Dependence of the superconducting transition temperature on the type and number of CuO_2 layers in $Tl_2Ba_2Ca_{n-1}Cu_nO_{2n+4-y}$

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The CuO_2 layer dependence of the superconducting transition temperature T_c at ambient pressure on the intrinsic transition temperature $T_c(i)$ of the type-I CuO_2 plane in which the copper atom has fivefold pyramid coordination of oxygen and the fourfold square coordinated type-II plane is studied in terms of the generalized Lawrence-Doniach theory. Calculations show that the increase of T_c with the number of CuO_2 layers benefits from the difference of the intrinsic $T_c(i)$ of the two types of CuO_2 layers and that interlayer coupling between the neighboring CuO_2 layers can enhance T_c for the multilayer cuprates. The upper limit of T_c is predicted to be 146 K for the bilayer thallium-based series. We present an extended pressure-induced charge transfer model for layered cuprate superconductors, assuming that the charge distribution among the crystallographically inequivalent CuO_2 layers is nonhomogeneous, which enables us to investigate the pressure effect on the intrinsic $T_c(i)$. The intrinsic $T_c(i)$ of the two types of CuO_2 layers is predicted to behave with pressure in a paraboliclike manner. For the optimally doped single, double, and triple CuO_2 sheets compounds, the saturation values of $T_c(I)$ of the type-I CuO_2 plane of 91.1, 119.6, and 133.9 K are obtained when $P=2.3$, 2.9, and 6.0 GPa, respectively. For the underdoped Tl-2234 compound with $T_c=113$ K, the calculated $T_c(I)$ of 118.5 K is obtained at $P=6.0$ GPa. Under the application of pressure, the intrinsic $T_c(II)$ of the type-II CuO_2 plane in Tl-2234 increases strongly compared with a modest increase of $T_c(II)$ in Tl-2223, possibly resulting from its underdoped nature. We suggest that at low pressure the T_c is the intrinsic $T_c(i)$ of the type-I CuO_2 plane, and at relatively high pressures the intrinsic effect of the type-II plane dominates. Our theoretical results are in agreement with experiments. [S0163-1829(99)04805-5]

I. INTRODUCTION

The bilayer thallium-based homologous series of $Tl_2Ba_2Ca_{n-1}Cu_nO_{2n+4-y}$ ($n=1$, 2, 3, and 4) [designated Tl-22$(n-1)n$] compounds has drawn particular interest because of the following features: (1) A negative pressure derivative of T_c was observed in the single layer compound Tl-2201,[1–3] in contrast to the positive derivatives generally found for hole-doped high-T_c superconductors (HTSC's).[4,5] (2) Multilayer material such as Tl-2223 or Tl-2234 shows a nonhomogeneous pressure effect on T_c,[6,7] initially increasing with increasing pressure, reaching a maximum, and then decreasing with further increasing pressure. After going through a minimum, T_c started to increase again. (3) The superconducting transition temperature T_c varies with the number n of CuO_2 layers. T_c is found to increase initially with n, with a maximum value of 90 K for Tl-2201,[8] 118 K for $n=2$ (Tl-2212),[9] and 128.5 K for $n=3$ (Tl-2223).[10,11] The fourth member (Tl-2234) has a somewhat lower T_c of 116 K.[12] There exist two types of CuO_2 planes which are crystallographically inequivalent when the number of CuO_2 layers is greater than 2. The one in which the Cu atom has a fivefold pyramidal coordination of oxygen is defined as a type-I CuO_2 plane. The other, that has a fourfold square coordinated CuO_2 plane, is called a type-II plane. Therefore, the bilayer thallium-based system provides a good opportunity to investigate the relationship between T_c and the number and the type of CuO_2 planes both at ambient pressure and under high pressure, i.e., the role played by each CuO_2 plane in the mechanism of high-T_c superconductivity.

The most obvious connecting link between T_c and the number or type of CuO_2 layers is the charge distribution among the CuO_2 layers, since these layers are generally believed to play the central role in high-T_c superconductivity. Kasowski, Hsu, and Herman[13] calculated the electronic structure of the four members of the bilayer thallium-based family, and found that the partial density of states of CuO_2 layers $N_{CuO_2}(E_F)$ is different for the two types of CuO_2 layers. Di Stasio, Müller, and Pietronero[14] obtained nonhomogeneous hole carrier distribution in the two types of CuO_2 layers within the sheet-charge model. Haines and Tallon[15] modified the sheet-charge model to a point-charge model, and concluded that the hole distribution is nearly uniform among the CuO_2 planes in Tl-2223 and inhomogeneous in Tl-2234. The ^{17}O NMR measurements on $(Bi,Pb)_2Sr_2Ca_2Cu_3O_{10-y}$ (Ref. 16) and ^{63}Cu NMR measurements on $HgBa_2Ca_2Cu_3O_{8+\delta}$ (Ref. 17) suggested that the hole concentration (n_H) is slightly higher in the type-I CuO_2 planes than that in the type-II plane. ^{63}Cu and ^{17}O NMR measurements on the triple-layered Tl-2223 brought controversial results. Early ^{17}O NMR measurements by Howes *et al.*[18] and ^{63}Cu NMR measurements by Han *et al.*[19] in optimal doped samples with $T_c=125$ K showed that there is a homogeneous charge distribution in the type-I and type-II CuO_2 planes. Recently, ^{63}Cu and ^{17}O NMR studies[20] in annealed ($T_c=123$ K) and as-grown ($T_c=115$ K) Tl-2223 sug-

gested that there are two different spin lattice relaxation rates for the type-I and type-II CuO_2 planes, suggesting a nonhomogeneous charge distribution in this system. Furthermore, the NMR data[21] of ^{63}Cu and ^{17}O in slightly overdoped ($T_c = 117$ K), nearly optimally doped ($T_c = 123$ K), and underdoped ($T_c = 104$ K) samples revealed that the hole concentration in the type-I CuO_2 plane is in each case larger than that in the type-II plane. The inhomogeneity of the charge distribution is weakened with the reduction of the hole concentration, and it may disappear in the highly underdoped regime. The calculation of the Cu valences for mercury-based[22,23] and bilayer thallium-based[24] series clearly demonstrates that the valences around Cu atoms in the type-I CuO_2 plane are slightly higher than that around Cu in the type-II plane. Recently, Zhou *et al.*[25] performed polarized micro-Raman-scattering measurements on the five members of the mercury-based family; they found that the oxygen-related phonon spectrum showed a systematic evolution with the number of CuO_2 layers. When increasing the layer number n above 2, the 590 cm^{-1} peak becomes unobservable while the 470 cm^{-1} peak begins to appear and eventually becomes dominant in Hg-1245. Using the self-consistent Born approximation, Yin and Gong[26] obtained quasiparticle bands for the multiple-layer superlattice t-J models, and found that the type-I CuO_2 planes are slightly less doped than the type-II ones.

