Specific-heat anomaly of overdoped $Bi_{1.8}Pb_{0.3}Sr_{1.9}CuO_y$

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The temperature dependence of the specific heat of $Bi_{1.8}Pb_{0.3}Sr_{1.9}CuO_y$ samples in the overdoped region has been measured between 1.5 and 8 K. Specific-heat anomalies were observed at T_c in two superconducting samples, while no anomalies were observed in a nonsuperconducting one. The values of $\Delta C/\gamma T_c$ were quite small in comparison to the BCS value, and the electronic specific heat in the superconducting state near T_c exhibited a weak temperature dependence. These findings are consistent with the impurity effects predicted by the model of d-wave superconductors proposed by Sun and Maki. Furthermore, we found that the Sommerfeld constant γ increases with increasing doped carrier from the superconducting region to the normal metal region, in contrast to the case of $La_{2-x}Sr_xCuO_4$. [S0163-1829(98)04813-9]

I. INTRODUCTION

When clarifying the mechanism of high-T_c cuprates, the symmetry of Cooper paring must be determined. Several different experiments involving angle-resolved photoemission,[1,2] the tricrystal superconducting ring,[3] NMR,[4] and the specific heat[5,6] have revealed that high-T_c cuprates behave as d-wave superconductors. Theories of d-wave superconductors[7–11] predict that the temperature dependence of the electronic specific heat C_{el} is proportional to T^2 at $T \ll T_c$. Furthermore, Maki and Won[12] have shown that as the impurity content increases, the temperature dependence of C_{el} changes from the T^2 type to the $(aT+bT^3)$ type. This behavior is quite different from the exponential dependence of C_{el} presented by BCS theory. In fact, Momono *et al.*[5,6] have observed that the temperature dependence of the C_{el} of $La_{2-x}Sr_xCu_{1-y}Zn_yO_4$ is proportional to T^2 at $T \ll T_c$ for low concentrations of Zn, while for high concentrations of Zn, the temperature dependence of C_{el} is described well by the sum of the aT and bT^3 terms. Thus the features of the electronic specific heat at $T \ll T_c$ have been theoretically and experimentally clarified.

Compared to the behavior of C_{el} at $T \ll T_c$ in d-wave superconductors, few studies have examined the behavior of C_{el} near T_c. Sun and Maki[10] have shown theoretically that for the C_{el} near T_c for d-wave superconductors, the value of the specific-heat anomaly at T_c, $\Delta C/\gamma T_c$, is significantly smaller than the BCS value ($\Delta C/\gamma T_c = 1.43$) and decreases markedly with increasing impurity content. Here ΔC is the jump in C_{el} at T_c and γ is the Sommerfeld constant. The measurement of ΔC is also important for the determination of the pairing symmetry and plays a significant role in the phase transition from the pseudogap state to the superconducting state. The existence of the pseudogap is well known in high-T_c cuprates,[2,13] especially in the underdoped region, where the formation of the pseudogap starts at a temperature well above T_c, increases in magnitude with decreasing temperature, and then finally agrees with the superconducting gap at T_c.[2] Therefore, the finite superconducting gap is already open at T_c, which is clearly different from the normal superconducting transition. To investigate the specific-heat anomaly at the superconducting transition from the pseudogap state, we first need to clarify the behavior of the jump in C_{el} at the superconducting transition from the normal state without the pseudogap for the d-wave superconductor.

To determine experimentally the value of $\Delta C/\gamma T_c$ for high-T_c cuprates, it is important to use samples with low T_c (~ 10 K), because in superconductors with a relatively high T_c of above 30 K, a contribution of the electronic term to the whole specific heat is so small that it is difficult to simultaneously determine ΔC and γ. Among high-T_c cuprates, the $Bi_2Sr_2CuO_y$ system is known to undergo the superconducting transition at low temperature ($T_c \sim 10$ K). So far, several groups[14–16] have reported the temperature dependence of the specific heat through the T_c of $Bi_2Sr_2CuO_y$ systems. Yamada *et al.*[14] and Yu and Frank[15] have observed no anomalies in the specific heat at T_c, while Nyeanchi *et al.*[16] have observed a significantly large jump in the specific heat at T_c. Since some contradictions exist among their results, the value of $\Delta C/\gamma T_c$ has not yet been experimentally defined.

