Electronic inhomogeneity in Pb-substituted Bi$_2$Sr$_2$CuO$_{6+\delta}$ studied by STM/STS measurements

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Abstract. Scanning tunneling microscopy/spectroscopy (STM/STS) studies on the slightly overdoped Pb-substituted Bi$_2$Sr$_2$CuO$_{6+\delta}$ (Bi$_{1.75}$Pb$_{0.32}$Sr$_{1.91}$CuO$_{6+\delta}$) with $T_c$ $\sim$ 19.5 K have been performed at 4.6 K in the modulation-free Bi(Pb)-O plane. Almost all dI/dV tunneling spectra exhibit clear peaks on both sides of the energy gap, in contrast to the lanthanoid (Ln) substituted Bi$_2$201 with the similar doping level in which V-shape spectra without peaks have been partially observed. The energy gap $\Delta_s$ distributes in the range from $\sim$5 meV to $\sim$20 meV which is much smaller than Ln-Bi$_2$201 with the similar doping level. It is found that the high-energy region of $\Delta_s$ disappears in the slightly overdoped Pb-substituted Bi$_2$Sr$_2$CuO$_{6+\delta}$.

1. Introduction

Intensive studies on the inhomogeneous electronic state in the high-$T_c$ cuprates have been conducted, triggered by the direct observation of the real-space segregation of the energy-gap by means of the scanning tunneling microscopy/spectroscopy (STM/STS)[1, 2]. It has been revealed that the characteristic length scale of the segregation is the order of nano meters. Since the length scale is comparable with the coherence length of superconductivity, such an inhomogeneous electronic state has been recognized as an intrinsic property of high-$T_c$ mechanism. On the other hand, recent reports[3, 4] have suggested that the inhomogeneous electronic state (the inhomogeneous energy gap) is regarded as being due to the either local electronic or structural perturbation. In Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ (Bi2212), McElroy et al.[3] have pointed out that the distribution of the excess oxygens in the Bi-O plane causes the spatially inhomogeneous energy gap. Furthermore, Sugimoto et al.[4] have reported that the inhomogeneity of the energy-gap is enhanced in the lanthanoid (Ln) substituted Bi2201 with increasing the mismatch of the ionic radius between Sr and the substituted Ln[5]. Accordingly, the relation between the inhomogeneous electronic state and the emergence of the high-$T_c$ superconductivity has not been settled yet. In order to clarify the relation, it is necessary to investigate the effect of substituted and/or interstitial atoms on the inhomogeneous electronic state systematically. In this paper, therefore, we perform STM/STS measurements for the Pb-substituted Bi$_2$Sr$_2$CuO$_{6+\delta}$ (Pbx-Bi2201) as another target to study the inhomogeneous electronic state.

2. Experimental

Single crystals of Pb-substituted Bi$_2$Sr$_2$CuO$_{6+\delta}$ (Pbx-Bi2201) were grown by the floating zone method[6]. Grown single crystals were characterized using the back-Laue x-ray photography and confirmed having single phase by means of the powder x-ray diffraction method. The chemical
compositions were determined by the inductively coupled plasma (ICP) spectroscopy and the electron-probe microanalysis (EPMA) to be Bi$_{1.75}$Pb$_{0.32}$Sr$_{1.91}$CuO$_{6+\delta}$ (Pb0.32-Bi2201). The doping level was controlled by adjusting the concentration of the excess oxygen by annealing in a flowing N$_2$. The superconducting transition temperature $T_c$ was determined to be 19.5 K in the slightly overdoping by measuring the temperature dependence of the magnetic susceptibility with a SQUID magnetometer (Quantum Design, MPMS-XL5) in a magnetic field of 1 Oe. STM/STS measurements were performed at 4.6 K using single crystals cleaved in the ultrahigh vacuum condition.

