

Two-magnon and two-phonon excitations in some parent insulating compounds of the high- T_c cuprates

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Scattering spectra for three types of parent insulators of high- T_c cuprates, $\text{YBa}_2\text{Cu}_3\text{O}_6$, $\text{Ca}_{0.85}\text{Sr}_{0.15}\text{CuO}_2$, and La_2CuO_4 due to phonons and magnons were investigated varying incident photon energies between 1.7 and 2.7 eV. The magnon scattering peaks of these materials were found to be suppressed when the incident photon energy approaches the charge-transfer gap energy. By contrast, peaks due to zone-boundary phonons appeared with strong second-order features at the same energies. Those peaks were ascribed to the bending and stretching vibrations of oxygens in the Cu-O_2 plane with anharmonic character.

I. INTRODUCTION

Since the discovery of superconductivity in various cuprates, the superconducting mechanism which brings about a high- T_c up to 100 K has attracted much interest. A possibility of superconductivity mediated by spin excitation¹ (magnon) has been investigated intensively as well as the usual BCS-type mechanism mediated by phonons.²

Phonons and magnons of both superconducting and semiconducting cuprates have been investigated intensively using Raman spectroscopy. In La_2CuO_4 with CuO_6 octahedron structure, several peaks due to Raman-forbidden phonons have been observed with strong second-order features when the polarization of light is in the Cu-O_2 plane.^{3,4} Those peaks show resonance enhancement at around the charge-transfer (CT) gap.⁵ We have reported that similar scattering peaks due to zone-boundary phonons are observed in $\text{Ca}_{1.8}\text{Sr}_{0.2}\text{CuO}_3$ with Cu-O chain structure, suggesting that anharmonic electron-phonon coupling is a characteristic feature of semiconducting cuprates.⁶

In the compounds with Cu-O_5 pyramid structure, e.g., $\text{YBa}_2\text{Cu}_3\text{O}_x$, such scattering peaks due to Raman-forbidden phonons have not been observed. However, since the CT-gap energy of $\text{YBa}_2\text{Cu}_3\text{O}_x$ (1.7 eV) is lower than that of La_2CuO_4 (2.0 eV) (Ref. 7) and/or $\text{Ca}_{1.8}\text{Sr}_{0.2}\text{CuO}_3$ (2.1 eV),⁶ resonance excitation of the CT gap has not been performed in experiments using Ar lasers (photon energy 2.41 eV) as exciting light sources.

Magnon-pair scattering peaks have been observed in several semiconducting cuprates.^{8,9} It has been found that the two-magnon peak energies depend strongly on the CuO_n structure.¹⁰ However, very little information on the incident-photon energy dependence of magnon scattering has been obtained.^{11,12}

In the present study, we have investigated the resonance behavior of magnon- and phonon-scattering peaks

at around the CT-gap energies in $\text{YBa}_2\text{Cu}_3\text{O}_6$ and La_2CuO_4 as well as $\text{Ca}_{0.85}\text{Sr}_{0.15}\text{CuO}_2$ with Cu-O_4 squares. Magnon peaks have been found to be suppressed when the incident-photon energy comes near the CT gap in these materials. On the other hand, peaks due to Raman-forbidden phonons have been observed with strong second-order features under the excitation around the CT-gap energies.

II. EXPERIMENTAL DETAILS

Single crystals of $\text{YBa}_2\text{Cu}_3\text{O}_6$, $\text{Ca}_{0.85}\text{Sr}_{0.15}\text{CuO}_2$, and La_2CuO_4 were obtained using the flux-growth method. To obtain single crystals of $\text{Ca}_{0.85}\text{Sr}_{0.15}\text{CuO}_2$, powders of Bi_2O_3 , SrCO_3 , CaCO_3 , and CuO were mixed with a cation ratio $\text{Bi}:\text{Sr}:\text{Ca}:\text{Cu}=1:1:1:1$. They were placed in platinum crucibles and heated to 1100°C in air, cooled down to 950°C at the rate of 5°C/min, and then cooled gradually to 700°C at the rate of 3.5°C/h. The compositions of the cations were determined by energy-dispersive x-ray (EDX) spectroscopy. Single crystals of $\text{YBa}_2\text{Cu}_3\text{O}_6$ and La_2CuO_4 were grown using CuO flux and annealed in a reducing atmosphere to get stoichiometric compounds.