In the present paper, we report the specific-heat anomalies observed at T_c in the overdoped region of $Bi_{1.8}Pb_{0.3}Sr_{1.9}CuO_y$ where the pseudogap is not formed above T_c. The values of $\Delta C/\gamma T_c$ were significantly smaller and the temperature dependence of the C_{el} below T_c was markedly weaker compared with those observed in the BCS superconductor. These results are consistent with the effect of impurities predicted in the model of d-wave superconductors proposed by Sun and Maki.[10] Using the specific-heat anomalies at T_c and the temperature dependence of C_{el} in the superconducting state near T_c, we show that the superconducting $Bi_{1.8}Pb_{0.3}Sr_{1.9}CuO_y$ in the overdoped region is a d-wave superconductor. In addition, the relationship between T_c, γ, and carrier concentration in the overdoped region of $Bi_{1.8}Pb_{0.3}Sr_{1.9}CuO_y$ behaves differently from that of the $La_{2-x}Sr_xCuO_4$ system.[5]

II. EXPERIMENTS

The $Bi_{1.8}Pb_{0.3}Sr_{1.9}CuO_y$ sample was prepared from Bi_2O_3, PbO_2, $SrCO_3$, and CuO powders of 99.99–99.999 % purity using a solid state reaction. The powders were mixed for 2 h

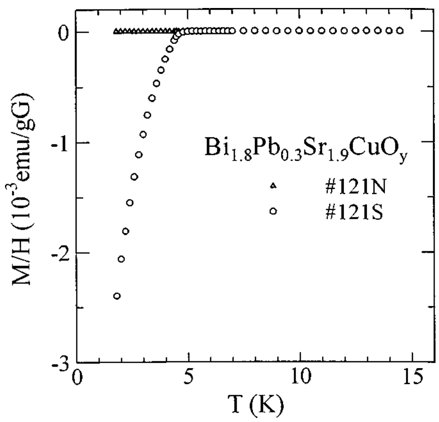

FIG. 1. Temperature dependence of the magnetic susceptibility for $Bi_{1.8}Pb_{0.3}Sr_{1.9}CuO_y$ superconducting (No. 121*S*, open circle) and nonsuperconducting (No. 121*N*, open triangle) samples.

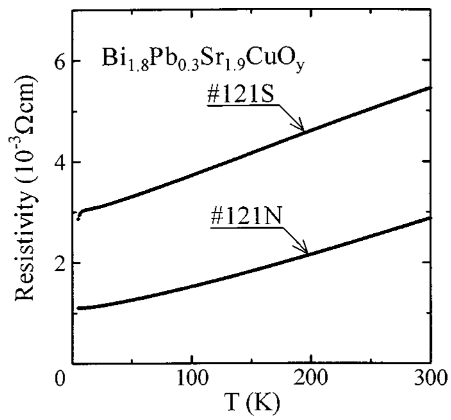

FIG. 2. Temperature dependence of the resistivity for $Bi_{1.8}Pb_{0.3}Sr_{1.9}CuO_y$ superconducting (No. 121*S*) and nonsuperconducting (No. 121*N*) samples.

and calcined at 815 °C for 8 h. The reacted material was ground and pressed into pellets 13 mm in diameter and 1 mm thick. The pellets were finally sintered to form the $Bi_{1.8}Pb_{0.3}Sr_{1.9}CuO_y$ single phase at 840 °C for 30 h in air. The formation of the single phase was confirmed by x-ray diffraction with Cu $K\alpha$ radiation. The resistivity measurement showed that the sintered sample was metallic, but not superconducting (nonsuperconducting sample). The superconducting sample was made from the nonsuperconducting sample by annealing at 450 °C for 700 h in an argon atmosphere. The superconductivity was determined by magnetic and electrical measurement. The magnetic susceptibility was measured between 1.5 and 15 K under a magnetic field of 0.5 G using a superconducting quantum interference device (SQUID) susceptometer. The resistivity was measured between 5 and 300 K using the four-probe method. The specific heat was measured by the thermal relaxation method in the temperature range from 1.5 to 8 K. The quantity of the samples used in the specific-heat measurement was about 15 mg.