3. Results and Discussion

Figure 1 shows the constant current STM image of the Bi(Pb)-O plane at 4.6 K in the slightly overdoped Pb0.32-Bi2201 ($T_c = 19.5$ K). The clear two-dimensional atomic arrangement without the structural modulation is found. The atomic arrangement consists of Bi and Pb atoms with the interatomic spacing of $\sim 3.8$ Å. The brighter and other spots are regarded as Pb and Bi atoms, respectively, since the number of the former corresponds to the concentration of Pb. The moderate contrast observed in the background may be attributed to the inhomogeneous electronic state[7]. In our previous reports[8, 9], we have reported that the Bi(Pb)-O plane exhibits a micro-phase separation consisting of the structural modulated and modulation-free domains around $x$(Pb) $\sim 0.3$. As for Pb0.32-Bi2201, sizes of the modulation-free domains have been estimated as $>(0.1 \mu m)^2$ which is much larger than those of the modulated domains[9]. The Bi(Pb)-O plane shown in Fig. 1 is a part of a such large-size modulation-free domain. In this study, the STM/STS measurement is performed on the structural modulation-free area.

Figure 2 shows the representative dI/dV tunneling spectra measured at 4.6 K in the same Bi(Pb)-O plane as shown in Fig. 1. Here, $\Delta_s$ denotes the energy gap estimated as one-half of the peak-to-peak value. It is found that the dI/dV tunneling spectra are asymmetric in shape and always have two peaks on both sides of the energy gap. The asymmetric shape is general in high-$T_c$ cuprates[1, 3, 4]. However, two peaks in all spectra is particular, because in fact Ln-
Figure 3. (a) The spatial mapping of the energy gap $\Delta_s$ in the slightly overdoped Pb0.32-Bi2201 ($T_c = 19.5$ K). (b) Histogram of $\Delta_s$. Ave. $\Delta_s$ and $\sigma$ denote the average and the standard deviation of $\Delta_s$, respectively. (c) The $dI/dV$ tunneling spectra picked up at regular intervals along the arrow in (a). The condition of measurements is the same as Fig. 2.

Bi2201 partially exhibits the V-shaped tunneling spectra without peaks in the similar doping level[4].

Figure 3(a) shows the spatial mapping of $\Delta_s$. $\Delta_s$ distributes spatially, as similar to other Bi-based high-$T_c$ cuprates[1, 2, 3, 4]. Figure 3(c) shows the $dI/dV$ tunneling spectra picked up at regular intervals along the arrow in Fig. 3(a). It is found that spatial variations in the value of energy gap and the shape of spectra are moderate. In order to evaluate inhomogeneity, we make the histogram of $\Delta_s$ as shown in Fig. 3(b). It is found that $\Delta_s$ distributes as $\sim 5$ meV $\leq \Delta_s \leq \sim 20$ meV with the average and the standard deviation of 10.72 meV and 2.41 meV, respectively. Here, it is worthwhile noting that the estimated average and standard deviation are much smaller than those of Ln-Bi2201 with the similar doping level. In fact, the average of energy gap in La-Bi2201 ($T_c = 34$ K) and Gd-Bi2201 ($T_c = 14$ K) with the similar doping level have been estimated as 36 meV and 48 meV with the standard deviation of 17 meV and 22 meV, respectively[4]. It appears that high energy region of $\Delta_s$ is anomalously suppressed in Pb0.32-Bi2201, compared with Ln-Bi2201 in which the large components in the high-energy gap makes the average and the standard deviation larger.

