

**$T_c(P)$ FROM MAGNETIC SUSCEPTIBILITY MEASUREMENTS IN HIGH TEMPERATURE
SUPERCONDUCTORS: $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ AND $\text{HgBa}_2\text{Ca}_2\text{Cu}_3\text{O}_{8+x}$**

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ABSTRACT

We have measured the pressure dependence of the critical temperature of the superconducting transition in $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ and $\text{HgBa}_2\text{Ca}_2\text{Cu}_3\text{O}_{8+x}$ using a highly sensitive diamagnetic susceptibility technique. Samples were investigated with quasihydrostatic (NaCl) and hydrostatic (He) pressure media. Our results for $\text{HgBa}_2\text{Ca}_2\text{Cu}_3\text{O}_{8+x}$ are consistent with previous measurements by resistive techniques under quasihydrostatic conditions. The results for $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ are much more sensitive to sample variability than to the pressure medium used. We observed the well-known rapid increase in T_c with increasing pressure for underdoped samples, with T_c reaching its maximum 98 K at 12 GPa. For overdoped samples, T_c has a maximum value about 92 K at 4 GPa. We fit our data for $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ using a phenomenological inverse parabolic dependence of T_c on hole concentration in CuO_2 planes and compare our high pressure results with previous low-pressure data.

Introduction

The pressure dependence of T_c in $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ has been measured many times using different techniques [1-6]. An approach proposed by Almasan et al. [2] and Neumeier and Zimmerman [3] incorporates essential features required to explain the available low-pressure experimental data. This approach is based on the phenomenological inverse parabolic dependence of T_c on carrier concentration, n , [7]

$$T_c/T_c^{\text{max}} = 1 - A(n - n_{\text{opt}})^2 \quad (1)$$

Here T_c^{max} is the critical temperature reached at optimum doping, n_{opt} . Carrier concentration is referred to a single CuO_2 unit in CuO_2 planes, and A is a universal constant. Tallon et al. [8] found recently that $A=82.6$ and $n_{\text{opt}} \approx 0.16$ holes/ CuO_2 for $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$. It was proposed in Refs. [2,9] that $n = n(0) + (dn/dP)P$, where dn/dP accounts for the charge transfer from CuO chains to CuO_2 planes, $A=16.9$ [2] or $A=27.7$ [9], $n_{\text{opt}} \approx 0.25$ and

$$T_c^{\text{max}}(P) = T_c^{\text{max}}(0) + (dT_c^{\text{max}}/dP)P \quad (2)$$

The motivation for the present work was (i) to examine the application of the low-pressure charge transfer model [2,3,9] at higher pressures, and (ii) to determine the value of the intrinsic pressure

derivative, dT_c^{max}/dP , for $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ and to compare it with data for the $\text{HgBa}_2\text{Ca}_2\text{Cu}_3\text{O}_{8+x}$.

Experiment

We have used a new versatile magnetic susceptibility technique proposed by Timofeev [10] to measure the superconducting transition temperature in diamond anvil cells. The information on the $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ samples is summarized in Tables 1 and 2. We state $x=0$ for our best untwinned sample C ($T_c(0) = 89.7$ K). We use the c-axis dependence on oxygen deficiency, x , from Refs. [11] to determine x in the samples A and B. Pressure was measured in situ using ruby pressure scale [12].

Table 1. Unit cell parameters from single crystal X-ray diffraction for $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ samples.

Sample	a, Å	b, Å	c, Å
A	3.823(1)	3.886(1)	11.705(1)
B	3.8186(6)	3.8861(9)	11.6960(9)
C	3.818(1)	3.887(1)	11.689(1)

Table 2. T_c (onset) versus estimated oxygen deficiency, and sample dimensions for $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ samples.

Sample	T_c , K	x	dimensions
A	93	0.11(2)	80x80x13 μ
B	92	0.04(2)	50x30x13 μ
C	89.7	0.00(2)	40x40x7 μ

The samples of $\text{HgBa}_2\text{Ca}_2\text{Cu}_3\text{O}_{8+x}$ used in this study were optimally doped polycrystalline samples

[13]. Pressures were determined at room temperature for $\text{HgBa}_2\text{Ca}_2\text{Cu}_3\text{O}_{8+x}$ and were corrected to low temperatures using previous in situ $P(T)$ measurements. This introduces an error about 1-2 GPa in pressure determination.

Results

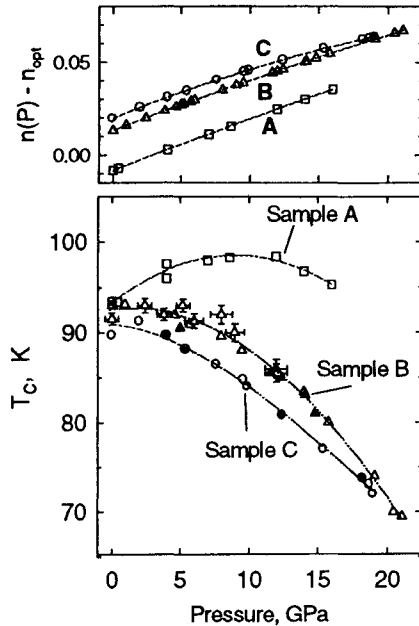


Fig.1. T_c versus pressure for several $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ samples. Sample A - NaCl medium (squares), sample B - He medium (triangles), NaCl medium (triangles with error bars), sample C - He medium (circles). Open symbols correspond to compression, and full symbols to decompression. The pressure dependence of the hole concentration is also shown (see text).

