Title
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Author(s)
Kambara, Hitoshi; Kakeya, Itsuhiro; Suzuki, Minoru

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Increase of superfluid density with growth of quasiparticle density of states probed by intrinsic tunneling spectroscopy in Bi$_{1.9}$Pb$_{0.1}$Sr$_2$CaCu$_2$O$_{8+\delta}$

Hitoshi Kambara,* Itsuhiro Kakeya,† and Minoru Suzuki
Department of Electronic Science and Engineering, Kyoto University, Kyoto, Japan
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Interlayer tunneling spectra, the Josephson critical current density $J_c$, and the normal tunneling resistance have been measured simultaneously for intrinsic Josephson junctions of Bi$_{1.9}$Pb$_{0.1}$Sr$_2$CaCu$_2$O$_{8+\delta}$ (BiPb2212) in the underdoped region using a small mesa structure ($S = 1 \times 1 \mu m^2$) and a short-pulse technique. $J_c$ was significantly enhanced when Bi was partially replaced with Pb. By comparing $J_c$ with the value estimated by the Ambegaokar-Baratoff formula, it was found that superfluid density doubles in the Pb-containing sample. In addition, Pb raises the superconducting coherence peak without changing the value of the superconducting gap $\Delta$. Based on a model that accounts for the inhomogeneity of $\Delta$ in $k$ space, we argue that the superfluid density increases because Pb substitution increases the homogeneity of $\Delta(k)$.

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I. INTRODUCTION

The crystal structures of cuprate superconductors comprise layers of superconducting CuO$_2$ atomic sheets interspersed with layers of insulating sheets. The interfaces between the layers are termed intrinsic Josephson junctions (IJJs). Intrinsic tunneling spectroscopy (ITS) is a technique by which information on the quasiparticle density of states (DOS) in the layers are termed intrinsic Josephson junctions (IJJs). Since an IJJ inside the crystal is protected by layers of superconducting CuO$_2$ atomic sheets interspersed (Bi$_{2212}$) have revealed that the pseudogap survives even in the electronic phase diagram of cuprates can be fully elucidated. Investigations on IJJ materials with varying $J_c$ would provide intriguing tests of this idea. Substituting Bi with Pb is known to drastically increase the critical current density $J_c$. $J_c$ is a macroscopic measure of the incompleteness of the Fermi surface. Given that $J_c$ suppression and shrinking Fermi arc depend similarly on the doping dependence, we infer that $J_c/J_{AB}$ is a macroscopic measure of the incompleteness of the Fermi surface. Investigations on IJJ materials with varying $J_c$ would provide intriguing tests of this idea. Substituting Bi with Pb is known to drastically increase the critical current density $J_c$. $J_c$ is a macroscopic measure of the incompleteness of the Fermi surface. Given that $J_c$ suppression and shrinking Fermi arc depend similarly on the doping dependence, we infer that $J_c/J_{AB}$ is a macroscopic measure of the incompleteness of the Fermi surface. Essentially, the Pb substitution orders the block layer and lowers the barrier height. We have previously found that Pb substitution increases $J_c$ beyond that expected from the change of resistivity.

In this paper, we discuss the effect of the Pb substitution on $J_c$, the quasiparticle DOS in the $k$ space, and the superconducting gap structure. We measured $J_c$, $\Delta$, and $R_N$ in Pb-substituted Bi2212 (BiPb2212) by ITS. We then estimated $J_c^{AB}$ from $\Delta$ and $R_N$, and determined $J_c/J_{AB}$. We found that the anomalously reduced $J_c$ was recovered by the Pb substitution. Moreover, by comparing the tunneling spectra with those of Bi2212 and calculations, we conclude that Pb substitution enlarges the Fermi arc and increases the $J_c$.

