Intrinsic Tunneling Spectroscopy for Pb-Substituted Bi2212 in the Underdoped Region

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Abstract. We measured intrinsic tunneling spectroscopy in mesa structures ($S = 1 \times 1 \mu m^2$) of Bi$_{1.9}$Pb$_{0.1}$Sr$_2$CaCu$_2$O$_{8+\delta}$ (PbBi2212) and Hall effect in PbBi2212 cleaved thin crystals. The doping level $p$ of the PbBi2212 is estimated quantitatively through the comparison with LSCO, which is in the underdoped region. It is found that the Josephson critical current density $J_c$ of PbBi2212 is less deviated from Ambegaokar Baratoff critical current density $J_{AB}^c \approx \frac{\pi \Delta}{2eR_N S}$ than the case of underdoped Bi2212. It is interpreted that the Pb substitution makes the tunnel barrier lower, resulting in a high superconducting pair density even with a lower doping.

1. Introduction
In some of the high-$T_c$ superconductors (HTSCs) such as Bi$_2$Sr$_2$Ca$_n$Cu$_n$O$_{2n+4+\delta}$ $(n = 1,2,3)$ which exhibits a significantly large anisotropy, the crystal structures are characterized by a layered structure of stacked CuO$_2$ superconducting atomic sheets with an insulating sheet in between, which are called intrinsic Josephson junctions (IJJs)[1]. IJJ provides us with various fields of research owing to almost ideal characteristics arising from the clean and flat barrier interfaces only attainable in the intrinsic crystal structure. An interesting application of IJJs is to use them as a probe into the superconducting properties of HTSCs, in particular to the quasiparticle density of states in the superconducting state[2], which is called intrinsic tunneling spectroscopy (ITS). We consider that experimental results gained through ITS, which probe further intrinsic properties of HTSCs than these through scanning tunneling spectroscopy (STS) in which the surface may deteriorate due to a high vacuum.

Recently, STS revealed the real-space inhomogeneous superconducting state in the $ab$-plane of underdoped Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$(Bi2212)[3]. This real-space inhomogeneity is a potential origin for the anomalous suppresion of the Josephson critical current density $J_c$ in underdoped Bi2212[4]. On the other hand, angle-resolved photoemission spectroscopy (ARPES) in underdoped Bi2212 showed that the density of states near the fermi level vanishes even at temperatures above $T_c$ except in the vicinity of $(\pi, \pi)$ direction[5]. This $k$-space heterogeneity is another potential origin for the suppresion because the conductive directions of carriers are limited to the zone near the nodal direction of $d$-wave superconducting order parameter and only a small portion of the carriers can form superconducting pairs[6].

We have performed ITS for a PbBi2212, which exhibits a high $J_c$ and a reduced anisotropy like overdoped Bi2212, whereas the temperature dependence of the $c$-axis resistivity $\rho_c$ shows strong upturn above $T_c$ commonly seen in underdoped Bi2212. Hall effect measurements for
the PbBi2212 were also performed to determine the doping level. It was found that the Pb substitution weakens the anisotropy and increases tunneling superfluid density even in the underdoped region.

2. Experiments

Single crystals of PbBi2212 were grown by the self-flux method. Electron dispersive spectroscopy analysis of a crystal shows that the distribution of Pb over the selected crystal is almost homogeneous and the composition is \(\text{Bi}_{1.9}\text{Pb}_{0.1}\text{Sr}_{2}\text{CaCu}_{2}\text{O}_{8+\delta}\).

Samples for the \(c\)-axis transport measurements were ultra-thin mesa structures fabricated on a crystal glued on a sapphire substrate. To reduce the contact resistance between the electrodes and the topmost IJJs, a high value of which always disturbs the precise measurement in this technique, we deposited an Ag thin film on the fresh surface cleaved inside the vacuum chamber (cleaving-in-vacuum method)[7]. The mesa structures were formed by electron-beam lithography and Ar-ion milling. A high resolution and a slow etching rate are necessary to make tiny volume mesas which enable precise measurement by reducing self-heating. The details of mesa-fabrication are described elsewhere[8]. ITS measurements were done by short-pulse bias method to reduce self-heating [9]. Voltage values were acquired at \(\sim 1\,\mu\text{s}\) from a pulse rise and then smoothed to numerically obtain \(dI/dV-V\) curves, which are refered as tunnel spectra. To measure \(J_c\), the current-voltage (\(I-V\)) characteristics were also recorded by taking traces on an analog oscilloscope under biasing triangular ac voltage.

