

High-pressure phase diagram of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ single crystalsXiao-Jia Chen, Viktor V. Struzhkin, Russell J. Hemley, and Ho-kwang Mao
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We have investigated the pressure dependence of the superconducting transition temperature T_c up to 18 GPa of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ single crystals ranging from the highly underdoped through the nearly optimally doped to the highly overdoped level. For all three samples studied, T_c is found to increase initially and then saturate at some critical pressure P_c , but decrease modestly with further increasing pressure. Oxygen doping has a tendency to reduce the increase of T_c and P_c . A high-pressure phase diagram between the saturated T_c and P_c is then obtained. Theoretical interpretation is given by using the competition between the hole carrier density and pairing interaction strength based on the high-pressure transport data of the resistivity and Hall coefficient in this system.

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I. INTRODUCTION

The doping dependence of the high-temperature superconductors (HTSCs) has become a significant issue due to pseudogap phenomena in underdoped compounds and to the peculiar influence of hole carrier density on properties of superconducting and normal states. Among the HTSCs discovered so far, $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$, $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$, and $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ are the most extensively studied systems due to available single crystals over a large range of doping levels and transition temperatures. $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ has a structural phase transformation between the underdoped and overdoped regimes.¹ Meanwhile, changing carrier density through cation substitution probably leads to local distortion due to the size effect.² Although the carrier density in $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ can be tuned by oxygen content, the occurrence of chain oxygen sites in this material complicates interpretation of much of the experimental data. Moreover, it is difficult to make even slightly overdoped samples in this system, only the underdoped part of the phase diagram can be thoroughly studied. By comparison, $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ is a perfect candidate for studying its physical properties over a wide range of the doping.

In order to get insight into the basic mechanism responsible for high-temperature superconductivity, one always strives to observe the possible change of the superconducting transition temperature T_c at fixed doping level. Pressure has been realized to be an effective way to change T_c for a compound. However, the effect of pressure on T_c is very complicated. The pressure derivative of T_c , dT_c/dP , can be varied from positive to negative, which strongly depends on the doping level. For most optimally doped compounds,³⁻⁹ T_c generally increases when pressure is applied at the initial stage, passes through a saturation at some critical pressure level P_c , then decreases modestly at high pressures. $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ is the only known system where the constantly positive dT_c/dP is not sensitive to oxygen content,¹⁰⁻¹² which makes it an ideal system to investigate the phase diagram between the saturated T_c and critical pres-

sure P_c over an entire doping regime. Early measurements on the 80~88 K samples^{13,14} have shown that for the sample with a relatively high T_c it is easy to reach the saturation at the beginning of the pressure range. It was also shown that the P_c shifts to higher pressure with additional oxygen content in the overdoped side.^{13,14} It remains unclear whether the saturation in the T_c versus P curve is present for samples away from optimal doping. Therefore, it is of interest to study the pressure dependence of T_c as well as the high-pressure phase diagram in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ over the entire doping range.

Experiments carried out on a variety of HTSCs^{5,15} have revealed that T_c is not a unique function of pressure, but depends strongly on the temperature at which the pressure is applied. For almost all systems, pressure-induced relaxation effects become activated for temperatures above 200 K. The pressure derivative dT_c/dP can be increased tenfold if the pressure is changed at room temperature rather than at low temperatures.¹⁶ Since the relaxation is believed to be frozen at low temperatures, the pressure dependence of T_c and the pressure derivative dT_c/dP basically represent the intrinsic pressure effect. Such an effect most directly reflects changes in the basic physical properties, including the pairing interaction itself, and thus is most valuable for comparison with theory. It is therefore of interest to perform detailed measurements of $T_c(P)$ under high pressure by changing pressure at low temperatures to elucidate the intrinsic pressure effects.

In this paper we present the results of a high-pressure study of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ single crystals in which the pressure is applied and measured at low temperatures. The characteristic nonmonotonic pressure dependence of the superconducting transition temperature is found in our crystals ranging from heavily underdoped to heavily overdoped samples. We obtain a high-pressure phase diagram between the saturation of T_c and critical pressure P_c at which T_c is saturated. We demonstrate that the nonmonotonic behavior originates from a competition between the hole carrier density and pairing interaction strength.

