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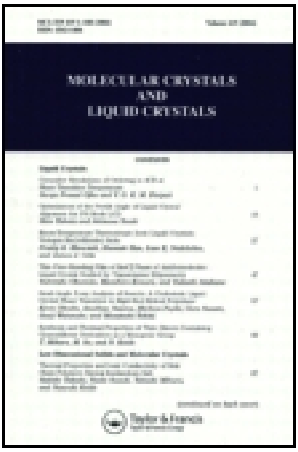

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Dependence of Raman Spectra of $YBa_2Cu_3O_6$ on Excitation Frequency

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DEPENDENCE OF RAMAN SPECTRA OF $YBa_2Cu_3O_6$ ON EXCITATION FREQUENCY

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ABSTRACT Raman spectra of semiconducting $YBa_2Cu_3O_6$ were measured as a function of wavelength of exciting radiation in the range from 441.6 nm to 632.8 nm. As the wavelength becomes longer, the Raman intensity of vibrational modes associated with the two-dimensional $Cu-O_2$ plane(451, 340 and 144 cm^{-1}) becomes larger. This effect is considered to be the resonance enhancement related to the charge transfer transition at around 1.75 eV in the $Cu-O_2$ network.

INTRODUCTION

The Raman scattering has been widely used to clarify the phonon structure of the high-T_c superconductors and related substances. In these investigations, radiations at 514.5 nm or 488.0 nm from Ar^+ laser have been preferentially employed. Raman spectra, however, may vary with the frequency of the exciting radiation due to the resonance effect. In these substances, electronic excited states are distributed continuously over the entire visible light region. Thus, we investigated Raman spectra of semiconducting $YBa_2Cu_3O_{\sim 6}$ by using several radiations in the range from 441.6 nm to 632.8 nm.

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EXPERIMENTAL

Polycrystalline samples of $YBa_2Cu_3O_{6.2}$ were prepared by heating $YBa_2Cu_3O_7$ up to 950°C in air followed by quenching to liquid nitrogen temperature. A fragment of the sample pellet was pulverized, mixed with a small amount of BaF_2 powder and applied for measurements of Raman scattering. The 245 cm^{-1} band of BaF_2 was used for the standard of the intensity. Since BaF_2 has the large band gap energy($\sim$10 eV[1]), Raman intensity could be assumed not to depend on the exciting frequency of visible region except for the forth power law dependence.

$YBa_2Cu_3O_{6.05}$ samples were prepared by heating $YBa_2Cu_3O_7$ in vacuum at 600° C. Sintered pellets were used for measurements.

Light sources used in the present study are radiations at 441.6 nm(2.81 eV) from a He-Cd laser, 476.5 nm(2.60 eV), 488.0(2.54 eV) nm and 514.5 nm(2.41 eV) from an Ar^+ laser, and 632.8 nm(1.96 eV) from a He-Ne laser.

RESULTS AND DISCUSSION

Figure 1 shows Raman spectra of the $O_{6.2}$ sample at various exciting frequencies. A trace d measured by 514.5 nm radiation agrees well with spectra of polycrystalline $YBa_2Cu_3O_6$ reported previously[2,3]. The bands at 453, 340 and 144 cm^{-1} have been assigned to the out-of-plane vibrations of the CuO_2 network[4,5]. The intensity of these three bands becomes weaker relatively to the 245 cm^{-1} band of BaF_2 and the 482 cm^{-1} band when the wavelength become shorter. Bands appear at 151 cm^{-1} and 657 cm^{-1} when the wavelength is shorter than 488 nm. At 632.8 nm excitation, on the other hand, the modes of the CuO_2 network are dominantly observed and a broad band is observed at 635 cm^{-1}.

It is reasonable that Raman intensity of the modes of the CuO_2 network is enhanced in the case of the long wavelength laser, considering the results of the investigation by Venkateswaran et al.[6] They observed an optical transition with significant intensity at 1.75 eV in $YBa_2Cu_3O_6$ and interpreted it as a charge-transfer transition in the CuO_2 network. The resonance effect may become significant when the radiation approaches the charge-transfer frequency.

