}$ compounds (R is a rare-earth element),[5] all of which has a T_c of about 90 K. The theory of the superconductivity-induced phonon self-energy effects has been further extended to the non-s-wave pairing systems.[6] The changes in the frequency and the linewidth between the normal and the superconducting state have been found to depend on the anisotropy of the superconducting gap $\Delta(\mathbf{k})$ and the presence of elastic or inelastic scattering.

For $YBa_2Cu_4O_8$ (Y1:2:4), the presence of two superconducting gaps was pointed out and their values were determined through phonon self-energy effects in the normal and superconducting states by Heyen *et al.*[7] They attributed these two gaps to $2\Delta_1$ (105 cm^{-1}) for the chain related bands and $2\Delta_2$ (320 cm^{-1}) for the plane related bands.

So far, to get further information about the superconducting related phonon anomalies, the substitution effects on Raman[8–10] and infrared (IR)[11] spectra have been investigated. In the YBCO compounds it is known that a $3d$ transition-metal substitution for Cu results in a reduction of T_c as the doping content is increased.[12] The decrease in T_c may change the position of the superconducting gap relative to the phonon energies. When the superconducting gap goes across the energy of a certain phonon, the frequency and the linewidth are to rapidly change. By investigating the temperature dependence of a certain phonon, we may get the information about the value and the symmetry of the superconducting gap. This method was applied to Pr-doped Y1:2:4 in the IR-active plane-oxygen phonon[11] and in the Raman-active Ba phonon.[8] Pr substitution for Y1:2:4 was found to give a constant ratio $2\Delta_2/kT_c=6.2$ for the large gap[11] and $2\Delta_1/kT_c=2.3$ for the small one.[8]

When YBCO compounds are doped with metal atoms, which copper site between the plane and the chain is preferentially substituted is crucial. It is well known that substitutions of divalent metals like Zn and Ni on the plane sites dramatically reduce the T_c, while those of trivalent Al, Fe, and Co on the chain sites show a similar but less dramatic effect.[12] Among the various $3d$ transition-metal substitutions in the YBCO compounds, the substitution of divalent metals such as Ni and Zn for Cu in plane sites is particularly interesting. This is because Ni or Zn substitution, in which the crystal structure is almost unaffected, causes a rapid drop in T_c. Noticeably, there is no significant qualitative difference in the suppression of T_c between substitutions of magnetic Ni and nonmagnetic Zn elements.

In this paper, we report on the temperature dependence of the frequencies and linewidths in the phonon Raman scattering for $YBa_2(Cu_{1-x}Ni_x)_4O_8$ system ($x=0-0.02$) and $YBa_2(Cu_{1-x}Zn_x)_4O_8$ system ($x=0-0.01$). We observed that the smaller gap $2\Delta_1$ is near or just above the Ba phonon frequency in undoped samples and its energy decreases with Ni or Zn doping.

II. EXPERIMENTAL DETAILS

Samples of $YBa_2(Cu_{1-x}M_x)_4O_8$ (M=Ni and Zn) with various contents of x were prepared by a conventional solid-

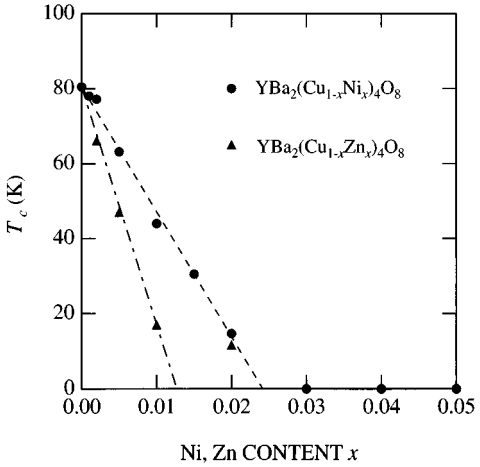

FIG. 1. Superconducting transition temperature T_c plotted as a function of M concentration x in $YBa_2(Cu_{1-x}M_x)_4O_8$ (M=Ni and Zn).

