Appearance of a maximum in the superconducting transition temperature of a $Bi_{2,2}Sr_{1,8}CaCu_2O_{8+\nu}$ single crystal under pressure

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(Received 26 April 1993)

The resistance and Hall coefficient of a $Bi_{2.2}Sr_{1.8}CaCu_2O_{8+y}$ single crystal have been measured under pressure. It is shown from our result that the pressure coefficient of dT_c/dP changes from positive to negative with increasing pressure and the maximum superconducting transition temperature appears at a pressure of about 1.05 GPa. The Hall-coefficient result shows a monotonic increase of hole concentration with pressure up to 1.19 GPa. The maximum of the superconducting transition temperature induced by pressure in $Bi_{2.2}Sr_{1.8}CaCu_2O_{8+y}$ can be interpreted in terms of the hole concentration passing through an optimal value under pressure.

The structure of the high- T_c superconductor Bi₂Sr₂CaCu₂O₈ (Bi 2:2:1:2) is strongly anisotropic along the *ab* plane and the *c*-axis direction. It is believed that the electrical-transport anisotropy in the *ab* plane and in the c-axis direction is derived from this structural anisotropy. Above the superconducting transition temperature, the resistivity in the ab plane varies linearly with temperature, whereas the resistivity along the c-axis direction almost displays semiconductive character,¹ and the ratio of these resistivities is about 10^4 . In addition, the compressibility of Bi 2:2:1:2 along the ab plane and the c-axis direction is also different due to its structure. X-ray diffraction experiments under high pressure up to 50 GPa (Ref. 2) shows that the relative contraction is linear below 10 GPa and the value of $d \ln c / dP$ is about -0.6%/GPa, whereas $d \ln a / dP$ is about -0.2%/GPa. In this pressure region, the structure is tetragonal and no phase change is observed. The reduction of the resistivities under pressure is also anisotropic.³ The value of $d \ln \rho_c / dP$ is about twice that of $d \ln \rho_a / dP$ at room temperature. The variation of the superconducting transition temperature with pressure in Bi 2:2:1:2 has been widely investigated by many groups.⁴ Both positive and negative values of dT_c/dP have been reported. Although the data in Ref. 4 were provided by different research groups measuring different samples, it is interesting to note that the positive value normally appears at lower pressure whereas the negative value appears at higher pressure.

The variation of the superconducting transition temperature with hole concentration was studied on the Bi 2:2:1:2 high- T_c superconductor. Allgeier and Schilling⁵ investigated the oxygen content in Bi₂Sr₂CaCu₂O_{8+y} and found that the curve of the oxygen-content dependence of T_c resembles a bell curve. As the oxygen content is decreased from y = 0.24, the superconducting transition temperature $T_c(y)$ increases initially, then passes through a maximum at y = 0.16 and decreases finally. However, research on the pressure dependence of the Hall coefficient⁶ reveals that the hole concentration in Bi 2:2:1:2 increases monotonically with pressure under 2.25 GPa.

Here we report the observation of the maximum of T_c

with increasing pressure up to 1.6 GPa in $Bi_{2.2}Sr_{1.8}CaCu_2O_{8+y}$ single crystals. In this pressure region, the pressure coefficient of dT_c/dP changes from positive to negative contrary to the previous report⁴ where the values of the pressure coefficient change monotonically. The measurements of the Hall coefficient in the *ab* plane imply that the hole concentration increases monotonically with increase of pressure. The change of pressure coefficient from positive to negative is directly relevant to the hole concentration in the *ab* plane passing through an optimal value under pressure.

A single crystal was prepared by the traveling solvent floating zone (TSFZ) method. Appropriate amounts of Bi_2O_3 (99.99%), $Sr(NO_3)_2$ (99.9%), $CaCO_3$ (99.9%), and CuO (99.99%) were stoichiometrically mixed in an agate mortar with ethanol. The ratio of [Bi]:[Sr]:[Ca]:[Cu] was 2.2:1.8:1:2 for feed rod and 2.4:1.5:1:1.8 for solvent rod, respectively. The powders of the mixture were calcined in air at 750 °C for 12 hours, then four cycles of grinding and recalcining at 860 °C for 12 hours in air were carried out. Finally, the product was pressed into rods with a diameter of 6 mm under hydrostatic pressure of 250 MPa and sintered at 860 °C for 12 hours in oxygen atmosphere. The crystal growth was undertaken using a single ellipsoidal infrared heating furnace (Nichiden Machinery Ltd.) in air and the speed of growth was chosen to be 0.5 mm/h. The grown crystals are sheetlike and can be easily laminated along the ab plane. The composition of the single crystal was determined by inductive coupled plasma (ICP, SEIKO model SPS 1500) analysis and the ratio was [Bi]:[Sr]:[Ca]:[Cu] = 2.2:1.8:1:2. The crystal was examined by x-ray diffraction and the lattice coefficients were $a = b = 0.513 \pm 0.007$ nm and $c = 3.054 \pm 0.05$ nm.