A variety of theoretical models have been proposed in the study of the connection between T_c and the number of CuO_2 layers in the layered superconductors within the framework of interlayer coupling.[27–34] The results provide strong support for an interlayer coupling between the adjacent CuO_2 planes as the driving force of the large T_c enhancement in multilayer compounds. However, in these theoretical analyses, the topologically inequivalent CuO_2 planes are usually treated as having an equal contribution to T_c, without considering the possible inhomogeneity of charge distribution in the two types of CuO_2 planes. Despite considerable theoretical and experimental works so far, a thorough understanding of the influence of the number and type of CuO_2 layers on the transition temperature in homologous layered cuprates series is still lacking.

The pressure dependence of T_c of cuprate superconductors is of fundamental interest for elucidating the microscopic mechanism of the superconducting state as well as for finding new materials with higher critical temperature by using the chemical pressure. For multilayer cuprates, since the intrinsic hole concentration $n_H(i)$ in the type-I and type-II CuO_2 planes may not be the same, the pressure dependence of the intrinsic transition temperature $T_c(i)$ of these planes is expected to be different. Tristan Jover and co-workers[35,36] measured resistively the pressure dependence of T_c of Hg-1223 and Hg-1234 under quasihydrostatic conditions up to 30 GPa. The results of several high-pressure experiments on different samples of Hg-1223 and Hg-1234 showed that at low pressures T_c increases relatively fast, while at higher pressures T_c may either increase further, saturate, or even decrease, depending on the particular sample. They interpreted this behavior in terms of the model of Haines and Tallon.[15] They proposed that at low pressures the T_c is determined by the type-II CuO_2 plane(s), while at high pressures T_c is determined by the type-I CuO_2 planes. For the present Tl-2223 and Tl-2234, Tristan Jover *et al.*[7] measured resistively the pressure dependence of T_c up to 21 GPa, and interpreted quantitatively the nonhomogeneous pressure effect on T_c by using the model of Haines and Tallon.[15] Interestingly, they suggested that at low pressures the T_c of samples is determined by the intrinsic T_c of the type-I CuO_2 layers, while at higher pressures it is determined by that of the type-II CuO_2 layer(s). It is interesting to verify the intrinsic behavior of the type-I and type-II CuO_2 planes under the application of pressure. Among the theoretical models proposed in the study of pressure effects on T_c,[4,37] the pressure-induced charge transfer (PICT) model has been proved to be a simple but powerful tool for cuprate superconductors. It has been used to interpret successfully the observed pressure effects on T_c in HTSCs.[44,38–45] In the present paper we propose an extended PICT model for layered superconductors which can be used to predict the pressure dependence of the intrinsic transition temperature $T_c(i)$ for the two types of CuO_2 planes.

The outline of this paper is as follows. In Sec. II we study the effect of the type and number of CuO_2 planes on transition temperature on the basis of the generalized Lawrence-Doniach theory. We discuss the influence of T_c on both the difference of the intrinsic transition temperature $T_c(i)$ of the two types of CuO_2 planes and interlayer coupling for the bilayer thallium-based series. In Sec. III we present an extended pressure-induced charge-transfer model for layered cuprate superconductors to extract the inequivalent CuO_2 layers contribution to T_c. We investigate the pressure effect on the intrinsic transition temperature $T_c(i)$ of two types of CuO_2 planes in the $Tl-12(n-1)n$ ($n=1-4$) superconductors. Section IV contains ours conclusions and a brief summary.

II. INEQUIVALENT CuO_2 LAYER DEPENDENCE OF T_c^{max} FOR BILAYER THALLIUM-BASED SERIES

In the phenomenological model of Klemm,[46] layered superconductors are viewed as a stacked array of two-dimensional conducting CuO_2 layers, coupled together by weak Josephson tunneling between adjacent layers. Assuming that the order parameter field ψ_i is taken to be spatially homogeneous, no vector potential is present, and the coupling between interlayers in adjacent unit cells is weak compared with that within the unit cell and can thus be neglected, one writes the free energy density per unit cell as

$$f=\sum_{i=1}^{n}\left[\alpha_i|\psi_i|^2+\tfrac{1}{2}\beta_i|\psi_i|^4\right]+\lambda\sum_{i=1}^{n-1}|\psi_i-\psi_{i+1}|^2, \quad (1)$$

where

$$\alpha_I=\mu[T_c(n)-T_c(I)]/T_c(I),$$

$$\alpha_{II}=\nu[T_c(n)-T_c(II)]/T_c(II),$$

and β_I and β_{II} are the Ginsburg-Landau parameters for the type-I and type-II CuO_2 planes, respectively, $T_c(i)$ is the distinct intrinsic critical temperature corresponding to each ψ_i, $T_c(n)$ is the predicted superconducting transition temperature for layered cuprates with n CuO_2 layers, and λ the intracell interlayer coupling.

Following Birman and Lu,[28] we neglect all but the diagonal terms in the right-hand side of Eq. (1), and obtain the free-energy density in another gauge invariant form:

$$f=\sum_{i=1}^{n}\left[\alpha_i|\psi_i|^2+\tfrac{1}{2}\beta_i|\psi_i|^4\right]-\lambda\sum_{i=1}^{n-1}\psi_i\cdot\psi_{i+1}^{*}+\text{c.c.} \quad (2)$$

Minimizing f with respect to a variation in ψ_i^*, we have the matrix equation

$$\sum_{i=1}^{n}\left[\alpha_i\psi_i+\beta_i|\psi_i|^2\psi_i-\lambda(\psi_{i-1}+\psi_{i+1})\right]=0. \quad (3)$$

Here $|\psi_i|^2$ denotes the carrier concentration of the ith layer. Suppose the maximum T_c is independent of the $|\psi_i|^2$ term when the layered compounds are optimally doped, we can take the value of β_i to be zero.[28] Then Eq. (3) reduces to

$$\sum_{i=1}^{n}\left[\alpha_i\psi_i-\lambda(\psi_{i-1}+\psi_{i+1})\right]=0. \quad (4)$$

The Ginsburg-Landau parameter α_i is defined by the determinant

$$\begin{vmatrix}
\alpha_I & -\lambda & 0 & \cdots & & & & \\
-\lambda & \alpha_{II} & -\lambda & 0 & \cdots & & & \\
0 & -\lambda & \alpha_{II} & -\lambda & \ddots & & & \\
\vdots & \ddots & -\lambda & \ddots & \ddots & & & \\
 & & & \ddots & \ddots & \ddots & \ddots & \vdots \\
 & & & & \ddots & \alpha_{II} & -\lambda & 0 \\
 & & & & \ddots & -\lambda & \alpha_{II} & -\lambda \\
 & & & & \cdots & 0 & -\lambda & \alpha_I
\end{vmatrix}=0. \quad (5)$$

