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Doping Dependence on Two Sizes of Superconducting Gaps on Tl1223 by Tunneling Spectroscopy at 4.2 K

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Abstract

We present tunneling results on tri-layered cuprate superconductors $\text{TlBa}_2\text{Ca}_2\text{Cu}_3\text{O}_{8.5+\delta}$ (Tl1223) with two different hole concentration, which are an over-doped Tl1223 with $T_C \sim 112$ K (OD-112K) and a slightly over-doped Tl1223 with $T_C \sim 126$ K (SOD-126K). The tunneling conductances on both samples exhibited two sizes of gaps originated from outer (OP) and inner (IP) CuO_2 planes. The superconducting gap at each planes, $\Delta_{(\text{OP})}/e \approx V_{\text{p}(\text{OP})}$ and $\Delta_{(\text{IP})}/e \approx V_{\text{p}(\text{IP})}$ on OD-112K are observed that $V_{\text{p}(\text{OP})}$ is 22 ± 2 mV and $V_{\text{p}(\text{IP})}$ is 37 ± 4 mV. Similarly, $V_{\text{p}(\text{OP})}$ is 26 ± 2 mV and $V_{\text{p}(\text{IP})}$ is 39 ± 3 mV on SOD-126K. Although both $V_{\text{p}(\text{OP})}$ and $V_{\text{p}(\text{IP})}$ decrease with increasing oxygen contents, $\Delta V_{\text{p}(\text{OP})} = V_{\text{p}(\text{OP})(\text{SOD-126K})} - V_{\text{p}(\text{OP})(\text{OD-112K})}$ is larger than $\Delta V_{\text{p}(\text{IP})}$ for IP. Moreover, $\Delta V_{\text{p}(\text{plane})} \equiv V_{\text{p}(\text{IP})} - V_{\text{p}(\text{OP})}$ increases with overdoping. These results as a function of doping implies the OP might control the variation of T_C dominantly.

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1. Introduction

Multilayer cuprates with $n \geq 3$ (n : the number of CuO_2 planes in a unit cell) have two kinds of crystallographically inequivalent CuO_2 planes (an inner plane (IP) with four-fold oxygen coordination and outer planes (OP) with five-fold oxygen coordination). The investigations of doping inhomogeneity in each CuO_2 planes for multilayer cuprates have been extensively performed by nuclear magnetic resonance (NMR) [1] and angle resolved photoemission spectroscopy (ARPES) [2] studies. Kotegawa *et al.* reported that the hole concentration in the OP is higher than that in IP [1], where each hole concentration in OP and IP were estimated by Knight shift measurement. Moreover, the local doping levels at each CuO_2 planes simultaneously increases with increasing the average doping levels, and the difference of doping levels between OP and IP also increases with increasing the average doping levels.

According to tunneling studies on $\text{Bi}_2\text{Sr}_2\text{Ca}_{n-1}\text{Cu}_n\text{O}_y$ ($\text{Bi}22(n-1)n$) crystals with $n=1$ and 2, the magnitude of superconducting gap Δ decreases with increasing the hole concentration [3, 4], and this behavior would be universal for

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all hole-doped cuprates. Combining these tunneling results with those by NMR, for multilayer cuprates with $n \geq 3$, we expect that the two kinds of gaps will be observed. Actually, we have succeeded in observing the two sizes of the gaps on a lot of multilayer cuprates [5-9]. Recently the similar results were reported by scanning tunneling microscopy/spectroscopy (STM/STS) studies [10] and ARPES studies [2]. Although these observations support strongly the results by NMR in which the hole concentration in OP is different from that in IP, there is no report for the doping dependence on the superconducting gaps Δ_{OP}/e and Δ_{IP}/e in OP and IP by the spectroscopic measurements. In this paper, we report the first investigation of the doping dependence on two sizes of gaps on the magnitude of gaps in OP and IP for $TlBa_2Ca_2Cu_3O_{8.5+\delta}$ (Tl1223) by the point contact tunneling spectroscopy.

