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TUNNELING STUDIES OF MULTILAYERED SUPERCONDUCTING CUPRATE $(Cu,C)Ba_2Ca_3Cu_4O_{12+\delta}$

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Point contact tunneling data are reported in a multilayered high-T_c cuprate $(Cu,C)Ba_2Ca_3Cu_4O_{12+\delta}$ with $T_c = 117$ K. The tunneling spectra in the superconducting state ($T \ll T_c$) display spectral features such as well-defined superconducting gap peak at $\pm\Delta$ as well as dip-hump structures beyond the peaks. In some cases, the spectra with two-gaps have been observed, indicating the coexistence of two inequivalent superconducting layers. The statistical distribution of superconducting gap magnitude suggests two distinct kinds of superconducting gaps that may originate from two inequivalent CuO_2 planes, a characteristics of multilayered cuprates with $n \geq 3$.

Keywords: Multilayered high-T_c cuprates; tunneling spectroscopy; inhomogeneity.

Extensive efforts to understand the mechanism of high-T_c superconductivity have been focussed on the doping dependence of superconducting (SC) and normal state properties. As a result an unusual phase diagram has been established in which SC critical temperature T_c varies as a bell-shaped curve with T_c maximum at hole concentration $p \sim 0.16$ for most of hole-doped cuprates,[1] and the SC gap magnitude, $\Delta(p)$, monotonically increases with decreasing hole concentration well into the underdoped region where T_c decreases.[2] The $\Delta(p)$ scales with the low-energy pseudogap temperature, $T^*(p)$,[3,4] indicating that the low energy pseudogap phenomenon

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is some type of precursor of superconductivity.[4–8] Furthermore recent tunneling studies on $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi2212) revealed that the dip structures have strong correlation with magnetic resonance mode observed by inelastic neutron studies.[9] However, these studies have been done mainly on a double CuO_2 layer cuprate, Bi2212, and it is not clear whether these characteristic features are generic or not.

We demonstrate here that dip-hump features are observed in a multilayered cuprate, (Cu,C)$Ba_2Ca_3Cu_4O_{12+\delta}$ (Cu1234) which bear a close resemblance to those found in Bi2212. However, unique to multilayered cuprates is the possibility of having up to two inequivalent Cu-O layers. The statistical distribution of Δ in Cu1234 suggests two distinct kinds of gaps, each of which can be linked to spectra found on Bi2212 at different doping. This linkage provides a more microscopic understanding of multilayered cuprates, showing most directly that the inequivalent Cu-O layers each have their own doping level and SC properties. Furthermore, these results provide additional evidence that the dip-hump features are generic to hole-doped high-T_c cuprates.

This SC family was discovered using a high-temperature and high-pressure technique.[10–12] Multilayered high-T_c cuprates that include three or more CuO_2 planes necessarily have two crystallographically-inequivalent kinds of superconducting CuO_2 planes. These are called as inner planes (IP) and as outer planes (OP) where the IP have Cu with fourfold-oxygen coordination and the OP have Cu with fivefold-oxygen coordination. It has been suggested that in the multilayered high-T_c cuprates, the hole concentration for each inequivalent Cu-O layer is different.[13–16] Here our interests are to investigate how these inequivalent CuO_2 planes are reflected in the quasiparticle density of states (DOS) as measured in tunneling.

Cu1234 polycrystalline samples were prepared by the high-temperature and high-pressure synthesis technique.[17] Our sample is $(Cu_{0.8}C_{0.1})Ba_2Ca_3Cu_4O_{12+\delta}$ as nominal composition. X-ray diffraction analysis shows the Cu1234 to be an almost single phase and lattice constant was 3.86 Å and 17.94 Å for *a*- and *c*-axis, respectively. The SC transition temperature T_c was determined as 117 K from zero resistance temperature, where transition width $\Delta T_c \sim 1$ K. Superconductor-insulator-normal metal (SIN) junctions were prepared by a point contact technique using a Au-tip.[2,5] Tunneling conductances were measured by standard ac lock-in technique.