This equation has n roots, and the superconducting state is defined by the lowest value of α_i^j $(j=1,2,\ldots,n)$.[32] If a homogeneous contribution to T_c is considered for the two types of CuO_2 planes, one can put $\alpha_I=\alpha_{II}$ [i.e., $\mu=\nu, T_c(I)=T_c(II)$]. Thus a remarkably simple expression for $T_c(n)$ is found,

$$T_c(n)=T_c(i)\left[1+2\frac{\lambda}{\mu}\cos\left(\frac{\pi}{n+1}\right)\right], \quad (6)$$

where $T_c(1)$ is the critical temperature of the single-layer compound, and its value is just equal to $T_c(i)$. It is interesting that our result agrees well with the work of Byczuk and Spaled based on microscopic theory.[33]

For the case $\alpha_I\neq\alpha_{II}$, that is, the superconducting transition temperature T_c of layered cuprates depends on the type of CuO_2 layers, one can rewrite Eq. (5) in the following form:

$$n=1, \quad \alpha_I=0,$$

$$n=2, \quad \alpha_I^2-\lambda^2=0,$$

$$n=3, \quad \alpha_I\alpha_{II}-2\lambda^2=0,$$

$$n\geq 4,$$

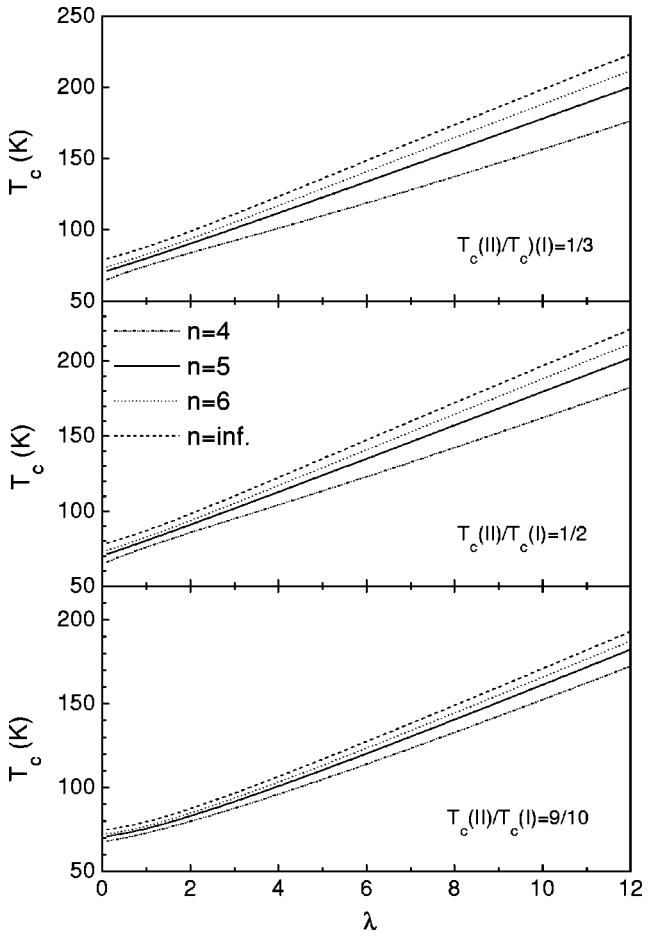

FIG. 1. Relationship between T_c and the interlayer coupling parameter for the ratio of distinct intrinsic transition temperature of inequivalent CuO_2 planes $T_c(II)/T_c(I)=\frac{1}{3}$, $\frac{1}{2}$, and $\frac{9}{10}$ as a function of the number of CuO_2 layers in bilayer thallium-based compounds.

$$\alpha_{II}(\alpha_I-\lambda^2)^2-2\lambda\left[\alpha_I^2\cos\left(\frac{\pi}{n-1}\right)-2\lambda^2\alpha_I\cos\left(\frac{\pi}{n-2}\right)+\lambda^2\cos\left(\frac{\pi}{n-3}\right)\right]=0, \quad (7)$$

To illustrate the above, we investigated the dependence of T_c on the type and number of CuO_2 layers at ambient pressure in multilayer cuprates. Based on the experimental values of T_c for the single CuO_2 layer compound Tl-2201 and double CuO_2 layers compound Tl-2212,[8,9] one can obtain the intrinsic $T_c(I)=T_c(1)=90$ K and the parameter λ/μ of 0.31 by using Eq. (7). In the multilayer cuprates with $n\geq 3$, there exist two types of inequivalent CuO_2 planes per unit cell. Choosing $T_c(II)/T_c(I)=\frac{1}{3}$, $\frac{1}{2}$, and $\frac{9}{10}$, Eq. (7) yields the corresponding λ/ν of 2.25, 1.27, and 0.40 according to the T_c (3) of 128.5 K for the triple Tl-2223 phase.[10] It is shown that for a given value of the number n of CuO_2 layers, T_c exhibits a significant variation with interlayer coupling λ.

In Fig. 1, T_c vs λ is plotted for different values of $T_c(II)/T_c(I)$. It is clear that the transition temperature T_c increases with λ. At any λ, T_c is an increasing function of n, reaching 95% of the maximum possible value $T_n(\infty)$ by $n=5$. The larger the difference between the intrinsic $T_c(II)$ and $T_c(I)$, the stronger the increase of T_c with n. This be-

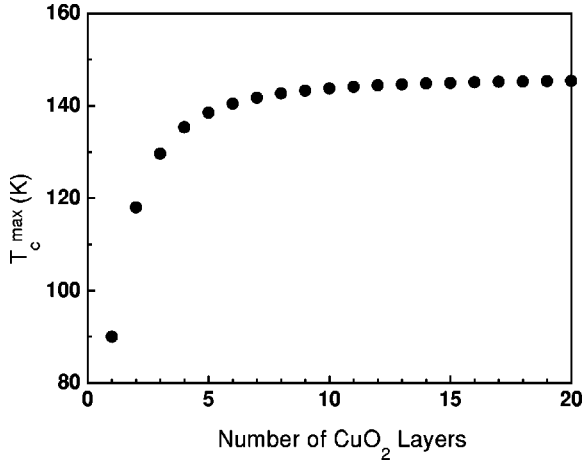

FIG. 2. Plot of the maximum T_c vs the number of CuO_2 layers in bilayer thallium-based compounds.

havior looks quite reasonable because the greater the inhomogeneity of carrier distribution among the inequivalent CuO_2 planes, the larger the difference between the distinct intrinsic $T_c(II)$ and $T_c(I)$.