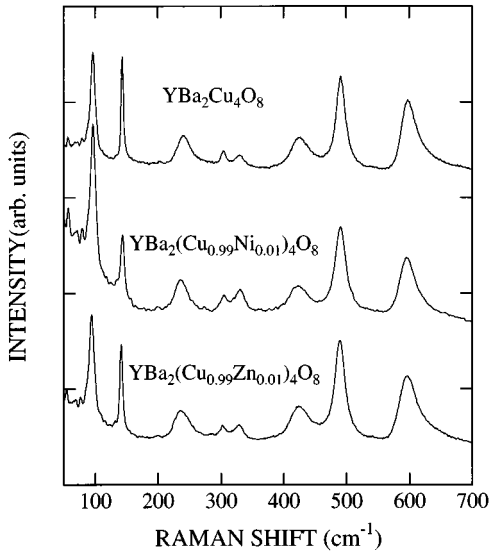

FIG. 2. Typical Raman spectra at room temperature for pure and Ni, Zn-doped samples.

state reaction method and a high-oxygen-pressure technique using a hot isostatic pressing apparatus in a mixed gas environment Ar+20%O_2 (O_2-HIP). Y_2O_3, $BaCO_3$, NiO, ZnO, and CuO powders were mixed to nominal compositions of $YBa_2(Cu_{1-x}M_x)_4O_8$. Powder mixtures were calcined at 860 °C for 12 h in air and then repeatedly sintered at 880–920 °C for 12 h several times with intermediate grindings. Sintered ceramics were finally treated by O_2-HIP at 1000–1100 °C for 40 h under an oxygen partial pressure of 20 MPa. The critical temperatures T_c determined by magnetic susceptibility were about 80.5, 78.0, 63.1, 44.0, and 14.7 K for nominal Ni content of $x=0$, 0.001, 0.005, 0.01, and 0.02, and 80.5, 66.1, and 47.0 K for Zn content of $x=0$, 0.002, 0.005, and 0.01, respectively. Details of the sample preparation as well as characterization (such as x-ray diffraction, resistivity, and magnetization) are described in Ref. 13.

Raman spectra were measured at various temperatures in the backscattering configuration. The 514.5 nm line of an Ar-ion laser was used for excitation. The incident laser beam from the Ar laser was focused on the sample surfaces with a diameter of about 8 μm and the power density was kept around 150 W/cm^2 to avoid heating effects. The temperatures were verified by using Stokes to anti-Stokes ratios of the spectral densities. The scattered light was detected with a Jasco NR-1800 triple monochromator and a charge-coupled-device detector. The samples were mounted on the cold finger of a liquid-He cryostat and the temperature was measured with a thermocouple placed near the sample.

III. RESULTS

Figure 1 shows the T_c plotted as a function of dopant M concentration x in $YBa_2(Cu_{1-x}M_x)_4O_8$ (M=Ni and Zn). T_c linearly decreases with Ni content, and the superconductivity disappears at a very small critical Ni concentration of 2.5%. The initial depression rates of T_c with increasing doping concentration x are about -32 and -67 K/at.% for Ni and Zn, respectively, which are larger than those in previous works.[14–17] The doping with Zn^{2+} (nonmagnetic ion) has a stronger effect on the suppression of T_c than that with Ni^{2+} (magnetic ion). These results are similar to those observed in $YBa_2(Cu_{1-x}M_x)_3O_{7-y}$ (M=Ni and Zn). The depression rate of T_c in $YBa_2(Cu_{1-x}M_x)_4O_8$ is larger than that in $YBa_2(Cu_{1-x}M_x)_3O_{7-y}$ (Ref. 12) for the same dopant. Since the structure of Y1:2:4 is similar to Y1:2:3 except for the double CuO chains, the larger depression rate of T_c in $YBa_2(Cu_{1-x}M_x)_4O_8$ than that in $YBa_2(Cu_{1-x}M_x)_3O_{7-y}$ should be related to the additional CuO chain in the double chain structure.