II. EXPERIMENTS

Single crystals (nominal composition of Bi$_{1.9}$Pb$_{0.1}$Sr$_2$CaCu$_2$O$_{8+\delta}$) used in the present work had been grown previously by the self-flux method. Electron dispersive spectroscopy (EDS) reveals a homogeneous distribution of Pb within the crystal, and a composition of Bi$_{1.9}$Pb$_{0.1}$Sr$_2$CaCu$_2$O$_{8+\delta}$. To ensure identical Pb concentrations among the samples, crystals with flat surfaces ($ab$ plane) were collected from aggregates grown in the same crucible. To obtain Hall-bar and mesa structures, crystals were fabricated under vacuum conditions. Basic properties of the samples are listed in Tables I and II.
Differences between samples are caused by changing oxygen concentrations throughout the processes in vacuum.

Samples H-1, H-2, and H-3 were used for \(ab\)-plane transport measurements. The Hall-bar patterns were formed by photolithography with either wet or dry etching in thin (\(<500\) nm) crystals cleaved from the single crystals. A prepared sample with silver electrodes is shown in Fig. 1(a). Resistances and Hall voltages are measured simultaneously with the ac standard bridge of the Physical Properties Measurement System (Quantum Design Inc.). The scanning external magnetic field was set perpendicular to the Hall-bar plane and was varied between \(\pm 1\) T. Samples M-1, M-2, and M-3 were used for the \(c\)-axis transport measurements. Mesa structures of a few unit cells thickness (surface area \(1 \times 1 \mu m^2\)) were fabricated by cleave-in-vacuum method\(^{21}\) and electron-beam lithography.\(^{20}\) Our method\(^{20}\) allows the thickness of the mesa to be controlled to half-unit-cell precision (1.5 nm). The mesas shown in Fig. 1(b) were measured by the three probe technique, assuming negligible contact resistance between the mesa and the upper electrode. The \(c\)-axis resistance was measured by standard dc methods, and the current-voltage (\(I-V\)) characteristics were collected under a constant-amplitude biasing triangular ac voltage. ITS measurements were performed with the short pulse (\(~1\mu s\)) bias method\(^2\) to maximally suppress self-heating in high bias regions. Details of sample preparation and measurement are described elsewhere.\(^{19,20}\)

### III. RESULTS AND DISCUSSION

#### A. \(ab\)-plane transport measurements

Figure 2 shows how the \(ab\)-plane resistivity \(\rho_{ab}(T)\) and the Hall coefficient \(R_H(T)\) vary with temperature. A typical Hall-bar sample (H-3) of thickness \(d=346\) nm was used. We define \(T_c\) as the midpoint of the resistance drop starting around \(100\) K. The obtained \(T_c\) values are consistent with those of bulk BiPbi2212 crystals obtained from the same crucible. \(R_H(T)\) exhibits a broad peak around \(130\) K associated with \(T^{-1}\)-linear behavior at a higher temperature region and a rapid decrease in the lower temperature region. These \(R_H(T)\) trends are commonly observed in cuprate superconductors.\(^{22}\) Among the samples, sample H-3 with the highest \(T_c\) shows the smallest \(R_H(300\) K) [see Table I], which tells that all of the samples are underdoped.

#### B. Estimation of hole doping

The extent of doping \(p\) in the mesa must be determined in this study, since we aim to compare the doping evolution of the superconducting gap and Josephson critical current between BiPb2212 and Bi2212. In BiPb2212, \(p\) cannot be directly determined by cation substitution because the excess oxygen located between the BiO planes introduces hole carriers into the CuO2 layers.\(^{23,24}\) According to Ando \textit{et al.}\(^{25}\) and Konstantinović \textit{et al.},\(^{26}\) \(p\) can be determined by comparing the hole number per Cu atom \(n_{\text{Cu}}^0\) at \(300\) K in various cuprates with that in La2-x, SrxCuO4 (LSCO), for which \(p\) is equal to \(x\). Applying this approach, we obtained \(p\) as 0.124–0.150 from the measured values of \(n_{\text{Cu}}^0 = 0.31–0.41\). From these results, we obtain the following empirical formula\(^2\) for determining \(T_c(p)\) of the BiPb2212:

\[
\frac{T_c}{T_{c\max}} = 1 - 82.6(p - 0.16)^2, \tag{2}
\]

where \(T_{c\max}\) is \(89.7 \pm 0.5\) K. The doping \(p\)'s of the mesas listed in Table II are estimated by Eq. (2) from the measured \(T_c\) vice versa. For all of the samples used in this study, \(p\) is below 0.16, inconsistent with an interpretation that substituting Bi\(^{3+}\) with Pb\(^{2+}\) in Bi2212 increases hole carriers in the CuO2 layers and makes the crystals overdoped. We posit instead that the Pb substitution decreases the volume of excess oxygen near the substitution site. In fact, the excess oxygen trapped between the BiO planes modifies the crystal structure,\(^{28,29}\) thus the

