Overdoping effect on pair breaking peak energy in the electronic Raman spectra of high-\(T_c\) cuprate superconductors

Takahiko Masui\(^1\), Kazutaka Nishikawa\(^1\), Takayuki Hiramachi\(^1\), Setsuko Tajima\(^1\), Hiroshi Eisisaki\(^2\), Hijiri Kito\(^2\) and Akira Iyo\(^2\)

\(^1\) Department of Physics, Graduate School of Science, Osaka University, Osaka 560-0043, Japan
\(^2\) Nanoelectronics Research Institute, National Institute of Advanced Industrial Science and Technology (AIST), Tsukuba 305-8568, Japan

E-mail: tmasui@phys.sci.osaka-u.ac.jp

Abstract. In the superconducting state of cuprates we have measured electronic Raman spectra (ERS) and plotted the pair breaking energy as a function of hole concentration in overdoped regime. The target materials in this study are Ca-substituted YBCO and Tl2201, both of which have similar \(T_c\) when it is optimally doped but different number of CuO\(_2\) planes in the unit cell. For both materials pair breaking peak energy (\(E_b\)) shows decrease with increasing carrier concentration. Interestingly \(E_b\) seems to show a rapid drop at a certain carrier concentration, where several measurements suggest the signature of quantum critical point. The similarity of the ERS between the two materials indicates that the behavior is not affected by the number of CuO\(_2\) planes in a unit cell, and that electronic state of cuprates may have a remarkable change at the critical hole concentration.

1. Introduction

One of the advantages of electronic Raman scattering study is to detect \(k\)-space information selectively. In high-\(T_c\) cuprate superconductors (HTSC), electronic Raman spectra (ERS) shows response of superconductivity, as a pair-breaking peak at around 2\(\Delta\) and the decrease of electronic scattering below 2\(\Delta\). In overdoped state of HTSC, \(T_c\) decreases with increasing hole concentration. Corresponding to this change, smooth decrease of 2\(\Delta\) is expected with increasing hole doping. However, the decrease of pair breaking peak energy in Tl\(_2\)Ba\(_2\)CuO\(_{6+\delta}\) (Tl2201) is much stronger than that expected from the decrease of \(T_c\) [1]. It is of great interest whether the behavior is common among HTSC or not. In this study we measure pair-breaking peaks of Tl2201 and Ca-substituted YBa\(_2\)Cu\(_3\)O\(_{7-\delta}\) (YCa123) single crystals, and discuss the similarity and the difference between the materials. These materials shows 90 K and 93 K if it is optimally doped, so it is suitable to compare the doping dependence between them.

2. Experimental

Tl2201 single crystals were grown by a flux method[2]. Since the oxygen contents of as-grown crystals were in most cases rather high, they were decreased by the annealing process in Ar atmosphere. The \(T_c\)s of all crystals were characterized by a SQUID magnetometer. Since
higher oxygen content gives lower $T_c$, all crystals are supposed to be overdoped. The highest $T_c$ achieved by annealing procedure is 90 K, which is almost the same value as previous reports. YCa123 single crystals were grown by a crystal pulling method. The impurity contents were estimated from inductively coupled plasma analysis. The samples were cut into pieces with rectangular shape and annealed in flowing oxygen. Some pieces of Ca substituted crystals were detwinned under uniaxial pressure. The oxygen deficiency $\delta$ for YCa123 were estimated according to reference [3]. The carrier concentration was estimated from the empirical relation $T_c/T_c\text{(max)} = 1 - 82.6(p - 0.16)^2$ [4].

We measured Raman scattering spectra by using T64000 Jobin Yvon triple spectrometer with a liquid nitrogen cooled CCD detector and Ar/Kr laser. The wavelength of excited beam is 514.5 nm. A closed cycle cryostat was used with temperature stabilization better than 1K. The crystal structure of Tl2201 belongs to the tetragonal $D_{4h}$ point group. We extracted $B_{1g}(XY')$, $B_{2g}(XY)$, and the $A_{1g}(X'X'Y')$ scattering component from various polarization spectra. The $X'$ and $Y'$ axes are rotated by 45 degree with respect to the $X$ and $Y$ axes, respectively, which are parallel to the CuO bonds in the unit cell. The crystal structure of YCa123 is orthorhombic, but we approximately treat the structure as tetragonal, as have been done in many previous reports.