II. EXPERIMENTAL

Samples of single crystal $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ were grown by a self-flux technique in a strong thermal gradient to stabilize the direction of solidification using a stoichiometric ratio ($\text{Bi}:\text{Sr}:\text{Ca}:\text{Cu}=2:2:1:2$) of cations, as described elsewhere.¹⁷ Vacuum anneals are carried out in sealed quartz tubes to obtain the underdoped crystals. Overdoping has been accomplished using stainless steel cells sealed with samples immersed in liquid oxygen. Crystals used in the high-pressure studies are the highly underdoped (“vacuum-annealed,” $T_c=48$ K), air-annealed ($T_c=86$ K) and overdoped (“oxygen-annealed,” $T_c=60$ K) samples. A 10–90% transition width of 0.5 K in susceptibility of our samples is indicative of very good quality and of high homogeneity in both cation and oxygen concentrations.¹⁷

We performed the measurements of T_c under high pressures using a highly sensitive magnetic susceptibility technique with diamond anvil cells.¹⁸ This technique has been proven to be especially successful in the discovery of superconductivity in compressed sulphur and lithium.¹⁹ The technique is based on the quenching of the superconductivity and suppression of the Meissner effect in the superconducting sample by an external magnetic field. The magnetic susceptibility of the metallic parts of the high-pressure cells is essentially independent of the external field. Therefore, the magnetic field applied to the sample inserted in the diamond cell mainly affects the change of the signal coming from the sample. If we apply an alternating low-frequency ($f=20$ Hz) magnetic field with an amplitude below 100 Oe, then the output signal changes twice per period of the magnetic field because the positive and negative magnetic fields act identically to destroy the superconducting state. The resulting high-frequency output signal is modulated in amplitude at frequency $2f$. This modulation is observed only in the narrow range of temperatures close to the temperature of superconducting transition. The T_c is then identified as the point where the signal goes to zero because of the disappearance of the Meissner effect.

Samples are loaded in Mao-Bell cells made from hardened Be-Cu alloy. The gaskets are made of nonmagnetic Ni-Cr alloy. The $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ crystal (typical size $50 \times 50 \times 10 \mu\text{m}^3$) together with ruby chips are placed in the sample chamber. NaCl is loaded into the gasket hole with diameter $200 \mu\text{m}$ to serve as pressure medium. The pressure is applied and measured at low temperatures and is gauged by the R_1 fluorescence line of ruby.²⁰ Each initial pressure run was performed as single run at low temperature, pressure was changed immediately after the temperature scan was finished, and the sample was taken back to low temperature to start another temperature scan.

III. RESULTS

Figure 1 shows the representative temperature scans at different applied pressures for an underdoped $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ single crystal. Superconducting transition is identified as the temperature where the signal goes to zero on the high-temperature side, which is the point at which

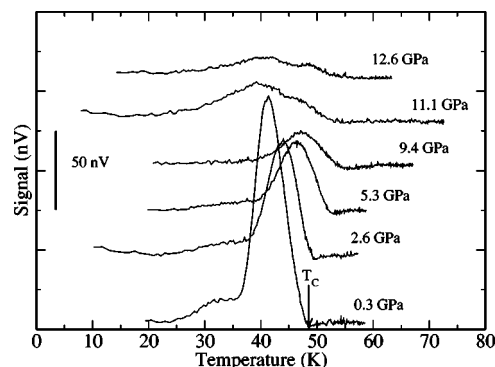


FIG. 1. Magnetic susceptibility signal in nV vs temperature for an underdoped $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ crystal at various pressures.

magnetic flux completely enters the sample. The superconducting transition of 48 K is obtained at ambient pressure. It is clear that at the pressure of 9.4 GPa the superconducting transition shifts to higher temperatures than at 1 atm, but it turns back beyond that pressure. The signal amplitude is suppressed by further increasing pressure. Also, at high pressures the shape of the signal broadens. In our measurements, we apply pressure in the increasing run and at low temperatures below 100 K. Therefore, we are confident for the existence of pressure medium over the whole measured range.