From Eq. (6), we have seen that $T_c(n)$ can be obtained from the observed values of the T_c's of single and double CuO_2 layer compounds. The predicted superconducting transition temperature T_c as a function of number of CuO_2 layers is presented in Fig. 2. As shown, T_c increases initially with increasing the layer number n, and reaches saturation when n tends to infinity. The upper limit of T_c for the bilayer thallium-based cuprates is 146 K. Our present theoretical study shows that interlayer coupling can enhance the superconducting transition temperature for the multilayer systems. It is therefore possible that, at large number n, T_c maintains a relatively high value because of the existence of the important coupling of CuO_2 planes, even in spite of the depletion of intrinsic hole concentration $n_H(II)$ in the type-II CuO_2 plane. This is supported by detailed experiments on the $T_c \sim n$ correlation in both thallium and mercury-based series.[47,48]

III. PRESSURE EFFECTS ON INTRINSIC $T_c(I)$ FOR TWO TYPES OF CuO_2 PLANES

A. Pressure-induced charge transfer model for layered cuprates

The general structure of hole-doped layered cuprates consists of an alternate stack of a charge reservoir (CR) block which contains $(M\text{-O})_m$ (e.g., Bi_2O_2, Tl_2O_2, TlO, HgO_δ, etc.; m is the number of M-O planes) planes and a so-called infinite-layer (IL) block in which n CuO_2 planes and $n-1$ alkaline-earth (typically Ca) atomic planes are alternately stacked. There are two R-O (R is Ba, Sr, La, etc.) planes with a rocksalt structure on both sides of the CR block. Thus all the layered cuprates can be represented by the general formula, $M-m2(n-1)n$. Taking δ to be the total charge of electrons in the $(M\text{-O})_m$ layer(s), one can obtain a density δ of holes to be shared among all the CuO_2 planes. If one defines $n_H(II)=\delta x$ as the hole carriers concentration on each of the type-II CuO_2 plane(s) in the layered cuprates, this gives $n(I)=[1-(n-1)x]\delta/2$ for the charge on each of the two type-I planes. The fraction of the total number of carriers on each of the type-II CuO_2 planes has a form within the framework of sheet-charge model for layered systems:[14,34]

$$x=\frac{1}{n+(n-2)A}, \tag{8}$$

where $A=4e^2d_0m^*/\varepsilon\hbar^2$. The geometric parameter d_0 denotes the distance between two adjacent CuO_2 layers within the unit cell, m^* is the effective mass of the electrons, and ε the medium dielectric constant.

It is now well known that the superconducting transition temperature T_c in cuprate superconductors varies approximately in an inverted parabolic manner with the hole concentration n_H in the CuO_2 planes.[49–51] Below a certain minimum hole concentration $n_H^{\min}$ the compounds are not superconducting, and in this region they show nonmetallic behavior. As the hole concentration n_H is increased, T_c increases, reaching a maximum value $T_c^{\max}$ at an optimal hole concentration $n_H^{\rm opt}$. For still higher values of the hole concentration, T_c starts to decrease and eventually goes to zero. For the layered cuprates, this behavior can be expressed by the following equation:

$$T_c(i)=T_c^{\max}(i)\{1-\beta[n_H^{\rm opt}-n_H(i)]^2\}. \tag{9}$$

Here $T_c(i)$ $(i=I,II)$ and $T_c^{\max}(i)$ $(i=I,II)$ are the intrinsic transition temperature and their maximum values of the type-I and type-II CuO_2 planes, respectively.

Considerable effort has been devoted in recent years to elucidate the pressure dependence of the superconducting transition temperature of cuprate superconductors. It is generally believed that the pressure dependence enters T_c through the two independent variables $T_c^{\max}$ and n_H. The variation in the maximum value of transition temperature at optimal doping ($T_c^{\max}$) among the various cuprate superconductors[52–54] indicates that $T_c^{\max}$ should be a pressure-sensitive variable. Some reasons such as interlayer coupling between the two neighboring CuO_2 layers, the effective interaction strength, phonon frequency, etc., are believed to contribute to changes in $T_c^{\max}$.[55–59] Measurements of Hall effect[60] and thermopower[61] under high pressure, pressure-induced structural changes,[62] bond-valence-sum calculations,[63] and electronic structure calculations[64–67] demonstrate that the mobile hole carrier concentration in the CuO_2 plane increases under the application of pressure. Assuming that β and $n_H^{\rm opt}$ are independent of pressure, the full derivative of T_c with respect to pressure P is obtained from Eq. (9)

$$\frac{dT_c(i)}{dP}=\frac{T_c(i)}{T_c^{\max}(i)}\frac{dT_c^{\max}(i)}{dP} + 2\beta T_c^{\max}(i)[n_H^{\rm opt}-n_H(i)]\frac{dn_H(i)}{dP}. \tag{10}$$

In order to generalize Eq. (9) to include the effect of pressure, we assume that both the $T_c^{\max}(i)(P)$ and $n_H(i)(P)$ vary with P in the following forms:

$$T_c^{\max}(i)(P) = T_c^{\max}(i) + \frac{dT_c^{\max}(i)}{dP} P \tag{11}$$

and

$$n_H(i)(P) = n_H(i) + \frac{dn_H(i)}{dP} P. \tag{12}$$

Hence the variation of the intrinsic $T_c(i)$ with pressure can be expressed as

$$T_c(i)(P) = T_c^{\max}(i)(P)\{1 - \beta[n_H^{\rm opt} - n_H(i)(P)]^2\}. \tag{13}$$

The pressure derivative of $T_c(i)(P)$ under pressure is then obtained from Eq. (13):

$$\frac{dT_c(i)(P)}{dP} = \frac{T_c(i)(P)}{T_c^{\max}(i)(P)} \frac{dT_c^{\max}(i)}{dP} + 2\beta T_c^{\max}(i)(P)[n_H^{opt} - n_H(i)(P)] \frac{dn_H(i)}{dP}. \tag{14}$$

From Eqs. (11)–(14), one notices that the change of pressure-induced maximum value of intrinsic $T_c(i)$ significantly affects the variation of $T_c(i)(P)$ and $dT_c(i)(P)/dP$ with pressure for the two types of CuO_2 layers. As a definition of $T_c^{\max}(i)$, one chooses the initial pressure derivative of $T_c^{\max}$ for layered cuprates as $dT_c^{\max}(I)/dP$ for the type-I CuO_2 planes. The crystal structure of the IL compound contains two-dimensional CuO_2 layers separated from each other by alkaline-earth atoms E [E=(Ca, Sr), Sr, (Sr, Ba), etc.].[68,69] In this structure the CuO_2 sheet has no apical oxygen, which is just like the type-II CuO_2 plane in layered cuprates. Thus it is reasonable to take the initial slope of $dT_c^{\max}/dP$ in hole-doped IL materials like $dT_c^{\max}(II)/dP$ for the type-II CuO_2 planes.[70]

