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Rapid Communication

Onset of Superconductivity at 107 K in $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ at High Pressure

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Abstract. Electrical resistivity ρ measurements under pressure have been carried out on the high-temperature superconductor $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ as a function of temperature T between 1 and 300 K at various pressures between 8 kbar and 149 kbar. The superconducting transition temperature T_c increases almost linearly with pressure at the rate $dT_c/dP \simeq 0.13$ K/kbar. The onset of T_c , defined as the temperature at which $\rho(T)$ drops to 90% of its extrapolated normal state value, increases from ~ 95 K at 8 kbar to 107 K at 149 kbar. These results suggest that higher pressures will yield yet higher values of T_c .

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During the past year, the maximum superconducting transition temperature T_c has risen from ~ 23 K for the A15 compound Nb_3Ge , where it had remained since 1973 [1], to $\simeq 95$ K for the compound $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ ($\delta \simeq 0.1$) [2], which has an orthorhombic oxygen-deficient perovskite-like crystal structure [3], and the isostructural series of rare earth (R) compounds $\text{RBa}_2\text{Cu}_3\text{O}_{7-\delta}$ (except for the compounds with $R = \text{Ce}$, Pr , and Tb , which are not superconducting above 4.2 K) [4]. The dramatic increase of the maximum T_c to its present value of ~ 95 K occurred through a series of developments starting with the discoveries of superconductivity with $T_c \simeq 30$ K in $(\text{La}_{1-x}\text{Ba}_x)_2\text{CuO}_{4-\delta}$ ($x \simeq 0.075$) [5,6] and $T_c \simeq 40$ K in $(\text{La}_{1-x}\text{Sr}_x)_2\text{CuO}_{4-\delta}$ ($x \simeq 0.075$) [7,8], and the increase of T_c with pressure to onset values, depending on the definition of the T_c onset,

ranging between $\simeq 40$ and $\simeq 50$ K, for materials in the La-Ba-Cu-O system [9,10]. Measurements of T_c under pressure have been reported for multiphase samples in the Y-Ba-Cu-O system [11] and $\text{RBa}_2\text{Cu}_3\text{O}_{7-\delta}$ compounds, with $R = \text{Y}$, up to $\simeq 10$ kbar [12], and $R = \text{Y}$, Gd , Er , and Yb , up to $\simeq 18$ kbar [13]. In the study the multiphase Y-Ba-Cu-O samples [11], T_c was found to increase from less than 90 K to about 91.5 K at 17.6 kbar, while for the studies on the $\text{RBa}_2\text{Cu}_3\text{O}_{7-\delta}$ compounds [12,13], the largest increase in T_c occurred for the compound $\text{YbBa}_2\text{Cu}_3\text{O}_{7-\delta}$ in which T_c increased from 88 K at zero pressure to 92.5 K at 18.2 kbar. In this communication, we report measurements of $T_c(P)$ in $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ to $\simeq 149$ kbar, using a clamped Bridgman anvil device, which reveal a nearly linear increase of T_c in which the T_c onset increases from $\simeq 95$ K at 8 kbar to $\simeq 107$ K at 149 kbar. To our knowledge, this is the highest value of T_c yet reported for a material known to

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exhibit a confirmed bulk superconducting transition. A brief account of this work was given in the Proceedings of a recent conference [14].

Experimental Details

Electrical resistivity measurements under pressure were made on a bar-shaped specimen of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ that was cut from a polycrystalline sintered pellet prepared in the manner described in [15]. A self-clamping Be-Cu device was used to generate and retain quasihydrostatic pressures up to 160 kbar between opposed tungsten-carbide Bridgman anvils. The sample and a Pb manometer were placed in series with one another between two disks of steatite, which served as the pressure transmitting medium, surrounded by a pyrophyllite ring-shaped gasket. The two current and four voltage leads of Pt were attached to the sample and Pb manometer with Ag loaded epoxy, and the voids between the steatite and the sample, manometer and electrical leads were filled with talc. Pressures were inferred from the T_c of the Pb manometer [16], and the temperature of the self-clamping high pressure device was varied between 1 and 300 K, using a He^4 cryostat. The four-lead electrical resistivity measurements were made using a standard phase sensitive detection technique at a frequency of 220 Hz.