<table>
<thead>
<tr>
<th>Sample</th>
<th>H-1</th>
<th>H-2</th>
<th>H-3</th>
</tr>
</thead>
<tbody>
<tr>
<td>(T_c) (K)</td>
<td>84.2</td>
<td>82.9</td>
<td>88</td>
</tr>
<tr>
<td>(R_H(300) K) (10^{-3}) cm(^3) C(^{-1})</td>
<td>2.21</td>
<td>2.12</td>
<td>1.72</td>
</tr>
<tr>
<td>(p)</td>
<td>0.124–0.140</td>
<td>0.126–0.142</td>
<td>0.138–0.150</td>
</tr>
<tr>
<td>Thickness: (d) (nm)</td>
<td>62.1</td>
<td>123.3</td>
<td>346.1</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Sample</th>
<th>M-1</th>
<th>M-2</th>
<th>M-3</th>
</tr>
</thead>
<tbody>
<tr>
<td>(T_c) (K)</td>
<td>63.5</td>
<td>77</td>
<td>86.5</td>
</tr>
<tr>
<td>(\Delta(T) at (10) K) (meV)</td>
<td>36</td>
<td>37.3</td>
<td>32.3</td>
</tr>
<tr>
<td>(J_c(10) K) (kA/cm(^2))</td>
<td>2.4</td>
<td>4</td>
<td>10</td>
</tr>
<tr>
<td>(J_{ab}(10) K) (kA/cm(^2))</td>
<td>43.6</td>
<td>48.2</td>
<td>51.1</td>
</tr>
<tr>
<td>(R_H(10) K) ((\Omega))</td>
<td>129.5</td>
<td>121.5</td>
<td>99.2</td>
</tr>
<tr>
<td>(\rho_{ab}(300) K) ((\Omega) cm)</td>
<td>11.3</td>
<td>10.3</td>
<td>7.88</td>
</tr>
<tr>
<td>(p)</td>
<td>0.098–0.103</td>
<td>0.115–0.123</td>
<td>0.132–0.148</td>
</tr>
<tr>
<td>Number of IJJs: (N)</td>
<td>5</td>
<td>4</td>
<td>4</td>
</tr>
<tr>
<td>Area: (S) ((\mu m^2))</td>
<td>1 × 1</td>
<td>1 × 1</td>
<td>1 × 1</td>
</tr>
</tbody>
</table>
presence of Pb reduces the significance of the modulation of the \(ab\) plane.\(^{16,30}\)

### C. ITS measurements

Figure 3(a) shows the relationship between temperature and \(c\)-axis resistivity \(\rho_c\) of the mesas. \(T_c\) is defined as the midpoint of the resistive transition. As temperature decreases up to \(T_c\), \(\rho_c\) increases to several times larger than that at room temperature, and suddenly drops thereafter. This remarkable change in \(\rho_c\) is characteristic of underdoped Bi2212.\(^{15,31}\) In addition, the magnitude of \(\rho_c\) and the extent of its rise increase with decreasing \(T_c\). These features further suggest that all of the mesas are underdoped. However, the \(\rho_c\) values are several times smaller than those of Bi2212, suggesting that the structural modifications induced by the Pb substitution\(^{18}\) lower the tunnel barrier of the IJJ.