III. RESULTS AND DISCUSSION

The formation of the single phase was confirmed by x-ray diffraction. The lattice constants ($a=5.382$ Å, $b=5.326$ Å, $c=24.627$ Å) of the nonsuperconducting sample agreed with those previously reported.[17]

Figure 1 shows the temperature dependence of the magnetization of $Bi_{1.8}Pb_{0.3}Sr_{1.9}CuO_y$. The open triangle and circle correspond to the data of the sample sintered in air (No. 121*N* sample) and that of the sample annealed in an argon atmosphere (No. 121*S* sample), respectively. The Meissner signal observed in the No. 121*S* sample was about 20% of the ideal value, which ensures the bulk nature of the superconductivity. From the onset of the Meissner signal, T_c was determined to be 4.7 K. Conversely, there is no indication of superconductivity for the No. 121*N* sample. This result demonstrates that the No. 121*N* sample is nonsuperconducting in the overdoped region, because the superconducting No. 121*S* sample was prepared by a reduction of carriers from the No. 121*N* sample.

Figure 2 shows the temperature dependence of the resistivity of the No. 121*N* and No. 121*S* samples. The resistivity of the No. 121*N* sample decreases with decreasing temperature. No rapid decrease in resistivity is observed in the temperature region above 4.2 K. On the other hand, the resistivity of the No. 121*S* sample decreases rapidly near the temperature corresponding to the onset of the Meissner signal, although the temperature dependence of the resistivity in the high-temperature range is almost the same as that of the No. 121*N* sample. The temperature dependences of resistivity at low temperature for both samples are consistent with the result of the Meissner signal shown in Fig. 1. The temperature dependence of resistivity for the No. 121*N* sample shows negative curvature, which is a common feature for samples in the overdoped region.[18,19] In the present study, the temperature dependence of resistivity exhibits negative curvature, even for the No. 121*S* sample, although this dependence is weaker than that seen for the No. 121*N* sample. This indicates that the No. 121*S* sample remains in the overdoped region when the carrier is reduced from the No. 121*N* sample in the overdoped region. Thus both No. 121*N* and No. 121*S* samples are in the overdoped region.

Figure 3 shows the temperature dependence of the specific heat of the No. 121*N* sample in which C/T is plotted as a function of T^2. As temperature decreases, the specific heat decreases smoothly without any anomalies, but cannot be described as a straight line of $C/T=\gamma+\beta T^2$ as shown in Fig. 3(a). The deviation from the straight line is observed in both the low- and high-temperature ranges, indicating that the specific heat of the No. 121*N* sample cannot be simply represented by only two terms of the electronic specific heat (γT) and the Debye specific heat (βT^3).

Assuming that the deviation in the low-temperature range is Schottky-type specific heat (A/T^2) due to magnetic impurities and that the deviation in the high-temperature range is the contribution from anharmonic lattice vibration (δT^5), the specific heat of the No. 121*N* sample is represented well by the following equation:

$$C=\gamma T+\beta T^3+\delta T^5+A/T^2. \tag{1}$$

The values of each coefficient calculated by Eq. (1) are $\gamma=8.7$ (mJ/mol K^2), $\beta=1.8$ (mJ/mol K^4), $\delta=1.5\times 10^{-2}$ (mJ/mol K^6), and $A=31.1$ (mJ/mol K^{-1}). Using these values, the temperature dependence of each contribution to the

