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Two distinct electronic contributions in the fully symmetric Raman response of high-T_c cuprates

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We show by resonance effects in $HgBa_2CuO_{4+\delta}$ (Hg-1201) and by Zn substitutions in $YBa_2Cu_3O_{7-\delta}$ (Y-123) compounds that the fully symmetric Raman spectrum has two distinct electronic contributions. The A_{1g} response consists of the superconducting pair-breaking peak at the 2Δ energy and a collective mode close to the magnetic-resonance energy. These experimental results reconcile the *d*-wave model to the A_{1g} Raman response function insofar as a collective mode that is distinct from the pair-breaking peak is present in the A_{1g} channel.

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In the last few years, it has been well established that the superconducting gap of the hole-doped cuprates at the optimal doping regime has the $d_{x^2-y^2}$ symmetry.[1–3]

This symmetry manifests itself in the low-energy part of the Raman spectra. In the B_{2g} channel[4] (probing the nodal directions), the electronic continuum behaves as a linear function of the Raman shift, while it follows a cubic law in the B_{1g} channel[4] (antinodal directions) (see Ref. 5). In the latter one, a well-defined pair-breaking peak near $2\Delta = 8k_BT_c$ is observed. However, existing theories based on the $d_{x^2-y^2}$ model fail to reproduce the position, the intensity, and the shape of the broad electronic peak observed in the fully symmetric A_{1g} channel.[3,4,6,7] Expansion of the Raman vertex to the second order of the Fermi surface harmonics[5] and resonant effects[8] have been proposed to reproduce the relative A_{1g} peak position and intensity with respect to that of B_{1g}. In these pictures, the A_{1g} peak is treated as another manifestation of the pair-breaking peak observed in the B_{1g} channel. Unfortunately, the backflow prevents the reproduction of the location, on one hand, and on the other hand, of the sharpness and the strong intensity of the A_{1g} peak. For a generic tight-binding model, the calculated screened A_{1g} channel is only a tiny fraction of the B_{1g} response.[6] This is in clear contradiction to all experiments and most studies showing the magnitude of the A_{1g} peak being even larger than the B_{1g} (Refs. 9 and 11–13). In this paper we show that the A_{1g} response has two components: one component originating from the pair breaking close to the 2Δ energy and the other from a collective mode which tracks the magnetic resonance.[9,10] In this sense, our experimental results reconcile the A_{1g} Raman response of the cuprates at the optimal doping regime with the *d*-wave model insofar as a collective mode is present in the A_{1g} channel.

Electronic Raman scattering (ERS) measurements have been carried out with a JY T64000 triple spectrometer in subtractive configuration using different lines of mixed argon-krypton laser gas. The Raman spectra were corrected for the spectrometer response, the Bose factor and the optical constants producing the imaginary part $\chi''(\omega)$ of the Raman response. The crystals were mounted in vacuum (10^{-6} mbar) on the cold finger of a liquid-helium flow cryostat. The power density was about 10 W/cm^2 on the sample surface, and the laser spot heating estimated from the anti-Stokes/Stokes intensity ratio of the Raman responses was less than 3 K.

Let us focus first on ERS measurements of optimally doped Hg1201 single crystals ($T_c = 95$ K). They have been grown by flux method whose detailed procedure is described elsewhere.[14] Figure 1 shows the superconducting Raman responses $\chi''_S(\omega)$ of Hg-1201 obtained for various excitations lines in A_{1g} and B_{1g} channels.

The Raman responses are composed of a broad electronic continuum surrounded by an assembly of narrow peaks corresponding to the well-identified phonons.[15] At first glance, the Raman responses for each excitation line (E.L.) reveal that the A_{1g} continuum exhibits a strong maximum around 330 cm^{-1}, with an asymmetric part in its high-energy side. This manifests itself as a bump for blue (488 nm) and green (514 nm) E.L., and as a "plateau" for yellow (568 nm) and red (647 nm) ones, which are around 520 cm^{-1} near the maximum of the B_{1g} continuum that corresponds to the pair-breaking peak.

