Superconducting-plasma resonance along the *c* axis in various copper oxide superconductors

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Superconducting plasma along the c axis in the far-infrared region are observed in various high-T_c cuprates including electron-doped and Bi cuprates. It is shown that the sphere resonance of the powder samples occurs near the frequency where the plasma along the c axis exists, since it is only sensitive to the c-axis dielectric constant in the far-infrared region due to the metal-like property in the ab plane. This method provides an alternative to the reflectivity measurement of single crystals for investigating the plasma along the c axis. By measuring down to 7.5 cm^{-1}, the resonance is observed in $Bi_{1.85}Pb_{0.35}Sr_2Ca_2Cu_{3.1}O_y$, $La_{1.82}Ca_{1.18}Cu_2O_{6+\delta}$, $(Nd_{0.66}Sr_{0.205}Ce_{0.135})_2CuO_4$ (T* phase), $(Ba_{0.56}Sr_{0.44})_2Cu_{1.1}O_{2.2+\delta}(CO_3)_{0.9}$, $Pr_{1.85}Ce_{0.15}CuO_4$, and $Nd_2CuO_{4-x}F_x$. Among the Bi cuprates, however, no resonance is observed in $Bi_2Sr_2CaCu_2O_y$ and $Bi_2Sr_{1.6}La_{0.4}CuO_y$ in this frequency region, which suggests that the resonance exists below 7.5 cm^{-1}. The resonance frequencies are discussed based on the Josephson-coupled layer model, and the c-axis penetration depth λ_c are estimated from the resonance frequencies. It appears that λ_c depends on the insulating nature of the barrier layer between CuO_2 planes. Substitution effects on the resonance are also investigated in $La_{2-x}Sr_x(Cu_{1-z}Zn_z)O_4$, $Nd_{1.85}Ce_{0.15}(Cu_{1-z}Zn_z)O_4$, and $YBa_2(Cu_{1-z}Co_z)_3O_{6+\delta}$, and compared to the results of $YBa_2(Cu_{1-z}Zn_z)_3O_{6+\delta}$. Through the substitution, the resonance frequencies strongly decrease for all samples, indicating the strong increase of λ_c. The frequency decrease in $YBa_2(Cu_{1-z}Co_z)_3O_{6+\delta}$ suggests that the resonance frequency is determined by the barrier at the (BaO)[Cu(1)O_δ](BaO) chain layer, not at the Y layer.
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I. INTRODUCTION

One of the unique features in high-temperature copper oxide superconductors is the appearance of sharp reflectance edge along the c axis below T_c, which is attributed to plasma excitation. To our knowledge, this feature was observed by both the transmission and reflectivity spectra of $La_{2-x}Sr_xCuO_4$ ceramics, which were wrongly interpreted as the superconducting gap.[1,2] Bonn *et al.* interpreted the reflectance edge of ceramics as the plasma excitations without assigning its direction,[3] and then Noh *et al.* correctly interpreted the powder transmission peak as the plasma excitation along the c axis.[4,5] The presence of the plasma excitation was widely recognized and established in 1992, when Tamasaku *et al.* directly measured the reflectivity along the c axis using the large $La_{2-x}Sr_xCuO_4$ single crystals and confirmed the above interpretation.[6] Due to the large anisotropy of the materials, the frequencies of the plasma are in the far-infrared region, which are extremely low compared to the plasma frequencies along other directions and in conventional metals. Since they are smaller than the superconducting energy gap and optical-phonon frequencies, the plasma damping process is almost totally prohibited and the edges become sharp. This behavior provides useful information such as c-axis penetration depth, and many exotic electromagnetic phenomena have been predicted and some of them have been observed.[7–10] For example, Matsuda *et al.* found a sharp microwave magnetoabsorption resonance in $Bi_2Sr_2CaCu_2O_y$ for **E**∥ **B**∥ **c**, whose frequencies reduce as the magnetic field increases.[9] This behavior is also concerned with the mechanism of high-temperature superconductivity, since the absence of this edge above T_c may be caused by the confinement mechanism.[11,12] The plasma has been observed in $La_{2-x}Sr_xCuO_4$, $YBa_2Cu_3O_{6+\delta}$, and $YBa_2Cu_4O_8$.[6,13,14] However, due to the limitation of the available sample size for the reflectivity measurement and the low-frequency limit of the infrared measurement, there are no reports of the observation of the plasma in other high-T_c cuprates. For example, the c-axis reflectivity measurements of $Bi_2Sr_2CaCu_2O_y$, $Tl_2Ba_2Ca_2Cu_3O_{10}$, and $La_2CuO_{4+\delta}$ did not show the reflectance edge in the measured frequency region.[15–17]