It should be noted that at present it is very hard to determine the intrinsic $dn_H(i)/dP$ value which possibly depends on both the type of CuO_2 planes and hole concentration. Gupta and Gupta[64] obtained $dn_H/dP = 1.13\times10^{-2}$ holes/GPa for $La_{1.85}Sr_{0.15}CuO_4$ calculated from structural data determined by neutron analysis using a cluster model. Jorgensen *et al.*[63] calculated $dn_H/dP = 6.5\times10^{-3}$ holes/GPa for $YBa_2Cu_3O_{6.93}$ using the bond valence sum method, while a modest dn_H/dP of 3.46×10^{-3} holes/GPa for the $YBa_2Cu_3O_7$ compound was reported by Gupta and Gupta[65] using a cluster model. The slightly lower value of dn_H/dP of 2.17×10^{-3} holes/GPa was obtained for the triple Hg-1223 system in a local-density approximation.[66] Recently, Chen and Jiao[43] reported a value of dn_H/dP of 1.51×10^{-3} holes/GPa for a $HgBa_2CaCu_2O_{6.22}$ compound using the bond-valence-sum model. Since there is no calculation for the value of dn_H/dP for the thallium-based class superconductors so far, we tentatively use $dn_H/dP = 5.0\times10^{-3}$ holes/GPa for the single- and double-sheet CuO_2 thallium-based compounds, in which there exists only the type-I CuO_2 plane. For the multilayer thallium-based compounds, we take the $dn_H(i)/dP$ value of 3.5×10^{-3} holes/GPa for each CuO_2 plane in our calculation.

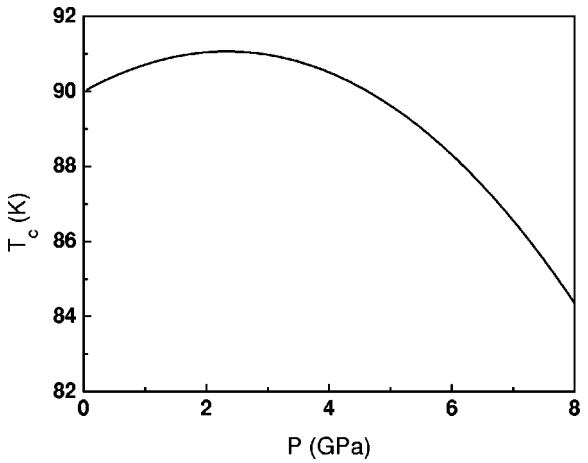

FIG. 3. Pressure dependence of T_c in the optimally doped Tl-2201 compound with $T_c = 90$ K up to 8 GPa.

In the following, we will concentrate on the charge distribution and pressure effects on the intrinsic T_c among the inequivalent CuO_2 layers in the typical bilayer thallium-based superconductors $Tl_2Ba_2Ca_{n-1}Cu_nO_{2n+4-y}$ ($n=1$, 2, 3, and 4). The hole sources in the bilayer thallium-based compounds contain some combination of thallium and calcium or thallium and copper, vacancies on the thallium site, and excess oxygen.[71] The partial replacement of trivalent thallium by divalent calcium and/or copper, or the deficiency of thallium, might explain the somewhat lower oxygen content values observed in this series. There are other possible origins of hole doping such as charge transfer between TlO and CuO_2 layers, which leads to a mixed-valency state of Tl^{3+} and Tl^{+}. Such coexistence of Tl^{3+} and Tl^{+} ions was proposed in an x-ray photoemission spectroscopy study of Tl-2223,[72] while only a Tl^{3+} signal was detected by ^{205}Tl NMR measurements.[73] Furthermore, both the wet chemical route[74] and x-ray absorption near edge spectra measurement[75] reveal that the thallium valence remains constant and close to +3.0. Wet chemical analyses show the optimal hole concentration $n_H^{\rm opt}$ is 0.14 holes in the thallium-based series.[50,76] Sinclair *et al.*[77] determined very precisely the cation content using combined powder neutron and resonant x-ray diffraction, and confirmed this value. Extensive experiments revealed that $\beta = 82.6/\text{holes}^2$ is universal for many hole-doped HTSC's.[51] In the present analysis we choose $n_H^{\rm opt} = 0.14$ holes and $\beta = 82.6/\text{holes}^2$ for bilayer thallium-based compounds. In this way, T_c appears to be maximized at $n_H^{\rm opt} = 0.14$ holes, and falls to zero on the underdoped and overdoped sides at $n_H^{\min} = 0.03$ holes and $n_H^{\max} = 0.25$ holes, respectively.

B. Results and discussion

1. $Tl_2Ba_2CuO_{6-\delta}$ and $Tl_2Ba_2CaCu_2O_8$ systems

Calculation of $T_c(P)$ was carried out for the optimally doped Tl-2201 compound with $T_c = 90$ K. Based on the work of Sieburger and Schilling,[2] we assume $dT_c^{\max}/dP = 0.9$ K/GPa. With these parameters, from Eq. (13) we can calculate the variation of T_c with pressure. The results are shown in Fig. 3. As can be seen, T_c increases modestly with pressure initially, and saturates at 91.1 K when $P = 2.3$ GPa. For

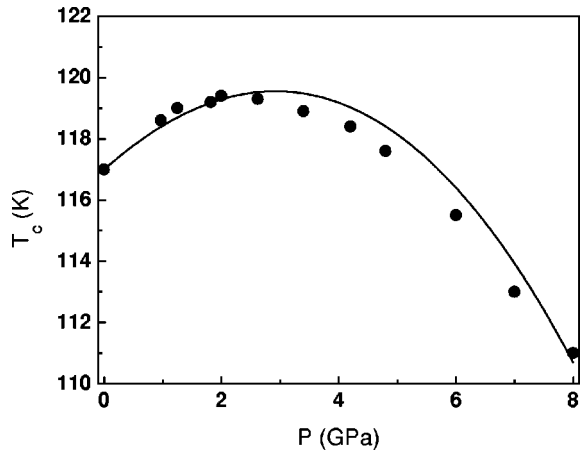

FIG. 4. Pressure dependence of T_c in the nearly optimally doped but slightly overdoped Tl-2212 compound with $T_c = 117$ K up to 8 GPa.