The Raman responses of the blue and green lines show strong phonon features superimposed to the electronic continuum near 520 cm^{-1}, which complicates the extraction of the electronic background. On the contrary, under the yellow and red E.L., the phonon modes are out of resonance; thus their structures are strongly reduced and the electronic contribution can be easily extracted. Subtractions of the normal $\chi''_N(\omega)$ response from the superconducting $\chi''_S(\omega)$ one are reported in the insets of Fig. 1. The Raman responses $\chi''_S(\omega)$-$\chi''_N(\omega)$ for the yellow and red lines are almost free of phonon contribution. The broad continua in the A_{1g} and B_{1g} channels correspond to the electronic contributions from the superconducting state, and the sharp features show misfits between the superconducting and normal phonon structures. After substraction of the normal-state contribution, the A_{1g} response is still asymmetric, and for each E.L., the high-energy part of this response is centered near the maximum of the B_{1g} superconducting gap. The asymmetry of the A_{1g} response is thus intrinsic to the superconducting state.

To go further and prove that the broad A_{1g} peak consists effectively of two distinct electronic components, we have performed ERS measurements on high quality, optimally

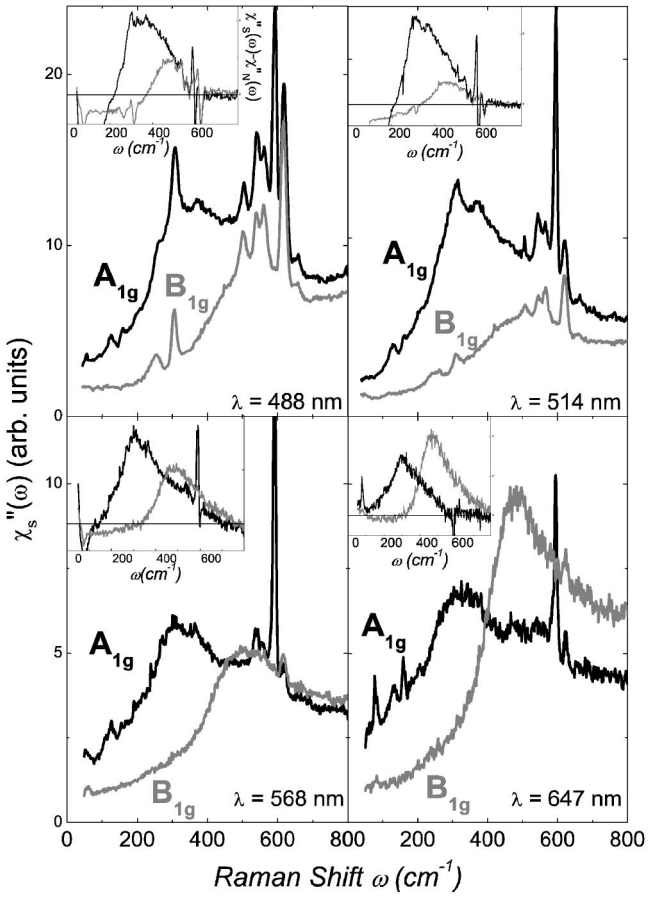

FIG. 1. Raman responses $\chi''_S(\omega)$ of optimally doped Hg-1201 for different excitations lines in the A_{1g} (black line) and B_{1g} (gray line) channels. The insets exhibit $\chi''_S(\omega)-\chi''_N(\omega)$ for both A_{1g} and B_{1g} channels.