2. Experimental details

We prepared two kinds of polycrystalline Tl1223 samples with different doping, which are an over-doped Tl1223 with $T_C \sim 112$ K (OD-112K) and a slightly over doped Tl1223 with $T_C \sim 126$ K (SOD-126K). The synthesis condition of the SOD-126K has been reported in Ref. 11. The OD-112K was synthesized at 800 °C for 2 hours in an O_2 atmosphere of approximately 0.1 MPa. To obtain an over-doped sample, the sample was annealed at 600 °C for 20 h in an O_2 atmosphere. X-ray diffraction patterns indicated that the samples were single phase. The T_C was determined as 126 and 112 K by resistivity and magnetic susceptibility measurements (Fig. 1(a, b)). The SIN tunneling junctions were prepared by a point contact tunneling method using an Au-tip. The dI/dV s were measured by ac lock-in technique at 4.2 K. Here, the negative (positive) bias in the tunneling conductance curves corresponds to the occupied (unoccupied) state of superconductors.

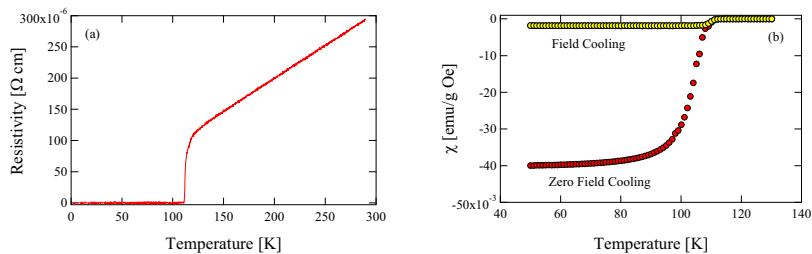


Fig. 1. (a): Resistivity vs temperature for OD-112K. (b): Magnetic susceptibility measurement of OD-112K. The T_C was determined as 112 K.

3. Results and discussion

As shown in Fig. 2, two kinds of spectra with respect to the magnitude of gap were observed for Tl1223. Figure 2 shows typical tunneling conductances on OP (Fig. 2a) and IP (Fig. 2b) in Tl1223 with different hole concentration. The shape in sub-gap region is similar to that of bilayer cuprates [3, 12] suggesting the d -wave order

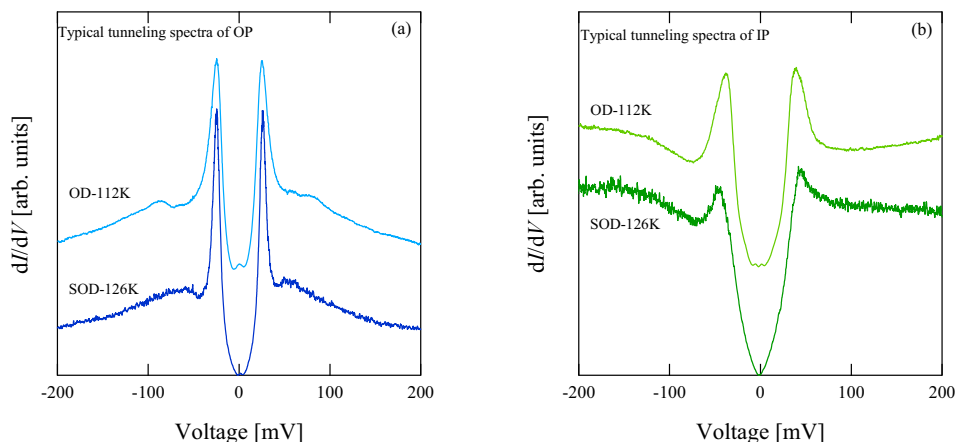


Fig. 2. Typical dI/dV curves on OD-112K and SOD-126K at 4.2 K. (a, b): Typical tunneling spectra of OP (a) and IP (b) on OD-112K (top) and SOD-126K (bottom).

parameter. In addition, we found that the peak structures in OP (Fig.2a) are sharper than those in IP (Fig. 2b). This feature is similar to the previous results for doping dependence in Bi2212 in which the gap peak is sharper in a material with higher carrier concentration [3]. This fact supports validity of our identification for two kinds of gaps, where we identified that the spectra displaying the smaller gap arise from OP. Furthermore the spectra on Tl1223 display a peak-dip-hump structure that is one of characteristic common features for hole-doped cuprates. In addition, the shape of tunneling conductances is asymmetric between occupied and unoccupied state, *i.e.* the strength of the dip and the height of the coherence peak is asymmetric. In particular, it is stronger on the tunneling spectra in IP than those in OP as shown in Fig. 2. These features are consistent with the behavior of the doping dependence on the tunneling spectra, here the spectra in under-doped state is much asymmetric.

