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Anomalous suppression of superconductivity in Zn-substituted $Bi_2Sr_2Ca_{1-x}Y_x(Cu_{1-y}Zn_y)_2O_{8+\delta}$

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We have found an anomalous suppression of superconductivity at $x=0.30$–0.35, where p (the hole concentration per Cu) $\sim$ 1/8, in the partially Zn-substituted compound $Bi_2Sr_2Ca_{1-x}Y_x(Cu_{1-y}Zn_y)_2O_{8+\delta}$ with $y=0.02$–0.03. In these samples with $p\sim 1/8$ and $y=0.02$–0.03, transport properties such as electrical resistivity and thermoelectric power exhibit less metallic behavior than usual. There is a possibility that a kind of order of holes and/or spins is stabilized owing to pinning by Zn, as in the La-based cuprate. It is likely that the so-called ''1/8 problem'' is not only characteristic of the La-based cuprate but also common to all high-T_c cuprates including CuO_2 planes in their crystal structures. [S0163-1829(98)06713-7]

I. INTRODUCTION

The anomalous suppression of superconductivity in the La-based cuprate with p (the hole concentration per Cu) $\sim 1/8$, namely, the so-called ''1/8 problem'' is a long-standing one.[1,2] It is known that both $La_{2-x}Ba_xCuO_4$ and $La_{2-x-y}R_ySr_xCuO_4$ (R indicates rare-earth elements) with $x=p\sim 1/8$ undergo a structural phase transition from the orthorhombic midtemperature phase to the tetragonal low-temperature (TLT) phase (space group : $P4_2/ncm$) at a low temperature, and have a local minimum of the superconducting transition temperature T_c as a function of x at $x=p\sim 1/8$.[3–8] As for $La_{2-x}Sr_xCuO_4$, on the other hand, it is known that the similar suppression of superconductivity at $x=p\sim 1/8$ is markedly enhanced through the partial substitution of Zn for Cu, though the TLT phase does not appear at low temperatures.[9] The role of the TLT structure or Zn substitution in the suppression of superconductivity has not been clarified for a long time, but the recent discovery of the stripe-patterned static order of holes and spins in $La_{1.48}Nd_{0.4}Sr_{0.12}CuO_4$ by Tranquada *et al.*[10] has thrown new light on the 1/8 problem. They have concluded that the role of the TLT structure in the suppression of superconductivity is to make the dynamical stripe order static, because the TLT structure is favorable for pinning of the stripe order. They have also pointed out that impurities will also be useful for the pinning. Accordingly, we guess that the enhancement of the suppression of superconductivity through the Zn substitution is due to the stripe-patterned static order pinned by Zn. In fact, we have found an anomaly in the thermoelectric power, which can be attributed to the static order, in the partially Zn-substituted $La_{2-x}Sr_xCu_{1-y}Zn_yO_4$ with $x\sim 1/8$ and $y=0.01$–0.02.[11,12]

If the dynamical stripe order of holes and spins is characteristic of the CuO_2 plane with $p\sim$ 1/8, it will exist not only in the La-based cuprate but also in the other high-T_c cuprates with $p\sim$ 1/8. Moreover, when some pinning centers are introduced into this kind of cuprate, the stripe order is expected to become static, leading to suppression of superconductivity.

In this paper, we take the $Bi_2Sr_2Ca_{1-x}Y_xCu_2O_{8+\delta}$ system of the Bi-2212 phase, whose x dependence of T_c is well defined and simple.[13,14] We investigate T_c of $Bi_2Sr_2Ca_{1-x}Y_x(Cu_{1-y}Zn_y)_2O_{8+\delta}$ in detail, focusing on samples with $p\sim$ 1/8. Here, Zn atoms are introduced into samples as pinning centers of the possible stripe order. We also investigate the electrical resistivity and thermoelectric power, in order to find some symptom of the possible static order of holes and spins.

II. EXPERIMENT

Sintered samples of $Bi_2Sr_2Ca_{1-x}Y_x(Cu_{1-y}Zn_y)_2O_{8+\delta}$ were prepared by the conventional solid-state reaction method. Raw materials of Bi_2O_3, $SrCO_3$, $CaCO_3$, Y_2O_3, CuO, and ZnO powders were used in the molar ratio of Bi : Sr : Ca : Y : Cu : Zn = 2 : 2 : $1-x$: x : $2(1-y)$: $2y$. The powders were mixed and prefired at 800 °C for 12 h in air. Then they were reground, pressed into pellets, and sintered for 24 h at temperatures between 860 and 880 °C. This sintering process was carried out once again so as to obtain homogeneous samples. All products were characterized by powder x-ray diffraction to be of the almost single-phase structure.