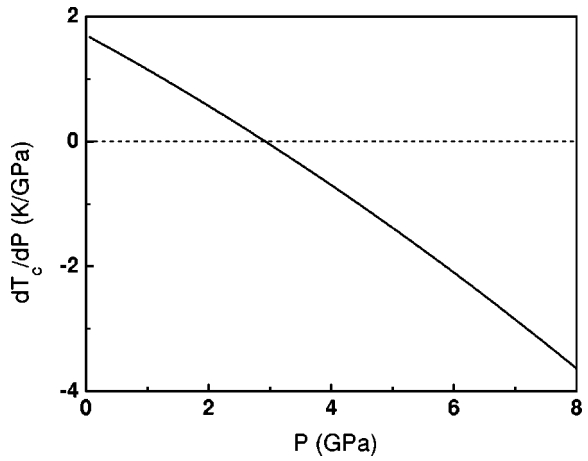

FIG. 5. Calculated values of $dT_c(P)/dP$ with pressure up to 8 GPa in the Tl-2212 compound with $T_c = 117$ K.

higher pressures T_c begins to decline, and reaches a value of 84.4 K at 8 GPa. In contrast, in a single CuO_2 layer Hg-1201 compound T_c strongly increases from 97 at $P=0$ to a maximum of 118 K at 24 GPa, with an initial slope of 1.7 K/GPa.[78] Since experimental values of T_c are not available beyond $P=0.6$ GPa in the present single CuO_2 layer Tl-2201 compound, further experiments are required to compare our theoretical prediction.

For the second member of bilayer thallium-based series, i.e., Tl-2212, we take the experimental data of $T_c(P)$ obtained by Môri *et al.*,[1] and compare with results of the following calculations. Now we evaluate $T_c(P)$ numerically as a function of pressure for a nearly optimally doped but slightly overdoped Tl-2212 compound with $T_c = 117$ K. The maximum $T_c^{\max}$ of 118 K (Ref. 9) is attained at an optimal hole concentration $n_H^{\rm opt} = 0.14$ holes. The hole concentration can be determined by Eq. (9) as $n_H = 0.15$ holes. The maximum pressure derivative $dT_c^{\max}/dP$ is then calculated from Eq. (10), which yields a value of 2.71 K/GPa. Based on these parameters, we calculated T_c from Eq. (13) as a function of pressure P in the range of $0 \leqslant P \leqslant 8$ GPa. In Fig. 4 we present the pressure dependence of T_c for Tl-2212. As can be seen, as pressure is increased T_c increases initially, then reaches a maximum at some pressure level, and decreases at higher pressures. For comparison, data from Môri *et al.*[1] are displayed. We found that the experimental data points roughly lie on our theoretical curve. T_c already reaches a maximum value of 118 K at normal pressure of the Tl-2212 cuprates. For a pressure of 2.0 GPa, T_c is calculated to be 119.3 K, in excellent agreement with the work of Môri *et al.*[1] The maximum T_c of 119.6 K on the $T_c \sim P$ curve occurs at 2.93 GPa. Susceptibility studies[79,80] revealed that $T_c(P)$ passes through a maximum at 3 GPa after increasing by ~ 2.5 K. Interestingly, our prediction coincides well with these experiments.

Figure 5 shows the pressure dependence of the pressure derivative of T_c evaluated from Eq. (14) for a nearly optimally doped but slightly overdoped Tl-2212 compound with $T_c = 117$ K. The initial pressure derivative dT_c/dP is 1.7 K/GPa. The value of $dT_c(P)/dP$ decreases with increasing pressure, and changes sign from positive to negative at the maximum $T_c(P)$. At the pressure level of 8 GPa, the calculated value of dT_c/dP is -3.6 K/GPa, compared to the experimental value of -2.3 K/GPa.[1]

2. $Tl_2Ba_2Ca_2Cu_3O_{10-y}$ system

According to Eqs. (8) and (9), one can determine the intrinsic $T_c(i)$ values of the two types of CuO_2 planes normalized with respect to $T_c^{\max}$. Parameter A in Eq. (8) denotes the relative contribution of the electrostatic and band energies. Substituting the values of $d_0 = 3.16$ Å,[71] and $m^*/m_e\varepsilon \simeq 0.33$,[14] 0.09, and 0.045 in the definition of A, one attains corresponding values of A of 8.8, 2.4, and 1.2, respectively. So far, there is not an effective way to distinguish the intrinsic $T_c^{\max}(i)$ among the inequivalent CuO_2 layers; we assume their values are equal at ambient pressure. In the $Tl_2Ba_2Ca_2Cu_3O_{10-y}$ system, the maximum $T_c^{\max}$ is 128.5 K (Ref. 10) obtained at $n_H^{\rm opt} = 0.14$ holes.[50,76] Figure 6 shows the normalized intrinsic $T_c(i)$ values for the type-I and type-II CuO_2 planes of Tl-2223 as a function of total hole concentration δ for $A = 8.8$, 2.4, and 1.2, respectively. Three clear parabolas are presented for the type-I CuO_2 planes, and the optimal carrier concentration $\delta^{\rm opt}$ pushes backward from 0.3 through 0.4 to 0.5 with decreasing the value of A. Additionally, the width of the parabola expands with the decrease in the value of A. One can also see that the type-II CuO_2 plane possesses a broad parabola, and the peak in the curve shifts to higher carrier concentration with increasing A. For the case of $A = 8.8$, superconductivity occurs in the type-II CuO_2 plane for values of δ above 0.35. With a further increase of δ above 0.7, the type-II CuO_2 plane is still superconducting while the type-I CuO_2 planes are no longer superconducting. For $A = 1.2$, the type-I CuO_2 planes become superconducting for δ values between 0.11 and 0.95, reaching a maximum T_c at $\delta = 0.5$. The type-II CuO_2 plane behaves in a similar way: it is superconducting in the broad range 0.12–1.05, with a maximum value of $T_c(II)$ at a large δ of 0.6. When $A = 1.2$, one finds $n_H(I) = 1.1 n_H(II)$, which is in agreement with the ^{63}Cu and ^{17}O NMR measurements.[16,17,20,21] Since there is not an available value of $m^*/m_e\varepsilon$ for cuprates, we choose $A = 1.2$ in the following.

Choosing the experimentally observed values as the intrinsic $T_c(I)$ and $dT_c(I)/dP$ of the type-I CuO_2 plane, one

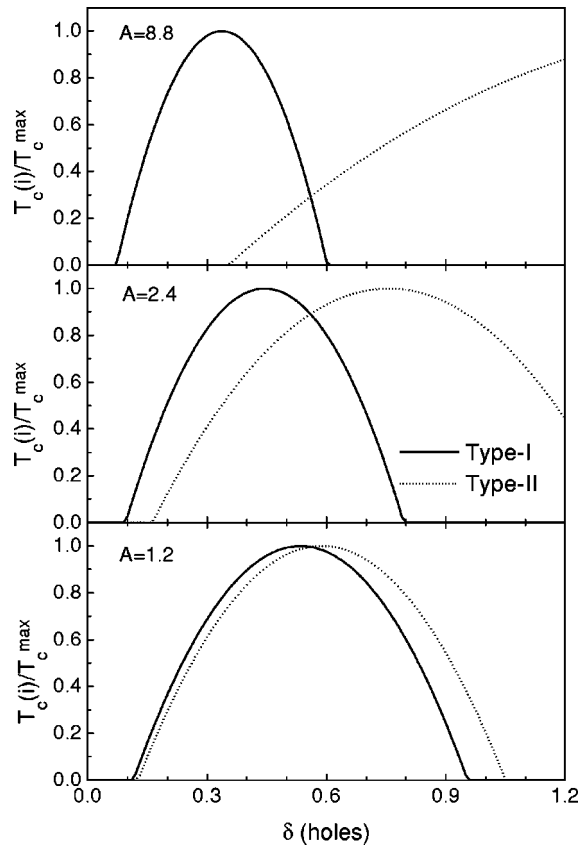

FIG. 6. Normalized intrinsic $T_c(i)$ values for the inequivalent CuO_2 planes in the Tl-2223 system as a function of carrier concentration for $A=8.8$, 2.4, and 1.2, respectively.

