Origin of positive out-of-plane magnetoconductivity in overdoped Bi$_{1.8}$Pb$_{0.4}$Sr$_2$CaCu$_{1.96}$Fe$_{0.04}$O$_{8+\delta}$

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I. INTRODUCTION

To determine the mechanism of high-$T_c$ superconductivity, we must understand the relationship between the pseudogap and superconductivity [1]. To date, different classes of theoretical models for high-$T_c$ superconductivity have proposed different pseudogap phase diagrams [2]. That is, if the pseudogap is a necessary ingredient for pairing, the pseudogap opening temperature $T^*$ may merge with $T_c$ in the overdoped state, whereas if the pseudogap is of a competing order, $T^*$ may cross the $T_c$-“dome” near the optimal doping. The difference especially appears in the overdoped region; hence, it is important to investigate this region. However, it is difficult to determine whether the observed effect is related to the pseudogap or the superconductivity when $T^*$ is close to $T_c$.

It is well known that $\rho_c(T)$ shows a typical upturn [3] and a negative $\rho_c(H)$ slope [4, 5] at high fields below $T^*$. Because $\rho_c$ probes the electronic density-of-states (DOS) around the Fermi level, reflecting the tunneling nature between CuO$_2$ planes in high-$T_c$ cuprates, the semiconductive upturn in $\rho_c(T)$ is attributed to the decrease of DOS caused by the pseudogap opening, and the negative $\rho_c(H)$ slope is attributed to the DOS recovery along with the suppression of the pseudogap under magnetic fields. These facts are well established, and thus, we can usually estimate $T^*$ by the $\rho_c$ behavior.

On the other hand, because superconductivity is also a phenomenon connected to the energy gap, there have been proposals from both experimental [6-8] and theoretical [9, 10] points of view that superconductivity causes similar effects on $\rho_c$ in a temperature and magnetic field range in which the DOS effect dominates over the Cooper pair tunneling effect. In such cases, it is difficult to distinguish $T^*$ from the onset temperature of the superconducting fluctuation $T_{scf}$. However, this interpretation for the superconductivity-originated anomalous $\rho_c$ behavior has not been generally accepted.

To address this issue, we focus on the magnetic field dependence of $\rho_c$ up to high fields, especially in the superconducting fluctuation regime. In general, the pseudogap is less sensitive to magnetic fields than superconductivity [11]. Therefore, if we can obtain a high enough field near the upper critical field $H_{c2}$, we can expect to observe the pseudogap and superconducting contributions separately for the magnetic field dependence of $\rho_c$. Hence, we measure $\rho_c(T, H)$ of Fe-substituted overdoped Bi-2212 under high pulsed magnetic fields up to 60 T. For comparison, we also measure $\rho_{ab}(T, H)$ of the same sample.

II. EXPERIMENT

Single crystals were grown in air using the traveling-solvent floating zone (TSFZ) method. The Bi site was partially substituted by Pb to overdope the sample. The Cu site was also substituted by 2% Fe to reduce $T_c$. The crystals were annealed under flowing oxygen at 400°C for 50 h to promote hole doping ($T_c = 50$ K, $p = 0.22$). In this study, $T_c$ was determined by the onset of zero resistivity. The doping level ($p$) was obtained using the empirical relation [12] with maximum $T_c = 71$ K for this sample.

The in-plane resistivity $\rho_{ab}(T, H)$ as well as the $\rho_c(T, H)$ measurements were performed using the DC four-terminal method [13]. Two types of magnets were used: the magnet at the Institute for Materials Research (IMR), Tohoku University, which provides steady magnetic fields up to 17.5 T, and the nondestructive pulsed magnet at the Institute for Solid State Physics (ISSP), University of Tokyo, which provides pulsed magnetic fields (36-ms pulse duration) up to 60 T. Magnetic fields were applied parallel to the c-axis. In the pulsed magnet measurements, we did not observe hysteresis behavior in high magnetic fields above the peak field $H_{peak}$ at which $\rho_c(H)$ is maximum. Furthermore, all down-ramped data coincided for measurements under several maximal fields, suggesting that the data were obtained with minimal eddy current influence.
In the following section, we will first estimate \( T^* \) and \( T_{scf} \) of this sample using the temperature dependence of \( \rho_c \) and \( \rho_{ab} \), respectively. Then, we will show that the out-of-plane magnetoconductivity (MC) is composed of two positive components below \( T_{scf} \). This two component analysis enables us to estimate \( H_{c2} \) at several temperatures. We will also estimate \( H_{c2} \) from the magnetic field dependence of \( \rho_{ab} \) in a standard manner. Consequently, an experimental \( H-T \) phase diagram of this compound will be used to validate the analysis. Finally, we will discuss the implications of the obtained results for the pseudogap phase diagram.