Figures 3(b)–3(d) exhibit the \(c\)-axis \(I-V\) characteristics for the respective mesas. The homogeneous critical current for all branches and equal voltage spacing between the branches demonstrate that the IJJs within the mesas possess identical properties. The number of junctions \(N\) included by each mesa is clearly indicated by the number of branches in the figures. Furthermore, because the first branch in each mesa is perpendicular to the voltage axis, the contact resistance of each mesa is sufficiently low. Therefore, we can extract quasiparticle tunneling properties for a single IJJ by dividing the observed voltage by the number of junctions. The Josephson critical current densities \(J_c\) ranged from 2 to 10 kA/cm\(^2\), much larger than those of Bi2212, which ranged from 0.2 to 1 kA/cm\(^2\) at \(p = 10^{-0.14}\)\(^{14,15}\).

Figure 4 displays the results of ITS measurements for the same mesas. The \(I-V\) curves are linear in the high voltage region (\(>120\) mV), implying that the self-heating induced by the bias current is sufficiently suppressed. Thus we can determine the normal tunneling resistance \(R_N\) from the slope of the \(I-V\) characteristics in the linear region. When self-heating is not negligible, sublinear behavior occurs in the high voltage region of the \(I-V\) curve.\(^{32}\) The \(dI/dV\) magnitudes of the tunneling spectra display sharp peaks, characteristic of superconducting gaps below \(T_c\). The superconducting gaps (\(\Delta = 30–40\) meV at \(T = 10\) K) are the same as those for underdoped Bi2212.\(^{15}\) However, as shown later, the peak magnitudes exceed those of Bi2212 in the same doping region. We also observe another gap structure (\(>2\Delta\)) that dips and rises below \(T_c\) and survives at temperatures exceeding \(T_c\). This can be interpreted as the pseudogap.\(^{15}\)

### D. Doping dependence of Josephson critical current density

As shown in Figs. 3(b)–3(d), since the Josephson critical current is homogenous within the stacked IJJs, \(J_c\) can measure bulk superconducting properties in addition to \(\Delta\). Here, the doping dependences of \(J_c\) and the superconducting gap structures of BiPb2212 and Bi2212 are compared. As already discussed, the Josephson critical current density \(J_c^{AB}\) in an \(s\)-wave superconductor at a low temperature is given by Eq. (1).\(^{14}\) Since the \(J_c^{AB}\) differs between \(s\) wave and \(d\) wave by an order of unity,\(^{33}\) Eq. (1) is applicable to our analysis. In
FIG. 3. (Color online) Temperature dependence of the $c$-axis resistivity $\rho_c$ (a) and $I$-$V$ characteristics obtained by an analog oscilloscope at 5 K (b)–(d) for the mesa samples.

The present study, $J_c$, $R_N$, and $\Delta$ are independently determined from Figs. 3(b)–3(d), 4(a)–4(c), and 4(d)–4(f), respectively. Figure 5 shows how $J^\text{AB}_c$, $J_c$, and the ratio $J_c/J^\text{AB}_c$ vary with $p$ in both Bi2212 and BiPb2212. Previous experiment has shown that $J_c$ in underdoped Bi2212 is more than an order of magnitude smaller than $J^\text{AB}_c$. It is also known that as $p$ decreases, $J_c$ becomes more suppressed. For instance, $J_c/J^\text{AB}_c$ decreases from 0.067 to 0.027 as $p$ decreases from 0.14 to 0.10. A similar result is obtained for BiPb2212, although the magnitudes of both $J^\text{AB}_c$ and $J_c$ are larger than those of Bi2212 due to the lowered $R_N$ values (relative to Bi2212). However, the extent of the suppression is quantitatively smaller in the

FIG. 4. (Color online) $I$-$V$ curves measured by short-pulse ITS (a)–(c) and tunneling spectra (d)–(f) for the mesa samples. In this figure, the horizontal axis is normalized to the single junction voltage by dividing the observed voltage by the number of junctions.
INCREASE OF SUPERFLUID DENSITY WITH GROWTH OF... PHYSICAL REVIEW B 87, 214521 (2013)

FIG. 5. (Color online) Plots for $J_c$, $J_c^{AB}$, and $J_c/J_c^{AB}$ as a function of $p$ for BiPb2212 (this work) and Bi2212 (Ref. 15).

present case than that for Bi2212. For example, around $p = 0.10$, $J_c/J_c^{AB}$ is 0.027 and 0.055 for Bi2212 and BiPb2212, respectively. Since the change in $J_c/J_c^{AB}$ is not explained by the reduced $R_N$, the Pb substitution must enhance the superfluid density by a factor of 2.

