RAMAN MODES AND STRONG INTRAPLANAR OXYGEN-OXYGEN CHARGE FLUCTUATIONS IN $YBa_2Cu_3O_7$

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Crystal field variation induced ep coupling is evaluated for all Raman modes in $YBa_2Cu_3O_7$. While the symmetric modes couple to the in-plane Cu(2)-O(2,3) charge transfer (CT) the antisymmetric 340 cm^{-1} mode couples to the O(2)-O(3) CT. This explains tentatively the different behavior of those modes below T_c.

Raman experiments represent a powerful tool in the examination of the electron correlations in the conducting materials, and in particular in the high-T_c superconductors. Specially interesting in this sense are the 116 cm^{-1} and 340 cm^{-1} modes in $YBa_2Cu_3O_7$ [1-3], with similar effects in other materials. The Fano resonances of those modes indicate the appreciable coupling to the continuum. That this continuum corresponds to the carrier excitations is indicated by the anomalous behavior of the 340 cm^{-1} frequency at T_c. While all Raman frequencies increase steadily towards low temperatures, the frequency of the 340 cm^{-1} mode starts to decrease below T_c. It is important to note in this respect that there is a symmetry difference between the 340 cm^{-1} mode which belongs to the antisymmetric B_{1g} representation of the (approximate) tetragonal group, while all other modes are A_{1g} symmetric. This difference leads to an essential difference in coupling to the carriers, as will be discussed below.

All Raman modes under consideration involve the displacements perpendicular to the conducting CuO_2 planes. Therefore the in-plane overlaps are not changed to the first order in deformation (disregarding the weak buckling of the plane[4]). However it has been realized some time ago that another, ionic mechanism of ep coupling is operative in high-T_c superconductors. The strongest effect of the deformation induced crystal field variation occurs in site energies [5,6]. Singling out the sites O(2), O(3) and Cu(2) in the conducting plane the corresponding ep coupling can be written as

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$$\delta\varepsilon_p^{(2)} n_p^{(2)} + \delta\varepsilon_p^{(3)} n_p^{(3)} + \delta\varepsilon_d^{(2)} n_d^{(2)} = \frac{1}{2}(\delta\varepsilon_p^{(2)} - \delta\varepsilon_p^{(3)})(n_p^{(2)} - n_p^{(3)})$$
$$+ \frac{1}{2}\left(\frac{\delta\varepsilon_p^{(2)} + \delta\varepsilon_p^{(3)}}{2} - \delta\varepsilon_d^{(2)}\right)(n_p^{(2)} + n_p^{(3)} - n_d^{(2)}) \qquad (1)$$
$$+ \frac{1}{2}\left(\frac{\delta\varepsilon_p^{(2)} + \delta\varepsilon_p^{(3)}}{2} + \delta\varepsilon_d^{(2)}\right)(n_p^{(2)} + n_p^{(3)} + n_d^{(2)}).$$

R. h. s. of Eq.(1) is written to emphasize the difference between A_{1g} and B_{1g} modes. While the latter (Jahn-Teller) splits the energy of the two in plane oxygens (neglecting the small orthorhombic splitting $\varepsilon_p^{(3)} - \varepsilon_p^{(2)}$) the former modes only change the already existing energy difference $\varepsilon_d^{(2)} - \varepsilon_p^{(2,3)}$.

The energy shifts

$$\delta\varepsilon_{i\nu} = \frac{e^2}{d_{cuo}} \cdot \alpha_{i\nu} u_\nu \qquad (2)$$

can be evaluated by the Ewald method for various modes ν and sites i. The corresponding α's are given in Table I, assuming $Y^{3+}Ba^{2+}Cu^{2+}(1)Cu^{2+}(2)O^{2-}(1)O^{1.75-}(2,3)O^{2-}(4)$ ionicities.

A_{1g} B_{1g}	$\omega(cm^{-1})$[1]	$\alpha_{O1\nu}$	$\alpha_{O2\nu}$	$\alpha_{O3\nu}$	$\alpha_{Cu2\nu}$
$O(2)^{\uparrow}O(3)_{\downarrow}$	340	0.01	0.81	-0.88	0.01
$Ba^{\uparrow}$	116	-1.41	2.23	2.25	1.94
$Cu(2)^{\uparrow}$	150	-2.21	2.10	2.07	2.35
$O(2)^{\uparrow}O(3)^{\uparrow}$	440	2.65	-1.81	-1.75	-1.42
$O(1)^{\uparrow}$	504	1.21	-1.97	-1.96	-2.39

TABLE I

In Table I it can be first noticed that motions of ions in the CuO_2 plane lead to large linear ep couplings. This is related to the absence of the mirror symmetry with respect to the CuO_2 plane (i.e. the corresponding α's vanish in La_2CuO_4). The effect is large due to the long range of ionic forces. Second, the motion of the out-of-plane ions is also coupled strongly to carriers for the same reason.

The ep couplings (2) lead to the renormalization of the phonon frequencies. In the adiabatic limit the second order deformation energy, corresponding to Eq.(1) for the B_{1g} and A_{1g} modes is respectively

$$\Delta E_B = -\frac{1}{8}\chi_{pp}(\delta\varepsilon_{pB}^{(2)} - \delta\varepsilon_{pB}^{(3)})^2, < 0 ,$$
$$\Delta E_A = -\frac{1}{32}\chi_{pd}(\delta\varepsilon_{pA}^{(2)} + \delta\varepsilon_{pA}^{(3)} - 2\delta\varepsilon_{dA}^{(2)})^2, < 0 , \qquad (3)$$

assuming that the total charge of the CuO_2 unit cell in Eq.(1) is conserved. Here χ_{pp} is the exact q = 0 O(2)-O(3) and χ_{pd} the O(2,3)-Cu(2) charge transfer (CT) correlation function. As the ep couplings are comparable in both cases, according to Table 1, the difference between B_{1g} and A_{1g} modes is related to the difference between χ_{pp} and χ_{pd} (it can be noted in this respect that the coupling between O(2,3) and the bridging O(1) is much weaker for the B_{1g} mode then for the A_{1g} modes). The essential difference between χ_{pp} and χ_{pd} is that unlike χ_{pd}, χ_{pp} corresponds to lifting the degeneracy of some high symmetry points in the Brillouin zone of the carriers (assuming the Fermi level either in the copper-oxygen or in the oxygen-copper bands). The site JT effect in Eq.(1) transforms into the intra- or inter-band JT effect in presence of hopings and interactions. The interband processes may also be important in presence of interactions, but tend to have [6] the same contribution to χ_{pp} and χ_{pd}.

It can be finally mentioned that Table I shows a particularly strong coupling between the 504 cm^{-1} Raman motion of the bridging O(1) and the CuO_2 plane. It is possible that in this case the dominant CT occurs along the c-axis rather than in the CuO_2 plane [7]. Eqs.(2) and (3) can be easily extended to cover this possibility too. On the other hand the Fano resonance observed in the Ba mode suggests that it is coupled to the ab plane. However the CT may well occur also along the c-axis.

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