Results and Discussion

Electrical resistance R vs temperature data for $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ at four different pressures ranging from 8 kbar to 149 kbar are shown in Fig. 1. In the normal state at temperatures greater than T_c , the $R(T)$ curves are nearly linear at all four pressures, with negative temperature coefficients of resistivity $\alpha \equiv (1/\rho)(d\rho/dT)$ at the two lower pressures and nearly zero α -values at the two higher pressures. The increase of α with pressure between 8 kbar and 90 kbar and the decrease of R with pressure may be partly due to changes in sample geometry, since the density of the sintered material is only about 80% of the theoretical density after the final sintering step. The decrease of the value of R with pressure reveals that the $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ compound becomes more metallic under pressure. The negative value of α at 8 kbar indicates that the $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ compound is slightly deficient in oxygen.

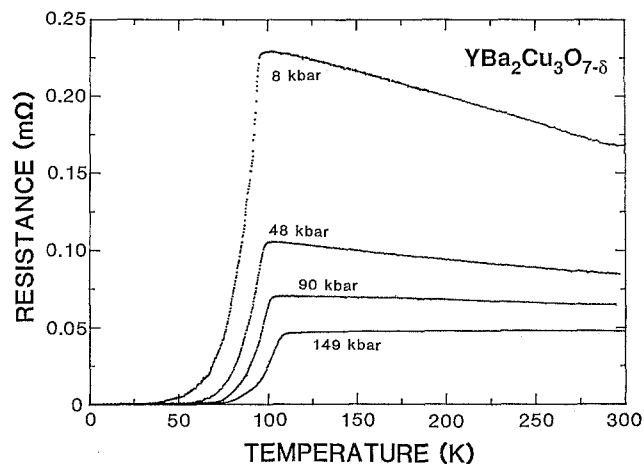


Fig.1. Electrical resistance vs temperature for $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ at pressures of 8, 48, 90, and 149 kbar

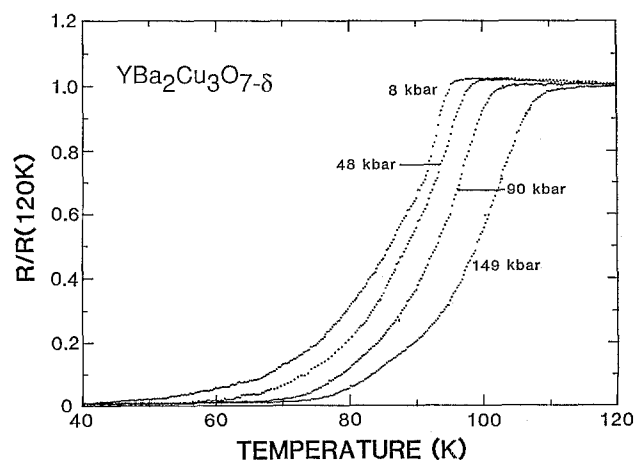


Fig.2. Electrical resistance R normalized to its value at 120 K, $R/R(120\text{ K})$, vs temperature at pressures of 8, 48, 90, and 149 kbar in the vicinity of T_c

The effect of pressure on the superconducting transition can be seen more clearly in Fig. 2 where the electrical resistivity ρ , normalized to its value at 120 K, is plotted vs temperature between 40 and 120 K. The shape of the resistive superconductive transition curve does not change with pressure, although the width of the transition decreases slightly with pressure. We define the transition temperature T_c as the value at which the resistivity drops to 50% of its extrapolated normal state value and the transition width ΔT_c as the difference between the temperatures at which ρ drops by 90% and 10% of its extrapolated normal state values; i.e., $T_c \equiv T(50\%)$ and $\Delta T_c \equiv T(90\%) - T(10\%)$. The transition width is rather large with $\Delta T_c \approx 20\text{ K}$, which is probably primarily due to the pressure inhomogeneity in the high pressure cell, since a

piece of the sample measured outside the cell at atmospheric pressure had a width $\Delta T_c \approx 5$ K.

Borges et al. [13] reported that the rate dT_c/dP varied from 0.09 K/kbar to 0.19 K/kbar for $\text{RBA}_2\text{Cu}_3\text{O}_{7-\delta}$ where $R = \text{Y, Gd, Er}$ and Yb . The value of 0.09 K/kbar for $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ between 0 and 16 kbar corresponds rather closely with the average rate of 0.085 K/kbar we have obtained from our resistivity measurements in the Bridgman anvil cell for pressures between 8 and 149 kbar. However, this value is about two times larger than the value $dT_c/dP = 0.043$ K/kbar which was recently reported by Driessen et al. [17] from resistive measurements of T_c up to 170 kbar in a beryllium-copper diamond anvil cell using epoxy plus Al_2O_3 powder as the pressure transmitting medium and the ruby fluorescence method to determine the pressure.

