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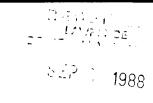


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Pressure Dependence of Superconductivity in Single-Crystal
Bi<sub>2</sub>Sr<sub>2</sub>CaCu<sub>2</sub>O<sub>8</sub>

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### **ABSTRACT**

We report the first high-pressure study of a single-crystal high- $T_{\rm C}$  superconductor. The pressure dependence of the superconducting transition temperature ( $T_{\rm C}$ ) of a single crystal sample of  ${\rm Bi}_2{\rm Sr}_2{\rm CaCu}_2{\rm O}_8$  was measured up to 42 Kbar.  $T_{\rm C}$  first increased slightly with pressure, reaching a maximum at around 10 Kbar, and then decreased with increasing pressure at a rate of about 0.05 K/Kbar. This non-linear pressure dependence of  $T_{\rm C}$  is discussed within a two-dimensional BCS model and within two of the resonating-valence-bond models of high- $T_{\rm C}$  superconductivity.

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Recently there has been much interest in superconductors with superconducting transition temperatures ( $T_c$ ) above 77 K, but containing no rare earth elements. One such family of the high  $T_c$  superconductors are the Bi-Sr-Ca-Cu-O compounds<sup>1,2</sup>. The pressure (P) dependence of  $T_c$  has always played an important role in the attempt to understand the physical mechanism of superconductivity in these materials and in the search for materials with even higher  $T_c$ 's For example, Chu et al.<sup>3</sup> found that the La-Ba-Cu-O compound has an unusually large and positive value of  $dT_c/dP$ . This finding led these authors to substitute La with atoms with smaller ionic radii, such as Y, leading to the discovery of the Y-Ba-Cu-O family of superconductors<sup>4</sup>.

In multi-phase samples of Bi-Sr-Ca-Cu-O, Chu et al. 5 found that  $T_c$ 's reached maxima at relatively low pressures of about 12 Kbar and decreased at higher pressures, for superconducting phases with Tc's between 81K and 114K. A decrease in Tc at pressures above 14 Kbar was also reported by Wijngaarden et al. 6 in a polycrystalline sample of Bi-Sr-Ca-Cu-O with a  $T_c$  of about 85K. To this date, all published experiments on the pressure dependence of  $T_c$  in the new superconductors have been performed on polycrystalline samples and have shown a wide range of values of  $dT_c/dP^7$ . The variability in the pressure measurements is probably due at least in part to the presence of grain boundaries, which can differ greatly from sample to sample, and whose effects would be most prominent in the low-pressure regime. In this paper we report the first measurement of the pressure dependence of Tc in a single crystal sample of Bi2Sr2CaCu2O8. We find that  $T_c$  first increases slightly with pressure, reaching a maximum at around 10 kbar, and then decreases gradually with pressure to at least 42 Kbar. Our results above 10 Kbar were

reproducible with pressure cycling, implying that the sample was not permanently altered by the high pressure. These results show that  $T_{\rm c}$  depends nonlinearly on the lattice constant in this superconductor.

The crystals of Bi<sub>2</sub>Sr<sub>2</sub>CaCu<sub>2</sub>O<sub>8</sub> were grown from a mixture of Bi<sub>2</sub>O<sub>3</sub>, CuO, SrCO<sub>3</sub> and CaCO<sub>3</sub> with molar percentages of respectively 22.4%, 32%, 26.9% and 18.7%. The powders were mixed in a ball mill with acetone, then placed in a gold crucible and heated at 920 C for 5 hours and cooled to 820 C at a rate of 3 C/hr in flowing oxygen. The result was a black, glassy mass that cleaved into micaceous sheets, with resistively determined T<sub>C</sub>'s of approximately 80K. X-ray analysis showed that the c axis was perpendicular to the cleavage plane and had a spacing of 3.0 nm, in agreement with the Bi<sub>2</sub>Sr<sub>2</sub>CaCu<sub>2</sub>O<sub>8</sub> compound identified by subramanian et al.<sup>8</sup>.

2 4 5 M

The sample was a thin platelet cleaved from the bulk crystalline mass, with approximate dimensions 200x100x10 (micron)<sup>3</sup>. Ohmic contacts were prepared by painting silver paint contacts on the crystal and baking them at 750 C for 20 minutes in oxygen. High-pressure measurements were made in a diamond anvil cell using an Inconel gasket and CaSO<sub>4</sub> as the pressure medium. Thin copper wires were introduced into the cell using the technique described by Erskine et al.<sup>9</sup>, and attached with silver paint to the contact pads in a two-probe configuration; the residual contact resistance in the superconducting state was about 10 ohms. The pressure was measured with the standard ruby fluorescence technique. Four ruby chips were placed around the sample, and

the pressure inhomogeneity was measured to be less than 10%. The sample temperature was determined with a calibrated Si diode thermometer in thermal contact with one of the diamonds. To minimize thermal lag, the temperature was changed slowly enough so that the resistance versus temperature curves during the cooling and warming cycles were identical. The sample pressure was first increased to 42 Kbar and then decreased to 14 Kbar with reproducible results, in contrast to some earlier experiments with polycrystalline high-T<sub>C</sub> superconductors that have shown irreversible changes in resistance with pressure cycling<sup>10</sup>. The experiment was discontinued when the wires were cut by the diamond anvils on repressurization. When the cell was opened, the sample was intact and showed no visible signs of damage.