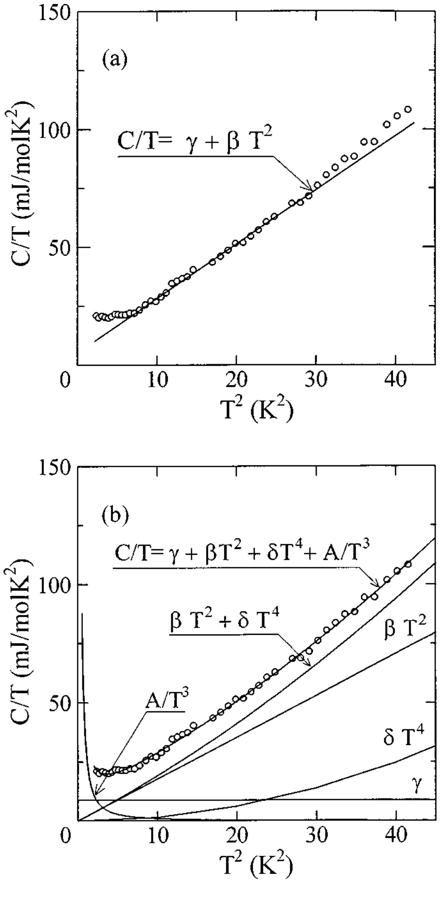

FIG. 3. Temperature dependence of the specific heat for a $Bi_{1.8}Pb_{0.3}Sr_{1.9}CuO_y$ nonsuperconducting sample (No. 121*N*) in a C/T vs T^2 plot. The solid lines are described by the function of $C/T=\gamma+\beta T^2$ (a) and by Eq. (1) (b).

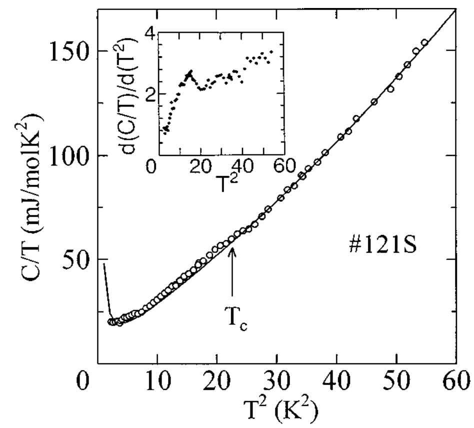

FIG. 5. Temperature dependence of the specific heat for a $Bi_{1.8}Pb_{0.3}Sr_{1.9}CuO_y$ superconducting sample (No. 121*S*). The inset shows the $d(C/T)/d(T^2)$ vs T^2 curve.

specific heat is calculated. In Fig. 3(b) the results are represented by the solid lines. We find that the contribution of the lattice specific heat ($\beta T^3+\delta T^5$) to the total is significantly larger than the others, especially in the high-temperature range above 5 K, where it is over 80%.

Figure 4 shows the temperature dependence of electronic specific heat C_{el}, which is obtained by subtracting the contributions of lattice specific heat and Schottky-type specific heat (A/T^2) from the total specific heat (C). The C_{el}/T obtained is constant within 8.7 ± 1.9 (mJ/mol K^2) over the whole temperature range measured. Wide scattering in the data of the electronic specific heat may be due to the large contribution of the lattice terms to the total specific heat.

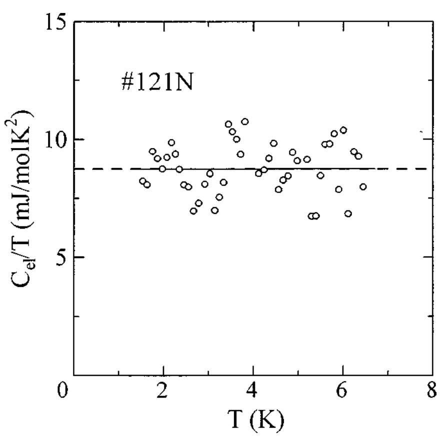

FIG. 4. Temperature dependence of the electronic specific heat for a $Bi_{1.8}Pb_{0.3}Sr_{1.9}CuO_y$ nonsuperconducting sample (No. 121*N*) in a C_{el}/T vs T^2 plot. The solid line is described by Eq. (1).

Figure 5 shows the temperature dependence of the specific heat of the superconducting sample (No. 121*S*) in which C/T is plotted as a function of T^2. As a whole, the C/T versus T^2 curve is similar to that obtained for the nonsuperconducting sample (No. 121*N*) except that a hump appeared around 4.5 K. In the inset of Fig. 5 in which $d(C/T)/d(T^2)$ is plotted as a function of T^2, a dip is clearly evident around $T^2=23$ K^2 and the corresponding temperature agrees with $T_c=4.7$ K. This indicates that the hump is the specific-heat anomaly due to the superconducting transition. The specific-heat anomaly with the hump is also observed in our other superconducting sample (No. 123*S*) with $T_c=4.8$ K. Our result of the specific-heat anomaly at T_c disagrees with the earlier works of Yamada *et al.*,[14] Yu and Frank,[15] and Nyeanchi *et al.*,[16] Yamada *et al.*,[14] and Yu and Frank[15] have reported that no anomaly in the specific heat appears at T_c in the $Bi_{2.1}Sr_{1.85}CuO_{6-\delta}$ and $Bi_{2.05}Pb_{0.05}Sr_{1.9}CuO_{6+\delta}$ systems, respectively, although the Meissner signal is observed at T_c. On the other hand, Nyeanchi *et al.*[16] have reported a significantly large jump in the specific heat at T_c in the $Bi_2Sr_2CuO_{6+\delta}$ system, whose value is one order larger than that observed here. These discrepancies will be discussed later in connection with the temperature dependence of the specific heat.