The orientation of the crystals was determined using the x-ray Laue method. Spectra were measured using the as-grown *ab* surfaces of $\text{YBa}_2\text{Cu}_3\text{O}_6$ and $\text{Ca}_{0.85}\text{Sr}_{0.15}\text{CuO}_2$ and the *ac* surface of La_2CuO_4 , which was cut from the ingot and polished by aluminum abscissa.

Raman spectra were measured in the backscattering configuration using Ar, He-Ne, and Ti-sapphire lasers as exciting light sources. The incident laser beams from the Ar and He-Ne lasers were focused on sample surfaces with a diameter of about 0.1 mm. The beam size of the Ti-sapphire laser was around 0.3 mm. The laser power was around 40 mW. The luminescence component from the Ti-sapphire laser was eliminated using a prism monochromator. The scattered light was detected with a Jobin

Yvon U-1000 double monochromator and a photon-counting detection system with a Hamamatsu R-943 photomultiplier.

III. RESULTS

Figures 1, 2, and 3 show the Raman spectra of $\text{YBa}_2\text{Cu}_3\text{O}_6$, $\text{Ca}_{0.85}\text{Sr}_{0.15}\text{CuO}_2$, and La_2CuO_4 , respectively, from 200 to 4000 cm^{-1} for the (X,X) configuration with excitation photon energies between 1.96 and 2.54 eV. The (X,X) means that the directions of the polarization of the incident and scattered lights are along the X direction, where X is the direction along the crystallographic a axis. In this configuration phonons and magnons with both A_{1g} and B_{1g} symmetries are Raman allowed. The spectra have been calibrated to a standard lamp to correct for the response of the spectrometer and detector, so that the spectra at different excitation frequencies may be directly compared in each figure. Effects due to the absorption and reflection of the incident and scattered lights were not corrected.

In the spectra of $\text{YBa}_2\text{Cu}_3\text{O}_6$ excited at 2.54 eV (Fig. 1), peaks are seen at 340, 450, 1290, and 2650 cm^{-1} . The 2650-cm^{-1} peak has been assigned to the magnon-pair scattering peak,⁹ while other peaks are assigned as phonon-scattering peaks.^{13,14} The 2650-cm^{-1} peak becomes weaker when excited at 2.41 eV and is hardly seen when excited at 1.96 eV, which is near the CT-gap energy (1.7 eV). On the other hand, several new peaks probably due to phonons appear below 1290 cm^{-1} when excited at 1.96 eV. The details of phonon-scattering spectra will be described later.

A similar behavior of the magnon-scattering peak is observed also in $\text{Ca}_{0.85}\text{Sr}_{0.15}\text{CuO}_2$. Figure 2 shows the Raman spectra of $\text{Ca}_{0.85}\text{Sr}_{0.15}\text{CuO}_2$ excited at 2.41 and 1.96 eV. A peak due to magnon-pair scattering is seen at

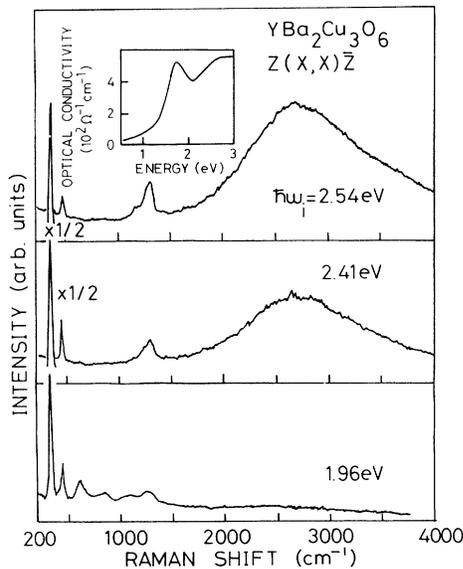


FIG. 1. Raman spectra of $\text{YBa}_2\text{Cu}_3\text{O}_6$ observed at room temperature with excitation photon energies at 1.96, 2.41, and 2.54 eV. The inset shows the optical-conductivity spectrum for polarization along the x direction.