In Fig. 2 typical Raman spectra at room temperature for pure and Ni, Zn-doped samples are shown. Raman spectra for doped samples that resemble undoped ones suggest that both Ni and Zn doping hardly affect the lattice dynamics. There are no signs of peaks from impurity phases or phonon scattering induced by structural disorder. The dependencies of the frequencies of these vibrational modes on both Ni and Zn substitution are reminiscent of the behavior of the corresponding modes in $YBa_2(Cu_{1-x}Ni_x)_3O_7$ (Refs. 18–21) and $YBa_2(Cu_{1-x}Zn_x)_3O_7$.[20]

When the temperature decreases, the Ba vibrational mode ($\sim$100 cm^{-1}) shows a remarkable change in its frequency while the Cu(2) vibrational mode ($\sim$150 cm^{-1}) exhibits a normal anharmonic behavior. Values for the frequencies and linewidths of the Ba mode were obtained by fitting the corresponding Ba mode feature to a Fano profile[2] with a linear background given by

$$I(\omega)=I_0\,\frac{(\varepsilon(\omega)+q)^2}{1+\varepsilon^2(\omega)}+\text{background}, \tag{1}$$

where

$$\varepsilon(\omega)=(\omega-\omega_0)/\gamma. \tag{2}$$

Here ω_0 is the phonon frequency, γ is the linewidth (half width at half maximum), q is a parameter that defines the asymmetry of the measured line profile, and the background is taken to be a linear term of the form $B\omega+C$, where B and C are adjustable parameters.

The temperature dependencies of the frequencies of the Ba vibrational mode in $YBa_2(Cu_{1-x}Ni_x)_4O_8$ and

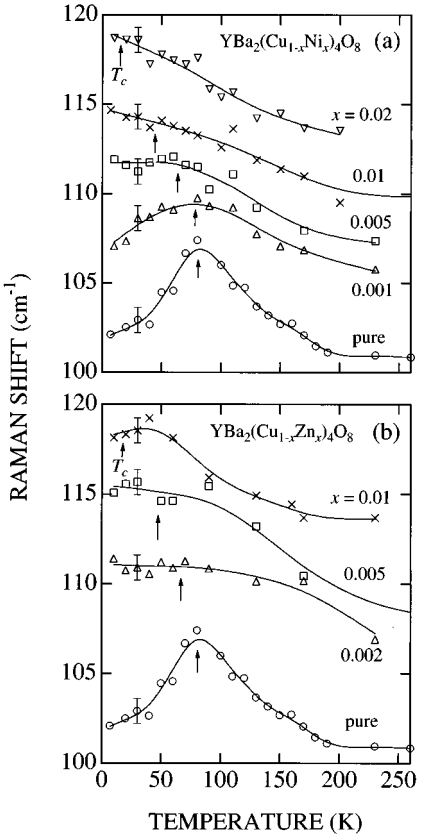

FIG. 3. Temperature dependence of the frequencies of the Ba mode for (a) $YBa_2(Cu_{1-x}Ni_x)_4O_8$ and (b) $YBa_2(Cu_{1-x}Zn_x)_4O_8$. The curves are offsets by (a) 12, 9, 6, 3, and 0 cm^{-1} and (b) 12, 8, 4, and 0 cm^{-1} from top to bottom. The solid lines are guides to the eye.