In Fig. 2, we plot the obtained T_c values in this underdoped $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ sample as a function of pressure. As shown, T_c is initially increased with applied pressure passing through a saturation value at around $P_c=10.5$ GPa and then decreases at higher pressures. A positive initial pressure derivative of 1.5 K/GPa is obviously consistent with the pure hydrostatic pressure measurements with the helium gas system.¹⁰ The nonmonotonic pressure dependence of T_c observed in the present heavily underdoped $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ is believed to be a common characteristic for all underdoped p -type copper oxides. This behavior has also been reported in the ~ 80 K $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ samples with nearly optimal dopings.^{13,14} For an underdoped $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ single crystal with $T_c=54$ K, which is close to our sample, Klotz and Schilling¹⁴ found an initial constancy of T_c up to 1.6 GPa as well as the absence of the saturation up to 6 GPa. This difference might be due to the difference of the samples and/or the difference in the hydrostaticity of the pressure medium used.

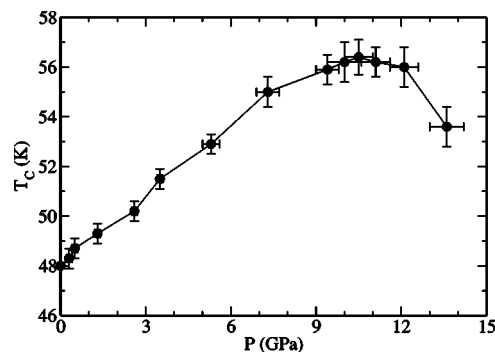


FIG. 2. Dependence of the superconducting transition temperature T_c on pressure in an underdoped $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ single crystal.

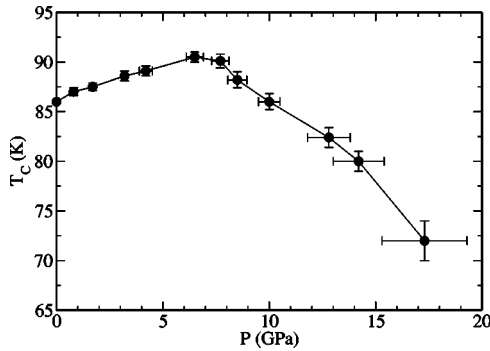


FIG. 3. Shift of the superconducting transition temperature T_c with applied pressure up to 18 GPa for a $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ single crystal, which is nearly optimally doped, but slightly overdoped.

The magnetic susceptibility curves for a nearly optimally doped, but slightly overdoped $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ single crystal, have a behavior similar to that in the underdoped sample. The superconducting transition is at 86 K at ambient pressure and shifts upward when pressure is applied at the beginning stage. Above 8.5 GPa, further pressure moves the transition to lower T_c values. The amplitude of the signal becomes weak with the applied pressure. It is hard to distinguish the signals from the sample or background at pressures above 18 GPa.

Shown in Fig. 3, the pressure dependence of T_c displays the characteristic behavior as is in other optimally doped HTSCs.³⁻⁸ Similar nonmonotonic pressure dependence of $T_c(P)$ with a saturation at a critical pressure near 2 GPa was reported in the sister bilayer $\text{Tl}_2\text{Ba}_2\text{CaCu}_2\text{O}_{8+\delta}$ system at the optimal doping.³ For the initial slope of the T_c versus P curve in Fig. 3 we derive $dT_c/dP=1.3$ K/GPa similar to that measured in the helium gas system.¹⁰ The saturation occurs at $P_c \approx 6.5$ GPa in our $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ sample at near optimal doping. This critical value is approximately the same as that in the $T_c \sim 80$ K crystal of Alekseeva *et al.*,¹³ but relatively higher than that for the $T_c=88$ K sample of Klotz and Schilling.¹⁴

We have measured the temperature dependence of magnetic susceptibilities near T_c for an overdoped $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ single crystal in both the compressing and decompressing runs. As plotted in Fig. 4, the susceptibility curves are nearly identical, regardless of the pressure history. In the decompressing run, T_c almost returns to its original ambient pressure value upon full release of the pressure. This is indicative of free relaxation in this sample.