Tunneling studies on multilayered high-T_c cuprate Cu1234 which has two IP and two OP, have only been done by Kane *et al.* to our knowledge.[18] They reported the overall quality of tunneling conductance varied from junction to junction, but the measured gap values remained almost constant, in the range of 27 to 30 meV. However, our tunneling results at 4.2 K showed a variety of gap magnitudes, which ranged from 5 mV to 72 mV in the peak position. Most of spectra ($\sim 70\%$) showed the larger gap whose magnitude Δ is about 40–70 meV and representative superconducting tunneling conductances are shown in Fig. 1(a). Most of the characteristic features including sharp coherence peaks and dip-hump structures

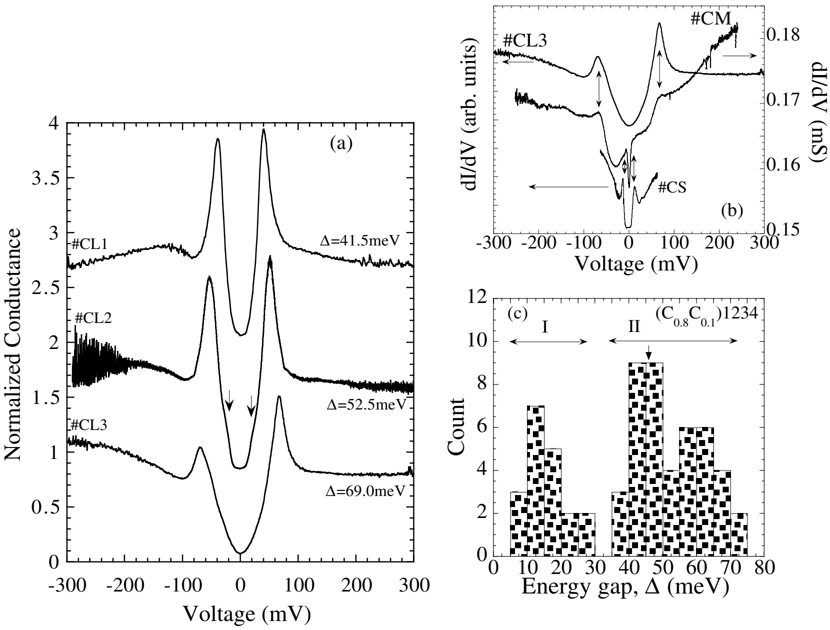

Fig. 1. (a) Typical dI/dV-V curves on $(Cu_{0.8}C_{0.1})1234$. Each peak position is described as Δ in the figure. (b) Representative tunneling conductance showing multigap features together with the spectra showing smaller and larger gaps. Each gap position of #CM corresponds to smaller gap and larger gap position. (c) Histogram showing the statistical distribution of superconducting gap, Δ of $(Cu_{0.8}C_{0.1})1234$ in our tunneling studies. The histogram indicates the coexistence of two kinds of gaps in the paring states, one is $10 \sim 20$ meV (denoted as I) and the another is $40 \sim 70$ meV (denoted as II) in the magnitude of Δ.

beyond the peaks are consistent with those observed on Bi2212,[5,6] $Tl_2Ba_2CuO_{6+\delta}$ (Tl2201)[19] and $Bi_2Sr_{2-x}La_xCuO_{6+\delta}$ (Bi2201).[20,21] Furthermore, the spectra showing the larger gap exhibit features which are consistent with doping dependent trends in Bi2212.[5] For example, note the developing asymmetry in peak height as the gap increases, which is exactly as found in Bi2212 in the underdoped region.[5]

However, we sometimes ($\sim 30\%$) observed the tunneling conductance with smaller Δ which are ranged from 7 to 27 meV as shown in #CS ($\Delta \sim 13$ meV) of Fig. 1(b), and the shape of tunneling spectra showing the small-gap magnitude varies from junction to junction. In addition, we also observed the two-gaps as shown in #CM of Fig. 1(b) and #CL2 in Fig. 1(a) as indicated by arrows. The junction #CM showed clearly two distinct features at ~ 10 mV and ~ 70 mV. In Fig. 1(b), the spectra showing larger gap (#CL3) and smaller gap (#CS) are shown together with #CM due to compare the spectrum. As clearly seen in Fig. 1(b), the position of notable features at $\sim$10mV and ~ 70 mV correspond to smaller gap and larger gap position, respectively. These two gap features have not been observed on single CuO_2 layer cuprates Bi2201,[20,21] Tl2201,[22] or double CuO_2 layer cuprate, Bi2212.[2,5,6] However the similar features have been observed on a triple