gains $dT_c^{max}(I)/dP=1.75$ K/GPa for the optimal material.[7] The effect of pressure on T_c in IL cuprates has been reported by various laboratories.[59] Takahashi *et al.*[70] showed that for $(Sr_{0.3}Ca_{0.7})_{0.95}CuO_2$, T_c increases up to 120 K for 8 GPa at a rate of $dT_c/dP=0.7$ K/GPa. It is therefore reasonable to take $dT_c^{max}(II)/dP=0.7$ K/GPa for the type-II CuO_2 plane in the layered cuprate superconductors due to the similar structure of the CuO_2 sheet for the IL and layered HTSC's.

The pressure dependence of the intrinsic $T_c(i)(P)$ for an optimally doped Tl-2223 compound with $T_c=128.5$ K can be evaluated by using Eqs. (11)–(13). Hence the intrinsic $T_c(I)=128.5$ K, and the corresponding intrinsic $n_H(I)=0.14$ holes for the type-I CuO_2 planes. The intrinsic hole concentration for the type-II CuO_2 plane can be determined by Eq. (8) as $n_H(II)=0.127$ holes; the intrinsic $T_c(II)$ is then calculated from Eq. (9), which yields a value of 126.7 K. From the parameter values determined above, we calculated T_c as a function of pressure P in the range of $0\leqslant P\leqslant 20$ GPa. Figure 7 gives the results for the inequivalent CuO_2 planes. We can see that the intrinsic $T_c(i)$ changes with pressure in a parabolic manner. There exist two different parabolas for the two different types of CuO_2 planes, which cross at 12 GPa. However, the maxima for the type-I and type-II CuO_2 planes occur at nearly the same pressure level. The maximum $T_c(I)$ of the type-I planes is 133.9 K at 6 GPa, in good agreement with Tristan Jover and co-workers,[7,11] who measured a maximum value of 133 K at 4.2 GPa. At 7.4 GPa, the calculated value of $T_c(I)$ is 133.6 K for the type-I CuO_2 planes, compared with 131.5 ± 0.5 K as

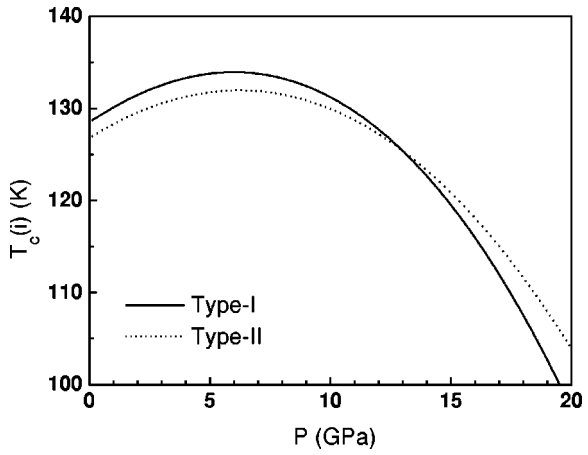

FIG. 7. Pressure dependence of the intrinsic $T_c(i)$ for Tl-2223 with $T_c=128.5$ K. The solid and dotted curves denote the predicted values of $T_c(i)(P)$ for type-I and type-II CuO_2 planes, respectively.

found by Berkley *et al.*[81] In the type-II CuO_2 plane, the maximum value of intrinsic $T_c(II)$ is 131.9 K at 6.2 GPa which is slightly lower than that in the type-I CuO_2 planes. The lower maximum $T_c(II)$ of the type-II CuO_2 plane compared with that of the type-I plane is possibly caused by the low value of $dT_c^{max}(II)/dP$ chosen for the type-II plane in the Tl-2223 phase. The difference in the $T_c(i)\sim P$ curve arises from the different intrinsic values of $T_c(i)$ and $n_H(i)$ among inequivalent CuO_2 planes. In recent pressure measurements on Tl-2223 by Tristan Jover *et al.*,[7] the variation of T_c with pressure clearly exhibits two regimes. Below 12 GPa the data points follow one parabola, while above 12 GPa they follow another parabola which has a slightly lower maximum T_c and a larger width. Both parabolas have maxima at approximately the same pressure. Furthermore, a kink on the $T_c\sim P$ curve was also observed at about 13 GPa for low-T_c Tl-2223 phase.[6] It follows that this kink may stem from the nonhomogeneous charge distribution among the two types of CuO_2 layers. Since the intrinsic value of $T_c(i)$ in the type-I CuO_2 planes is the exact value of T_c at ambient pressure for superconductors, the variation of T_c with pressure is expected to be followed from the curve of the intrinsic $T_c(I)(P)$ for the type-I CuO_2 planes under low pressures. Above the pressure level of the occurrence of the kink, the effect of the type-II CuO_2 plane dominates. Interestingly, our extended PICT model for layered cuprates can be used to explain this pressure dependence of T_c successfully.

3. $Tl_2Ba_2Ca_3Cu_4O_{12-y}$ system

In the $Tl_2Ba_2Ca_3Cu_4O_{12-y}$ system, we obtained a theoretical maximum value of T_c of 135.3 K within the framework of the Lawrence-Doniach theory. Following the above treatment, we also suppose that the intrinsic maximum $T_c^{max}(i)$ is the same for the two types of CuO_2 layers, that is, $T_c^{max}(i)=135.3$ K. The normalized intrinsic $T_c(i)$ values for the type-I and type-II CuO_2 planes as a function of total hole concentration δ calculated from Eqs. (8) and (9) are plotted in Fig. 8 for $A=8.8$, 2.4, and 1.2, respectively. Three clear parabolas are presented for the type-I CuO_2 planes, and the optimal carrier concentration δ^{opt} moves back from 0.51 through 0.41 to 0.31 with the increase of A. On the other

