Evidence for Two Distinct Energy Scales in the Raman Spectra of YBa$_2$(Cu$_{1-x}$Ni$_x$)$_3$O$_{6.95}$

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We report electronic Raman scattering from Ni-substituted YBa$_2$Cu$_3$O$_{6.95}$ single crystals with $T_c$ ranging from 92.5 to 78 K. The fully symmetrical $A_{1g}$ channel and the $B_{1g}$ channel which is sensitive to the $d_{x^2-y^2}$ gap maximum have been explored. The energy of the $B_{1g}$ pair-breaking peak remains constant under Ni doping while the energy of the $A_{1g}$ peak scales with $T_c$ ($E_{A_{1g}}/k_BT_c = 5$). Our data show that the $A_{1g}$ peak tracks the magnetic resonance peak observed in inelastic neutron scattering yielding a key explanation to the long-standing problem of the origin of the $A_{1g}$ peak.

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The pairing mechanism that leads to high temperature superconductivity in the cuprates is still a subject of intense controversy. Electronic Raman scattering (ERS) has been revealed as a very powerful tool for probing selectively the electronic excitations in different regions of the Fermi surface of the cuprates by orienting the incoming and outgoing light polarizations [1]. In particular, ERS results in the superconducting state of nearly stoichiometric cuprates going light polarizations [1].

Recent experimental findings suggest that the $A_{1g}$ peak is controlled by a collective spin fluctuation mode which is identified with the $Q = (\pm \pi, \pm \pi)$ resonance observed by inelastic neutron scattering (INS) in the superconducting state [19]. However, no experimental proof has been provided until now.

In this Letter, we report ERS experiments on Ni-substituted YBa$_2$Cu$_3$O$_{6.95}$ (YBCO) systems, where Ni impurity is used as a probe for testing the physical origin of the $A_{1g}$ peak with respect to the $B_{1g}$ pair-breaking peak. Our most striking result is that the $A_{1g}$ peak, contrary to the $B_{1g}$ one, scales with $T_c$ with the same energy scale as the magnetic resonance peak detected by INS [7,20–22]. A thorough examination shows that the temperature dependence of the $A_{1g}$ peak follows the same behavior as the INS resonance peak in contrast to the $B_{1g}$ peak. The question of a possible link between the $A_{1g}$ peak and the INS peak will be addressed. In particular, the energy, the symmetry, and the nonscreening of the $A_{1g}$ peak are consistent with a spin fluctuation origin for this peak.

ERS measurements were carried out on optimally doped Ni-substituted YBCO single crystals. They were grown in a gold crucible using a flux technique [23]. The Ni diluent is believed to substitute preferentially for divalent copper in the CuO$_2$ plane of YBCO [24]. Therefore Ni substitution offers a particularly attractive way to reduce $T_c$ while preserving the carrier concentration [25].

As a consequence, Ni substitution in YBCO allows us to investigate the $A_{1g}$ and the $B_{1g}$ peaks as a function of $T_c$ near the optimal doping, where the $A_{1g}$ and the $B_{1g}$ peaks are experimentally the most easily observable. The $T_c$ values were defined from $x$ contents of $x = 0$, $x = 0.01$, and $x = 0.03$ with $T_c = 92.5$, 88, and 78 K, respectively.

Raman measurements were performed with a double monochromator using a single channel detection and the...
TABLE I. An overview of the peak locations of the ERS $A_{1g}$ ($E_{A_{1g}}$) and $B_{1g}$ ($E_{B_{1g}}$) and the INS resonance ($E_R$).