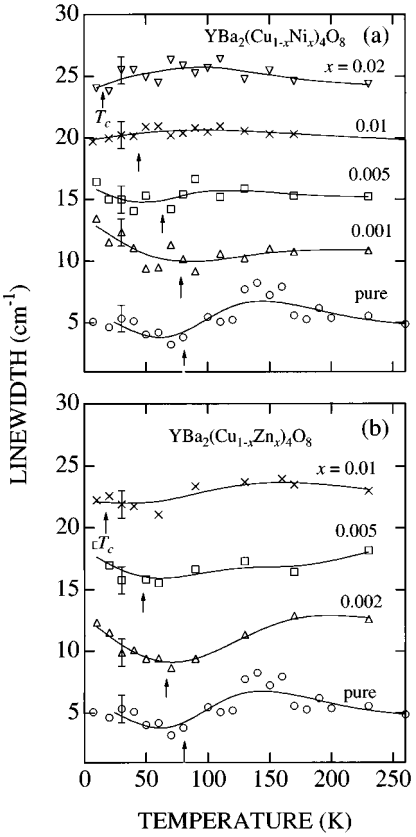

FIG. 4. Temperature dependence of the linewidths of the Ba mode for (a) $YBa_2(Cu_{1-x}Ni_x)_4O_8$ and (b) $YBa_2(Cu_{1-x}Zn_x)_4O_8$. The curves are offsets by (a) 20, 15, 10, 5, and 0 cm^{-1} and (b) 18, 12, 6, and 0 cm^{-1} from top to bottom. The solid lines are guides to the eye. Linewidths were determined by numerical fits to Fano (Ref. 2) profiles with a linear background.

$YBa_2(Cu_{1-x}Zn_x)_4O_8$ are summarized in Fig. 3. Above T_c this mode displays the normal slight shift to higher frequencies. Below T_c, however, this mode displays an abrupt mode softening for the samples with $x=0-0.005$. The largest shift is observed in the pure Y1:2:4 sample in which the frequency softens by approximately 5.5 cm^{-1} between T_c and 10 K. This value is consistent with results on other Y1:2:4 single crystals.[7] Our results show that Ni and Zn significantly suppress the magnitude of the superconductivity-induced phonon softening of the Ba vibrational mode at 100 cm^{-1}. When the Ni concentration x reaches 0.01, this mode slightly exhibits hardening. Unlike Ni, in the Zn-doped samples this hardening was not clearly exhibited. Though it is not clear whether this difference between the effects of Ni and Zn impurities is intrinsic or not for Y1:2:4, one possible explanation is that the weak chain disorder that may be due to a small Zn substitution at the chain-Cu(1) site reduces the self-energy effect. Actually the linewidths of the Cu(1) and O(1) phonons are increased at higher dopant concentrations in the Zn-doped Y1:2:4, while this is not the case in the Ni-doped one.[13] It should be noted that Käll *et al.*[10] observed a phonon softening below T_c in experiments carried out on the Zn-doped one, a result that is in sharp contrast to our results. These discrepancies may be caused by the difference of sample properties and/or preparation.

In Fig. 4, the temperature dependencies of the linewidths of the Ba vibrational mode are plotted for $YBa_2(Cu_{1-x}Ni_x)_4O_8$ and $YBa_2(Cu_{1-x}Zn_x)_4O_8$. The linewidth shows an unusual increase from room temperature down to about 130–150 K, then decreases rapidly to reach a minimum around T_c, where it starts to increase slightly again, as reported by Käll *et al.* A broadening of the phonon at low temperatures is observed for the samples with $x=0-0.005$. The largest broadening is observed at $x=0.001$ for Ni and 0.002 for Zn. For these samples the linewidth broadens by approximately 4.5 cm^{-1} between T_c and 10 K.

IV. DISCUSSION

The Ba phonon softens below T_c by as much as 5 cm^{-1}, corresponding to 5% of its frequency. With respect to the relative frequency change, this is the largest phonon anomaly reported so far for high-T_c superconductors. The large softening of the Ba mode may be explained by either a very strong coupling to the higher energy gap $2\Delta_2$ or by a coupling to the lower gap $2\Delta_1$ with an energy only slightly higher than the Ba mode frequency. It should be observed that the anomalies of Ba phonons consistently suggest the existence of a gap near $2\Delta_1 \sim 100$ cm^{-1}. This is because the dependence of the frequency on temperature appears to be normal in the Cu(2) phonon at about 150 cm^{-1}.

It is of interest to compare the observed superconductivity-induced change in frequency and line-

width to the predictions of strong-coupling models[4] in an attempt to ascertain the assumption that $T_c \propto \Delta_1$ in these systems.