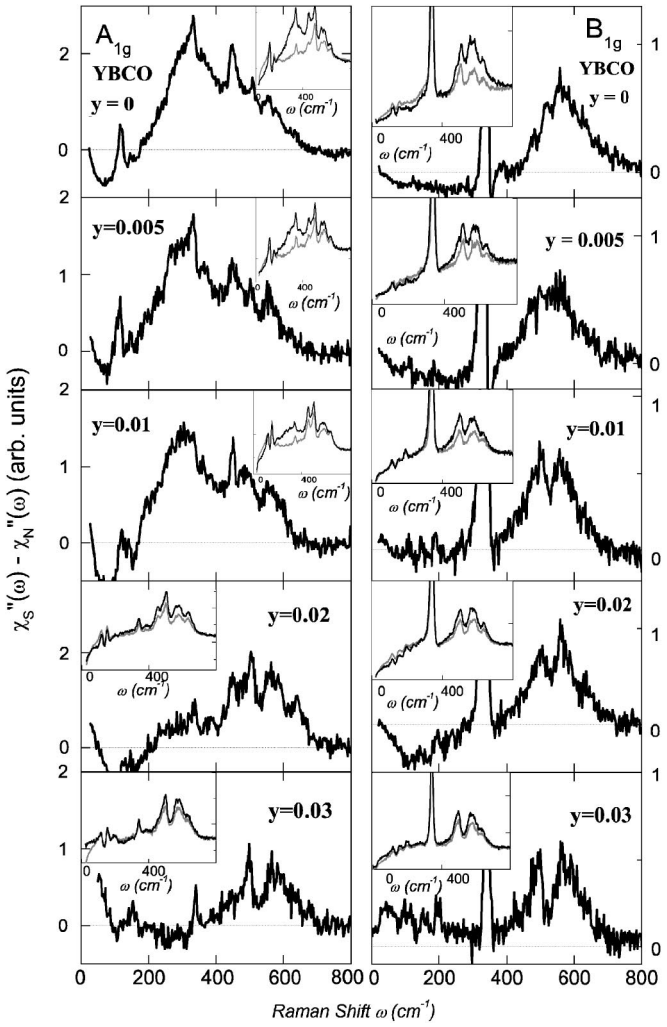

FIG. 2. $\chi''_S(\omega)$-$\chi''_N(\omega)$ A_{1g} (left panel) and B_{1g} (right panel) Raman responses of optimally doped $YBa_2(Cu_{1-y}Zn_y)_3O_{7-\delta}$ for various Zn concentrations y. $\lambda=514$ nm. In each inset are plotted the Raman response functions in the normal (gray line) and superconducting (black line) states.

doped, YBCO single crystals grown by the self-flux method,[16] where copper is substituted by zinc. Zn is a divalent ion known to substitute preferentially in the CuO_2 layers without altering the carrier concentration.[17] In addition to the pure $YBa_2Cu_3O_{7-\delta}$ (Y-123, $T_c=92$ K), we have studied $YBa_2(Cu_{1-y}Zn_y)_3O_{7-\delta}$ single crystals with $y=0.005$ ($T_c=87$ K), $y=0.01$ ($T_c=83$ K), $y=0.02$ ($T_c=73$ K), and $y=0.03$ ($T_c=64$ K). Zn concentration was verified by chemical analysis using an electron probe. T_c measurements were obtained from dc magnetization and we found $dT_c/dy\sim -10$ K/%.

Figure 2 shows the $\chi''_S(\omega)-\chi''_N(\omega)$ Raman responses in A_{1g} and B_{1g} channels in Y-123 for various Zn contents. The insets exhibit the A_{1g} and B_{1g} Raman responses in the normal and superconducting states before subtraction. The A_{1g} and B_{1g} Raman responses show a set of sharp phonon peaks lying on a strong electronic background. In the A_{1g} channel, for pure YBCO, the $\chi''_S(\omega)-\chi''_N(\omega)$ Raman response shows a broad and strong asymmetric peak which spreads out in the high-energy side and reaches its maximum close to 330 cm^{-1}. In the B_{1g} channel, the $\chi''_S(\omega)-\chi''_N(\omega)$ response for pure YBCO, exhibits a well defined and nearly symmetric peak close to 530 cm^{-1}. These A_{1g} and B_{1g} superconducting spectra are very similar to those obtained from Hg-1201 at the optimal doping. Here again, the A_{1g} response exhibits a maximum around 330 cm^{-1} with an asymmetric part which extends up to the pair-breaking peak near 530 cm^{-1}. The positions of the A_{1g} and B_{1g} peaks are nearly the same for both Y-123 and Hg-1201. The changes in the band structure induced by two CuO_2 layers instead of one CuO_2 layer do not affect the A_{1g} and B_{1g} peak positions; rather, the critical temperature (92 K for Y-123 and 95 K for Hg-1201) seems to govern the A_{1g} and B_{1g} peak energies at the optimal doping. This is observed for many cuprates where the B_{1g} peak is found close to $8k_BT_c$ and the A_{1g} peak maximum close to $5k_BT_c$ at the optimal doping (see Table I of Refs. 9 and 10). Adding some Zn in the pure Y-123, one can see that the intensity of the pair-breaking peak seen in the B_{1g} channel decreases, but the peak does not disappear and is still present even in the sample with $y=0.03$ ($T_c=63$ K), contrary to what is suggested in Ref. 18. The B_{1g} peak does not shift in energy, and thus does not follow T_c, but this effect and other related to nonmagnetic impurity substitutions in YBCO will be discuss in a next paper.