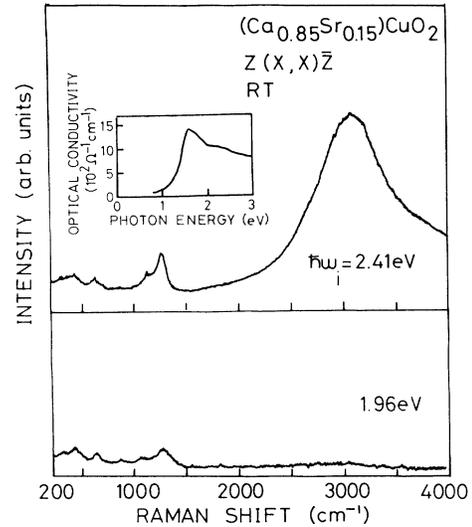


FIG. 2. Raman spectra of $\text{Ca}_{0.85}\text{Sr}_{0.15}\text{CuO}_2$ observed at room temperature for the (X,X) configuration with excitation photon energies at 1.96 and 2.41 eV. The inset shows the optical conductivity for polarization along the x direction.

3100 cm^{-1} when excited at 2.41 eV, as reported by Tokura *et al.*¹⁰ On the other hand, when excited at 1.96 eV, the magnon-pair scattering peak is hardly seen, though the photon energy of the magnon peak nearly agrees with the CT gap energy, 1.6 eV (outgoing resonance). The correction due to the absorption and reflection of the incident and scattered light or the ω^4 factor in the dipole matrix element cannot explain such a drastic decrease of the intensity of the magnon peak in the spectrum with excitation photon energy of 1.96 eV in comparison with that of 2.41 eV.

Figure 3 shows the Raman spectra of La_2CuO_4 with

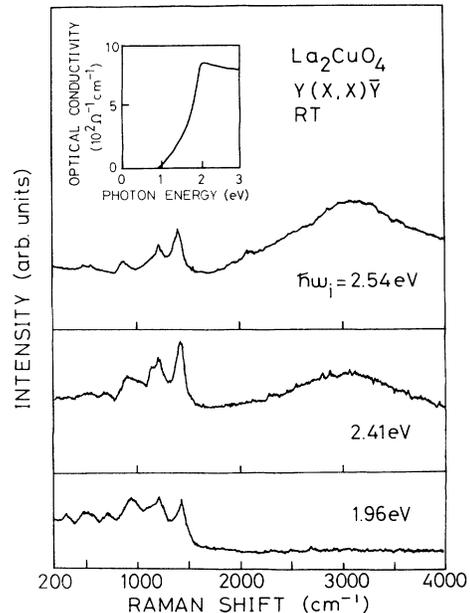


FIG. 3. Raman spectra of La_2CuO_4 observed at room temperature for the (X,X) configuration with excitation photon energies between 1.96 and 2.54 eV. The inset shows the optical-conductivity spectrum for polarization along the x direction.

the largest CT-gap energy (2.0 eV) among the three. In Fig. 3 the magnon-scattering peak is seen at 3100 cm^{-1} when excited at 2.54 and 2.41 eV. The photon energy of the magnon peak agrees with the CT-gap energy when excited at 2.41 eV. However, the intensity of the magnon peak becomes about a half of that excited at 2.54 eV. In the spectrum excited at 1.96 eV (resonance excitation of the CT gap), the magnon-scattering peak is not seen.

It is clarified that, in those three semiconducting cuprates, the magnon-scattering peaks do not show resonance enhancement at the CT gap. Next, we see the resonance behavior of the phonon-scattering peaks in detail.