In order to obtain each contribution to the total specific heat, the temperature dependence of the specific heat was fitted using Eq. (1) except for the temperature range near T_c where the specific-heat anomaly appears. The values obtained were $\gamma=6.0$ (mJ/mol K^2), $\beta=2.1$ (mJ/mol K^4), $\delta=1.1\times10^{-2}$ (mJ/mol K^6), and $A=40.0$ (mJ/mol K^{-1}). It is worth comparing the values obtained here with those reported previously by Yamada *et al.*[14] and Yu and Frank,[15] because the temperature range measured is almost the same (from 1.5 to ca. 10 K) and the temperature dependence of C is well represented by Eq. (1) or by an equation of $C=\gamma T+\beta T^3+A/T^2$. Our value for A is less than half of their values, although values for γ, β, and δ are almost the same.

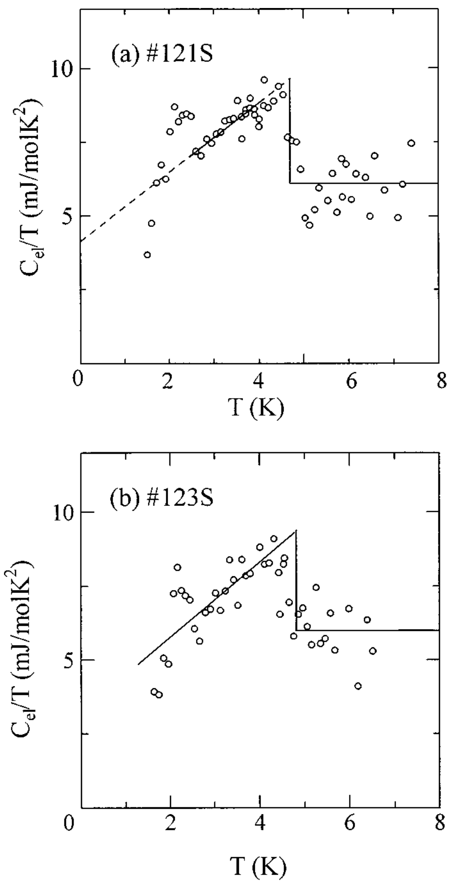

FIG. 6. Temperature dependence of the electronic specific heat for a $Bi_{1.8}Pb_{0.3}Sr_{1.9}CuO_y$ superconducting sample, No. 121*S* (a) and No. 123*S* (b). The values of $\Delta C/\gamma T_c$ are 0.60 and 0.55 for No. 121*S* and No. 123*S*, respectively.

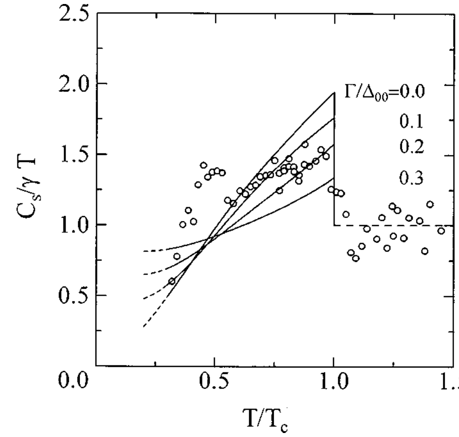

FIG. 7. Normalized temperature dependence of the ratio of the electronic specific heat in the superconducting state and γT for various concentrations of impurities. The solid lines represent the results calculated using the theory of Sun and Maki (Ref. 10). The values of $\Delta C/\gamma T_c$ are 0.95, 0.77, 0.57, and 0.34 for $\Gamma/\Delta_{00}=0$, 0.1, 0.2, and 0.3, respectively. The electronic specific heat for the $Bi_{1.8}Pb_{0.3}Sr_{1.9}CuO_y$ superconducting sample (No. 121*S*) is shown as open circles.