In this paper, we show that the measurement of the sphere resonance of powder samples is an alternative to the reflectivity measurement of single crystals to observe the plasma along the c axis, since it is only sensitive to the c-axis dielectric constant in the far-infrared region. It is free from the limitation of sample size and the difficulty of crystal growth. Our measurements, extending down to 7.5 cm^{-1}, show the resonance in $Bi_{1.85}Pb_{0.35}Sr_2Ca_2Cu_{3.1}O_y$, $La_{1.82}Ca_{1.18}Cu_2O_{6+\delta}$, $(Nd_{0.66}Sr_{0.205}Ce_{0.135})_2CuO_4$, $(Ba_{0.56}Sr_{0.44})_2Cu_{1.1}O_{2.2+\delta}(CO_3)_{0.9}$, $Pr_{1.85}Ce_{0.15}CuO_4$, and $Nd_2CuO_{4-x}F_x$, but not in $Bi_2Sr_{1.6}La_{0.4}CuO_y$ and $Bi_2Sr_2CaCu_2O_y$. Substitution effects on the resonance frequencies are also investigated.

II. EXPERIMENTAL

Ceramic samples were synthesized by conventional solid-state reactions. After multiple calcinations with intermediate grinding, the powders were pressed into pellets and sintered. The powder x-ray diffraction, electrical resistivity, and mag-

TABLE I. The critical temperatures, resonance frequencies, and London penetration depths of various copper oxide superconductors. λ_c is estimated assuming the listed values for ϵ_∞.

Chemical formula	T_c (K)	ω_{peak} (5 K)(cm^{-1})	λ_c (5 K) (μm)
$Bi_{1.85}Pb_{0.35}Sr_2Ca_2Cu_{3.1}O_y$	107	17.0	23 (ϵ_∞=12)
$La_{1.82}Ca_{1.18}Cu_2O_{6+\delta}$	52	22.0	15 (ϵ_∞=20)
$(Nd_{0.66}Sr_{0.205}Ce_{0.135})_2CuO_4$	24	12.0	29 (ϵ_∞=17)
$(Ba_{0.56}Sr_{0.44})_2Cu_{1.1}O_{2.2+\delta}(CO_3)_{0.9}$	35	38.5	8 (ϵ_∞=20)
$Pr_{1.85}Ce_{0.15}CuO_4$	20	7.5	50 (ϵ_∞=12.6)
$Nd_2CuO_{4-x}F_x$	22	8.5	40 (ϵ_∞=17.0)

netic susceptibility measurements showed that the pellets had single phases, large Meissner signals and high transition temperatures comparable to the published data.[18–20] The transition temperatures determined by the midpoint of resistivity curves are summarized in Table I. The oxygen contents of $YBa_2\,[Cu_{1-z}(Zn,Co)_z]_3O_{6+\delta}$ were determined to be 6.66 $\pm$0.02 by iodometric measurements. For $Bi_2Sr_2CaCu_2O_y$, single crystals were used instead of ceramics. These pellets were ground into fine particles about $\sim$ 2 μm in diameter, mixed with Teflon or polyethylene powder in a volume fraction f from 0.1 to 1% and pressed into pellets about 2 mm thick. Since the grain size of the ceramics before grinding were about 10 to 100 μm, we assume that most of the particles in the composite pellets were single crystals.

Transmission spectra were measured between 7.5 and 200 cm^{-1} using a rapid scan interferometer. Below 20 cm^{-1}, a Si Bolometer operated at 1.5 K was used. Background spectrum were measured just before the sample spectrum measurement at each temperature, which were between 5 and 120 K. For each composition of the samples, samples measured in Teflon and polyethylene pellets were measured and it was confirmed that essentially the same resonance is observed in both samples.