III. RESULTS AND DISCUSSION

Figure 1(a) shows \( \rho_c(T) \) for a \( \text{Bi}_{1.8}\text{Pb}_{0.2}\text{Sr}_2\text{CaCu}_1.96\text{Fe}_{0.04}\text{O}_{8+\delta} \) crystal with and without a steady 17.5-T magnetic field. For zero field, \( \rho_c(T) \) is metallic \((d\rho_c(T)/dT>0)\) over a wide temperature region, showing a slight upward trend with decreasing temperature. From this result, we estimate the pseudogap opening temperature \( T^* \) to be 166 K. Here, \( T^* \) is defined as the temperature at which \( \rho_c \) increases by 1% from that for high-temperature linear behavior [5][13][14].

Figure 1(b) shows the temperature dependence of the out-of-plane magnetoresistance (MR) for several magnetic fields \( H \). A small negative MR is observed below 170 K, and the magnitude increases with increasing magnetic field. This negative MR confirms that the pseudogap opens below this temperature [4][7]. However, for a zero field, \( \rho_c(T) \) shows a steep increase near \( T_c \) (Fig. 1(a)). When a 17.5-T magnetic field is applied, \( \rho_c(T) \) increases below \( T_c \), showing a sharp maximum around 30 K and then decreases to zero (Fig. 1(a)). The inset of Fig. 1(a) shows an enlarged view of the out-of-plane MR at 17.5 T. The small negative MR starting below \( T^* \) rapidly increases near \( T_c \). These results suggest that the superconducting DOS fluctuation effect is added to the pseudogap effect below the onset temperature of the superconducting fluctuation \( T_{scf} \) [13].

To estimate \( T_{scf} \), \( \rho_{ab}(T) \) was measured with and without a steady 17.5-T magnetic field (Fig. 1(c)). The upward curvature for the temperature dependence is evident, further indicating that the sample is in an overdoped state [13][14]. However, a small negative MR, whose origin is unknown, was observed above \( T_c \) (details will be published elsewhere). Therefore, to estimate \( T_{scf} \), we cannot use our standard method of determining the onset temperature for the rapid increase of positive MR [13]. Instead, the temperature derivative \( d\rho_{ab}/dT \) is compared for a zero field and a 17.5-T field (inset of Fig. 1(c)). The difference in \( d\rho_{ab}/dT \) appears below 70–75 K. Because the Aslamazov-Larkin (AL)-type superconducting fluctuation effect (paraconductivity) is expected to affect the slope of \( \rho_{ab}(T) \) below \( T_{scf} \), the onset temperature for the change may be assigned as \( T_{scf} \). Note that \( T^* \) is far greater than \( T_{scf} \), even in this overdoped state (\( \rho = 0.22 \) [15]).

Figure 2(a) shows the high pulsed magnetic field data for \( \rho_c(H) \) above and below \( T_c \). At 1.4 K, \( \rho_c(H) \) remains at zero up to 55 T, indicating that \( H_{c2} \) is above 55 T. At other temperatures below \( T_c \), \( \rho_c(H) \) shows a typical peak structure; it changes from zero to a resistive state at a vortex melting field, \( H_m \), reaching a maximum at \( H_{peak} \) before decreasing to a constant value. This peak structure in \( \rho_c(H) \) can be understood by parallel two-channel tunneling conductivity [8]: Cooper pair tunneling, \( \sigma_{c}^{Cooper}(H) \), and quasiparticle tunneling, \( \sigma_{qp}^{c}(H) \). That is, \( \rho_c(H) = 1/(\sigma_{c}^{Cooper}(H) + \sigma_{qp}^{c}(H)) \). The behavior under weak magnetic fields is controlled by \( \sigma_{c}^{Cooper}(H) \), which arises due to phase slips between neighboring \( \text{CuO}_2 \) planes caused by the motion of mobile pancake vortices, and the Josephson contribution decreases with increasing field strength [17]. However, \( \sigma_{qp}^{c}(H) \) increases under high magnetic fields, causing a negative \( \rho_c(H) \) slope, and \( H_{peak} \) is a crossover field for both contributions. In the following section, we concentrate on the analysis of the negative \( \rho_c(H) \) slope.