This suggests a small amount of magnetic impurity in our samples, which may have resulted in the appearance of the specific-heat anomaly at T_c. On the other hand, Nyeanchi *et al.*[16] have reported that the C measured from 0.1 to 5 K is represented well by the equation $C=aT^n+\beta T^3+A/T^2$ with $n=0.75$. However, because the temperature range measured and the temperature dependence of C in their study differ markedly from ours, no direct comparison can be made. At the present time, therefore, the reason for anomalous large jump in C at T_c reported by Nyeanchi *et al.*[16] remains unclear.

The C_{el} of the superconducting sample (No. 121*S*) was evaluated by subtracting the contributions of lattice and Schottky anomalies from the total specific heat, which is the same method used for the nonsuperconducting sample. Figure 6 shows the temperature dependence of C_{el}/T of No. 121*S*. The data show scattering due to a large contribution of lattice specific heat as in the case of the nonsuperconducting sample. However, we see that the C_{el}/T changes significantly around T_c; above T_c, C_{el}/T $(=\gamma)$ is constant within 6.0 ± 1.4 (mJ/mol K^2), but at T_c a rapid increase in C_{el} is observed. When the shape of the specific-heat anomaly at T_c is approximated as a step, the value of the specific-heat jump ΔC is estimated to be 17.2 (mJ/mol K) and, correspondingly, $\Delta C/\gamma T_c$ is 0.60. The value of ΔC is clearly larger than the scattering of C_{el} measured in the normal state. A similar result was obtained for our other superconducting sample (No. 123*S*) as shown in Fig. 6(b), in which the values for ΔC and $\Delta C/\gamma T_c$ are 15.6 (mJ/mol K) and 0.55, respectively. These results indicate that the anomaly in C_{el} observed here is due to the superconducting transition of $Bi_{1.8}Pb_{0.3}Sr_{1.9}CuO_y$. Notably, the value of $\Delta C/\gamma T_c$ obtained here is quite small, less than half of the BCS value (1.43).

The theory of d-wave superconductors proposed by Sun and Maki[10] can be used to help explain the small value of $\Delta C/\gamma T_c$. They investigated theoretically the relationship between $C_s/\gamma T_{c0}$ and T/T_{c0}, where C_s, γ, and T_{c0} are the electronic specific heat in the superconducting state, the Sommerfeld constant, and the superconducting transition temperature in the absence of impurities, respectively. Their result shows that the magnitudes of $\Delta C/\gamma T_c$ and T_c decrease remarkably with increasing impurities. The calculated value of $\Delta C/\gamma T_c$ is very small (0.95) even for a clean d-wave superconductor compared to the BCS value (1.43) and decreases to 0.5 for samples containing impurities with $\Gamma/\Delta_{00}=0.2$, where Γ is given by the ratio $[\Gamma=n_i/\pi N(0)]$ of the impurity concentration n_i and the density of states at the Fermi energy per spin, $N(0)$, and Δ_{00} is the energy gap at $T=0$ K for a clean d-wave superconductor. Our small value of $\Delta C/\gamma T_c$ $(=0.55\text{–}0.60)$ just lies on impurity concentrations of between $\Gamma/\Delta_{00}=0.1$ and 0.2. Since the Schottky anomaly observed in the specific heat suggests the existence of magnetic impurities in our sample, our experimental results are consistent with the theoretical ones for the impurity effects on d-wave superconductors proposed by Sun and Maki.[10]

To compare our experimental result on the temperature dependence of $C_{el}/\gamma T$ in the superconducting state with the theoretical behavior proposed by Sun and Maki, $C_{el}/\gamma T$ as a function of the normalized temperature T/T_c was recalculated for various impurity contents using the relationship between $C_s/\gamma T_{c0}$ and T/T_{c0} shown by Sun and Maki (see Fig. 5 in Ref. 10). In Fig. 7, the calculated results are shown with our experimental result and show that the temperature dependence of $C_s/\gamma T$ becomes weak with increasing impurity concentration. Our result near T_c agrees with the calculated one between $\Gamma/\Delta_{00}=0.1$ and 0.2, although it deviates from the theoretical results in the low-temperature region due to the influence of the Schottky anomaly. The temperature dependence of $C_{\mathrm{el}}/\gamma T$ in the superconducting state is also consistent with the impurity effects for d-wave superconductors proposed by Sun and Maki,[10] in addition to the result of the jump in C_{el} at T_c. Thus the jump in C_{el} at T_c and the temperature dependence of C_{el} near T_c as well as the temperature dependence of C_{el} at $T \ll T_c$,[5,6] which has been reported previously, argue for high-T_c cuprates being d-wave superconductors.