<table>
<thead>
<tr>
<th>Cuprates</th>
<th>Y-123</th>
<th>Bi-2212</th>
<th>La-214</th>
<th>TI-2201</th>
<th>Hg-1223</th>
</tr>
</thead>
<tbody>
<tr>
<td>$T_c$ (K)</td>
<td>92.5 ± 0.5$^a$</td>
<td>89 ± 1$^c$</td>
<td>37 ± 1$^e$</td>
<td>90 ± 1$^f$</td>
<td>130 ± 1$^h$</td>
</tr>
<tr>
<td>$E_{B_{1g}}$ (meV)</td>
<td>67 ± 0.5$^a$</td>
<td>62 ± 2$^c$</td>
<td>25 ± 2$^e$</td>
<td>58 ± 2$^f$</td>
<td>100 ± 2$^h$</td>
</tr>
<tr>
<td>$E_{A_{1g}}$ (meV)</td>
<td>40 ± 0.5$^a$</td>
<td>43 ± 2$^c$</td>
<td>16 ± 2$^e$</td>
<td>44 ± 2$^f$</td>
<td>67 ± 2$^h$</td>
</tr>
<tr>
<td>$E_{B_{1g}}/k_BT_c$</td>
<td>8.4 ± 0.1</td>
<td>8.1 ± 0.4</td>
<td>7.8 ± 0.8</td>
<td>7.5 ± 0.3</td>
<td>8.9 ± 0.2</td>
</tr>
<tr>
<td>$E_{A_{1g}}/k_BT_c$</td>
<td>5.0 ± 0.1</td>
<td>5.6 ± 0.3</td>
<td>5.0 ± 0.8</td>
<td>5.7 ± 0.3</td>
<td>6.0 ± 0.2</td>
</tr>
<tr>
<td>$E_R$ (meV)</td>
<td>40 ± 0.5$^b$</td>
<td>43 ± 0.5$^d$</td>
<td>8$^i$</td>
<td>47 ± 1$^g$</td>
<td></td>
</tr>
<tr>
<td>$E_R/k_BT_c$</td>
<td>5.0 ± 0.1</td>
<td>5.4 ± 0.1</td>
<td>5.8 ± 0.2</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

$^a$This study. $^b$Ref. [7]. $^c$Ref. [8]. $^d$Ref. [9]. $^e$Ref. [6]. $^f$Ref. [10]. $^g$Ref. [11] ($T_c = 91$ K). $^h$Ref. [3]. $^i$The $^*$ indicates that the neutron magnetic resonance peak has never been reported in La-214. However, the temperature dependence of the neutron magnetic intensity observed for $x = 0.14$ at the incommensurate wave vector around 9–16 meV [12] exhibits a certain similarity with that of the neutron resonance observed in the other high-$T_c$ cuprates.

Ar$^+$ and Kr$^+$ laser lines. The imaginary parts of the Raman susceptibilities $\chi''(\omega)$ are deduced from the raw spectra using the same procedure as in Ref. [3].

The difference between the Raman responses obtained from $T = 130$ K (normal state) and $T = 35$ K (superconducting state) in the $B_{1g}$ and the $A_{1g}$ channels as a function of Ni content are displayed in Fig. 1. The $\Delta \chi'' = \chi''_S - \chi''_N$ spectra show for both $B_{1g}$ and $A_{1g}$ channels a set of sharp phonon peaks (typical width at half maximum: $\sim 2$ meV) lying on a strong electronic background. In the $B_{1g}$ channel [Fig. 1(a)], for $x = 0$, we observe in the $\Delta \chi''$ spectrum a strong and well-defined electronic peak at 67 meV which is not present in the normal state. On the other hand, the $\Delta \chi''$ spectrum exhibits a negative intensity part below 40 meV which corresponds to a decrease of the electronic spectral weight in the superconducting state as compared to the normal one. These two observations are the signature of the opening of the superconducting gap along the $k_x = 0$ and $k_y = 0$ directions. The intensity of the $B_{1g}$ peak [defined as the value of $\Delta \chi''(\omega)$ at the maximum of the peak] decreases with Ni content by a factor of 2 for $x = 0.01$ and 3 for $x = 0.03$. A striking feature is that the $B_{1g}$ peak does not shift in energy with increasing Ni content. Even though the $B_{1g}$ peak becomes broader (width at half maximum increases by a factor of 1.5), its position remains centered at 67 meV which suggests that

![FIG. 1. Raman response functions difference between the superconducting and the normal states of YBa$_2$(Cu$_{1-x}$Ni$_x$)$_3$O$_{6.95}$ for $x = 0$, 0.01, and 0.03 in the $B_{1g}$ channel (a) and the $A_{1g}$ channel (b). The 514.52 nm laser line was used. The peaks were fit to a Gaussian shape in their upper parts (bold lines) to define their positions (the mathematical continuation is shown as a dotted line).](177401-2)
the gap maximum is unaffected despite the introduction of $x = 0.03$ Ni impurities and the $T_c$ decrease of 14 K.