III. RESULTS AND DISCUSSION

Sphere resonance is the absorption of small particles in a host material and its analysis is the conventional way to investigate the phonon spectrum of a solid sample. As shown below, the resonance occurs at the frequency where $\epsilon_{1c}(\omega)\approx -2\epsilon_m$, where $\epsilon_{1c}(\omega)$ is the real part of the c axis dielectric constant of the superconductor and $\epsilon_m(\omega)$ is that of the host material. The resonance frequency is a little smaller than the sharp reflectivity edge along the c axis that occurs at $\epsilon_{1c}(\omega)=0$. The effective dielectric constant of the medium, $\epsilon^{av}(\omega)$, including the isolated sphere anisotropic crystals, which are smaller than the wavelength, is calculated according to the relation

$$\frac{\epsilon^{av}(\omega)-\epsilon_m}{\epsilon^{av}(\omega)+2\epsilon_m}=\frac{f}{3}\sum_i \frac{\epsilon_i(\omega)-\epsilon_m}{\epsilon_i(\omega)+2\epsilon_m}, \tag{1}$$

where $\epsilon_i(\omega)$ and f are the dielectric constants of a single crystal along the ith crystal axis and the volume fraction of the particles.[4,5,21] In the far-infrared region, since ϵ_m is negligible compared to $\epsilon_{1a}(\omega)$ and $\epsilon_{1b}(\omega)$ in the high-T_c cuprates, Eq. (1) becomes

$$\epsilon^{av}(\omega)=\epsilon_m\frac{(1+2f)\epsilon_c(\omega)+2(1+f)\epsilon_m}{(1-f)\epsilon_c(\omega)+(2-f)\epsilon_m}. \tag{2}$$

At $\epsilon_{1c}(\omega)\approx -2\epsilon_m$, the real part of the denominator of Eq. (2) becomes zero and sphere resonance occurs. The resonance determined by $\epsilon_a(\omega)$ and $\epsilon_b(\omega)$ have no effect in this frequency region, since they are shifted to the near-infrared region due to the metal-like property in these directions.

Figure 1 shows the absorption spectra of the media composed of $La_{2-x}Sr_xCuO_4$, $YBa_2Cu_3O_{6.66}$, and $YBa_2Cu_4O_8$particles in polyethylene. The low-temperature absorption peaks shift to higher frequencies as the doping increases, and shift to smaller frequencies and weaken as the temperature approaches T_c. No peaks are observed in the normal state. The absorption peak frequencies and their temperature dependency correspond well to the changes in the point where $\epsilon_{1c}(\omega)$ crosses zero, which is determined by the c-axis reflectivity measurement of single crystals.[6,13,14] These observations confirm that the absorption peaks of the medium are determined by $\epsilon_c(\omega)\approx -2\epsilon_m$ and the plasma resonance along the c axis can be observed by this method.

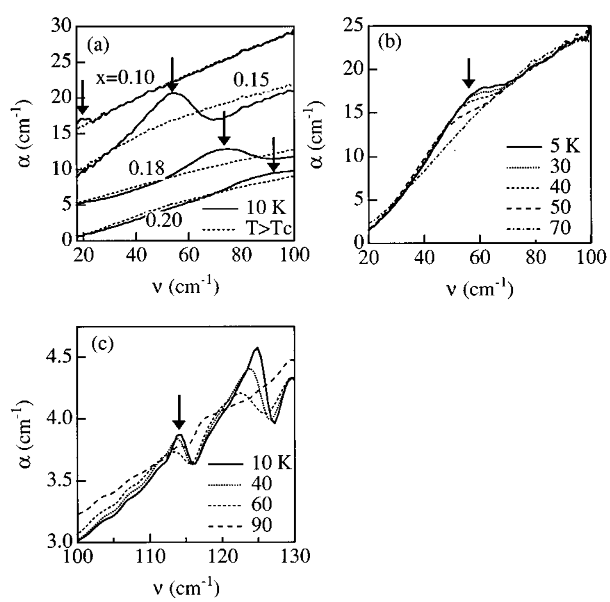

FIG. 1. Absorption coefficients of the mediums composed of (a) $La_{2-x}Sr_xCuO_4$, (b) $YBa_2Cu_3O_{6.66}$, and (c) $YBa_2Cu_4O_8$ particles in polyethylene. The arrows indicate the peaks. The peak at 127 cm^{-1} for $YBa_2Cu_4O_8$ may arise from the coupling between the plasma and a phonon at 132 cm^{-1} (Ref. 14).