A typical resistance versus temperature curve for the sample is shown in figure 1. At each pressure we define T<sub>CO</sub>, T<sub>CM</sub> and T<sub>Cf</sub> as the temperatures where the sample resistance has dropped respectively by 10%, 50% and 90% of the total resistance decrease of the superconducting transition. The variation of these temperatures with pressure is shown in Figure 2. The horizontal error bars represent the variation in pressure as determined from the ruby fluorescence. The open squares are data points obtained with increasing pressure, and the triangles with decreasing pressure. The circle is the data point obtained on the second pressurization, before the wires broke. As can be seen in Figure 2, the data for increasing and decreasing pressures are quite reproducible.

The results presented in Figure 2 are in general agreement with previous results in polycrystalline samples of Bi-Sr-Ca-Cu-O.

At pressures below about 10 Kbar, we find that  $T_{\rm cm}$  increases at the rate of about 0.17 K/Kbar. Chu et al.<sup>5</sup> have reported an increase in  $T_{\rm c}$  with pressure in Bi-Ca-Sr-Cu-O compounds at the rate of about 0.3 K/Kbar. Although this is a significantly larger rate, the uncertainty in both measurements could be substantial because  $T_{\rm c}$  varies nonlinearly with pressure in this region. Above 10 Kbar, we find that  $T_{\rm cm}$  changes at a rate of about -0.05 K/Kbar, with a pressure-induced broadening of the transition giving rates of -0.03 K/Kbar for  $T_{\rm co}$  and -0.17 K/Kbar for  $T_{\rm cf}$ . Winjgaarden et al.<sup>6</sup> report pressure dependences in  $T_{\rm cm}$ ,  $T_{\rm co}$  and  $T_{\rm cf}$  of respectively -0.16, -0.09 and -0.21 K/Kbar. The increased slope and additional broadening of their data is most likely due to the polycrystalline nature of their sample and to the higher pressures obtained (80 Kbar).

Recently, Griessen published a survey of the pressure coefficients  $dT_C/dP$  in all of the high  $T_C$  superconductors reported up to that time, and compared the data with predictions based on some of the proposed superconducting models. Only three of the models were considered to be consistent with the pressure measurements as well as with other experimental data: the two-dimensional BCS theory of Labbe and Bok<sup>11</sup>, and the resonating-valence-bond (RVB) models of Cyrot<sup>12</sup> and of Fukuyama and Yosida<sup>13</sup>. Nonlinear dependences of  $T_C$  on pressure were not considered. Here we will discuss briefly how the nonlinearities observed in our data may be understood within these models.

Within the two-dimensional BCS model of Labbe and  $Bok^{11}$ ,  $T_c$  is

determined by the usual electron-phonon interaction parameter  $\lambda$  and by D, the "width" of the two-dimensional saddle point  $E_s$  in the electronic density-of-states, via the equation:

$$k_B T_C = 1.13 \text{MDexp} [(-1/\sqrt{\lambda})]$$
 (1)

The expression for the volume (V) dependence of  $T_C$  is derived from Eq. (1) as

$$\frac{d\ln T}{d\ln V} c = \frac{d\ln D}{d\ln V} + \frac{1}{2\sqrt{\lambda}} \frac{d\ln \lambda}{d\ln V}.$$
 (2)

To explain the saturation and nonlinear dependence of  $T_c$  on P, one or both terms in Eq. (2) has to be strongly dependent on V. Since there is no evidence of lattice instability or softening of the Cu-O vibrational mode involved in  $\lambda$  , there is no reason for it to have a strong nonlinear dependence on V. On the other hand, it is possible that D does have a strong nonlinear dependence on V. Labbe and Bok 11 assumed an exactly half-filled two-dimensional band so that the Fermi level coincided with Es. This assumption would no longer be valid if pressure caused electrons in adjacent Cu-O planes to overlap, thus modifying the van Hove singularity and in addition shifting the Fermi level. In general, we expect that pressure will tend to cause any two-dimensional system to behave more like a three-dimensional system, so the singularity at the saddle point in any two-dimensional model should be very sensitive to pressure. In addition we note that the saturation and nonlinear dependence in Tc with pressure has been observed in conventional superconductors, such as La<sub>3</sub>S<sub>4</sub> and La<sub>3</sub>Se<sub>4</sub><sup>14</sup>, where the data were interpreted as a pressure-induced shift of the Fermi level through a sharp peak in the density of states. Similar pressure-induced shifts of the Fermi level relative to the

singularity may also contribute to the pressure effects in Bi<sub>2</sub>Sr<sub>2</sub>CaCu<sub>2</sub>O<sub>8</sub>.