From Fig. 4, it is found that as the applied pressure is increased, T_c continues to increase until it reaches a saturation value near 4 GPa. Previous studies¹⁴ on an overdoped $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ single crystal with a relatively high $T_c=80$ K have shown that the T_c also has a saturation value near $P_c=4$ GPa. Compared to the maximum value of $T_c \approx 90$ K in flux-grown $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ at optimal doping, the present sample with a T_c of 60 K is heavily overdoped. The occurrence of the saturation in the $T_c(P)$ curve in our sample is very interesting because it has not been reported in such a heavily overdoped cuprate where the significant change of T_c is tuned only through oxygen content rather

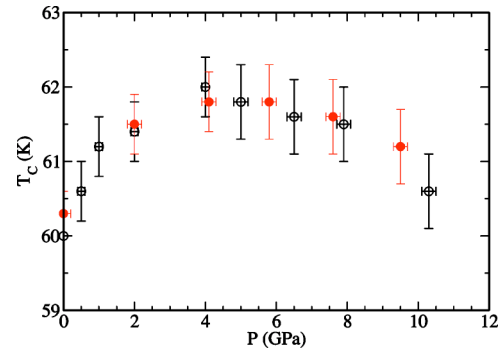


FIG. 4. (Color online) Superconducting transition temperature T_c for an overdoped $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ sample as a function of pressure. The open and solid circles represent the measurements in the compressing and decompressing run, respectively.

than cation substitution. Considering the fact that T_c would decrease with further adding oxygen content in the overdoped regime, this result clearly indicates that neither the pressure-induced charge transfer²¹ nor the pressure-induced oxygen doping²² is solely responsible for the pressure effect on T_c . The observed saturation in the $T_c(P)$ curve also suggests that another intrinsic variable independent of the hole carrier density is playing a significant role in enhancing T_c at even the heavily overdoped region.

Among the results obtained in these experiments, the most important finding is that the saturation with larger value than the ambient T_c occurs in the $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ samples over the entire doping range from the heavily underdoped through the optimally doped to the heavily overdoped. Both the critical pressure P_c and difference between the T_c and its saturated value decrease with the hole carrier density created by adding oxygen into the $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ system. This behavior is similar to that reported for the $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ system in the overdoped side²³ where the pressure-induced transition between the orthorhombic and tetragonal phases is accompanied when T_c reaches saturation.²⁴ Furthermore, we find the similar trend of $T_c(P_c)$ in the present $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ samples at different doping levels as observed in other optimally doped compounds.³⁻⁹ That is, the higher the P_c , the larger the saturation value of the T_c . We thus obtain an extended phase diagram at both the ambient condition and critical pressure, which is presented in Fig. 5. The saturation of T_c under the critical pressure of a hole-doped cuprate has been shown to cover the heavily underdoped to heavily overdoped regime. Despite extensive effort to reach saturation at the critical pressure, such a high-pressure phase diagram over the whole doping regime has not been established previously. The saturation $T_c(P_c)$ was only found in either the underdoped side⁵ or the overdoped side.^{14,23}

IV. DISCUSSION

To understand the experimentally observed nonmonotonic behavior of $T_c(P)$ of these samples, we use theoretical model for the layered d -wave superconductors.²⁵ For simplicity, we

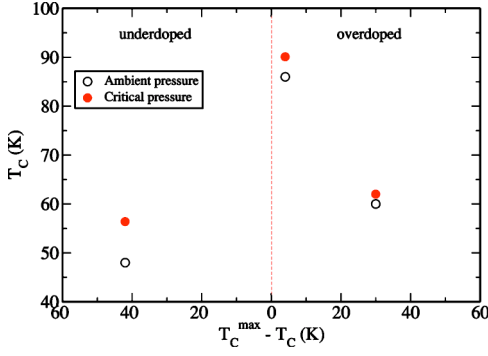


FIG. 5. (Color online) Plot of the superconducting transition temperature T_c at the ambient pressure (open circles) and critical pressure P_c (solid circles) in flux-grown $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ single crystals as a function of T_c referenced to the maximum value of 90 K.