Next, we discuss the relationship between T_c, γ, and doped carrier content. According to the results obtained by Yamada *et al.*,[14] as the carrier content increases from the underdoped region to the overdoped one, T_c increases initially and then decreases; it shows the typical curve of T_c versus carrier in high-T_c cuprates, while γ increases smoothly with increasing carrier (see Figs. 6 and 8 in Ref. 14). In our case, as shown in Figs. 4 and 6, the value of γ of the superconducting sample is smaller than that of the nonsuperconducting sample. Since our superconducting sample in the overdoped region was produced by the reduction of carriers from the nonsuperconducting sample, γ increases with increasing doped carrier in contrast with the decrease in T_c. Thus we find that our result of the relationship between T_c, γ, and carrier content is in good agreement with that obtained by Yamada *et al.*[14] Therefore, in the nonsuperconducting overdoped region of $Bi_{1.8}Pb_{0.3}Sr_{1.9}CuO_y$, γ increases with increasing carrier content. This is inconsistent with the result[5] in the nonsuperconducting overdoped region of $La_{2-x}Sr_xCuO_4$, in which γ decreases with increasing carrier content. This suggests that the relationship between γ and carrier content is not given universally, at least in the overdoped region of high-T_c cuprates.

Finally, it should be emphasized that the specific-heat anomaly obtained here is due to the superconducting transition from the normal state, but not from the pseudogap state. If the superconducting transition occurs in the underdoped region where the pseudogap state is formed well above T_c, the behavior of the specific-heat anomaly at T_c might be different from the behavior predicted by the Sun-Maki theory[10] which applies to a d-wave superconductor without the pseudogap. It is widely accepted that understanding the pseudogap is important in relation to the mechanism of high-T_c cuprates. However, at present, the specific-heat anomaly at the superconducting transition from the pseudogap state is not sufficiently clear. We believe that the present result will give us a starting point to investigate superconductivity from the pseudogap state. Current research is focusing on the specific-heat measurement of $Bi_{1.8}Pb_{0.3}Sr_{1.9}CuO_y$ in the underdoped region.

IV. CONCLUSION

We have investigated the temperature dependence of the specific heat of superconducting and nonsuperconducting samples of $Bi_{1.8}Pb_{0.3}Sr_{1.9}CuO_y$ in the overdoped region. The temperature dependence of the specific heat of the nonsuperconducting sample is accurately presented as $C=\gamma T+\beta T^3+\delta T^5+A/T^2$, while in the superconducting sample, a specific-heat anomaly is observed at T_c, in addition to the temperature dependence of C seen for the nonsuperconducting sample. The magnitude of the specific-heat anomaly at T_c, $\Delta C/\gamma T_c$, is 0.55–0.60, which is less than half of the BCS value. The electronic specific heat in the superconducting state near T_c shows a weak temperature dependence compared with BCS behavior. The quite small value of $\Delta C/\gamma T_c$ and the weak temperature dependence of the electronic specific heat in the superconducting state are consistent with the effects of impurities predicted by the model of d-wave superconductors proposed by Sun and Maki. These findings demonstrate that superconducting $Bi_{1.8}Pb_{0.3}Sr_{1.9}CuO_y$ in the overdoped region is a d-wave superconductor.

The value of γ for the superconducting sample was smaller than that for the nonsuperconducting sample; that is, γ increased with increasing doped carrier, because the superconducting sample was prepared by the reduction of carrier content from the nonsuperconducting sample. The relationship between T_c, γ, and carrier concentration in the overdoped region of $Bi_{1.8}Pb_{0.3}Sr_{1.9}CuO_y$ shows behavior that contrasts with the $La_{2-x}Sr_xCuO_4$ system. This suggests that the relationship between T_c and carrier content is similar for many high-T_c cuprates, but that between γ and carrier content is not.

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