CuO_2 layer cuprate, $TlBa_2Ca_2Cu_3O_{10-\delta}$ (Tl1223) whose results will be published on separate paper.[23] Thus the coexistence of two gaps may be a generic feature for multilayered cuprates with the number of CuO_2 planes $n \geq 3$. In order to clearly see the coexistence of two kinds of gaps, we summarized our tunneling results using $(Cu_{0.8}C_{0.1})$1234-Au point contact junctions in Fig. 1(c), which is a histogram showing the statistical distribution of Δ. From Fig. 1(c), one can notice that the gap distribution consists of two regions, that is, one region (I) is about 10–25 meV and the another region (II) is about 40 $\sim$ 70 meV. This result strongly suggests that Cu1234 has two distinct energy gaps originating from distinct Cu-O planes.

We now discuss why the multilayered cuprates might display two distinct kinds of gaps in the quasiparticle DOS. The major difference between multilayered cuprates ($n \geq 3$) and single- or double-CuO_2 layer cuprates ($n = 1, 2$) is the crystallographical structure of the CuO_2 planes. The cuprates with $n = 1, 2$ have only equivalent CuO_2 planes, but the cuprates with $n \geq 3$ must have two inequivalent CuO_2 planes. That is, the IP and OP of multilayered cuprates most probably have different electronic properties due to differences in bonding, doping etc. There are some reports to support this assertion. For example, ^{63}Cu-NMR studies of $(Cu_{0.6}C_{0.4})$1234 by Tokunaga *et al.* showed that OP and IP have different electronic states, that is, the results on $1/T_1T$ and Knight shift of OP showed the characteristic features of heavily overdoped and those of IP showed those of underdoped, and they suggested that the bulk SC transition at $T_c = 117$ K is triggered by the underdoped IP in Cu1234.[24] Previous our tunneling studies on cuprates with $n = 1, 2$ showed that $\Delta(p)$ monotonically increases with decreasing p on Bi2212[5,6] and La-doped Bi2201.[20,21] Furthermore tunneling study on LSCO also showed the similar $\Delta(p)$.[25] Thus this unusual $\Delta(p)$ is most probably generic feature for all hole-doped cuprates. If we assume this unusual $\Delta(p)$ is realized for multilayered cuprates, our tunneling results suggest that the spectra with larger gap corresponds to the electronic state of IP but those with smaller gap corresponds to those of OP, by linking with the results observed by NMR studies.[24] In addition, if we assume that the ratio of superconducting gap and T_c in Cu1234 with $T_c = 117$ K is roughly same as that of optimally-doped Bi2212 with $T_c = 95$ K and $\Delta \sim 38$ meV,[5] the superconducting gap Δ will be 46.8 meV whose position is indicated by arrow in Fig. 1(c). Thus this result suggests that the electronic state showing larger gaps (IP) has a role producing a T_c as high as 117 K. On the other hand, OP has a role to absorb the carrier supplying from charge reservoir layers because bulk T_c of Cu1234 does not change even hole concentration is changed.[17] Furthermore, concerning to the gap distribution within the each region (I & II) in Fig. 1(c), it may originate intrinsic inhomogeneity of IP and OP as suggested by Pan *et al*.[26] Based on these discussion, we suggest that the IP corresponds to optimally-doped or underdoped states and the OP to heavily overdoped states. The Δ of IP is roughly 40–70 meV, that of OP is roughly 10–20 meV.

In summary, we have measured tunneling conductance on multilayered cuprates, Cu1234 and reported the quasiparticle DOS at IP and OP. We found that multilayer cuprates have intrinsic electronic inhomogeneity between chemically distinct CuO_2 layers as well as intrinsic inhomogeneity within each CuO_2 plane. The IP correspond to optimal/underdoped regime and produces a T_c as high as 117 K and the OP is heavily overdoped by absorbing the majority of carriers supplied from the charge reservoir layers. Furthermore, we find that peak-dip-hump structure is qualitatively similar to Bi2212 and is therefore a generic feature of hole-doped high-T_c cuprates.

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