The temperature dependence of the electric resistance under pressure was measured on two crystals for the abplane and the *c*-axis direction by DC standard-four-probe and pseudo-four-probe techniques,⁷ respectively. The lead wires were connected by indium to the samples. Hydrostatic pressure was generated by a Cu-Be pistoncylinder cell up to 1.6 GPa and the pressure in the cell between 60 K to room temperature was determined by the change of manganin resistance.⁸ The temperature of the sample was detected by a chromel-constantan thermocouple situated next to the sample and the error of temperature induced by pressure was checked⁹ within the accuracy of the thermocouple calibration.

The temperature dependence of electric resistance in the c-axis direction at various pressures for Bi 2.2:1.8:1:2 is plotted in Fig. 1(a). Because the way we measured the resistivity in the c-axis direction is not standard, in this paper, we give the resistance instead of the resistivity. In the c-axis direction, the resistance, under different pressure, exhibits semiconductive character in the normal state with a maximal value at the onset temperature. The pressures shown in the figure are the values at the superconducting transition temperature. In this paper the superconducting transition temperature in the c-axis direction was defined as the temperature at which the electric resistance drops to 50% of its maximal value. It is clear that the resistance curves in Fig. 1(a) shift to high temperature with an increase of pressure below 1.05 GPa and turn back above this pressure. Figure 1(b) shows the pressure dependence of the superconducting transition temperature in the c-axis direction. In the figure the solid line is a guide to the eye. The onset and the zeroresistance temperatures also shift with pressure in a similar way to T_c . It can be seen from the curve that the pressure coefficient changes from positive to negative at

about 1.05 GPa and at this pressure the superconducting transition temperature has a maximum. Figures 2(a) and 2(b) show the temperature dependence of electric resistance and the pressure dependence of T_c in the *ab* plane, respectively. In Fig. 2(a), the resistance in the normal state, contrary to that in Fig. 1(a), has metallic character. T_c in the *ab* plane is defined as the temperature at which the resistance drops to the half of its extrapolated normal state value. It can be seen from Fig. 2, that the variations of the resistance and T_c have the same tendency as in the *c*-axis direction although the measurement is carried out on two different crystals.

The change of pressure coefficient from positive to negative and the maximum of T_c induced by pressure have been reported on $La_{2-x}Sr_2CuO_4$ when x = 0.15,¹⁰ YBa₂Cu₃O₇,¹¹ YBa₂Cu₄O₈,¹² Y₂Ba₄Cu₇O_{15.32},¹³ Tl₂CaBa₂Cu₂O₈,^{14,15} and Tl₂Ba₂Ca₂Cu₃O_{10-x}.¹⁶ It is interesting to note that T_c is also observed to pass through a maximum as a function of the hole concentration in $La_{2-x}Sr_2CuO_{4+y}$,¹⁷ YBa₂Cu₃O_{7-y},¹⁸ Bi₂Sr₂CaCu₂O_{8+y}^{5,19} and Tl₂Ba₂CuO_{6+y}.⁵ It is reasonable to assume that a bell-shaped $T_c(h)$ curve is characteristic for all hole-doped cuprate superconductors. For a certain hole-doped cuprate superconductor, there is an optimal hole concentration at which the superconducting transition temperature has a maximum. The deviation





FIG. 1. (a) Temperature dependence of electric resistance in the *c*-axis direction at various pressure for $Bi_{2,2}Sr_{1,8}CaCu_2O_{8+y}$. (b) Pressure dependence of the superconducting transition temperature in the *c*-axis direction. The solid line through the data is a guide to the eye.

FIG. 2. (a) Temperature dependence of electric resistance in the *ab* plane at various pressure for $Bi_{2.2}Sr_{1.8}CaCu_2O_{8+y}$. (b) Pressure dependence of the superconducting transition temperature in the *ab* plane. The solid line through the data is a guide to the eye.

from the optimal value of the hole concentration would cause the decrease of T_c . We can understand from this parallel variation of $T_c(P)$ and $T_c(h)$, that the change of pressure coefficient from positive to negative may be due to the increase of hole concentration in CuO₂ planes under pressure.