We notice that the observed peak voltages on both samples spread from ~20 to ~50 mV. If we plot the histograms on superconducting gap $\Delta/e \approx V_p$, one may notice that the statistical distribution of the V_p consists of two-peaks, as shown in Fig. 3(a, b). The mean value of the OP-gap, $\langle V_{p(OP)} \rangle$, are observed at 22 ± 2 mV and 26 ± 2 mV for OD-112K and SOD-126K, respectively. Similarly, the mean value of the IP-gap, $\langle V_{p(IP)} \rangle$, are observed at 37 ± 4 mV and 39 ± 3 mV for OD-112K and SOD-126K, respectively. The results on Tl1223 are similar to those that have been previously reported on the other multilayered cuprates, such as heavily overoped Tl1223 [5], $(Cu,C)Ba_2Ca_3Cu_4O_y$ [6], $HgBa_2Ca_2Cu_3O_y$ [7], $Ba_2Ca_3Cu_4O_{y-x}F_x$ [8] and Bi2223 [9]. In these previous papers [5-9], it was reported that the two kinds of gaps arise from two crystallographically in-equivalent CuO_2 planes, thus OP and IP, where it is known that they have different local carrier concentrations [1]. Therefore, we similarly identify them that the smaller (larger) gap is due to an OP(IP)-gap.

The average $\langle V_p \rangle$ were plotted in Fig. 3c. Although both $V_{p(OP)}$ and $V_{p(IP)}$ decreases with increasing the doping, the variation ratio of $V_{p(OP)}$ from SOD-126K is larger than that of $V_{p(IP)}$, as shown in Fig. 3d, where the “variation ratio” is defined as $[(V_{p(OD-112K)} - V_{p(SOD-126K)}) / V_{p(SOD-126K)}] / [(T_{C(OD-112K)} - T_{C(SOD-126K)}) / T_{C(SOD-126K)}]$. Moreover we found that