It is interesting to compare this method with the reflectivity measurement of ceramic samples. The strong edge in the far-infrared region below T_c has also been reported in the reflectivity measurement of $La_{2-x}Sr_xCuO_4$ ceramic samples.[3,22,23] If the spectra can be analyzed by a short-wavelength approximation, it may be possible to assign that the edge comes from the plasma along c axis. However, since the grain size of the ceramic is comparable to the wavelength, we can neither apply the short-wavelength approximation nor long-wavelength effective-medium approximation. In this case, the low-frequency reflectivity depends on the grain size, ab plane, and c-axis contribution, and it is quite difficult to deduce a c-axis response from the spectra. In contrast to the reflectivity measurements of ceramics, the present method can determine the plasma along the c axis unambiguously. Since the size of the particles in the composite pellet is about 2 μm, we can apply the long-wavelength electrostatic approximation. In this approximation, the ab-plane contribution has no effect in the far-infrared region, as shown above.

Figures 2(a)–2(f) show the absorption coefficients of mediums composed of $Bi_{1.85}Pb_{0.35}Sr_2Ca_2Cu_{3.1}O_y$, $La_{1.82}Ca_{1.18}Cu_2O_{6+\delta}$, $(Nd_{0.66}Sr_{0.205}Ce_{0.135})_2CuO_4$, $(Ba_{0.56}Sr_{0.44})_2Cu_{1.1}O_{2.2+\delta}(CO_3)_{0.9}$, $Pr_{1.85}Ce_{0.15}CuO_4$, and $Nd_2CuO_{4-x}F_x$ particles in polyethylene. All of them show peaks below T_c, and the peaks shift to higher frequencies as the temperature decreases. These changes are the same as those observed for $La_{2-x}Sr_xCuO_4$, $YBa_2Cu_3O_{6.66}$, $YBa_2Cu_4O_8$, and confirm that the peaks are plasma resonance along the c axis. The appearance of plasma in the superconducting state and the absence of it in the normal state seem a common feature of most high-T_c cuprates.

In Fig. 2, the peaks of $La_{1.82}Ca_{1.18}Cu_2O_{6+\delta}$ and $(Ba_{0.56}Sr_{0.44})_2Cu_{1.1}O_{2.2+\delta}(CO_3)_{0.9}$ are broad compared to the others. This suggests the existence of unpaired carriers below T_c in these materials, which is also seen in $La_{1.8}Sr_{0.2}CuO_4$ in Fig. 1(a). The reflectivity measurement of $La_{1.8}Sr_{0.2}CuO_4$ single crystal has revealed that the existence of the unpaired carriers is intrinsic in highly doped cuprates, which was explained by the electronical phase separation, not by the effect of strong disorder or by an anisotropic gap.[24] In the present experiment, however, it seems that the broadenings are mainly due to sample inhomogeneity rather than the above explanation, since it is difficult to make good samples of $La_{1.82}Ca_{1.18}Cu_2O_{6+\delta}$ and $(Ba_{0.56}Sr_{0.44})_2Cu_{1.1}O_{2.2+\delta}(CO_3)_{0.9}$.[18,19]

If we neglect the unpaired carriers in the superconducting states, the low-frequency dielectric function of the superconductor can be written as $\epsilon_{1c}(\omega)=\epsilon_\infty-\omega_{ps}^2/\omega^2$, where ϵ_∞ and ω_{ps} are the high-frequency dielectric constant and plasma frequency of the condensed carrier. In this approximation, the London penetration depth λ_c becomes

$$\lambda_c=\frac{c}{\omega_{ps}}=\frac{c}{\sqrt{\epsilon_\infty+2\epsilon_m}\,\omega_{\rm peak}}. \tag{3}$$

