

Experimental determination of partial density of vibrational states of Cu and O atoms in $\text{YBa}_2\text{Cu}_3\text{O}_{7-y}$

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The partial densities of vibrational states of Cu and O atoms and their changes with the oxygen concentration in $\text{YBa}_2\text{Cu}_3\text{O}_{6.93}$ and $\text{YBa}_2\text{Cu}_3\text{O}_{6.07}$ have been determined by the method of isotopic contrast in inelastic neutron scattering.

The Cu and O atoms, which form Cu1-O4 chains and Cu2-O2, O3 planes in the 1-2-3 compounds, are known to play a governing role in the superconductivity of ceramic cuprates with high superconducting transition temperatures. It is thus worthwhile to carry out studies to obtain information about partial vibration spectra of the Cu and O atoms. A generalized density of vibrational states,¹ $\theta(E)$, has been found for $\text{YBa}_2\text{Cu}_3\text{O}_{7-y}$ from data on inelastic neutron scattering.²⁻⁴ Only theoretical data^{4,5} based on various models are available for the partial densities of the vibrations for all the atoms of $\text{YBa}_2\text{Cu}_3\text{O}_7$. The dependence of the probability for inelastic neutron scattering on the ratio σ/M (σ is the neutron scattering cross section, and M the nuclear mass) opens up the possibility in principle of experimentally determining partial vibrational spectra of the individual elements. For the isotope Cu^{65} , the factor σ/M is more than twice that for the natural mixture of Cu isotopes. The difference between the spectra found for two samples of $\text{YBa}_2\text{Cu}_3\text{O}_{7-y}$ with these isotopic compositions will thus be proportional to the partial density of vibrations of Cu atoms.⁶ In the present study we have worked from the results of such measurements to reconstruct the vibration spectra of copper in $\text{YBa}_2\text{Cu}_3\text{O}_{6.07}$ and of oxygen in $\text{YBa}_2\text{Cu}_3\text{O}_{6.93}$ and $\text{YBa}_2\text{Cu}_3\text{O}_{6.07}$.

The spectra of vibrational states were determined by inelastic neutron scattering on a time-of-flight spectrometer.⁷ For the experiments we used ceramic samples at room temperature. The experimental data, which were obtained at five scattering angles in the range 30–90°, were analyzed in the incoherent approximation, with allowance for multiphoton scattering processes.

The phase composition and structure of the samples were monitored by the x-ray structural method. The composition in terms of oxygen was varied by annealing in gaseous medium with the appropriate oxygen partial pressure. The sample was then quenched in a jet of gaseous helium.⁸

The partial vibrational spectrum of Cu atoms (Fig. 1) was found as the difference between the functions $\theta(E)$ for $\text{YBa}_2\text{Cu}_3\text{O}_{6.07}$ and $\text{YBa}_2\text{Cu}_3\text{O}_{6.93}$, normalized to the weight of the sample and to the incident neutron flux density. Shown for comparison in this figure is the vibration spectrum of Cu atoms in $\text{YBa}_2\text{Cu}_3\text{O}_{6.93}$ according to Ref. 6. The highest density of vibrational states of copper is seen to occur in the energy interval 10–30 meV, in which we see some clearly defined peaks. The upper boundary