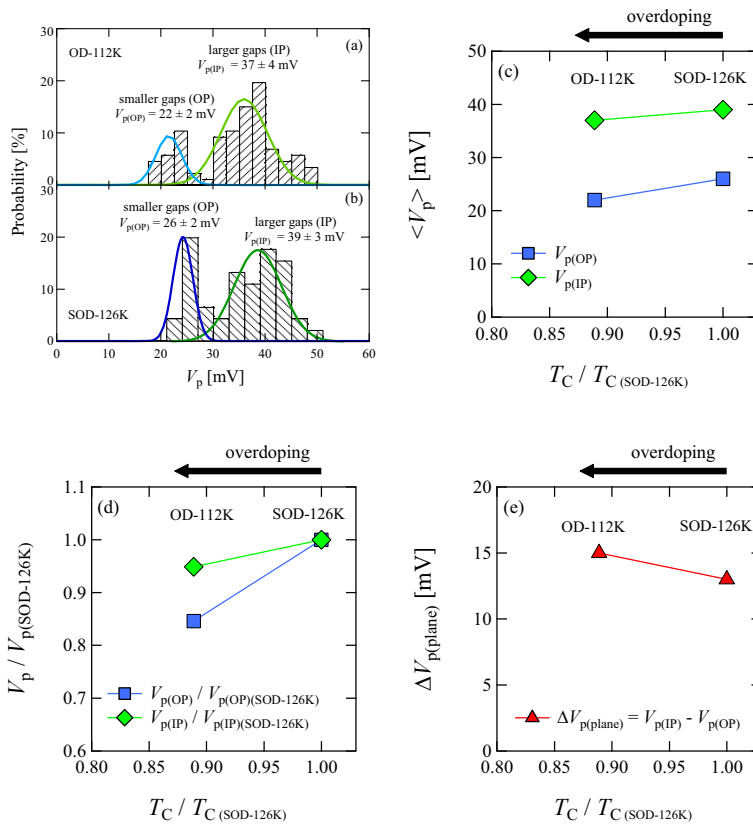


Fig. 3. (a, b): Distribution of magnitude of half peak-to-peak gap V_p on OD-112K (a) and SOD-126K (b). Solid lines are fitted Gaussian distribution. The mean value of V_p on OP (IP), $V_{p(OP)}$ ($V_{p(IP)}$) of 22 ± 2 (37 ± 4) mV and 26 ± 2 (39 ± 3) mV are observed for OD-112K and SOD-126K, respectively. (c, d): Doping dependence on V_p and $V_p / V_{p(SOD-126K)}$ between OP and IP. Green and blue marks are the mean values of $V_{p(OP)}$ and $V_{p(IP)}$, respectively. The variation ratio, $((V_{p(OD-112K)} - V_{p(SOD-126K)}) / V_{p(SOD-126K)}) / ((T_{C(OD-112K)} - T_{C(SOD-126K)}) / T_{C(SOD-126K)})$, of OP (IP) is 1.38 (0.46). (e): Doping dependence on $\Delta V_{p(plane)} = V_{p(IP)} - V_{p(OP)}$.

$\Delta V_{p(\text{plane})} = V_{p(\text{IP})} - V_{p(\text{OP})}$ increases with increasing oxygen content. This result suggest that inhomogeneity of doping along the *c*-axis is enhanced with over-doping. These behaviors can be understood by combining the doping dependence on Δ [3] and on the local doping levels both OP and IP by NMR study [1]. That is the magnitude of gap decreases with increasing doping. Furthermore, $\langle V_{p(\text{OP})} \rangle$ strongly decreases with doping than that at IP, suggesting the carrier at OP strongly increase with doping than that at IP. This is consistent with the results in Ref. 1, the local doping levels in OP and IP, $N_h(\text{OP})$ and $N_h(\text{IP})$, increase with increasing the average doping levels, and $N_h(\text{OP})$ is always larger than $N_h(\text{IP})$. The increasing rate of the $N_h(\text{OP})$ with doping is larger than that of the $N_h(\text{IP})$.

Now, question is “which CuO_2 plane dominantly affects T_C ?” As shown in Fig. 3d, the variation of $V_{p(\text{IP})}$ is slight (the variation ratio of IP is 0.46), and this result suggests that a local doping level in IP is almost constant between SOD-126K and OD-112K. Furthermore, the variation ratio of IP of 0.46 means that $2\Delta/k_B T_C$ decreases with decreasing the hole concentration. This behaviour is inconsistent with that in other hole-doped cuprates. On the other hand, the $V_{p(\text{OP})}$ certainly varied with doping, and also T_C varied. Here, the variation ratio of OP is 1.38, and this value is three times value of IP. The variation ratio of OP of 1.38 means that $2\Delta/k_B T_C$ increases with decreasing the hole concentration and this behavior is consistent with the relationship between $2\Delta/k_B T_C$ and the hole concentration on Bi2212. Namely, it is understood that the variation of the hole concentration in OP leads to the change of $V_{p(\text{OP})}$ and T_C . Therefore, this result suggests that T_C in the range of from 112 K to 126 K on Tl1223 over-doped regime is controlled by outer CuO_2 planes.

In summary, we performed the point contact tunneling measurement on OD-112K and on SOD-126K. The tunneling conductances on Tl1223 exhibited two sizes of superconducting gaps, *i.e.* $V_{p(\text{OP})}$ and $V_{p(\text{IP})}$. The average magnitude of the gap on OP, $\langle V_{p(\text{OP})} \rangle$, are observed at 22 ± 2 mV and 26 ± 2 mV for OD-112K and SOD-126K, respectively. Similarly, the average magnitude of the gap on IP, $\langle V_{p(\text{IP})} \rangle$, are observed at 37 ± 4 mV and 39 ± 3 mV for OD-112K and SOD-126K, respectively. Both $\langle V_{p(\text{OP})} \rangle$ and $\langle V_{p(\text{IP})} \rangle$ are decreasing with decreasing T_C (increasing the doping), the variation ratio from $V_{p(\text{OP})}$ is stronger than that of $V_{p(\text{IP})}$. These results are suggest that OP might control the variation of T_C dominantly.

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