In order to know the variation of carrier concentration in the ab plane the Hall coefficient of Bi 2.2:1.8:1:2 was measured at various pressures. The pressure dependence of the Hall coefficient was detected at room temperature along the *ab* plane. The measured crystal is cut in a cross shape with 0.0234 mm thickness. High pressure was generated by the same device used for the resistance measurement. The electric current and the magnetic field were chosen to be 75 mA and 1.2 T, respectively. The magnetic field was produced by an electromagnet and checked within the error of 5% in the high pressure cell. Figure 3 shows the pressure dependence of the Hall coefficients for the ab plane in our crystal. The line in the figure is obtained from the least-square-method and is within the statistical errors of our measurement. With increasing pressure, the Hall coefficient decreases monotonically up to 1.19 GPa and in reverse the hole concentration increases with pressure. This result reveals that the pressure can induce the hole redistribution in the crystal and the hole concentration increases in the ab plane. In the figure, there seems to be no indication that the Hall coefficient changes in the vicinity of 1 GPa where the pressure coefficient of dT_c/dP changes from positive to negative. At ambient pressure the Hall coefficient was also measured by others^{6,20} on Bi 2:2:1:2 polycrystal and single crystal. The values in Refs. 6 and 20 are $R_H = 1.41 \times 10^{-3}$ (cm³/C) (250 K) and 3×10^{-3} (cm^{3}/C) (extrapolated value at 300 K), respectively, whereas our datum is 2.85×10^{-3} (cm³/C) (300 K). The difference of these data, probably, is mainly due to the different oxygen content and the deviation from stoichiometry in the samples. It was proven²⁰ that the Hall coefficient was reduced to only half of its original value after the Bi 2:2:1:2 single crystal annealed in oxygen at lower temperature. The relative change of R_H induced by pressure is $d \ln R_H / dP = -8\% / \text{GPa}$ which is



FIG. 3. Pressure dependence of the Hall coefficient for the *ab* plane in $Bi_{2.2}Sr_{1.8}CaCu_2O_{8+y}$. The line is obtained by the least-square-method.

smaller than that of -10.8%/GPa in Ref. 6. Maybe, this deviation is from the different hole concentration in the samples at ambient pressure.

The Hall coefficient dependence of superconducting transition temperature is plotted in Fig. 4. The upper scale shows the hole concentration which is based on an assumption: the hole concentration and the Hall coefficient satisfy the simplest relation of $h = 1/eR_H$. It is realized in the figure that the optimal hole concentration appears at 2.4×10^{21} /cm³. The positive or negative pressure coefficient only appears when the hole concentration is less than or greater than this value. If all the holes are considered to come from copper cations, the optimal number of holes per copper is 0.24 which deviates from the value of 0.16 obtained from the oxygen content determination.⁵ This difference probably indicates that the idea, whereby the holes are only contributed by copper, is unrealistic in Bi 2.2:1.8:1:2. It was pointed out²¹ that if the Fermi surface possess complex curvatures, which usually appear in materials with multiple carriers, the Hall coefficient R_H should be determined by the integration of different parts of the Fermi surface. In hole-doped cuprate superconductors, normally, the multiple carriers are present. In $YBa_2Cu_3O_7$ and YBa₂Cu₄O₈, for example, both CuO₂ plane layers and CuO chain layers exist. For the Bi 2:2:1:2 system, besides the CuO_2 layers, the Bi_2O_2 layers can be thought of as another origin supplying the carriers in the ab plane.²² Otherwise, there is 0.2 per molecular formula of surplus bismuth in our crystal. It is most probable that the surplus bismuth is situated at the position of the strontium in the structure and forms $Sr_{1.8}Bi_{0.2}O_2$ layers. Thus, the electrons are provided in these layers after doping bismuth at the site of strontium. The measured Hall coefficient is the total contribution of all these layers rather than only CuO₂ layers.

In conclusion, we have reported the observation of the



FIG. 4. Hall coefficient dependence of the superconducting transition temperature in $Bi_{2.2}Sr_{1.8}CaCu_2O_{8+y}$. Upper scale shows the hole concentration assuming that the simplest relation of Hall coefficient and the hole concentration $h = 1/eR_H$ is satisfied. The solid line through the data is a guide to the eye.

maximal T_c induced by pressure in Bi_{2.2}Sr_{1.8}CaCu₂O_{8+y} single crystals. The result of pressure research on the Hall coefficient measurement shows the monotonic increase of the hole concentration with increasing pressure. The evidence above implies that the maximum of the superconducting transition temperature induced by pres-

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sure in $Bi_{2,2}Sr_{1.8}CaCu_2O_{8+y}$ is due to the hole concentration passing through an optimal value under pressure.

Part of this work was made possible by a Grant-in Aid for Scientific Research, Science of New Superconductors from Ministry of Education, Science and Culture.

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