Electrical resistivity measurements were carried out by the standard dc four-point probe method. The thermoelectric power was measured by the dc method with a temperature gradient of $\sim$ 0.5 K across a sample.

III. RESULTS

Figure 1 displays the temperature dependence of the electrical resistivity ρ. The x dependence of T_c, defined as the midpoint of the superconducting transition in the ρ vs T plot, is shown in Fig. 2. These values of T_c are in good correspondence with the onset temperature of the Meissner effect, estimated from the magnetic-susceptibility measurements. The x dependence of T_c in the nonsubstituted samples with $y=0$ is the same as described in the literature.[13,14] As well known in the high-T_c cuprates, the value of T_c decreases through the partial substitution of Zn for Cu. What is remarkable in the x dependence of T_c is that a plateau appears at $x=0.30$–0.35 for $y=0.02$ and that a local minimum of T_c is

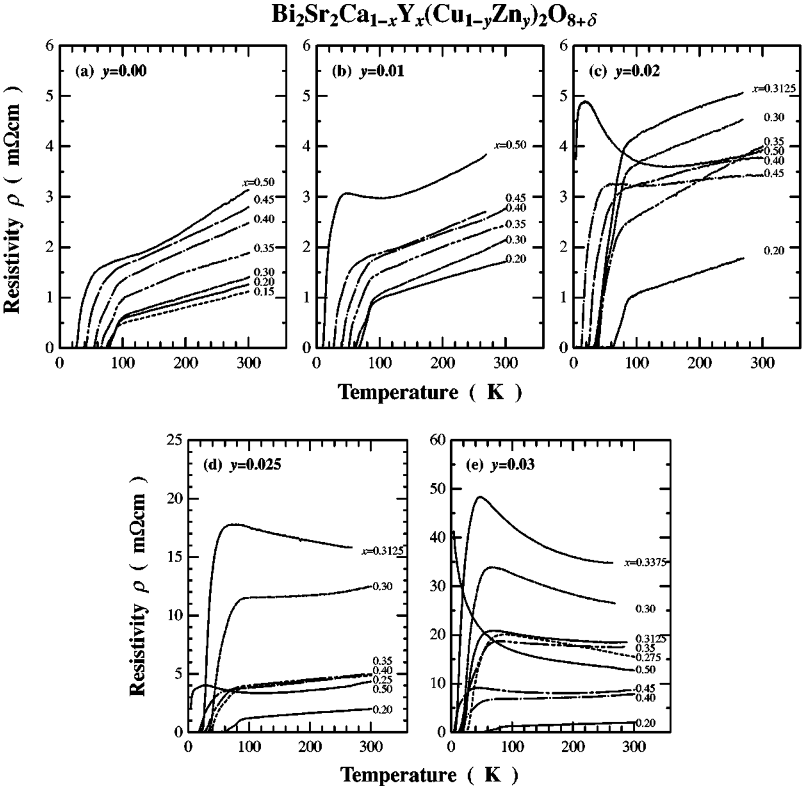

FIG. 1. Temperature dependence of the electrical resistivity ρ for $Bi_2Sr_2Ca_{1-x}Y_x(Cu_{1-y}Zn_y)_2O_{8+\delta}$. (a) $y=0.00$, (b) $y=0.01$, (c) $y=0.02$, (d) $y=0.025$, (e) $y=0.03$.

observed at $x=0.30$–0.35 for $y=0.025$–0.03. Although the data of T_c scatter a little, the plateau and local minimum of T_c are very reproducible in our repeated experiments.

As for ρ in the normal state shown in Fig. 1, the value of ρ increases monotonously with increasing x for $y=0$. This is reasonable, for the hole concentration decreases through the substitution of Y^{3+} for Ca^{2+}. The value of ρ increases through the Zn substitution. For $y=0.02$–0.03, to the surprise, values of ρ at $x=0.30$–0.35 are larger than those at $x=0.40$–0.45. Especially for $y=0.03$, values of ρ at $x=0.30$–0.35 are extraordinarily large and the temperature dependence of ρ is semiconductorlike.