So λ_c can be calculated from the peak frequency $\omega_{\rm peak}$ and ϵ_∞. ϵ_∞ can be estimated by the single-crystal value of the materials for Pr_2CuO_4, Nd_2CuO_4,[25] and ϵ_∞ is assumed to be 17 for $(Nd_{0.66}Sr_{0.205}Ce_{0.135})_2CuO_4$, 20 for $La_{1.82}Ca_{1.18}Cu_2O_{6+\delta}$ and $(Ba_{0.56}Sr_{0.44})_2Cu_{1.1}O_{2.2+\delta}(CO_3)_{0.9}$, and 12 for $Bi_{1.85}Pb_{0.35}Sr_2Ca_2Cu_{3.1}O_y$. Table I summarizes the peak frequencies and penetration depths for these compounds. It is clear that λ_c depends on the insulating nature of the buffer layer between CuO_2 planes: it becomes progressively shorter for $(Pr, Ce)_2O_2$, Nd_2O_2, $(Ba_{0.55}Sr_{0.45})_2Cu_{0.1}O_{0.2+\delta}(CO_3)_{0.9}$, $(La, Sr)_2O_2$ layers, which is the same order in which c-axis resistivity decreases.[26] It is known that λ_c^2 is proportional to ρ_c and Δ^{-1} in the Josephson-coupled layer model along the c axis, where ρ_c and Δ are the c-axis resistivity and gap value, respectively.[14,27] Our result shows that the model qualitatively explains the ρ_c dependence of λ_c in the present single-layered high-T_c cuprates, so it is used as the basis for the following discussion.