Let us focus now on the A_{1g} channel. As Zn content increases, the low-energy contribution in the A_{1g} response becomes broader, shifts to a lower energy (from 330 to 300 cm^{-1}), strongly decreases in its intensity, and finally disappears. On the contrary, the high-energy contribution in the A_{1g} response moderately decreases in its intensity but keeps the same position at 530 cm^{-1}.

For $y=0.01$, the $\chi''_S(\omega)$-$\chi''_N(\omega)$ A_{1g} response, clearly shows two components. The first is centered at 300 cm^{-1} and the second is close to 530 cm^{-1}. For higher Zn concentrations, the intensity ratio of the upper- and the lower-energy parts of the A_{1g} response is reversed in such a way that for $y=0.03$, the lower-energy component completely disappears while the upper component persists. A straightforward comparison between the left and right panels reveals that the high-energy component (530 cm^{-1}) in the A_{1g} response tracks the B_{1g} peak. For both $y=0.02$ and $y=0.03$, when the lower-energy component in the A_{1g} response is weak and no longer mixes with the higher-energy component, the ratio of the spectral weight between the higher-energy component in the A_{1g} response and the pair-breaking peak in the B_{1g} response remains constant. In these cases the peaks observed in A_{1g} and B_{1g} channels are located at the same energy[19] and correspond both to the pair-breaking peak.

This gives experimental evidence that the A_{1g} response has two distinct components and that the one of higher energy corresponds to the pair-breaking peak. As the B_{1g} one probes the antinodal directions of the $d_{x^2-y^2}$ superconducting gap, and the A_{1g} Raman response has no symmetry restriction, it is therefore not surprising to observe the pair-breaking peak in both A_{1g} and B_{1g} channels. The low-energy component of the A_{1g} Raman response corresponding to the maximum of the electronic continuum is intrinsic to the superconducting state and disappears above T_c as it was already pointed out in previous works.[9,10] The A_{1g} peak is located at $5k_BT_c$ well below the 2Δ energy gap ($8k_BT_c$) and therefore cannot be induced by individual electronic excitations which required energies beyond 2Δ. As a consequence, the A_{1g} mode has to be a bound state of quasiparticle pairs at an energy less than 2Δ and refers to a collective mode. We have not yet identified the origin of the A_{1g} mode but several scenarios can be figured out. Among them, the Bogoliubov-Anderson collective mode[20] calculated in the framework of the *d*-wave model merits to be considered as well as a double magnon with a zero-spin-flip energy as suggested by Demler.[21] Zero-spin-flip energy is possible for a *d*-wave superconductor if we consider spin-flip excitations over two nodal regions.

Zero-spin-flip quasiparticle excitations have already been invoked to explain the quadratic increase of the[17] O spin-lattice relaxation rate under magnetic fields across the vortex lattice NMR spectrum in YBCO.[22] In our case, the A_{1g} peak tracks the acoustic magnetic resonance detected by inelastic neutron scattering[23] at $\mathbf{Q}=(\pi,\pi)$ as previously shown.[9,10] A double spin flip of transfered momenta $\mathbf{Q}=(\pi,\pi)$ and $\mathbf{Q}=(-\pi,-\pi)$ is then needed for preserving the total transfer momentum close to zero in the Raman-scattering process. The first spin flip is over two antinodal regions and costs the magnetic-resonance energy, whereas the second spin flip over two nodal regions costs zero energy. In this Raman process the A_{1g} mode takes the same energy as the magnetic resonance as expected experimentally. Theoretical investigations of this last scenario are in progress.

In summary, the ERS spectra of Hg-1201, free of phonon peaks, reveal that the A_{1g} response and its asymmetry near the pair-breaking peak are of electronic origin. Moreover, ERS in Y-123 substituted with Zn shows that the A_{1g} peak has two distinct components: one at the higher energy corresponding to the pair-breaking peak observed in the B_{1g} channel, and the other at lower energy corresponding to another electronic contribution that is distinct from the pair-breaking peak. This study reconciles the A_{1g} Raman response function with the *d*-wave model, where the pair-breaking peak manifests itself in both B_{1g} and A_{1g} channels. This implies the existence of a charge collective mode below the pair-breaking peak energy which we have previously related to the magnetic resonance.

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