We consider \( \sigma_{c}(H) \) above \( T_c \). Figures 2(b)–(c) show the
FIG. 2. (Color online) (a) Magnetic field dependence of $\rho_c$ for Bi$_{1.8}$Pb$_{0.2}$Sr$_2$CaCu$_{2.96}$Fe$_{0.04}$O$_{8+\delta}$ below and above $T_c$. The arrows indicate the peak field $H_{peak}$, and the vortex melting field $H_m$. $H_m$ is defined as a field in which $\rho_c = 0.01\rho_c^0$. (b)–(c) Out-of-plane magnetoconductivity (MC), $\sigma_c(H) - \sigma_c(0)$, at 50 and 60 K, respectively. In (b), the normal state $\sigma_c(0)$ was assumed as an extrapolated value to the zero field of the high-field data. (d) Out-of-plane MC above 65 K for steady magnetic fields. (b)–(d) The solid lines are linear extrapolations of MC at low and high fields. Here, $H_{c2}$ is defined as the field at which MC deviates 1% as an average from these lines. (e) Schematic representation for the two component analysis used above $T_c$. MC, $\Delta\sigma_c(H) = \sigma_c(H) - \sigma_c(0)$, as a function of $H^2$, at 50 and 60 K, respectively. Above 65 K, we additionally measure the MC using a steady magnet up to 16 T (Fig. 2(d)). The MC rapidly increases with increasing magnetic fields, approaching $\propto H^2$ for higher fields. With increasing temperature, the field interval MC, which follows $\propto H^2$, extends to lower fields, and the slope, $a$, decreases. These data imply that the MC comprises two positive components: one component gradually increases with $aH^2$ (hereafter, we denote this component as $\Delta\sigma_c^{PG}$), and the other component rapidly increases with increasing magnetic field but tends to saturate at higher fields (hereafter, we denote this component as $\Delta\sigma_c^{SC-DOS}$). This two component analysis is schematically shown in Fig. 2(e). Figure 2(d) shows that the $H^2$ component, $\Delta\sigma_c^{PG}$, is present both below and above $T_{scf}$, while the component with a different (faster) $H$-dependence, $\Delta\sigma_c^{SC-DOS}$, decreases with increasing temperature and vanishes around $T_{scf}$ ($= 75$ K). Thus, the former component ($\Delta\sigma_c^{PG}$) is attributed to the pseudogap effect, and the latter component ($\Delta\sigma_c^{SC-DOS}$) is attributed to the superconducting DOS depletion effect. This result verifies that the positive superconducting contribution in MC appears below $T_{scf}$. Furthermore, the observation of $H^2$ MC at high temperatures above $T_{scf}$ (when superconducting fluctuations are presumably gone) justifies the following analysis in Figs. 2(b)–(d), which separate the pseudogap components from its $H^2$ dependence.

This two component analysis enables us to estimate the weight of the superconducting contribution, $W_{SC}$, for the total quasiparticle tunneling MC. The saturated value of $\Delta\sigma_c^{SC-DOS}$ is given by the $y$-intercepts, $A$, of the high field $H^2$-linear be-
havior as shown in Fig. 2(e). The total quasiparticle contributions, $\Delta\sigma^{qp}_c = \Delta\sigma^{SC-DOS}_c + \Delta\sigma^{PG}_c$, are given by $B$ of the figure. Then, we can estimate $W_{SC} = \Delta\sigma^{SC-DOS}_c / (\Delta\sigma^{SC-DOS}_c + \Delta\sigma^{PG}_c)$, by $A/B$. At 50 K, $W_{SC}$ is estimated as 66% at 55 T. The estimated value is reliable in that it can be obtained directly from the raw MC data. Considering that the data are obtained at $T_c$, this value is fairly large. Furthermore, the field at which $\Delta\sigma^{SC-DOS}_c$ saturates is used to determine $H_{c2}$. Thus, $H_{c2}$ is estimated as 41, 22, 13, 12, and 10 T at 50, 60, 65, 70, and 75 K, respectively. However, $\Delta\sigma^{PG}_c$ does not saturate even under high fields up to 55 T. This indicates that the field needed to close the pseudogap, $H_{pg}$ [4], is very high (far greater than 55 T).