Zeyher and Zwicknagl calculated the change of the complex self-energy, $\Delta\Sigma_v = \Delta\omega_v - i\Delta\gamma_v$, due to the electron-phonon coupling through redistribution of the electron density of states (DOS) assuming an isotropic s-wave symmetry for pair function.[4] Here $\Delta\omega_v$ is the change in the phonon frequency and $\Delta\gamma_v$ is the change in the phonon linewidth for mode v. The phonon frequency change is given by

$$\Delta\omega_v = \frac{\lambda_v \omega_v}{2} \mathrm{Re}(\Pi/N), \tag{3}$$

where λ_v is the electron-phonon coupling constant for the vth phonon. Π is the polarization, and N is the normal-state DOS per spin.

The change in the phonon linewidth due to superconductivity is

$$\Delta\gamma_v = -\frac{\lambda_v \omega_v}{2} \mathrm{Im}(\Pi/N). \tag{4}$$

ZZ have calculated the real and imaginary parts of Π/N for various temperatures below T_c and for different impurity scattering rates.

To compare the experimental results with their calculations using Eqs. (3) and (4), we normalized the experimental values of $\Delta\omega$, $\Delta\gamma$ by $\lambda_v\omega_v/2$. On the assumption that $T_c \propto \Delta_1$ (Δ_1: the superconducting gap energy) in $YBa_2(Cu_{1-x}M_x)_4O_8$ (M=Ni or Zn) and the gap value $2\Delta_1 = 105\ \mathrm{cm}^{-1}$ at $x=0$ (after Ref. 7), $\omega/2\Delta_1$ for the Ba phonon varies from 1.03 to 5.64 as x increases from 0 to 0.02 for Ni. In the same way it varies from 1.03 to 4.88 as x increases from 0 to 0.01 for Zn. As for the coupling constant λ_ν for the Ba mode, we used a value $\lambda_\nu = 0.034$ obtained by a local-density approximation frozen-phonon calculation.[22] We plotted the normalized $\Delta\omega$ and $\Delta\gamma$ in Figs. 5 and 6 as a function of the $\omega/(2\Delta_1)$ (ω is the Ba phonon frequency, $2\Delta_1 \propto T_c$), respectively. The dashed curve is the theoretical result[4] for $T/T_c = 0.16$ and an impurity scattering rate of $\tau^{-1} = 2\Delta_1$ (taken from IR measurements[23–25]). As for $x=0.02$ (Ni) and 0.01 (Zn) samples, we left these data out of Figs. 5 and 6 because their T_c are too low to distinguish the phonon anomaly from a normal anharmonic behavior below T_c.

It turns out that the decrease in the magnitude of softening between T_c and 10 K with increasing the dopant content is explained quite well by the ZZ model assuming an isotropic s-wave gap. The main prediction of the ZZ theory[4] that is based on the strong electron-phonon coupling is as follows: the phonon peaks with energies below 2Δ soften and do not change in the width, while those with energies above 2Δ harden and broaden. In addition, the most remarkable change should occur for $\omega \sim 2\Delta$ due to the divergence of density of states. The crossover point from softening to hardening is expected to lie near 1.1 for $\omega/2\Delta$. The sample just at this crossover point should have no anomaly in its frequency but have the largest phonon broadening. If the value of $2\Delta_1$ varies with the M-doping level, the temperature dependencies of frequency and linewidth for a certain phonon should be changed with doping. According to this prediction, the softening upon cooling of the phonon at 100 cm^{-1} for three samples with Ni content $x<0.005$ suggests the presence of the gap at higher energies. For higher Ni content of $x=0.01$ this mode appears to start to harden below T_c.