In our previous ITS studies on Bi2212, we posited that such anomalous suppressions of $J_c$ result from inhomogeneous superconductivity. The Josephson current flows only when both junction electrodes are in the superconducting (SC) state. When this is not the case, the junction is resistive and no Josephson current flows. If the SC state is inhomogeneous, the Josephson current will flow in some regions but not in others. When a sheet of the CuO$_2$ plane appears randomly fractionalized into SC and nonsuperconducting (NSC) regions, the $J_c$ decreases according to the degree of fractionalization. This model is supported by microscopic spatial inhomogeneity of the $\Delta$ values on the surface $ab$ planes of pristine Bi2212 crystals, revealed by STS experiments. Similar inhomogeneity has been reported in the $\Delta$ of BiPb2212. The standard deviation of the $\Delta$ distribution is very similar in BiPb2212 and Bi2212. According to the real space inhomogeneity model, the extent of $J_c$ suppression is mirrored in the spatial distribution of SC and NSC regions. However, our results showed that the suppression is significantly weakened in BiPb2212. Thus we argue that inhomogeneous superconductivity in real space cannot fully account for the increased $J_c/J_c^{AB}$ in BiPb2212.

We suggest instead that the Pb substitution increases the length of the Fermi arc, which weakens the extent of $J_c$ suppression. This suggestion is justified by a model that explains the anomalous suppression of $J_c$ in underdoped Bi2212, in terms of $k$-space inhomogeneity in the superconducting gap. This $k$-space inhomogeneity model predicts that superfluid density and $J_c$ in a $d$-wave superconductor drastically decrease when the $\Delta(k)$ around the antinodal, $(\pi, 0)$ and $(0, \pi)$, directions in the $k$ space disappears. ARPES experiments have revealed that $\Delta$ grows in regions of $k$ space where the Fermi arc exists. As $p$ decreases, the Fermi arc shrinks in the nodal ($\pi, \pi$) direction. This model sufficiently explains the anomalous $J_c$ suppression in underdoped Bi2212. According to the $k$-space inhomogeneity model, the enlarged $J_c/J_c^{AB}$ found in BiPb2212 implies a longer Fermi arc for BiPb2212 than that for Bi2212. Suzuki et al. found that the length of Fermi arc influences the observed tunneling spectra; as Fermi arc increases, the heights of superconducting peaks increase and the dip-hump characteristic becomes less pronounced. Figure 6 shows the comparison between normalized tunneling spectra of a Bi2212 sample (black line; the sample C in Ref. 15) and a BiPb2212 sample (red-dotted line; the sample M-2 in this work) with close $p$. Each tunneling spectrum is normalized by dividing each spectrum by each value at $V = 150$ mV because the values at high voltage correspond to the density of states in the normal states.

![Normalized tunneling spectra comparison](image)

FIG. 6. (Color online) Comparison between normalized tunneling spectra of a Bi2212 sample (black line; the sample C in Ref. 15) and a BiPb2212 sample (red-dotted line; the sample M-2 in this work) with close $p$. Each tunneling spectrum is normalized by dividing each spectrum by each value at $V = 150$ mV because the values at high voltage correspond to the density of states in the normal states.

IV. CONCLUSIONS

We have analyzed the BiPb2212 system in the underdoped region by intrinsic tunneling spectroscopy. The $\Delta$ are identical to those of Bi2212, although the $J_c$’s are significantly higher. The deviation of $J_c$ from $J_c^{AB}$ was found to be smaller than that for Bi2212. By comparing the tunneling spectra between BiPb2212 and Bi2212 with the numerical results, we conclude that the observed $J_c$ increase in Pb-substituted Bi cuprates results from not merely the reduced tunnel barrier but also elongation of the Fermi arc in the $k$ space.
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