The pressure dependence of T_c for $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ samples is shown in Fig. 1. $T_c(P)$ apparently follows an inverse parabolic dependence, and we can apply Eqs.(1,2) to calculate the pressure dependence of T_c^{max} in linear approximation (see Eq.(2)). We use the charge transfer term in the form

$$n(P) = n_0 + (dn/dP)P + (d^2n/dP^2)P^2, \quad (3)$$

where $n_0 = n(0) - n_{\text{opt}}$. We obtain

$$T_c(P) = (T_c^{\text{max}}(0) + (dT_c^{\text{max}}/dP)P)(1 - 82.6 n^2(P)) \quad (4)$$

We have included pressure derivatives up to second order into the charge transfer term (Eq.(3)) because a linear approximation was not sufficient to fit all three data sets. The resulting parameters obtained from fitting Eq.(4) to the experimental data are summarized in Table 3. The fits and the calculated pressure dependence of the hole concentration $n(P)$ for all three samples are shown in Fig. 1.

Table 3. Parameters are determined by fitting Eq. (4) to experimental data in Fig. 1. The values $T_c^{\text{max}}(0) = 93.89(10)$ K and $dT_c^{\text{max}}/dP = 0.77(5)$ K/GPa were constrained to be equal for all three samples and were determined from the fit.

Sample	n_0 , holes/ CuO_2	$dn/dP \times 10^3$, holes/ CuO_2 1/GPa	$d^2n/dP^2 \times 10^5$, holes/ CuO_2 1/GPa ²
A	-0.008(3)	2.8(6)	-0.6(30)
B	0.013(2)	2.8(2)	-1.2(7)
C	0.020(2)	3.0(2)	-3.4(9)

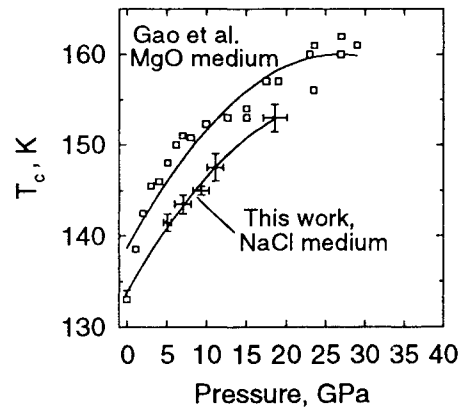


Fig.2. T_c versus pressure for $\text{HgBa}_2\text{Ca}_2\text{Cu}_3\text{O}_{8+x}$ samples.

We have also measured $T_c(P)$ in optimally doped $\text{HgBa}_2\text{Ca}_2\text{Cu}_3\text{O}_{8+x}$ samples in a NaCl medium. The results are shown in Fig. 2. There is an offset of about 6-7 K between previous resistivity data [13] and our measurements at $P > 0$. We attribute this offset to the procedure of T_c determination used by Gao et al. [13]. They determined T_c as the onset of the drop in resistivity. However, T_c could have been overestimated due to contributions from inter-

grain boundaries. Grain boundaries are stressed more than the rest of the sample volume and could have higher T_c values. Our data at room pressure agree with the data of Gao et al. ($T_c=134$ K). It is evident from Fig. 2, that the offset between resistivity and susceptibility data is rapidly increasing at low pressures. This supports our hypothesis concerning the contribution from grain boundaries to resistivity onset of T_c . We obtain $(dT_c/dP)_{P=0} = 1.4$ K/GPa.

Discussion

There is a striking difference in $T_c(P)$ behavior between $\text{HgBa}_2\text{Ca}_2\text{Cu}_3\text{O}_{8+x}$ and $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ samples. $T_c(P)$ in $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ is governed mainly by the charge transfer term dn/dP and decreases rapidly with pressure for nearly optimally doped samples, because the samples become heavily overdoped with increasing pressure. For $\text{HgBa}_2\text{Ca}_2\text{Cu}_3\text{O}_{8+x}$, we probably have the case, where the contribution from charge transfer is small and the intrinsic dT_c^{max}/dP dominates the pressure dependence of T_c [13,14]. If this indeed is the case, Hg-based superconductors are the best candidates to study the pressure effect on intrinsic properties of high- T_c superconductors. Further pressure studies of underdoped and overdoped Hg-based superconductors would be very helpful in providing information on the charge transfer contribution to $T_c(P)$. If this contribution is appreciable at 30-40 GPa, T_c values even higher than 164 K [13] can be reached for properly doped samples. dT_c^{max}/dP is not known yet for $\text{HgBa}_2\text{Ca}_2\text{Cu}_3\text{O}_{8+x}$, but it should not be less than our measured $(dT_c/dP)_{P=0} = 1.4$ K/GPa for optimally doped samples. This value is even higher than dT_c^{max}/dP for $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$.

Comparing our data for sample C with Fig. 3 in Refs. [9], which predicts $T_c=80.5$ K at $P=25$ GPa for $\text{YBa}_2\text{Cu}_3\text{O}_7$, we immediately come to the conclusion that the parameters used by Almasan et al. [2] and Gupta and Gupta [9] fail to reproduce the high-pressure data. However, our charge transfer term $dn/dP=3.0 \times 10^{-3}$ (holes/ CuO_2) GPa^{-1} at $x=0$ is the same as that given by Almasan et al. [2]. This value is close to the results from theoretical calculations and to estimates from bond-valence-sum analyses (for further references see [9]). Almasan et al. [2] fit their data to the charge transfer term dn/dP given by bond-valence-

sum analysis. However, they underestimated the parameter A (Eq.(1)), which results in failure to describe the high-pressure behavior determined here.

Acknowledgments

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