Figure 4 shows the Raman spectra of $\text{YBa}_2\text{Cu}_3\text{O}_6$ observed at room temperature for the (X,X) configuration with excitation energies between 1.69 and 2.71 eV. In the spectra excited at 2.71 and 2.54 eV, peaks are seen at 140, 340, 450, 640, and 1290 cm^{-1} . The 140-, 340-, and 450 cm^{-1} peaks are due to Raman-allowed A_{1g} -mode phonons as reported by Burns *et al.*,¹³ while the 640 cm^{-1} peak is probably due to a Raman-forbidden phonon. The 1290 cm^{-1} peak is broad and has a tail on the lower-energy side. This peak is supposed to be due to two-phonon scattering, as pointed out by Friedle *et al.*¹⁴

When excited at 1.96 eV, the 640 cm^{-1} peak becomes strong. In addition, broad peaks appear at around 850 and 1080 cm^{-1} . When excited at 1.69 eV, the 440-, 640-, 850-, 1080-, and 1290 cm^{-1} peaks become considerably stronger. This enhancement is probably due to the resonance with the CT gap. These peaks became weak in the (X',Y') or (X,Y) configurations showing A_{1g} character, where X' (Y') is the direction rotated 45° from the X (Y) direction. The 450 cm^{-1} peak in the spectra excited at 2.54 and 2.71 eV shifts to the lower-energy side by about 10 cm^{-1} when excited at 1.69 eV. So probably these peaks have different origins. Raman shifts of the 850- and 1290 cm^{-1} peaks are about twice of those of the 440- and 640 cm^{-1} peaks and that of the 1080 cm^{-1} peak sum of

the 440- and 640 cm^{-1} peaks, suggesting that these peaks are due to two-phonon scattering of corresponding phonons. It should be noted that, under resonance excitation of the CT gap, the intensity of the 850- and 1080 cm^{-1} peaks becomes nearly equal to that of the 1290 cm^{-1} peak, while only the 1290 cm^{-1} peak is seen when excited at 2.71 and/or 2.54 eV.

Figure 5 shows the Raman spectra of $\text{Ca}_{0.85}\text{Sr}_{0.15}\text{CuO}_2$ observed for the (X,X) configuration with excitation photon energies at 1.96, 2.41, and 2.54 eV. In the spectra excited at 2.54 and 2.41 eV, peaks are seen at 630 and 1260 cm^{-1} and a broad feature at around 300 cm^{-1} . There is no Raman-allowed phonon in the $(\text{Ca,Sr})\text{CuO}_2$ -type structure.¹⁵ Thus the 630 cm^{-1} peak is probably due to a Raman-forbidden phonon. The 1260 cm^{-1} peak is broad and has a tail on the lower-energy side as the 1290 cm^{-1} peak in $\text{YBa}_2\text{Cu}_3\text{O}_6$. In the spectra excited at 1.96 eV, new peaks are seen at 420, 850, and 1050 cm^{-1} , probably due to the resonance with the CT gap at 1.7 eV.

Figure 6 shows the Raman spectra of La_2CuO_4 observed for the (X,X) configuration with excitation photon energies between 1.96 and 2.71 eV. In the spectrum excited at 2.71 eV, peaks are seen extending up to 1415 cm^{-1} . Peaks at 230 and 430 cm^{-1} are due to the A_{1g} -mode phonons,¹⁶ while other peaks are due to Raman-forbidden phonons.⁴ These Raman-forbidden peaks become stronger when excited at 2.41 and 1.96 eV, indicating resonance enhancement at the CT gap at 2.0 eV. The intensity of the peaks around 900 and 1200 cm^{-1} becomes nearly the same as that of the 1415 cm^{-1} peak when excited at 1.96 eV, while, when excited at 2.71 eV, the 1415 cm^{-1} peak is considerably stronger than the other peaks.

As shown here, the common feature observed in these three compounds is that many scattering peaks due to Raman-forbidden phonons appear and become strong

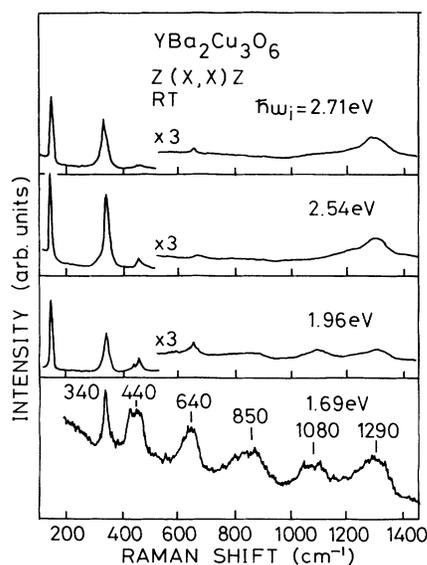


FIG. 4. Raman spectra of $\text{YBa}_2\text{Cu}_3\text{O}_6$ observed at room temperature for the (X,X) configuration with excitation photon energies between 1.69 and 2.71 eV.