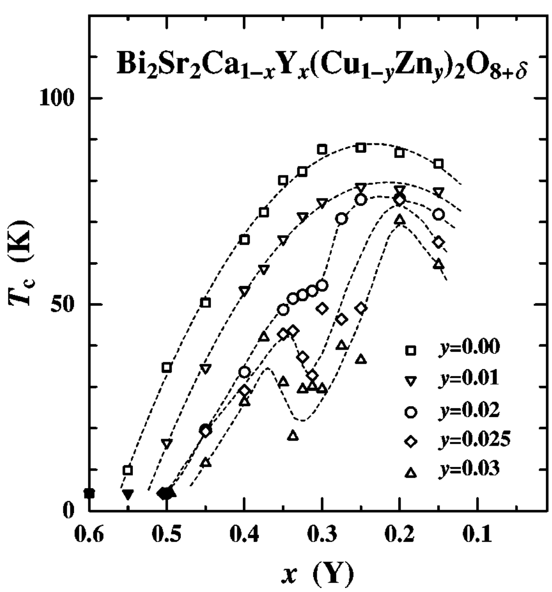

FIG. 2. Y-concentration x dependence of T_c, defined as the midpoint of the superconducting transition curve in the ρ vs T plot, for $Bi_2Sr_2Ca_{1-x}Y_x(Cu_{1-y}Zn_y)_2O_{8+\delta}$. Closed symbols indicate samples which are not superconducting above 4.2 K. Dashed lines are guides to the eye.

Figure 3 shows the temperature dependence of the thermoelectric power S. Although the temperature dependence of S has not yet been understood clearly for the high-T_c cuprates, it is empirically known that S at 290 K, $S_{290\,K}$, decreases with increasing p universally for the high-T_c cuprates.[15,16] In fact, one can confirm that this empirical law holds good for $y=0$–0.02. That is, $S_{290\,K}$ increases with increasing x, namely, with decreasing p. For $y=0.025$–0.03, however, the empirical law appears not to hold good. Values of $S_{290\,K}$ around $x=0.3$ are a little too large.

IV. DISCUSSION

According to the empirical law of $S_{290\,K}$,[15] the value of p is estimated as 0.120–0.136 at $x=0.30$–0.35 in $Bi_2Sr_2Ca_{1-x}Y_xCu_2O_{8+\delta}$. Referring to the chemical-titration analysis in $Bi_2Sr_2Ca_{1-x}Y_xCu_2O_{8+\delta}$ by Kawano *et al.*,[17] it is also estimated as 0.124–0.118 at $x=0.30$–0.35. It is improbable that the value of p changes markedly through the partial substitution of Zn^{2+} for Cu^{2+}. Consequently, it is concluded that the plateau or local minimum of T_c at $x=0.30$–0.35 for $y=0.02$–0.03 is related to $p\sim$ 1/8. For these samples with $p\sim$ 1/8 and $y=0.02$–0.03, the electrical resistivity is singularly less metallic and the value of $S_{290\,K}$ tends to be larger than usual. This indicates that the mobility of holes or the number of mobile holes decreases in these samples. These anomalies may be called a 1/8 problem in the Bi-based cuprate, which we have expected to find.

As for S in the high-T_c cuprates, another interpretation has been proposed by Sera *et al.*[18] They insist that S is given