The origin of the 635 cm^{-1} band is not clear, but it

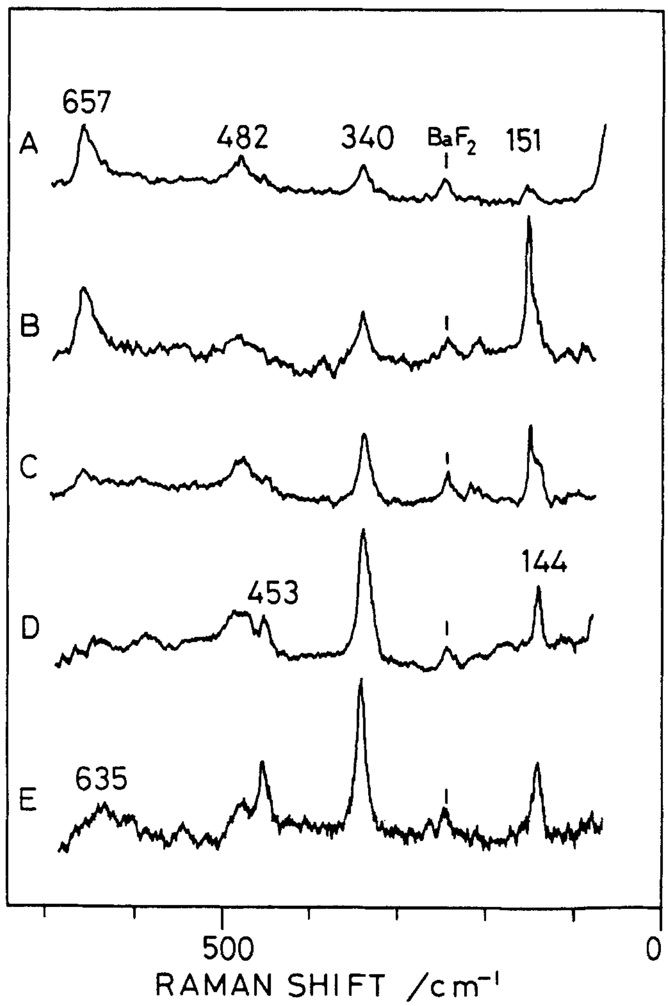

Fig. 1 Raman spectra of $YBa_2Cu_3O_{6.2}$ at 30 K obtained with 441.6 nm(A), 457.9 nm(B), 488.0 nm(C), 514.5 nm(D), and 632.8 nm(E). As to the marked peak at 245 cm^{-1}, see text.

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may be possible to assign it to the in-plane Cu-O stretching mode of the two-dimensional network.

It is known that the 482 cm^{-1} band is affected markedly by the oxygen content in both of frequency and intensity[7]. In the previous paper, we have shown that this band is not observed in the $O_{6.05}$ sample at least under 514.5 nm excitation[8]. We certified this result by several exciting frequencies in the present study. Observed spectra are reproduced partly in Fig. 2.

Next we will discuss the 151 and 657 cm^{-1} bands.

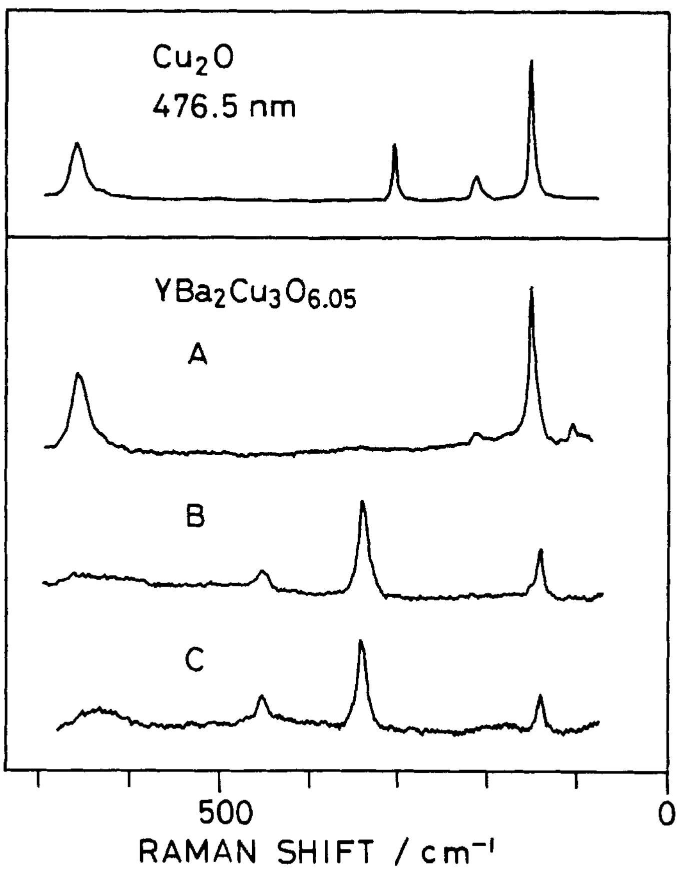

Fig 2. Raman spectra of $YBa_2Cu_3O_{6.05}$ at 30 K obtained with 476.5 nm(A), 514.5 nm(B) and 632.8 nm(C). The trace at the top is the spectrum of Cu_2O at 476.5 nm.

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These bands are observed when wavelength shorter than 488 nm is employed. Liu et al. observed this phenomenon and ascribed it to the resonance Raman scattering from a small amount of Cu_2O which was formed in the deoxygenation process, because the frequencies and the excitation profile coincide with those of Cu_2O[9]. It should be noted, however, that the 308 cm^{-1} band of Cu_2O is not observed in $YBa_2Cu_3O_6$. Although the band assignment is controversial, the 308 cm^{-1} band has been confirmed by many researchers[10]. In Fig. 2, the Raman spectrum of the Cu_2O powder at 476.5 nm excitation is shown for comparison with those of $YBa_2Cu_3O_{6.05}$. Thus it can be assumed that the 151 and 657 cm^{-1} might be intrinsic bands of $YBa_2Cu_3O_6$.

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