The specific of Pb0.32-Bi2201 is clearly seen in Fig. 4. Figure 4 shows $T_c$ vs the standard deviation $\sigma$ plot for various Bi-based high-$T_c$ cuprates with the slightly underdoping, the optimally doping and the slightly overdoping. Sugimoto et al. have proposed this plot and indicated the linear relation between $T_c$ and $\sigma$ shown by the broken line. However, it is found that Pb0.32-Bi2201 does not follow the linear relation because of the very small $\sigma$. It appears that there are three factors which control the amplitude of $\sigma$. The first is the structural modulation in the Bi-O plane. Recently, Slezak et al.[13] have shown that the energy gap of Bi2212 varies spatially with the same periodicity as the structural modulation of the Bi-O plane, by means of STM/STS measurements. They have proposed that the spatial variation of the energy gap is directly correlated with a modulation of the apical oxygen position controlled by the structural modulation in the Bi-O plane. Moreover, a recent theoretical work by Mori et al.[14] has also
pointed out that the change of the apical oxygen position due the structural modulation in the 
Bi-O plane can modulate the pairing interaction in the CuO$_2$ planes. These explain why $\sigma$
 of Pb$_{0.32}$-Bi$_{2201}$ is smaller than that of Bi$_{2212}$. That is, the flatness of the modulation-free Bi(Pb)-O plane must make the variation of the energy gap small. The second is the disorder
due to the mismatch of the ionic radius between Sr and the substituted Ln[4, 5]. This is typical
in the Ln-Bi$_{2201}$. $\sigma$ of Ln-Bi$_{2201}$ becomes pretty large because of both the mismatch in the Sr
site and the structural modulation. The third is the inhomogeneous distribution of the excess oxygens in Bi-O plane[3]. This effect is common among the Bi-based high-$T_c$
cuprates. It is highly possible that the small $\sigma$ of Pb$_{0.32}$-Bi$_{2201}$ is realized because the factor is limited to this
effect.

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References
[1] Pan S H, O’Neal J P, Badzey R L, Chamon C, Ding H, Engelbrecht J R, Wang Z, Eisaki H, Uchida S, Gupta 
A K, Ng K-W, Hudson E W, Lang K M, Davis J C 2001 Nature 413 282
[2] Lang K M, Madhavan V, Hoffman J E, Hudson E W, Eisaki H, Uchida S, Davis J C 2002 Nature 415 412
[3] McElroy K, Lee J, Sleazak J A, Lee D-H, Eisaki H, Uchida S and Davis J C 2005 Science 309 1048
[4] Sugimoto A, Kashiwaya S, Eisaki H, Kashiwaya H, Tsuchiura H, Tanaka Y, Fujita K and Uchida S 2006 Phys. 
Rev. B 74 094503
[5] Eisaki H, Kaneko N, Feng D L, Damascelli A, Mang P K, Shen K M, Shen Z-X and Greven M 2004 Phys. 
Rev. B 69 064512
[6] Kudo K, Miyoshi Y, Sasaki T, Nishizaki T and Kobayashi N 2006 J. Phys. Soc. Jpn 75 124710
[7] Kudo K, Nishizaki T, Okumura N and Kobayashi N 2007 Physica C 460–462 948
[8] Nishizaki T, Nishizaki T, Okumura N and Kobayashi N 2007 Physica C 460–462 156
[9] Kudo K, Nishizaki T, Okumura N and Kobayashi N 2007 Physica C 463–465 40
[10] McElroy K, Lee D-H, Hoffman J E, Lang K M, Lee J, Hudson E W, Eisaki H, Uchida S and Davis J C 2005 
Phys. Rev. Lett. 94 197005
[11] Hudson E W, Lang K M, Madhavan V, Pan S H, Eisaki H, Uchida S and Davis J C 2001 Nature 411 920
[12] Kinoda G, Hasegawa T, Nakao S, Hanaguri T, Kitazawa K, Shimizu K, Shimoyama J and Kishio K 2003 
Appl. Phys. Lett. 83 1178
[13] Sleazak J A, Lee J, Wang M, McElroy K, Fujita K, Andersen B M, Hirschfeld P J, H. Eisaki H, Uchida S and 
Davis J C 2008 Natl. Acad. Sci. USA 105 3203
[14] Mori M, Khaliullin G, Tohyama T and Maekawa S Preprint arXiv:0805.1281