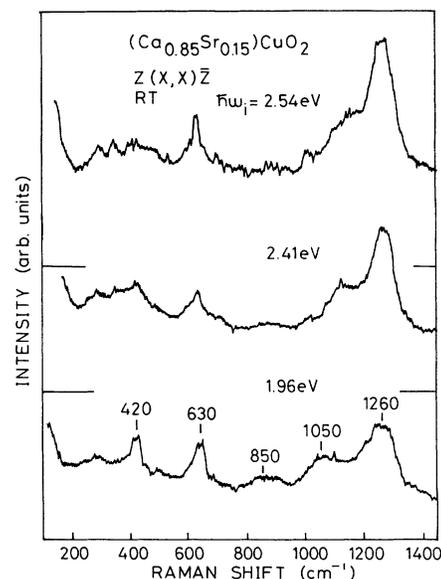


FIG. 5. Raman spectra of $\text{Ca}_{0.85}\text{Sr}_{0.15}\text{CuO}_2$ observed at room temperature for the (X,X) configuration with excitation photon energies between 1.96 and 2.54 eV.

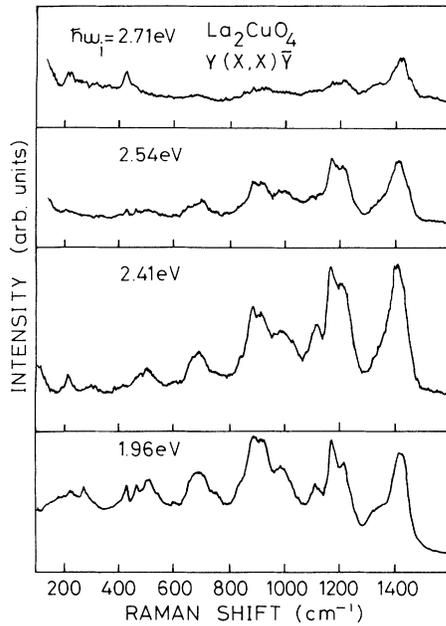


FIG. 6. Raman spectra of La_2CuO_4 observed at room temperature for the (X,X) configuration with excitation photon energies between 1.96 and 2.71 eV.

when the excitation photon energy becomes close to the CT-gap energy.

IV. DISCUSSION

A. Suppression of the magnon-scattering peak at the CT gap

In the previous section, it was shown that phonon-scattering peaks were enhanced while magnon peaks were suppressed at around the CT-gap energies.

The theory of light scattering by two-magnon excitation was investigated intensively two decades ago.¹⁷ It was found that, in the antiferromagnetic compounds, e.g., MnF_2 and FeF_2 , magnon-pair scattering occurs through the excited-state nondiagonal exchange interaction under the intra-atomic excitation of transition-metal ions. Recently, a theory of Raman scattering in a Mott-Hubbard system was developed.¹⁸ Those theories, however, seem not to be available for the two-magnon scattering under the resonance excitation of the CT gap. So we can only make two possible speculations on the suppression of magnon scattering around the CT gap energy.