calculate T_c for a given chemical potential μ by the following self-consistent equation:

$$1 = \frac{1}{2N} \sum_k \frac{Vg^2(k)}{|\varepsilon_k - \mu|} \tanh\left(\frac{|\varepsilon_k - \mu|}{2k_B T_c}\right), \quad (1)$$

where $g(k) = \cos k_x - \cos k_y$, ε_k is the quasiparticle dispersion, N is the number of k vectors, k_B is the Boltzmann constant, and V the in-plane pairing interaction strength. The constraint condition for the hole carrier density n_H in conjunction with μ is given by

$$n_H = \frac{1}{2} - \frac{1}{2N} \sum_k \tanh\left(\frac{|\varepsilon_k - \mu|}{2k_B T_c}\right). \quad (2)$$

The validity of this kind of d -wave BCS formalism in describing the superconducting state of HTSCs has been confirmed by recent angle-resolved photoemission spectroscopy (ARPES)²⁶ and transport^{27,28} measurements. As quasiparticle dispersion, we use²⁹ $\varepsilon_k = 0.166 \cos k_x \cos k_y + 0.046(\cos 2k_x + \cos 2k_y)$ eV, which corresponds to the hole dispersion in an antiferromagnetic background at half-filling, but should be approximately valid also at finite n_H as long as the antiferromagnetic correlation length is not negligible. A characteristic feature of this dispersion is the near flatness of the energy in the vicinity of $(\pi, 0)$, which is in good agreement with ARPES results for $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$.

The experimentally observed parabolic relation between T_c and n_H can be obtained by combining Eqs. (1) and (2) for given V . If one would consider the interlayer effect, the interlayer tunneling strength T_J is usually expressed as $T_J = t_\perp^2 / (16t)$ (Ref. 30), with t and t_\perp being the nearest-neighbor hopping integral between the in-plane and out-of-plane Cu atoms, respectively. For $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$, $t = 0.4$ eV and $t_\perp = 55$ meV are chosen from recent ARPES experiments.³¹ Taking the maximum T_c of 90 K for flux-grown $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$, we deduce $V = 0.03782$ eV. The resulting carrier densities n_H 's are then determined for the samples with various T_c values.

The temperature dependences of both the in-plane resistivity ρ_{ab} and Hall coefficient R_H under high pressures have

been suspected of giving clues to the pressure effects on T_c . For optimally doped samples at ambient pressure, the basic experimental trends are $\rho_{ab} \propto T$ and $R_H \propto 1/T$ in the high-temperature regime around room temperature. The quantity $R_H e z / v$ is inversely proportional to the hole carrier density n_H , where z is the number of Cu atoms per unit cell of volume v and e the carrier charge. In the high-temperature T -linear region, the resistivity satisfies the simple form $\rho_{ab} = (4\pi / \omega_p^2) \tau^{-1}$, where ω_p is the plasma frequency and the scattering rate $\tau^{-1} = 2\pi\Lambda T$ with Λ being the coupling strength between the charge carriers and some excitations. We take the Drude spectral weight $\omega_p^2 \sim n_H / m^*$ with m^* being the carrier effective mass. Systematic resistivity studies suggest that both the carrier scattering rate and coupling strength are independent of doping and nearly the same for a superconductor.³² The pairing interaction strength V appeared in Eq. (1) should have the same physical meaning as Λ . Therefore, we have $\rho_{ab} \propto (m^* / n_H) V T$ and $R_H \propto v / (n_H e z)$, which should account for the resistivity and Hall data at room temperature and under high pressures. Taking the pressure derivative of the quantities in the expressions of ρ_{ab} and R_H , we have the following relations:

$$\frac{d \ln \rho_{ab}}{dP} = \frac{d \ln m^*}{dP} - \frac{d \ln n_H}{dP} + \frac{d \ln V}{dP}, \quad (3)$$

$$\frac{d \ln R_H}{dP} = -\kappa_v - \frac{d \ln n_H}{dP}, \quad (4)$$

where the volume compressibility $\kappa_v \equiv 1/B_0 = -d \ln v / dP$ and B_0 is the bulk modulus.