In the RVB models,  $T_C$  depends only on the transfer integral  $t_D$  and the electron-electron interaction U. It is not obvious why these quantities should possess strong nonlinear dependences on V. However, we note that the expression derived by Cyrot for  $T_C$  depends on an additional parameter  $\delta$ :

 $T_c = t_b \delta \exp(-U\delta/t_b)$  (3) where  $\delta$  is the fractional doping which creates some  $Cu^{3+}$  ions. In this model  $T_c$  depends very strongly on  $\delta$  and in a very nonlinear way (see, for example, Fig. 3 of Ref. 7). Griessen assumed that only  $t_b$  depended on V, while U and  $\delta$  were both independent of V. If  $\delta$  is dependent on pressure, perhaps through a charge-transfer mechanism, it could account for the nonlinear behavior of  $dT_c/dP$ .

In summary, we find a nonlinear dependence of  $T_{\rm C}$  on pressure in single-crystal  ${\rm Bi_2Sr_2CaCu_2O_8}$ , with a negative  ${\rm dT_C/dP}$  at pressures above about 10 Kbar. We have shown how these results may be understood within a two-dimensional BCS model and within two of the RVB models of high- $T_{\rm C}$  superconductivity.

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### FIGURE CAPTIONS

- Figure 1. A typical resistance vs. temperature curve showing the superconducting transition in Bi<sub>2</sub>Sr<sub>2</sub>CaCu<sub>2</sub>O<sub>8</sub> at high pressure.
- Figure 2. The pressure dependence of  $T_c$  in  $Bi_2Sr_2CaCu_2O_8$ , showing  $T_{co}$  (open symbols),  $T_{cm}$  (dark shading), and  $T_{cf}$  (light shading). The differences in the symbols are explained in the text.

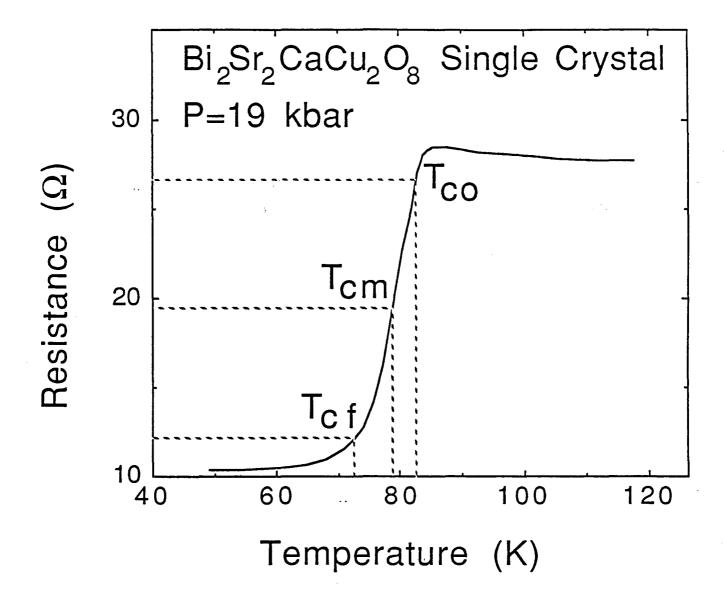


Fig. 1

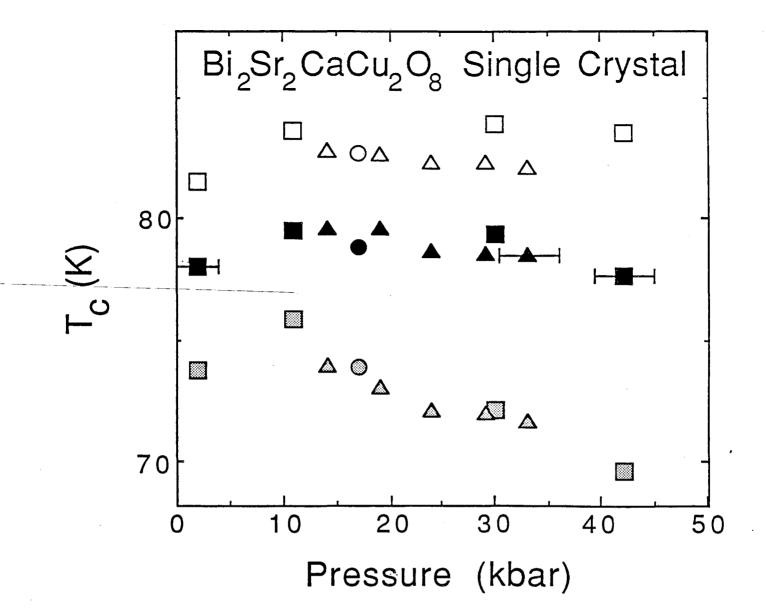


Fig. 2

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