Samples for the \(ab\)-plane transport measurements were Hall-bar structures fabricated from mechanically cleaved crystals, which were taken from the same batch of crystals used for the \(c\)-axis transport measurements. The Hall-bar pattern was formed by photolithography and either by wet etching with diluted nitric acid or by Ar-ion milling. To obtain the data precisely, the thickness of Hall-bar structures is thinned down less than 500 nm.

3. Results and discussion

Figure 1 shows the quasiparticle \(I-V\) curves and the tunneling spectra of the mesa sample obtained by short-pulse ITS measurement at various temperatures. The junction number and the junction area of the mesa are \(N = 5\) and \(S = 1\times1\,\mu\text{m}^2\), respectively. The spectrum at \(10\,\text{K}\) shows distinct peaks of quasiparticle density of states that enable us to determine the superconducting gap \(2\Delta = 72\,\text{meV}\). The value of \(2\Delta\) corresponds to the ones of Bi2212 in the underdoped region. It is noted that increase of \(dI/dV\) outside \(2\Delta\) is considered as a symptom of the pseudogap.

Figure 2a shows the \(I-V\) characteristics at \(5\,\text{K}\). It is clear that critical currents of all IJJs are uniform and the separations between adjacent branches are equivalent, indicating that the mesa can be considered as a stack of five identical IJJs. This allows us to estimate \(2\Delta\) by measuring the \(c\)-axis quasiparticle conductance of the mesa. Figure 2b shows the temperature dependence of the \(c\)-axis resistivity \(\rho_c\) of the same mesa. With decreasing temperature, \(\rho_c\) increases up to the value several times larger than the value at room temperature, and suddenly drops to the zero resistance with \(T_c = 63.5\,\text{K}\). This significant upturn in \(\rho_c\) is known as a characteristic feature of underdoped Bi2212 and is consistent with a symptom of a large pseudogap in the spectra. In addition, the upturn of \(\rho_c\) in this sample is more significant than the one of previous PbBi2212 sample with \(T_c = 77\,\text{K}\)[10]. This is consistent with the difference in \(T_c\) and the mesa in this work is in the underdoped regime. However, the values are one order smaller than those in optimally-doped Bi2212 and \(J_c\) is much larger than ones in Bi2212. This contradiction between the temperature dependence and the values of \(\rho_c\) is explained by the structural modifications induced by the Pb substitution[11]. Thus the mesa samples are in the underdoped region.

The doping level \(p\) of mesas change from the one of the as-grown crystal due to the reduction during the fabrication process, which made the difference in properties among the mesas.
\[ T_c = 63.5 \text{ K} \]

**Figure 1.** The quasiparticle \( I-V \) curves obtained by short-pulse ITS measurements at various temperatures (a), and the inter-layer tunneling spectra derived by applying numeric calculations (b). Abscissas of both (a) and (b) are normalized to the single junction voltage by dividing the values by the junction number of the sample. The linearity of the \( I-V \) characteristics in the high voltage regime, which indicates that the self-heating was suppressed enough, enables us to derive the normal tunneling resistance \( R_N \). The superconducting gap \( 2\Delta \) is determined by the peak-to-peak width of the tunneling spectrum.

**Figure 2.** The \( I-V \) characteristics at 5 K obtained by using an analog oscilloscope (a), and the temperature dependence of the \( c \)-axis resistivity \( \rho_c \) of the mesa sample (b).