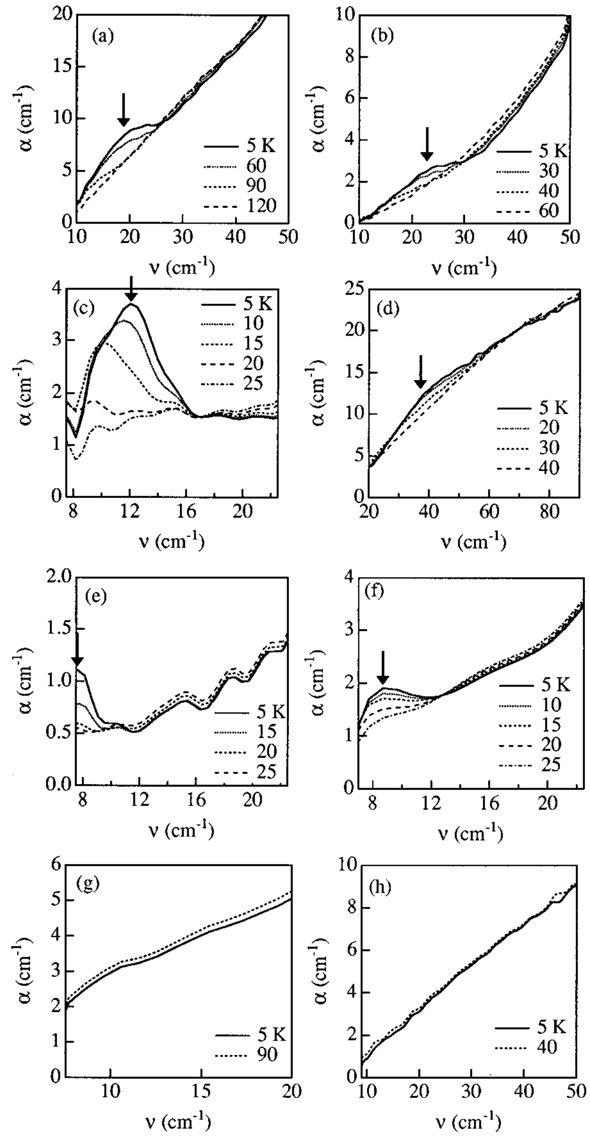

FIG. 2. Absorption coefficients of mediums composed of (a) $Bi_{1.85}Pb_{0.35}Sr_2Ca_2Cu_{3.1}O_y$, (b) $La_{1.82}Ca_{1.18}Cu_2O_{6+\delta}$, (c) $(Nd_{0.66}Sr_{0.205}Ce_{0.135})_2CuO_4$ (T^* phase), (d) $(Ba_{0.56}Sr_{0.44})_2Cu_{1.1}O_{2.2+\delta}(CO_3)_{0.9}$, (e) $Pr_{1.85}Ce_{0.15}CuO_4$, (f) $Nd_2CuO_{4-x}F_x$, (g) $Bi_2Sr_2CaCu_2O_y$, and (h) $Bi_2Sr_{1.6}La_{0.4}CuO_y$ particles in polyethylene. The arrows indicate the peaks.

It has been reported that the c-axis reflectivity of Bi2212 is essentially independent of temperature in a frequency range down to 30 cm^{-1}.[15] We also measured the absorption coefficients of mediums composed of $Bi_2Sr_2CaCu_2O_y$ ($T_c \approx$ 88 K) and $Bi_2Sr_{1.6}La_{0.4}CuO_y$ ($T_c \approx$ 28 K) particles in polyethylene. As shown in Figs. 2(g) and 2(h), they show no remarkable changes down to 7.5 cm^{-1}. However, the decrease of the absorption coefficients in the superconducting state in this frequency region suggests that peaks exist below 7.5 cm^{-1}. The difference in the plasma frequency among Bi2201, Bi2212, and Bi2223 can be explained by the larger anisotropy of Bi2201 and Bi2212 compared to Bi2223: the anisotropy parameter determined from the upper critical field in Bi2212 is 50–140, which is much larger than that of Bi2223, which is 31.[26] From this difference, it is roughly estimated that the plasma of Bi2212 may exist between 4 to 10 cm^{-1}, which is difficult to measure in the present experiment.[28,29] This is consistent with the results of the microwave absorption experiment under magnetic field, which predicts that the plasma of Bi2212 exists at 5.5 cm^{-1} at zero magnetic field.[10] The same explanation also applies to the Bi2201 case.

In Figs. 3(a)–3(e), the effects of Zn substitution on the resonance in $La_{2-x}Sr_x(Cu_{1-z}Zn_z)O_4$, $Nd_{1.85}Ce_{0.15}(Cu_{1-z}Zn_z)O_4$, and $YBa_2(Cu_{1-z}Zn_z)_3O_{6+\delta}$ are summarized. For unsubstituted $Nd_{1.85}Ce_{0.15}CuO_4$, essentially the same resonance has been reported previously.[5] Through Zn substitution, the resonance frequencies strongly decrease for all samples. Fukuzumi *et al.* found a similar strong shift for the reflectivity measurements of $YBa_2(Cu_{1-z}Zn_z)_3O_{6.63}$ single crystals, and explained the shift as follows:[30] In the Josephson-coupled layer model, the plasma frequency is proportional to $\rho_c^{-1/2}$ and $\Delta^{1/2}$ as discussed above. Assuming that $\Delta \sim$const and taking the temperature-dependent value of ρ_c just above T_c, the strong shift of the plasma frequencies can be explained by the increase of ρ_c through Zn substitution, since ρ_c shows semiconducting behavior ($d\rho_c/dT<0$) and follows the same line for all Zn concentrations ($z \leqslant$ 0.02). Although we cannot measure ρ_c in the experiment, the strong shift of the plasma frequencies through substitution also seems to be explained by the increase of ρ_c. In this case, the increase of ρ_c may be due to the increase in the residual resistivity and/or the semiconductive temperature dependence of the sample.