For the present $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ system, we take $d \ln R_H / dP = -10.8 \times 10^{-2} \text{ GPa}^{-1}$ and $\kappa_v = 16.7 \times 10^{-3} \text{ GPa}^{-1}$ from the Hall coefficient³³ and neutron diffraction³⁴ measurements, respectively. Substituting these values into Eq. (4), we obtain $d \ln n_H / dP = 9.1 \times 10^{-2} \text{ GPa}^{-1}$. Studies of pressure effects on the irreversibility line³⁵ in a nearly optimally doped $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ reveal that the in-plane penetration depth λ_{ab} decreases from 220 to 205 nm when the applied pressure increases from 0 to 1.5 GPa, yielding $d \ln \lambda_{ab} / dP = -4.55 \times 10^{-2} \text{ GPa}^{-1}$. As it is known λ_{ab} can be expressed in the following relation: $\lambda_{ab}^2 = m^* / n_H$. We then get $d \ln m^* / dP = 2d \ln \lambda_{ab} / dP + d \ln n_H / dP$. According to this simple relation, we obtain the value of $d \ln m^* / dP$ of zero by using the experimental data for $d \ln \lambda_{ab} / dP$ and $d \ln n_H / dP$. It is therefore reasonable to neglect the pressure dependence of m^* . This indicates that the hole carrier density n_H and pairing interaction strength V are the two main intrinsic pressure variables responsible for pressure effect besides the cell volume v . This conclusion drawn from the normal-state transport properties agrees very well with the assumption in interpreting the pressure effect in the Y-Ba-Cu-O systems.³⁶

High-pressure transport measurements give $d \ln \rho_{ab} / dP = -7.5 \times 10^{-2} \text{ GPa}^{-1}$ for a nearly optimally doped $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ single crystal at 300 K.¹² Then we have $d \ln V / dP = 1.6 \times 10^{-2} \text{ GPa}^{-1}$ by using Eq. (3). This result implies $d \ln V / dP \approx \kappa_v$ in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$. Noting that the pressure-induced relative change of V satisfies a simple for-

malism $d \ln V/dP=(2\sim 4)\kappa_a$ in the Y-Ba-Cu-O systems,³⁶ where $\kappa_a \equiv -d \ln a/dP$ being the lattice compressibility along the a axis. The relationship between the pairing interaction strength and cell volume is then expressed by $\partial \ln V/\partial \ln v \approx -1$. We would like to mention that this probably universal relation does not rely on any model, at least in these two most generally studied HTSCs.

Since n_H and V are two intrinsic pressure variables, the variation of T_c with pressure is believed to be due to the change of the competition between them. Assuming that the pressure dependence of both n_H and V are not as linear in pressure P , but in the relative volume decrease given by $1-v(P)/v_0$, we can write $n_H(P)$ and $V(P)$ as

$$n_H(P) = n_H(0) \left[1 + \frac{d \ln n_H}{dP} B_0 \left(1 - \frac{v(P)}{v_0} \right) \right],$$

$$V(P) = V(0) \left[1 + \frac{d \ln V}{dP} B_0 \left(1 - \frac{v(P)}{v_0} \right) \right]. \quad (5)$$

The pressure dependence of the relative volume can be well described by the first-order Murnaghan equation of state $v(P)/v_0 = (1 + B'_0 P/B_0)^{-1/B'_0}$, where B'_0 is the pressure derivative of B_0 and v_0 the cell volume at ambient pressure. It has been shown that the construction of the pressure dependence of the intrinsic pressure variable through the relative volume change always reproduces an excellent agreement of the resulting curve with the experimental values.^{37,38} Olsen *et al.*³⁹ obtained $B'_0 = 6.0$ for Bi-based cuprates by fitting their high-pressure X-ray diffraction data up to 50 GPa. Their bulk modulus B_0 coincides well with that derived from the neutron diffraction technique.³⁴

Using the determined parameters B_0 , B'_0 , $d \ln n_H/dP$, and $d \ln V/dP$, we are able to evaluate the pressure dependence of T_c in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ samples in terms of Eqs. (1), (2), and (5). Here we have neglected the pressure dependence of the interlayer tunneling strength T_J . In the study of the c -axis pressure effect on T_c , Chen *et al.*⁴⁰ found that the pressure-induced change of T_J is relatively weak and negligible, compared to the pressure-induced change of n_H . Thus, the pressure enters the T_c equations only through $n_H(P)$ and $V(P)$.