The doping level \( p \) was estimated by comparing the hole numbers per Cu atom \( n_{\text{H}}^{\text{Cu}} \) with those in \( \text{La}_{2-x}\text{Sr}_x\text{CuO}_4 \)[12] in which \( p \) is equal to \( x \). \( n_{\text{H}}^{\text{Cu}} \) obtained by the Hall coefficient as \( n_{\text{H}}^{\text{Cu}} = V_{\text{Cu}}/eR_{\text{H}} \), where \( V_{\text{Cu}} \) is the volume occupied by a Cu atom, and \( e \) is the elementary charge. In this work \( n_{\text{H}}^{\text{Cu}} \) ranges from 0.31 to 0.41, which corresponds to \( p = 0.124 \sim 0.150 \). These results enable us to derive a function \( T_c(p) \) of the PbBi2212 through the Tallon’s empirical formula[13]. The doping level of the mesa was estimated by its \( T_c \) vice versa.

Figure 3 shows the dependence of the experimental \( J_c \) and the theoretical estimate, \( J_c^{\text{AB}} \approx \pi\Delta/2eR_N S \) which is derived from the Ambegaokar-Baratoff model[14], on the doping level. Here \( R_N \) is the normal tunneling resistance obtained from high voltage extrapolation of the current-voltage characteristics. Due to the reduced \( \rho_c \) by the Pb substitution, both of the values are greater than those of Bi2212 in the same doping region. Moreover, the comparison of the ratio of \( J_c \) to \( J_c^{\text{AB}} \) with the ones of Bi2212 indicates that the Pb substitution relieves the anomalous suppression of \( J_c \) in the underdoped region. So far, we do not have a clear suggestion
to explain this increase of $J_c/J_{AB}$ ratio by substituting Pb for Bi. STS reveals inhomogenous superconducting gap in PbBi2212 as well as Bi2212[15]. This result excludes the scenario that the increase of the superconducting region in the $ab$-plane increases tunneling superfluid density along the $c$-axis.

Figure 3. The dependence of the actual Josephson critical current density and the theoritical estimate, which are $J_c$ and $J_{AB}$, respectively, on the doping level compared with that of Bi2212.

4. Conclusion

We have measured the superconducting gap $2\Delta$ and the critical current density $J_c$ in an underdoped PbBi2212 mesa. The Hall coefficient measurements reveal that the doping level of the mesa turns to be 0.10, which is consistent with the large $2\Delta = 72$ meV and the strong upturn in $\rho_c(T)$ above $T_c$. The suppression of $J_c$ comparing with the value estimated from the superconducting gap $J_{AB}$ is less pronounced than in Pb-free Bi2212.

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References

[1] Kleiner R and Müller P 1994 Phys. Rev. B 49 1327
[2] Suzuki M, Watanabe T and Matsuda A 1999 Phys. Rev. Lett. 82 5361
[3] 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 and Davis J C 2001 Nature 413 282
[4] Suzuki M, Hamatani T, Yamada Y, Anagawa K and Watanabe T 2006 J. Phys. Conf. Ser. 43 1110
[5] Lee W S, Vishik I M, Tanaka K, Lu D H, Sasagawa T, Nagaosa N, Devereaux T P, Hassain Z and Shen Z-X 2007 Nature Lett. 450 81
[6] Suzuki M, Hamatani T, Yamada Y, Anagawa K and Watanabe T 2009 J. Phys. Conf. Ser. 150 052252
[7] Zhao S P, Zhu X B, Wei Y F, Chen G H, Yang Q S, Lin C T 2005 Phys. Rev. B 72 184511
[8] Kakeya I, Hamada K, Tachiki T, Watanabe T, Suzuki M 2009 Supercond. Sci. Technol. 22 114014
[9] Anagawa K, Yamada Y, Shibauchi T, Suzuki M 2003 Appl. Phys. Lett. 83 2381
[10] Kambara H, Kakeya I and Suzuki M to be published in Physica C
[11] Gladyshevskii R, Musolin N and Flikiger R 2004 Phys. Rev. B 70 184522
[12] Ando Y, Kurita Y, Koniya S, Ono S and Segawa K 2004 Phys. Rev. Lett. 92 197001
[13] Tallon J L, Cooper J R, De Silva P S I P N, Williams G V M and Loram J W 1995 Phys. Rev. Lett. 75 4114
[14] Ambeagaokar V and Baratoff A 1963 Phys. Rev. Lett. 10 486; Erratum 11 104
[15] Kinoda G and Hasegawa T 2003 Phys. Rev. B 67 224509