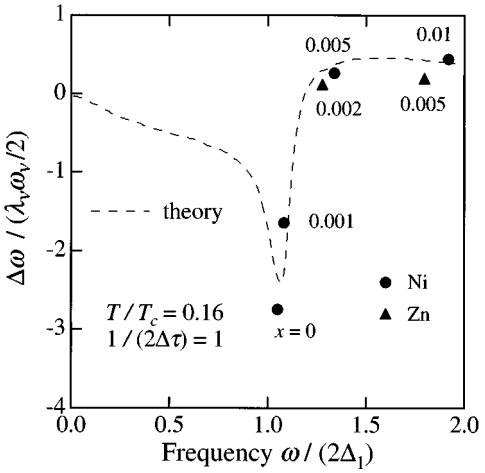

FIG. 5. A comparison of the measured frequency shifts with the theoretical curves (dashed line) of the real part of Π/N (Ref. 4) in the $YBa_2(Cu_{1-x}M_x)_4O_8$ (M=Ni,Zn) system. The experimental data have been normalized to $\lambda_v\omega_v/2$ ($\lambda_v = 0.034$, Ref. 22) on the assumption that $T_c \propto \Delta$ and the gap value $2\Delta_1 = 105\ \mathrm{cm}^{-1}$ at $x=0$ (Ref. 7). The parameters taken for the theoretical curve were scattering rate $\tau^{-1} = 2\Delta$ and $T/T_c = 0.16$.

The change in magnitude of the linewidth below T_c with increasing dopant content can also be fitted to the ZZ prediction. The largest broadening observed at $x=0.001$ for Ni and 0.002 for Zn indicates that the energy gap goes across the Ba phonon energy with doping. Concerning the data for Zn-doped samples, the discrepancy between the experimental results and the ZZ theory is somehow large. Though it is not

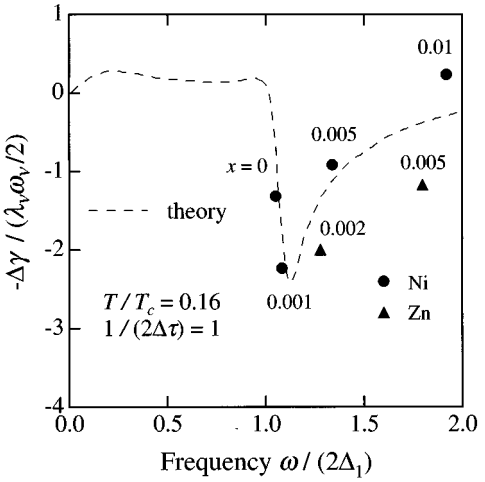

FIG. 6. A comparison of the measured shifts in linewidth with the theoretical curves (dashed line) of the imaginary part of Π/N (Ref. 4) in the $YBa_2(Cu_{1-x}M_x)_4O_8$ (M=Ni,Zn) system. The experimental data have been normalized to $\lambda_v\omega_v/2$ ($\lambda_v = 0.034$, Ref. 22) on the assumption that $T_c \propto \Delta$ and the gap value $2\Delta_1 = 105\ \mathrm{cm}^{-1}$ at $x=0$ (Ref. 7). The parameters taken for the theoretical curve were scattering rate $\tau^{-1} = 2\Delta$ and $T/T_c = 0.16$.

clear whether the discrepancy is intrinsic or not for Zn-doped Y 1:2:4, the reduction of the broadening is qualitatively consistent with the theory.

These results suggest that the gap symmetry is consistent with the s-wave model and the gap value $2\Delta_1$ scales linearly with T_c for $YBa_2(Cu_{1-x}M_x)_4O_8$ (M=Ni and Zn). Contrary, as for $\Delta\omega_\nu$ and $\Delta\gamma_\nu$ the two-peaks structure that was predicted by the d-wave model[6] is not observed. It is also noted that, as regards to the effect of the reduction of T_c on the Raman-active phonon self-energies, qualitatively similar results have been observed for Pr-substituted $YBa_2Cu_4O_8$.[8]

The s-wave symmetry for the smaller gap deduced from present Raman measurements is consistent with the results of the tunneling data along the c axis[26] and the microwave surface impedance measurements in Y 1:2:3.[27] On the contrary, an increasing number of experiments such as the tunneling along the a or b axis,[28] penetration depth,[29] microwave surface resistance,[30] NMR,[31] and specific heat[32] support the picture of a d-wave symmetry for the CuO_2 plane site. These apparently conflicting results may be explained if we can consider that the smaller gap in the YBCO systems has a different gap structure with a different symmetry. Note that the results on the scaling of the superconducting gap with T_c do not rule out the possibility of a d-wave gap in the large gap of YBCO systems.