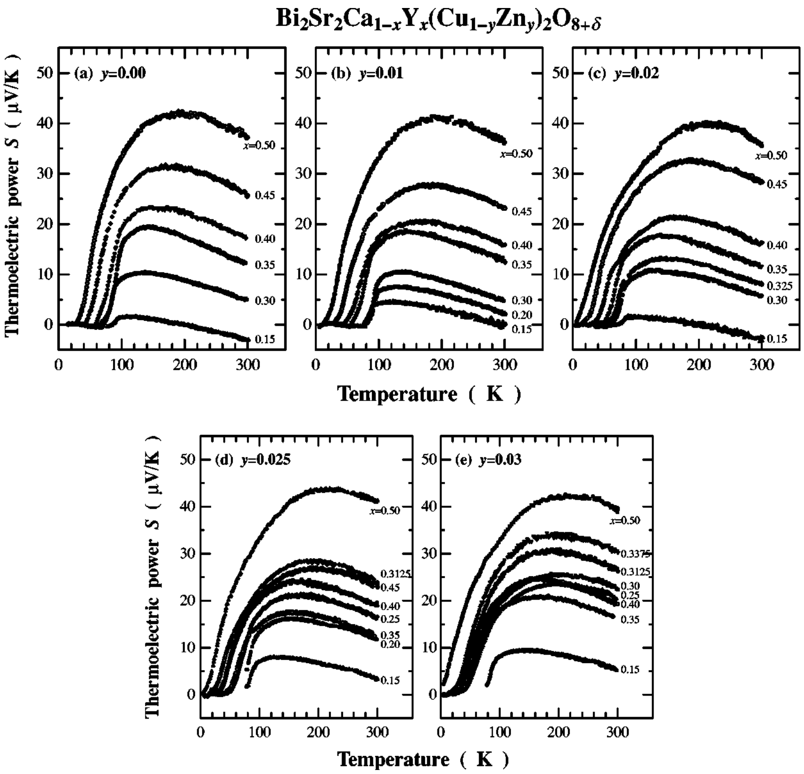

FIG. 3. Temperature dependence of the thermoelectric power S for $Bi_2Sr_2Ca_{1-x}Y_x(Cu_{1-y}Zn_y)_2O_{8+\delta}$. (a) $y=0.00$, (b) $y=0.01$, (c) $y=0.02$, (d) $y=0.025$, (e) $y=0.03$.

by the sum of the T-linear term and the anomalous term. The T-linear term is usual and ascribed to electron diffusion in metal. The anomalous term is due to spin fluctuation or spin correlation. In the high-T_c cuprates, it is well known that the value of S decreases through the partial substitution of Zn for Cu.[18–20] They explain this result as being due to decrease of the anomalous term, owing to suppression of the spin fluctuation or spin correlation. According to their interpretation, the increase in S for the samples with $p\sim 1/8$ and $y=0.02$–0.03 may be explained as being due to increase of the anomalous term, owing to enhancement of the spin fluctuation or spin correlation.

According to the conclusions by Tranquada *et al.*,[10] the following scenario may be described. That is to say, the stripe-patterned dynamical order of holes and spins exists also in the Bi-based cuprate with $p\sim 1/8$. The dynamical order becomes rather static on account of pinning by not the TLT structure but Zn partially substituted for Cu.[21] Consequently, samples with $p\sim 1/8$ and $y=0.02-0.03$ become less metallic and their superconductivity is suppressed. The increase in S of these samples may be due to enhancement of the spin fluctuation in a region where the stripe-patterned dynamical order is changing to be rather static. This scenario is supported by some theoretical and experimental results for $Bi_2Sr_2CaCu_2O_{8+\delta}$.[22–25] One is a theoretical calculation[22] on the assumption of the existence of a fluctuating stripe order, which well explains the experimental results of angle-resolved photoemission spectroscopy and optical conductivity.[23] Another is a polaronic quantum stripe model based on the experimental result of extended x-ray-absorption fine-structure.[24,25] At present, however, it is hasty to regard this scenario as conclusive, because the stripe order of holes and spins has not yet been found directly in the Bi-based cuprate. There remains a possibility that a kind of charge-density wave except the stripe order may be pinned by Zn.

V. CONCLUSION

We have found anomalous suppression of superconductivity at $x=0.30$–0.35, where $p\sim 1/8$, in the partially Zn-substituted $Bi_2Sr_2Ca_{1-x}Y_x(Cu_{1-y}Zn_y)_2O_{8+\delta}$ with $y=0.02$–0.03. In these samples with $p\sim 1/8$ and $y=0.02$–0.03, both electrical resistivity and thermoelectric power exhibit less metallic behaviors than usual. On the analogy of the 1/8 problem in the La-based cuprate, there is a possibility a kind of order of holes and/or spins exists also in the Bi-based cuprate with $p\sim 1/8$ and that it is pinned by Zn, leading to the less metallic behaviors and the anomalous suppression of superconductivity at $p\sim 1/8$. This is a 1/8 problem in the Bi-based cuprate. It is likely that the 1/8 problem is not only characteristic of the La-based cuprate but common to all high-T_c cuprates including CuO_2 planes in their crystal structures.

To be more conclusive, further experiments such as direct observation of the possible order of holes and/or spins by means of electron diffraction and neutron diffraction are necessary in the Zn-substituted $Bi_2Sr_2Ca_{1-x}Y_x(Cu_{1-y}Zn_y)_2O_{8+\delta}$ with $x=0.30$–0.35 and $y=0.02$–0.03.

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