One possibility is as follows. It is well known that, in the superconducting cuprates, the antiferromagnetic order of Cu spin is destroyed by the ferromagnetic interaction of Cu spins with oxygen hole spins. In the Raman spectra of superconducting cuprates, a broad continuum with nearly constant ω dependence has been observed in contrast with a two-magnon scattering peak in semiconducting cuprates.¹¹ Under resonance excitation of the CT gap, a hole is created on the oxygen $2p$ state and an electron on the Cu $3d_{x^2-y^2}$ upper Hubbard state.¹⁹ The antiferromagnetic order may be destroyed by the optical-

ly excited oxygen holes even though the excitation density in the present experiments is not so high as to create the photoconductivity observed recently by Yu *et al.*²⁰

Another possibility is simply due to the Pauli exclusion principle. By the CT excitation, electrons are created on the Cu $3d$ upper Hubbard state as described above. Then the Cu $3d$ state is occupied by two electrons with both up and down directions, which inhibits the change of the spin component due to a (super)exchange interaction between nearest-neighbor Cu ions.

Anyhow, it is supposed that the fact that the magnon-scattering peak does not show resonance enhancement at the CT gap is in close relationship to the fact that the CT gap corresponds to a charge-transfer-type transition of electrons from oxygen to copper.

B. Identification of phonon peaks

In the Raman spectra of $\text{YBa}_2\text{Cu}_3\text{O}_6$, $\text{Ca}_{0.85}\text{Sr}_{0.15}\text{CuO}_2$, and La_2CuO_4 , peaks probably due to Raman-forbidden phonons were observed when excited at around the CT gaps. We give the identification of those peaks below.

Phonon energies of La_2CuO_4 and $\text{YBa}_2\text{Cu}_3\text{O}_6$ were obtained in the entire Brillouin zone using inelastic neutron scattering.²¹ Figure 7 shows the dispersions of La_2CuO_4 and $\text{YBa}_2\text{Cu}_3\text{O}_6$, which were traced from Ref. 21. In both compounds there are three phonon branches between 400 and 700 cm^{-1} . The highest- and second-highest-energy branches are due to phonons with the vibration mainly of the oxygens in the Cu-O₂ plane where the former is of the stretching type and the latter of the bending type. The third branch corresponds to the phonon with the vibration mainly of the apical oxygens. Since, by resonance excitation of the CT gap holes are created on the oxygens in the Cu-O₂ plane, it is expected that scattering due to the phonons associated with those

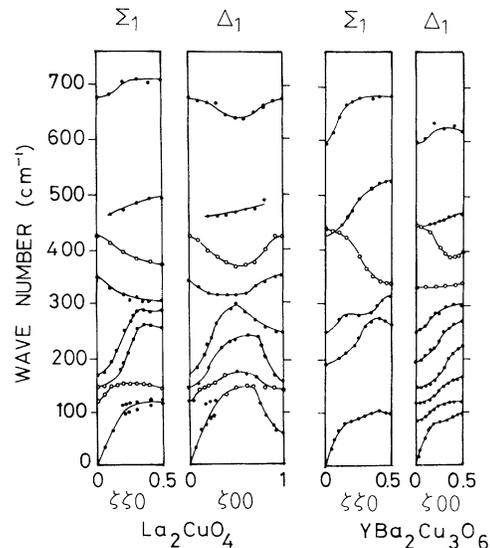


FIG. 7. Phonon-dispersion curves of Σ_1 and Δ_1 modes of La_2CuO_4 and $\text{YBa}_2\text{Cu}_3\text{O}_6$ obtained by inelastic neutron scattering (Ref. 21). The solid and open symbols refer to modes with elongations predominantly in the basal plane and along the c axis, respectively. Lines are a guide to the eye.

oxygen atoms is strong.

The highest-energy branches of La_2CuO_4 extend from 670 cm^{-1} (Γ point) to 710 cm^{-1} along the Σ axis or to 650 cm^{-1} along the Δ axis. An expanded figure of the spectrum of La_2CuO_4 excited at 1.96 eV is given in Fig. 8. In this figure the highest-energy peak is located at 1415 cm^{-1} with a shoulder at around 1330 cm^{-1} . The 1415-cm^{-1} Raman peak can be ascribed to the two-phonon scattering of the highest-energy branch with a \mathbf{k} vector around $(0.5,0.5,0)$, and the shoulder at around 1330 cm^{-1} can be ascribed to the phonon with a \mathbf{k} vector around $(0.5,0,0)$.

