δ-Influence on the Pressure-Effect on Tc of HgBa2CuO4+δ and the Inverse Parabolic Tc-Relation

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We have observed a δ-independent linear pressure effect on Tc of HgBa2CuO4+δ under hydrostatic pressures up to 1.7 GPa for δ ≤ 0.30 but a nonlinear effect for δ > 0.30. These results, together with previous doping data, show a possible non-rigidity of the electron bands of the compounds, based on which previous phenomenological models of the pressure effect on Tc are built, and thus suggest only a restricted universality for the inverse parabolic relation between Tc and charge carriers per CuO2-layer. In other words, factors in addition to charge carriers can affect high temperature superconductivity.

KEY WORDS: high temperature superconductivity, pressure effect

1. INTRODUCTION

Immediately after the discovery of high temperature superconductivity (HTSΔ), extensive efforts were devoted to the search for systematic trends of the evolution of HTSΔ with various physical parameters. These trends form a useful basis for the development of a comprehensive microscopic theory of HTSΔ and for the continued quest for high temperature superconductors (HTSΔ's) with a higher transition temperature (Tc). One of the trends observed in the layered cuprates is the universal relationship between Tc and the number of charge carriers per CuO2-layer (n). It is known as the inverse parabolic universal relation \[ T_c(n) = T_{c,\text{max}}[1-a(n-n_{\text{opt}})^2], \] where \( T_{c,\text{max}} \) is the maximum Tc of the cuprate with an optimal n (n_{\text{opt}}). In other words, HTSΔ occurs only over a limited doping range for (n_{\text{opt}} - 1/\sqrt{a}) < n < (n_{\text{opt}} + 1/\sqrt{a}). Compounds with n < n_{\text{opt}} are known as underdoped, n > n_{\text{opt}} as overdoped and n = n_{\text{opt}} as optimally doped. The Tc(n) above with constants a = 82.6 and n_{\text{opt}} = 0.16 describes a wide variety of high temperature superconducting (HTSΔ) compounds examined [1] and therefore has been considered universal [1].

High pressure has been employed extensively [2] to obtain information about the occurrence of HTSΔ and to provide tests and to impose constraints on the universal Tc-relations and HTSΔ models. Most of the experiments were made on optimally doped compounds with a small but non-negligible pressure effect on their Tc (dTc/dP). Only a few were done on non-optimally doped samples. The results can be described by the phenomenological model on Tc(P) based on the combined pressure effects on n [3] and \( T_{c,\text{max}} \) [4], which give rise to an ever-decreasing dTc/dP with increasing n due to the former effect superimposed on a constant d\( T_{c,\text{max}}/dP \), consistent with the inverse parabolic universal relation Tc(n) [1]. Unfortunately, the large ultrahigh pressure-induced Tc-enhancement detected [5] in HgBa2CaM-1CuM O2M+2+δ [Hg-12(m-1)m] is too large to be accounted for by the phenomenological model on Tc(P) [3,4].

Recently, a large Tc-variation was achieved [6] in the newly discovered HgBa2CuO4+δ (Hg-1201) by anion-doping only without introducing possible chemical complications [7] by the combined cation- and anion-dopings adopted usually to acquire the similarly large Tc-change in other HTSΔ-system. The Tc of Hg-1201 varies with δ as \( T_c(\delta) = 97[1.26(\delta-0.22)^2] \) throughout the doping range of 0.03 ≤ δ ≤ 0.4 examined. In contrast to the expected similar parabolic relation of Tc(n) over the same δ-range due to the usual linear dependence of n on δ, such a behavior was observed [6] only over a limited range of δ ≤ 0.3, i.e. \( T_c(n) = 97[1.50(n-0.16)^2] \), for 0.02 ≤ n ≤ 0.23 only. The deviation of Tc(n) from the parabolic
dependence has been attributed to the possible new oxygen site being filled at high $\delta$. It should be noted that $n$ here was determined from the thermoelectric power measurements as was previously done [1]. Although the $n_{op} = 0.16$ for Hg-1201 is the same as for other HTSg compound systems, superconductivity takes place at a lower $n \sim 0.02$ in Hg-1201 than that of $\sim 0.05$ according to the universal inverse parabolic $T_c(n)$-relation.