In the $A_{1g}$ channel [Fig. 1(b)], we clearly detect in the $\Delta \chi''$ spectra for $x = 0$ a broad and intense peak in the continuum centered around 40 meV. The $A_{1g}$ peak, similar to the $B_{1g}$ one, appears only in the superconducting state. However, it is much more robust than the $B_{1g}$ peak and is still well defined even for the highest nickel concentration ($x = 0.03$) with an intensity decrease less than a factor of 1.6. In contrast with the $B_{1g}$ peak which does not change its position, the $A_{1g}$ peak shifts significantly to lower energy with increasing Ni content. It softens from 40 meV at $x = 0$ down to 38 and 33 meV for $x = 0.01$ and $x = 0.03$, respectively.

Figure 2 displays the ERS $A_{1g}$ and $B_{1g}$ peak energies as a function of $T_c$. Our data clearly show that, while the $B_{1g}$ peak energy remains constant upon Ni substitution [i.e., the magnitude of the gap at $(\pi, 0)$ does not scale with $T_c$], the $A_{1g}$ peak energy scales with $T_c$. In fact, if we compare our results with INS data obtained from Ni-substituted optimally doped YBCO [21], we find quantitative correspondence between the $A_{1g}$ peak and the INS magnetic resonance. The ERS $A_{1g}$ peak and the INS resonance energies both scale with $T_c$ and can be plotted along the same line corresponding to $E/k_B T_c = 5$. This similarity does not hold only for the Y-123 system, since a quite good correspondence between the energies of the $A_{1g}$ peak and the INS resonance is also achieved for the Bi-2212 and Tl-2201 systems as shown in Table I.

To go further, we have investigated the temperature dependence of the ERS $A_{1g}$ and $B_{1g}$ peaks with respect to their intensity and their energy location in the pure optimally doped YBCO system ($x = 0$) and performed a comparative analysis with the INS resonance. Figure 3(a) shows that the ERS $B_{1g}$ peak intensity is linear as a function of temperature, whereas the ERS $A_{1g}$ peak and INS resonance [22] intensities decrease in the form of a step function and vanish at $T_c$. In addition, the energy positions of the $A_{1g}$ peak and the INS resonance remain constant with the same value under the temperature variation as shown in Fig. 3(b).

In Fig. 4, the $A_{1g}$ responses as a function of various excitation lines are displayed. It appears that the $A_{1g}$ peak position is not sensitive to the laser line excitations and therefore cannot be attributed to a resonance effect of the $A_{1g}$ Raman vertex.

In light of these new results, we discuss the origin of the $A_{1g}$ peak. On one hand, our data have clearly shown that the ERS $A_{1g}$ and $B_{1g}$ peaks exhibit drastically different behaviors as a function of $T_c$ (for Ni-substituted YBCO systems) but also as a function of temperature (for pure YBCO system). On the other hand, we have clearly
established quantitative correspondences between the ERS $A_{1g}$ peak and the INS magnetic resonance peak. These experimental results suggest that, in contrast to the attribution of the $B_{1g}$ peak as arising from the pair-breaking process in the charge channel, the $A_{1g}$ peak could be coming from a different origin. The idea of a magnetic origin for the $A_{1g}$ peak associated with the antiferromagnetic (AF) spin fluctuations is very appealing for three main reasons: first, because we have introduced the experimental evidence that the $A_{1g}$ peak and the INS magnetic resonance are deeply related; second, the Coulomb screening is expected to be inefficient because AF fluctuations are not coupled with the electron density. This explains why the $A_{1g}$ peak detected in the Raman spectra is unscreened. Finally, the resonance peak has the fourfold symmetry in the $k$ space which corresponds to a fully symmetrical $A_{1g}$ configuration. It follows that the magnetic resonance contribution to the Raman response will be relevant in the $A_{1g}$ channel only [19].

In conclusion, through Ni substitution we have clearly established the same energy scale $5k_BT_c$ for the INS resonance and the ERS $A_{1g}$ peak, and we have shown that the ERS $A_{1g}$ and INS resonance peaks follow the same temperature dependence. This $A_{1g}$ energy scale is different from the one of the pair-breaking process related to the $B_{1g}$ peak. The existence of an additional energy scale in the superconducting state, observed both in ERS and INS experiments, can provide the key towards an understanding of the pairing mechanism in high-$T_c$ superconducting cuprates.

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