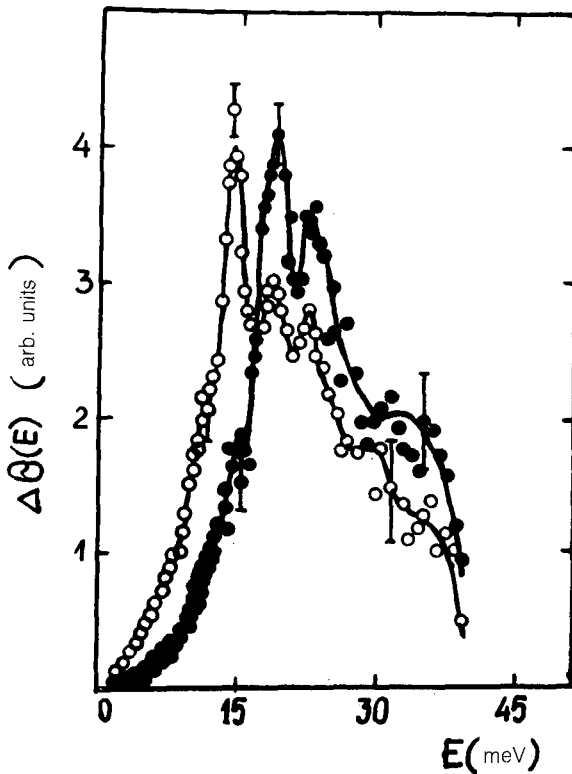


FIG. 1. Partial density of vibrations of Cu atoms. \circ — $\text{YBa}_2\text{Cu}_3\text{O}_{6.07}$; \bullet — $\text{YBa}_3\text{Cu}_3\text{O}_{6.93}$.

of the spectrum is at $E_{\text{up}} \sim 40$ meV. A reduction of the oxygen concentration softens the vibration spectrum of the Cu atoms, and an additional band appears at $E = 15$ meV. This band is not found in $\text{YBa}_2\text{Cu}_3\text{O}_{6.93}$. There is no change in the upper boundary of the energy spectrum. It might be suggested that the softening of the spectrum results from a weakening of the force interaction as a result of a change in the surroundings of the copper atoms and that the band at $E = 15$ meV corresponds to vibrations of Cu1 in a plane devoid of oxygen.

Because of the relatively large mass of the Y and Ba atoms and the small value of the factor σ/M for these elements in comparison with oxygen, we can ignore the contribution of Y and Ba vibrations to the spectrum at $E > 20$ meV (Refs. 4 and 5). Consequently, the difference between the overall vibration spectrum of $\text{YBa}_2\text{Cu}_3\text{O}_{7-y}$ and the partial vibration spectrum of Cu atoms is the partial vibration spectrum of O atoms at $E > 20$ meV (Fig. 2). The most pronounced changes in the vibration spectrum of the O atoms, which result from the removal of oxygen from O4 positions, are observed near energies of 25, 45, and 80 meV. In particular, the upper boundary of the energy spectrum of the vibrations hardens. As was shown in Ref. 9, a concentration dependence of this upper boundary is observed only in compounds with an orthorhombic lattice which exhibit a metallic conductivity. Consequently, by analogy with metals and alloys, it is natural to suggest that the hardening of the vibration spectrum

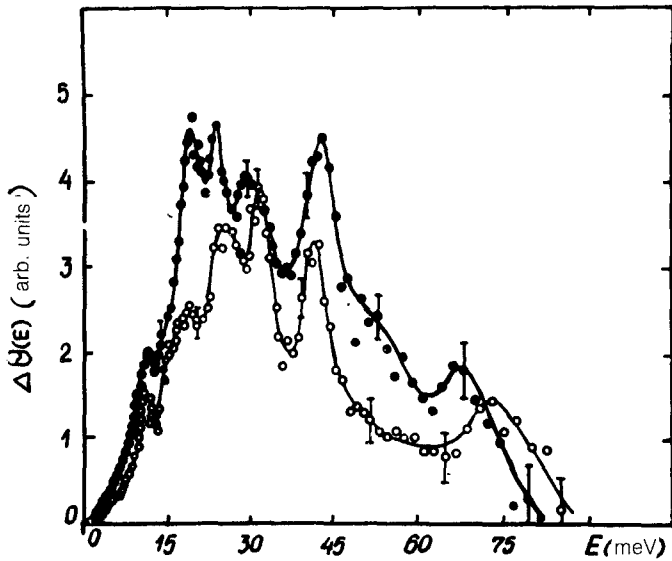


FIG. 2. Partial density of vibrations of O atoms. ○— $\text{YBa}_2\text{Cu}_3\text{O}_{6.07}$; ●— $\text{YBa}_2\text{Cu}_3\text{O}_{6.93}$.

of the O atoms is a consequence of a weakening of the screening of the ion-ion interaction by free carriers. In particular, as follows from Ref. 10, this effect could result from a lowering of the density of electron states at the Fermi level upon a decrease in the oxygen concentration. The difference in the effects of the O4 oxygen on the upper boundaries of the vibration spectra of the Cu and O atoms correlates with the conclusion¹⁰ that there are two electronic subsystems, associated with Cu and O atoms. The existence of two electronic subsystems was also suggested in Refs. 11 and 12, which dealt with various physical properties of YBCO compounds.

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