We have therefore tested the universal $T_c(n)$ proposed by investigating the $\delta$-dependence of the hydrostatic pressure effect on $T_c$ of Hg-1201 with $0.07 \leq \delta \leq 0.39$ up to 1.7 GPa. We find a constant linear pressure effect on $T_c$ for $\delta < 0.30$ and a nonlinear effect for $\delta \geq 0.30$. The results raise questions concerning the universality of the inverse parabolic $T_c(n)$-relation [1] and the phenomenological $T_c(P)$-models [3,4]. The results suggest that the dopant affects $T_c$ through channels in addition to varying $n$, and that the electron-bands of HTSs may be less rigid than previously thought. Some of the results to be discussed have been recently submitted for publication elsewhere [8].

2. EXPERIMENTAL

The Hg-1201 samples investigated here were prepared by the controlled solid-vapor reaction technique [9]. The excess oxygen content $\delta$ was varied by annealing the Hg-1201 compound at a temperature between 240° and 500° C in an oxygen atmosphere of pressure ranging from 10$^{-8}$ to 500 bar. Detailed sample synthesis steps to achieve various dopings have been published previously [6].

The structure and phase purity were characterized by powder X-ray diffraction, employing a Rigaku D-MAX/BIII diffractometer. The $T_c$ at ambient was determined both resistively by the four-lead method using a Linear Research LR-400 bridge, and magnetically using a Quantum Design SQUID magnetometer. Under pressure, the $T_c$ was measured mostly resistively and for a few by the ac susceptibility technique. The hydrostatic pressure environment was generated by the modified Be-Cu clamp technique [10] inside a Teflon cup using the 3M fluorinert as the pressure medium. The pressure was measured with a superconducting Pb-manometer situated next to the sample inside the Teflon cell and the temperature by a chromel-alumel thermocouple above 30 K or a Ge-thermometer below 30 K.

3. RESULTS AND DISCUSSION

All Hg-1201 samples investigated are single-phase within the resolution of $\sim 4\%$ of our X-ray diffraction. The superconducting transition is narrow for the optimally doped sample with a width of $\sim 1 K$° and is broadened with a width of $\sim 10 K$° for the heavily underdoped sample, or $\sim 5 K$° for the heavily overdoped sample. The midpoint transition temperature ($T_c$), where the resistivity drops by 50% during the transition, is shown as a function of $\delta$ in Fig. 1. The value of $\delta$ is determined according to the $T_c(\delta)$-curve previously obtained [6], once the $T_c$ is measured.

![Fig. 1. $T_c(\delta)$ for the Hg-1201 examined.](image)

Under pressure, the superconducting transition width increases slightly for the sample with $\delta = \delta_o = 0.22$ but remains constant for those with $\delta \neq 0.22$. The onset, midpoint, and the offset temperatures of a sample respond to pressure in a similar general fashion. Therefore, we shall consider below only the pressure effect on the midpoint transition temperature $T_c$ of the samples with different $\delta$'s. The results are summarized in Fig. 2a for the underdoped (Nos. 1-3) and the optimally doped (No. 4) samples, and in Fig. 2b for the overdoped ones (Nos. 5-11). The numbers in these figures stand for the sequential order of increasing $\delta$ in the samples. They are, respectively, 0.07, 0.12, 0.19, 0.22, 0.27, 0.30, 0.31, 0.34, 0.36, 0.37, and 0.39. It is evident that pressure enhances $T_c$ linearly for samples with $\delta < 0.30$ and that it affects $T_c$ nonlinearly for those with $\delta \geq 0.30$. The data can be fitted as $T_c(P) = T_c(0) + \alpha P + \beta P^2$ and the linear pressure terms $\alpha$ and the quadratic pressure term $\beta$ can be obtained. They are displayed in Figs. 3a and 3b for different $\delta$'s. $\alpha$ is a constant of $2.0\pm0.2 K/GPa$ for $\delta \leq 0.27$, decreases rapidly to 0 near $\delta = 0.30$ and turns...