The peaks below 1215 cm^{-1} can also be explained on the basis of the results of neutron-scattering measurements. The peaks at 910 and 990 cm^{-1} can be assigned to the two-phonon scattering of the phonons in the second highest branch and the peaks between 1100 and 1215 cm^{-1} the combination of phonons in the highest and the second highest branch.

The peaks at 465 , 505 , and 685 cm^{-1} are supposed as the single-phonon scattering peaks of phonons described above. The broad width of these peaks are probably due to the contribution from the entire Brillouin zone, though only zone-center phonons are Raman allowed in the \mathbf{k} -selection rule of single-phonon scattering. The reason is not clear why single-phonon scattering peaks of the zone-boundary phonons appear in the Raman scattering. It may be caused by the orthorhombic distortion of the lattice, as pointed out in Ref. 4, or by some kind of lattice defects.

The Raman spectra of $\text{YBa}_2\text{Cu}_3\text{O}_6$ can also be explained using the results of neutron-scattering measurements. The 1290- and 850-cm^{-1} peaks can be ascribed to the highest and second highest branches, respectively, and the 1080-cm^{-1} peak the combination of them.

The origin of the 440- and 640-cm^{-1} peaks is rather uncertain. They may be ascribed to zone-boundary phonons or ir-active LO phonons, which become allowed through a Fröhlich-type interaction.²² However, we want to point out that the peaks above 850 cm^{-1} cannot be explained as the two-phonon scattering of LO phonons, because it cannot explain the following: (1) The 1290-cm^{-1} peak and the 850- or 1050-cm^{-1} peaks show different resonance behavior, and (2) the 1290-cm^{-1} peak has a tail on the lower-energy side (see Fig. 4) as the 1415-cm^{-1} peak of La_2CuO_4 .

In the spectra of $\text{Ca}_{0.85}\text{Sr}_{0.15}\text{CuO}_2$, peaks were observed at 850 , 1050 , and 1260 cm^{-1} . Neutron-scattering measurements of this material have not been performed. However, the 850- and 1260-cm^{-1} peaks may be ascribed to the two-phonon scatterings of bending- and

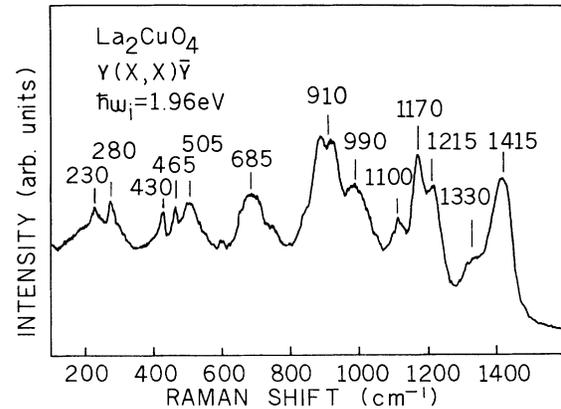


FIG. 8. Expanded spectra of La_2CuO_4 with excitation photon energy at 1.96 eV shown in Fig. 6.

stretching-type vibrations of oxygen in the Cu-O_2 plane, respectively, and the 1050-cm^{-1} peak the combination of them.

In the present study, it was shown that, in the Raman spectra of $\text{YBa}_2\text{Cu}_3\text{O}_6$, $\text{Ca}_{0.85}\text{Sr}_{0.15}\text{CuO}_2$, and La_2CuO_4 , scattering peaks due to zone-boundary phonons appear with strong second-order features, suggesting strong coupling of electron-hole pairs (excitons) with those phonons. It was shown that both stretching- and bending-type phonons couple strongly with excitons created under the CT-gap excitation. It is well known that, when the extension of the electronic wave becomes the same order of the lattice spacing, coupling with zone-boundary phonons becomes strong.²³ Finally, we want to point out that the strong coupling of excitons with zone-boundary phonons may be caused by the formation of small-radius excitons under resonance excitation of the CT gap of cuprates.

V. SUMMARY

It has been confirmed that in the Raman spectra of undoped cuprates, the two-magnon scattering peak has been found to become weak as the incident and scattered photon energy approaches the CT-gap energy. On the other hand, strong second-order features due to zone-boundary phonons have been observed when excited at around the CT gap.

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