Primarily, the two-gap structure, the chain and plane bands in the YBCO compounds was suggested by Kresin and Wolf.[33] They claimed that the charge transfer between these subsystems along with its uniquely short coherence length leads to the appearance of a two-gap structure. The hole doped CuO planes, which are thought to be the major structural and conducting unit for all cuprate superconductors, are by themselves intrinsically superconducting. As for the chains, the conducting state is intrinsically normal. The superconductivity in the chains is induced by the proximity effect due to the presence of the superconductivity in the neighboring layers. Thus the density of states of this compound in the superconducting state has the two-gap-like structure. Atkinson and Carbotte[34] have also studied the effect of proximity coupling of chains and planes in Y1:2:3.

In Y1:2:3, the Ba phonon exhibits a characteristic Fano line shape similar to that in Y1:2:4, but no anomaly is observed at T_c.[35] Heyen *et al.*[7] suggest that this result may be interpreted with different two superconducting gap values from those of Y1:2:4. Though the phonon Raman scattering hardly distinguish the chain band contribution from the plane one, the only difference between Y1:2:4 and Y1:2:3 is in the different character of the chains and the respective electronic chain bands. They also suppose that the so-called z-polarized experiments just probe this chain gap, whose wave functions contain mostly $O(4)p_z$-$Cu(1)d_{z^2-y^2}$-$O(1)p_y$ character. If their interpretation is correct, our results can be consistent with the two-gap structure models.

We finally note the effect of the impurity scattering for self-energy effect. Zeyher and Zwicknagl[4] mentioned that the change in the superconductivity-induced phonon self-energy may vanish in the extreme dirty limit. They showed that the superconductivity-induced shifts in phonon frequencies are very sensitive to impurities, especially in the case of $\omega/2\Delta > 1$. The structure around $\omega \sim 2\Delta$ is not much washed out by impurities. However, there is a dramatic decrease of the positive values of $\mathrm{Re}(\Pi/N)$ above the gap. This means that the phonon hardening above the gap in the clean case becomes a phonon softening if impurity scattering is important. Experimentally resistivity and infrared reflectance measurements provide an estimate the scattering rate of τ^{-1}. Sumner and co-workers[24] and Kim *et al.*[25] studied the resistivity and infrared reflectance for $YBa_2(Cu_{1-x}M_x)_3O_7$ (M=Ni and Zn), and found that τ^{-1} is roughly linear in the Ni concentration, and it reaches 95 cm^{-1} ($\sim 2\times 10^{13}$/s–140 K) at $x=4$ at. %, where $T_c=76$ K. The scattering rate decreases smoothly as T decreases. It is linear for $T \geqslant T_c$ and constant below T_c. Further, Ni and Zn produce the same scattering rate, proportional to x, within the $\pm 30\%$ uncertainty in determining $\tau^{-1}(x)$ from infrared reflectance measurements. In our case, even when the concentration x reaches 0.01 in Zn-doped samples, the Ba mode hardly exhibits hardening. It may be due to the fact that the scattering rate τ^{-1} should increase with increasing x then the self-energy effect may be reduced. (However, since the pure Y 1:2:4 system is not thought to be in the dirty limit, the impurity scattering mechanism does not seem to be appropriate for our results on the lower impurity samples.)

V. CONCLUSIONS

We investigated the temperature dependence of Raman spectra for polycrystalline $YBa_2(Cu_{1-x}M_x)_4O_8$ (M=Ni and Zn). We found that when the temperature decreases below T_c, the Ba vibrational mode at 100 cm^{-1} shows softening and broadening for $x=0$, while the amount of softening is greatly reduced with increasing dopant M concentration. This can be interpreted by assuming that the superconducting gap energy decreases with Ni and Zn doping and is consistent with the ZZ model assuming the s-wave symmetry for pair function.

ACKNOWLEDGMENT

This work was partially supported by NEDO for the R&D of Industrial Science and Technology Frontier Program.

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