In the $B_{1g}$ spectra of YCa123, there appears a large phonon peak at around 340 cm$^{-1}$, which strongly interacts with electronic Raman component. It sometimes prevents us to distinguish pair-breaking peak. To overcome the point, we extracted the ERS, according to a Green function method [5]. On the other hand, no such phonon peak appears for the case of Tl2201. Accordingly the spectra with phonons are enough to distinguish pair-breaking peaks. The difference comes from the crystal structures; Tl2201 possess one CuO$_2$ plane in a unit cell, while YBCO possesses two CuO$_2$ planes in a unit cell.

3. Results and discussion

Figure 1 shows $B_{1g}$ spectra of Tl2201 single crystals at 10 K. A broad bump appears in Raman spectra below $T_c$, which is assigned as cooper pair-breaking peak. The pair-breaking peak energy ($E_b$) resides around 380 cm$^{-1}$ for the sample with $T_c=80$ K. With increasing hole concentration $E_b$ shifts to lower energy, and the energy reaches to 140 cm$^{-1}$ for the sample with $T_c=52$ K. Although the $T_c=52$ K is 65 percent of 80 K, the pair-breaking energy is almost one-third of the result for the case with $T_c=80$ K. The results are reasonably reproduce the previous reports[1, 6]. For comparison, ERS of YCa123 are shown in figure 2. Pair-breaking peaks also shift to lower energy with increasing $p$. The tendency of decreasing $E_b$ is quite similar to the case of Tl2201. However, it should be remarked that the $T_c$, in other word, hole concentration $p$ is quite different between Tl2201 and YCa123. For example, if the samples with $T_c=80$ K are compared between Tl2201 and YCa123, the pair-breaking peak energy for Tl-2201 is 50 percent larger than that of YCa123. This is strange enough if pair-breaking peak for $B_{1g}$ polarization spectra simply reflects the maximum of superconducting gap $2\Delta$ with $d_{x^2−y^2}$-symmetry.

The difference of pair-breaking peak energy between the materials are clearly seen in the doping dependence (figure 3). For YCa123, peak energy decreases with increasing hole concentration. The decrease is much larger if we compare the change with $k_BT_c$. At around $p\sim0.19$, the decrease shows kink-like anomaly[7]. On the other hand, the doping dependence of Tl2201 shifts toward higher doping region. Kink-like anomaly is not seen for the case of Tl2201, probably due to insufficient hole concentration.

The rapid decrease of pair-breaking peak energy in overdoped regime seems common among HTSC[8]. Therefore it is natural to expect a common mechanism in ERS of HTSC. In order to explain the rapid decrease, it is currently most plausible to consider the change of superconducting order parameter from $d$-wave to $d + s$-wave symmetry[9]. In this model, pair breaking peak in ERS shifts from $2\Delta$ to lower energy with increasing $s$-wave component. The
mixing of $s$-wave symmetry into pure $d$-wave superconductivity is reasonably explained if lattice distort from tetragonal symmetry to orthorhombic. However, since the crystal structure of Tl2201 is tetragonal, such a simple explanation through lattice distortion collapses.

It has been pointed out that the hole concentration $p \sim 0.19$ for YCa123 is quantum critical point (QCP) in the phase diagram[10]. It seems natural to consider the relation between the anomaly of pair-breaking peak energy and quantum critical behavior. The change of electronic state of HTSC at QCP is the closing pseudogap. A simple viewpoint is that closing pseudogap
may lead to simple $d$-wave superconductivity below $T_c$, but the observed behavior in ERS is quite different from such a simple view. It may be better to consider the anomalies of electronic state in overdoped region of HTSC, such as the existence of unpaired carriers[11].

The difference of doping dependence between Tl2201 and YCa123 may mean that difference of QCP in the phase diagram. In this study, optimally doped hole concentration and $T_c$ dependence on hole concentration are assumed to be common among HTSC. However, if the observed anomaly in this study indicates that the QCP is dependent on material, we should reconsider such a common picture of phase diagram, and the origin of pseudogap.

In summary, $B_{1g}$ electronic Raman spectra of Tl2201 and YCa123 in overdoped region have been presented at 10 K. With increasing hole concentration, a rapid decrease of Cooper-pair breaking peak was observed for both materials. The critical doping where decrease of pair breaking peak energy shows a rapid drop is quite different between them. The origin was discussed in relation to QCP in the phase diagram of HTSC.

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