The competition between the $n_H(P)$ and $V(P)$ provides a natural explanation for the observed high-pressure phase diagram as shown in Fig. 5. $V(P)$ is a favorable factor for the increase of T_c with pressure. While, $n_H(P)$ favors T_c only for the compound in the underdoped regime, and it becomes a negative factor in the optimally doped or the overdoped region. For the underdoped compound, $n_H(P)$ would be the negative factor once the pressure-induced increase of n_H passes through the optimal doping. The crucial parameter P_c is the crossover pressure where the factor(s) increasing T_c is(are) equal to that(those) decreasing T_c . A higher P_c in our heavily underdoped sample arises from both the positive contribution to T_c from $V(P)$ as well as the relatively long way for n_H to go before passing through the optimal doping. The nearly optimally doped compound has a modest P_c due to the favorable factor $V(P)$ but the unfavorable $n_H(P)$. Although $V(P)$ still factors the T_c increase for the heavily over-

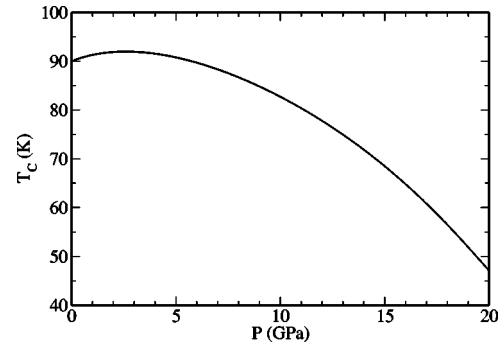


FIG. 6. Calculated variation of the superconducting transition temperature T_c with pressure for the optimally doped $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$.

doped material, the pressure-induced increase of n_H strongly suppresses such T_c increase. As a result, there only exists a smaller P_c for the heavily overdoped material. Therefore, the extended high-pressure phase diagram can be well understood on the basis of the competition of the two intrinsic pressure variables.

In Fig. 6 we plot the calculated variation of T_c as a function of pressure up to 20 GPa for the optimally doped $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ compound with $T_c = 90$ K at ambient condition. As can be seen, as pressure is increased, T_c increases initially until passing a saturation at critical pressure P_c of 2.7 GPa, and at higher pressures T_c decreases. This non-monotonic behavior of $T_c(P)$ of the optimally doped compound is consistent with the measurements in our nearly optimally doped sample as well as the 88 K sample of Klotz and Schilling.¹⁴ The theoretical value of the initial dT_c/dP is 1.69 K/GPa for the optimally doped material, which is in excellent agreement with the reported 1.5 ± 0.2 K/GPa from various measurements.¹⁰⁻¹² It is apparent that the competition between the hole carrier density and pairing interaction strength indeed captures the essential physics of pressure effect on T_c .

Figure 7 shows the critical pressure P_c dependence of the difference between the superconducting transition temperatures ΔT_c at the ambient pressure and critical pressure in the optimally doped $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ with those in some other optimally doped compounds.³⁻⁸ It can be seen that there exists the same trend of increasing the ΔT_c value with the increase of P_c in these optimally doped compounds. ΔT_c increases under P_c at the rate of near 1 K/GPa. It is indicated that any optimally doped compound has a saturation value of the maximum T_c which can be reached by applied pressure at critical P_c value. Moreover, it is confirmed that pressure is a most effective way to enhance T_c in optimally hole-doped cuprates. These results also suggest that the enhancement of the maximum T_c with pressure in the optimally doped HTSCs is not due to the increase of the pressure-induced hole carrier density.