Next, we consider $\sigma_c(H)$ below $T_c$. Here, the $\sigma_c(H)$ behavior [positive $\sigma_c(H)$ slope] above $T_c$ is continuously observed above $H_{peak}$. However, in this case, as MC does not exhibit $H^2$ behavior for whole fields, it is difficult to estimate the pseudogap contribution directly from the MC data. Therefore, we assume it from the data above $T_c$. Figure 3(a) shows the $a$ values obtained in Figs. 2(b)–(d) for the pseudogap contribution above $T_c$. The accuracy of the data is not sufficient to determine the precise functional form (either sublinear or superlinear) for the temperature dependence of $a$. Thus, the $a$ value near $T_c$ was simply assumed to be linear in temperature. Note that this is an approximation near $T_c$ to estimate $a$ in the superconducting state. At higher temperatures ($\geq 80$ K), the slope becomes smaller, and the $a$ value approaches zero at around $T^* = 170$ K. Figure 3(b) and (c) show the magnetic field dependence of $d\sigma_c/dH$ at 40 and 30 K, respectively. To determine only the superconducting contribution to the data, the above pseudogap contribution is subtracted from the raw data. We find that the pseudogap contribution is not very large. In this case, $H_{c2}$ may correspond to the field at which the superconducting $d\sigma_c/dH$ is extrapolated to zero. Thus, $H_{c2}$ is estimated as 51 and 60 T for 40 and 30 K, respectively.

We then investigate the weight of superconducting contribution $W_{SC}$ below $T_c$. For this purpose, the magnetic field dependences of $\sigma_c$ and $\sigma_c - \Delta\sigma^{PG}_c$ are plotted in Figs. 3(d)–(f) at 40, 30, and 20 K, respectively. At the highest field (55 T), we consider $\sigma_c$ as $\sigma_c^{pp}$ because $\sigma_c^{Cooper}$ may be negligible. Then, $\sigma_c - \Delta\sigma^{PG}_c$ is expressed as $\Delta\sigma^{SC-DOS}_c + \sigma_c^{pp}(0T)$. Assuming that $\sigma_c^{pp}(0T)$ at 20 K is negligible, the result in Fig. 3(f) implies that $W_{SC}$ accounts for $\approx 80$% of the total MC. This indicates that the positive slope for the MC primarily originates from superconductivity.

Figure 4 shows the high pulsed magnetic field data for $\rho_{ab}(H)$ at several temperatures above and below $T_c$. With increasing magnetic fields, $\rho_{ab}(H)$ changes from zero to a resistive state at $H_m$ and rapidly increases in a similar manner as $\rho_c(H)$ (Fig. 2(a)). Then it gradually approaches a constant value, keeping a positive slope. This in-plane behavior contrasts the out-of-plane behavior, in which $\rho_c(H)$ shows a typical negative slope at higher fields, indicating that, in Fig. 2(a), we have succeeded in detecting a property peculiar to $\rho_c(H)$. The superconducting fluctuation theory [19] including the DOS contribution in highly anisotropic materials predicts that, for the out-of-plane conduction, the negative DOS contribution dominates over the positive Aslamazov-Larkin (AL) contribution near $H_{c2}$, resulting in a negative slope for $\rho_c(H)$. For the in-plane conduction, the AL contribution dominates, causing the positive slope for $\rho_{ab}(H)$. The experimental observations in this study are consistent with this theory. Then, it may be natural to ascribe the origin of the negative $\rho_c(H)$ slope (positive slope for the out-of-plane MC) primarily to the superconducting DOS fluctuation effect. Supposing that $\rho_{ab}(H)$ is constant above $H_{c2}$, $H_{c2}$ is estimated as 21, 39 and 48 T for 60, 50 and 40 K, respectively. These values agree with those obtained by $\rho_c(H)$ measurements, supporting the validity of the above analysis for $\rho_c(H)$.