The rather large positive dependence of T_c on P in both the $(\text{La}_{1-x}\text{A}_x)_2\text{CuO}_{4-\delta}$ ($A = \text{Ba}$ and Sr) and $\text{RBA}_2\text{Cu}_3\text{O}_{7-\delta}$ systems suggests that a common mechanism is responsible for the high T_c superconductivity of these two superconducting phases. Moreover, such a strong positive dependence of T_c on P is unusual among superconducting materials in general.

Recently, Griessen [18] has discussed the pressure dependence of T_c of the high- T_c copper oxide superconductors within the context of several models for superconductivity: the standard Bardeen-Cooper-Schrieffer (BCS) electron-phonon theory, the two-dimensional BCS electron-phonon theory, resonating valence bond models, and various bipolaronic models. Based on the pressure dependence of T_c of the $(\text{La}_{1-x}\text{A}_x)_2\text{Cu}_{4-\delta}$ ($A = \text{Ba}$ and Sr) and $\text{RBA}_2\text{Cu}_3\text{O}_{7-\delta}$ compounds, as well as the small isotope effect in the $\text{RBA}_2\text{Cu}_3\text{O}_{7-\delta}$ compounds with $R = \text{Y}$ and Eu , Griessen concluded that the resonating valence bond type models, as treated by Fukuyama and Yosida [19] and by Cyrot [20], seem to be the most appropriate of these models for describing the superconductivity of these materials. Kaneko et al. [21] have shown that there is a linear relationship between the pressure dependence of T_c or the Néel temperature T_N of various superconducting, antiferromagnetic and ferrimagnetic oxides and sulfides and $\kappa_v T_c$ or $\kappa_v T_N$, where κ_v is the volume compressibility, and have agreed that Cyrot's model appears to be capable of explaining the high T_c 's of the copper oxide superconductors.

In summary, the behavior of the electrical resistivity as a function of temperature and pres-

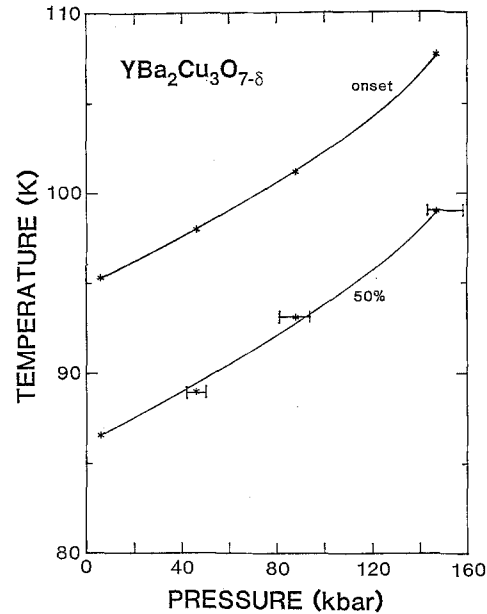


Fig.3. Onset (90%) and midpoint (50%) values of T_c , as defined in the text, vs pressure for $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$

sure, shown in Figs. 1 and 2, indicates that applied pressure causes $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ to become more metallic and increases T_c . The application of pressure has the same qualitative effect as increasing the oxygen concentration x in $\text{YBa}_2\text{Cu}_3\text{O}_x$ which gradually evolves from semi-conducting with $T_c \approx 0$ from $x \approx 6.5$ to metallic with $T_c \approx 95$ K with $x \approx 6.9$; the more metallic, the higher T_c . The results as plotted in Fig. 3 show no evidence of a maximum or decrease in dT_c/dP suggesting that even higher T_c 's could be attained at higher applied pressures. X-ray diffraction measurements under pressure [22,23] to 130 kbar at 15 K, 150 K and 300 K, reveal an approximately linear decrease of the unit cell volume which reaches $\approx 9\%$ at 130 kbar, with no indication of a structural transition within this pressure range. This suggests that higher T_c materials of the same general structure may be possible with appropriate changes in the substituents.

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