As the above determined parameters are available only for the optimally or nearly optimally doped materials, we cannot calculate the change of T_c with pressure for various dopings by using the same parameters, especially for $d \ln n_H/dP$ and $d \ln V/dP$. There are not enough high-pressure transport data available in obtaining the information on these parameters at

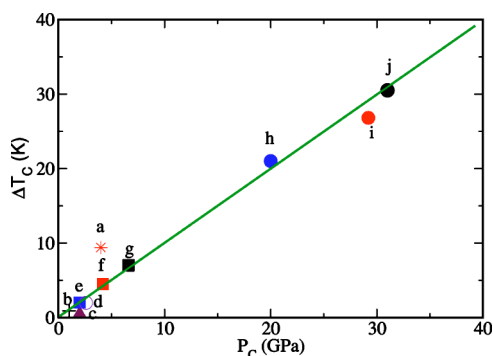


FIG. 7. (Color online) Critical pressure P_c dependence of the difference between the superconducting transition temperatures (ΔT_c) at the ambient pressure and critical pressure of optimally hole-doped HTSCs: a, $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ (star, Ref. 4); b, $\text{La}_{2-x}\text{Ca}_{1+x}\text{Cu}_2\text{O}_6$ (plus, Ref. 6); c, $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ (triangle, Ref. 5); d, $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ (open circle, this work); e, $\text{Tl}_2\text{Ba}_2\text{CaCu}_2\text{O}_{8+\delta}$ (Ref. 3); f, $\text{Tl}_2\text{Ba}_2\text{Ca}_2\text{Cu}_3\text{O}_{10+\delta}$ (Ref. 7); g, $\text{Tl}_2\text{Ba}_2\text{Ca}_3\text{Cu}_4\text{O}_{12+\delta}$ (Ref. 7); h, $\text{HgBa}_2\text{CuO}_{4+\delta}$ (Ref. 8); i, $\text{HgBa}_2\text{CaCu}_2\text{O}_{6+\delta}$ (Ref. 8); j, $\text{HgBa}_2\text{Ca}_2\text{Cu}_3\text{O}_{8+\delta}$ (Ref. 8). The line is a guide to the eyes.

the moment. However, we can estimate the pressure-induced change of carrier density n_H , if we assume that $d \ln V/dP$ is independent of oxygen content just as the interaction strength V is. Substituting the experimentally determined dT_c/dP of 1.5 K/GPa for $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$,¹⁰ we get the $d \ln n_H/dP$ values of 5.0×10^{-3} , 6.3×10^{-3} , and $2.5 \times 10^{-3} \text{ GPa}^{-1}$ for the underdoped, nearly optimally doped, and overdoped samples with $T_c=48$, 86, and 60 K, respectively. For comparison, the high-pressure transport data provide the $d \ln n_H/dP$ value of $9.1 \times 10^{-2} \text{ GPa}^{-1}$ for the optimally doped compound with $T_c=90$ K. This means that the optimally doped compound has the largest amount of the pressure-induced increase of the hole carrier density among

the samples with different doping level. However, as discussed above, the pressure effect on T_c in this material is still dominated by the pairing interaction strength.

V. CONCLUSION

We have measured the pressure dependence of the superconducting transition temperature T_c in flux-grown $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ single crystals by using a highly sensitive magnetic susceptibility technique with diamond anvil cells. In order to avoid the possible contribution from the pressure relaxation effect, we apply and measure pressure at low temperatures. The nonmonotonic pressure behavior of T_c is observed for all the samples, from the heavily underdoped through the nearly optimally doped to the heavily overdoped regime. We therefore obtain a high-pressure phase diagram between the saturation value of T_c and critical pressure P_c . Theoretical calculation based on a d -wave BCS formalism reproduces well this nonmonotonic behavior in terms of the high-pressure transport data. We have demonstrated that the pressure dependence of T_c in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ is the result of the competition between the hole carrier density and pairing interaction strength. The generally observed $T_c(P_c)$ in our samples indicates that the pairing interaction strength is mainly responsible for the pressure effect in this system.

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