Figure 5 shows $H_m$, $H_{peak}$, and $H_{c2}$ obtained by $\rho_c(H)$ measurements as well as $H_{c2}$ obtained by $\rho_{ab}(H)$ measurements as a function of temperature. In highly two-dimensional (2D) superconductors such as high-$T_c$ cuprates, the vortex system transforms from a liquid state to a 2D solid state upon cooling [13]. The transition temperature is represented as $T_m^p$. Here, the field at which the vortex melting line extrapolates to 0 K is simply considered to be $H_{c2}(0)$ at $T = 0$. To estimate $H_{c2}(0)$, we adopted the 2D vortex solid state formula, $H_{c2} = (c^2_l/2\Delta)^{1/2}(1 - H/H_{c2}(0))^2$ [18]. Here, we tentatively used the same parameter values, namely the Ginzburg number $\Delta = 0.05$ and the Lindemann number $c_l = 0.14$, as in the previous report [5] with the exception of $T_c (= 50$ K). The fitting to the experimental data is very good between 4.2 K to 15 K, probably better than that of ref. [5], and the fitting gives $H_{c2}(0) = 70$ T. The extrapolation of $H_{peak}$ to 0 K gives $H_{peak} = 69$ T; here, we assumed $H_{peak}$ at low temperatures is exponential in $T$ [4]. On the other hand, $H_{c2}(0)$ is directly estimated as 70 T by fitting the obtained $H_{c2}(T)$ using the empirical formula $H_{c2}(T) = H_{c2}(0)(1 - (T/T_c)^2$); here, we assumed the $T_{c^{f}}$ value ( = 75 K) for $T_c$. All three superconductivity-related characteristic fields roughly coincide at $T = 0$, indicating that the obtained $H_{c2}(T)$ contain information on the upper critical field. Hence, the above procedure (two component analysis in MC) for estimating $H_{c2}(T)$ from $\rho_c(H)$ data should be appropriate. In superconductors with strong fluctuation, the mean-field upper critical field, $H_0$, and the mean-field superconducting transition temperature in $H = 0$, $T_0$, are shown to be the onset of superconducting fluctuation [19](see Fig. 3 in ref. [19]). We consider that $H_{c2}$ and $H_{c^{f}}$ in this study correspond to $H_0$ and $T_0$, respectively.

In a pioneering report [5], an unusually large quantum-dissipative pseudogapped state was shown to exist near $T = 0$. However, we did not observe such behavior in our sample (Fig. 5). Note that $H_m$ is not observed until 55 T at 1.4 K (Fig. 2(a) and Fig. 4). We assume that our sample acquired a strong vortex pinning effect through the co-doping of Pb and Fe, and thus, such anomalous behavior has been suppressed compared to that in the pristine Bi-2212 sample of ref. [5].

Finally, we briefly discuss the implication of our result on the pseudogap problem. In this study, we have revealed that the superconducting contribution to the positive slope for the
out-of-plane MC evolves below $T_{\text{scf}}$ in addition to the pseudogap contribution. This result, on one hand, confirms that the pseudogap and superconductivity are distinct, as many authors suggest [20–22]. On the other hand, this result verifies that superconductivity is another cause for the anomalous $\rho_c(T, H)$ behavior. Recently, we have observed a pseudogap-like upturn and negative MR in $\rho_c(T, H)$ just above $T_c$ for heavily overdoped Bi-2212 ($p = 0.23$) [13]. Thus, this behavior should be attributed to superconductivity; the pseudogap does not open above $T_c$ in this doping state. Therefore, if we could know whether or not the pseudogap opens below $T_c$ in this heavily overdoped sample, we would understand the pseudogap phase diagram completely.

IV. CONCLUSIONS

In summary, to determine the origin of the positive slope in interlayer MC in high-$T_c$ cuprates, $\rho_c(T, H)$ as well as $\rho_{ab}(T, H)$ measurements under high pulsed magnetic fields were performed on Pb and Fe co-doped Bi-2212. The results not only establish that the positive MC and the correlated upturn for $\rho_c(T)$ near $T_c$ are a sign of superconductivity but also enable us to estimate $H_{c2}$ and the superconducting contribution on the positive MC quantitatively. This finding may pro-
vide an important clue toward understanding the pseudogap phenomenon in high-$T_c$ superconductivity.

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