There seem to be two Josephson junctions along the c axis in $YBa_2Cu_3O_{6.66}$: one at the (BaO)[Cu(1)O_δ](BaO) chain layer and the other at the Y layer. So, it would be interesting to know which junction determines the plasma frequency. Figure 3(f) shows the absorption of $YBa_2(Cu_{1-z}Co_z)_3O_{6.66}$. It also shows a strong frequency decreases through Co substitution, which suggests that the above explanation may be applicable to this case. In this case, the increase of ρ_c may be due to an increase of junction resistivity at the (BaO)[Cu(1)O_δ](BaO) chain layer, since Co substitutes primarily the chain copper site Cu(1). This indicates that the junction that determines the plasma frequency is at the chain layer, not at the Y layer. If the latter junction determined the plasma frequency, a small frequency shift corresponding to the slight T_c reduction would occur, since the junction resistivity does not change through Co substitution.

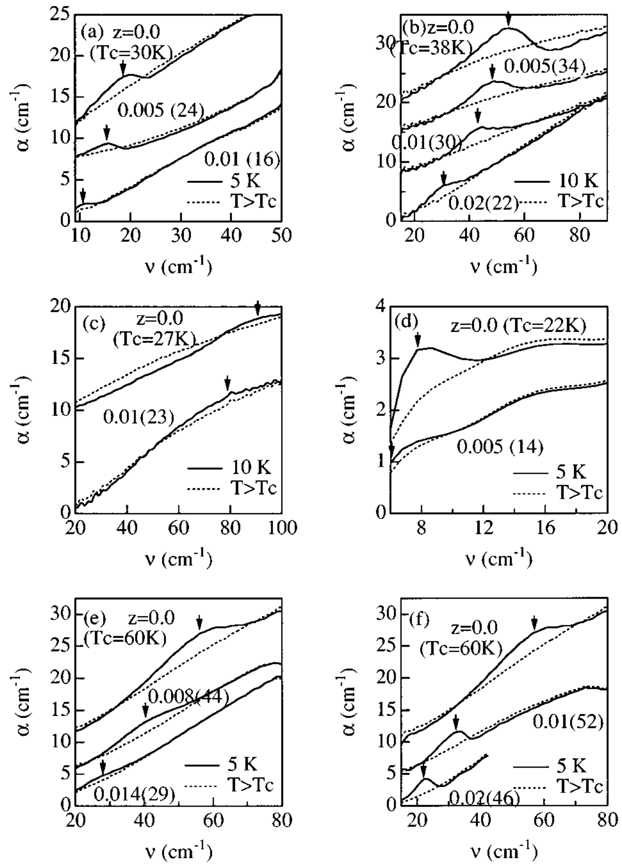

FIG. 3. Absorption coefficients of mediums composed of (a) $La_{1.9}Sr_{0.1}Cu_{1-z}Zn_zO_4$, (b) $La_{1.85}Sr_{0.15}Cu_{1-z}Zn_zO_4$, (c) $La_{1.8}Sr_{0.2}Cu_{1-z}Zn_zO_4$, (d) $Nd_{1.85}Ce_{0.15}Cu_{1-z}Zn_zO_4$, (e) $YBa_2(Cu_{1-z}Zn_z)_3O_{6.66}$, and (f) $YBa_2(Cu_{1-z}Co_z)_3O_{6.66}$ particles in polyethylene. The arrows indicate the peaks. All successive spectra are shifted up.

In summary, we showed that the measurement of sphere resonance is an easy way to determine the plasma along the c axis, and found that the resonance exists in $Bi_{1.85}Pb_{0.35}Sr_2Ca_2Cu_{3.1}O_y$, $La_{1.82}Ca_{1.18}Cu_2O_{6+\delta}$, $(Nd_{0.66}Sr_{0.205}Ce_{0.135})_2CuO_4$, $(Ba_{0.56}Sr_{0.44})_2Cu_{1.1}O_{2.2+\delta}(CO_3)_{0.9}$, $Pr_{1.85}Ce_{0.15}CuO_4$, and $Nd_2CuO_{4-x}F_x$. λ_c was estimated from the resonance frequencies. No resonance was observed in $Bi_2Sr_2CaCu_2O_y$ and $Bi_2Sr_{1.6}La_{0.4}CuO_y$; it may exist below 7.5 cm^{-1}. A strong plasma frequency decrease was observed through Zn and Co substitution, and was discussed on the basis of the Josephson-coupled layer model. From the analysis, it was found that the chain layer determines the junction resistivity in the model.

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