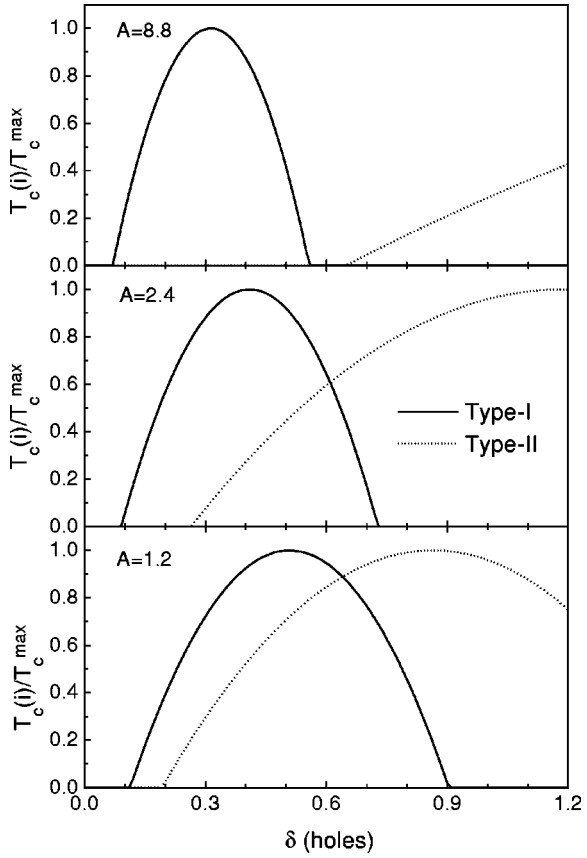

FIG. 8. Normalized intrinsic $T_c(i)$ values for the inequivalent CuO_2 planes in the Tl-2234 system as a function of carrier concentration for $A=8.8$, 2.4, and 1.2, respectively.

hand, the minimum carrier concentration $\delta^{\min}$ in the type-II CuO_2 plane increases with an increasing value of A. However, the $\delta^{\min}$ in the type-I CuO_2 planes scarcely change. For the case of $A=8.8$, superconductivity occurs in the type-II CuO_2 plane for values of δ above 0.65 compared with 0.35 in the Tl-2223 system. To our surprise, there is a regime of 0.55–0.65 where superconductivity would vanish completely due to the high inhomogeneity of charge distribution between the two types of CuO_2 layers. For $A=1.2$, the type-I CuO_2 plane is superconducting for δ values between 0.12 and 0.90, reaching a maximum T_c at $\delta=0.51$. When the hole concentration is over 0.9, the type-I CuO_2 planes are no longer superconducting while the intrinsic $T_c(II)$ reaches nearly the maximum in the type-II CuO_2 plane.

To our knowledge, there are few reports on the pressure dependence of the superconducting transition temperature in Tl-2234. Recently, Tristan Jover *et al.*[7] investigated the pressure effect on T_c for the Tl-2234 compound with $T_c=113$ K up to 14 GPa, and reported an initial pressure derivative dT_c/dP of 2.0 K/GPa. Here we use our extended PICT model to interpret their results. For the type-I CuO_2 plane, the intrinsic $T_c(I)$ and intrinsic initial pressure derivative $dT_c(I)/dP$ are 113 K and 2.0 K/GPa, respectively.[7] The corresponding intrinsic $n_H(I)$ is obtained to be 0.095 holes according to Eq. (9). Then, the intrinsic hole concentration for the type-II CuO_2 plane can be determined by Eq. (8) as $n_H(II)=0.056$ holes, and Eq. (9) yields an intrinsic $T_c(II)$ value of 56.6 K. Using Eq. (10), one can obtain the intrinsic

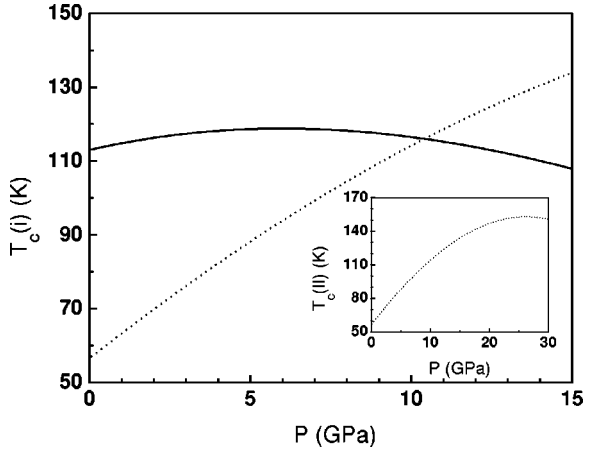

FIG. 9. Pressure dependence of the intrinsic $T_c(i)$ for type-I (solid) and type-II (dotted) CuO_2 planes in the Tl-2234 phase with $T_c=113$ K up to 15 GPa. Inset: A plot of the intrinsic $T_c(II)$ vs pressure in the range from 0 to 30 GPa.

maximum pressure derivative $dT_c^{\max}(I)/dP$ of -1.79 K/GPa in the type-I CuO_2 plane. The pressure derivative $dT_c^{\max}(II)/dP$ for the type-II CuO_2 plane is chosen to be 0.7 K/GPa.[70]

In Fig. 9 we present the pressure dependence of the intrinsic $T_c(i)$ for Tl-2234. As can be seen, the two types of CuO_2 planes have themselves paraboliclike curves. As pressure is increased the intrinsic $T_c(i)$ increases initially until it reaches a maximum at some pressure level, and at higher pressures $T_c(i)$ decreases. The maximum intrinsic $T_c(I)$ of 118.8 K on the $T_c(I)\sim P$ curve is exhibited at 6.0 GPa in the type-I CuO_2 plane. For a pressure of 6.6 GPa, the intrinsic $T_c(I)$ is calculated to be 118.7 K, in good agreement with Tristan Jover *et al.*[7] The inset of Fig. 9 gives the variation of intrinsic $T_c(II)$ with pressure up to 30 GPa for the type-II CuO_2 plane. The intrinsic $T_c(II)$ increases in a regular manner with P. It already attains 113 K, the value for the underdoped Tl-2234 compound at normal pressure, at $P\sim 9.7$ GPa. Further increasing pressure to 15.4 GPa, it reaches the theoretical maximum value of 135.3 K at normal pressure for the Tl-2234 cuprates. At higher pressures it considerably exceeds this value and the maximum is achieved for $P\sim 26$ GPa with intrinsic $T_c(II)(P)$ of 152.8 K, which is larger than the theoretical saturation value of T_c of 146 K at ambient pressure for the bilayer thallium-based series. It is suggested that the type-II CuO_2 plane becomes active under high pressure. It is possible to attain higher critical temperatures for the multilayer superconductors by using chemical pressure to increase the mobile carrier concentration in the type-II CuO_2 plane, which may mimic the effect of a true pressure. Atomic substitution introduces off-diagonal or nonhydrostatic stresses which have a large effect on T_c, as has been found in Y-123 (σ_{aa} vs σ_{bb}). It should be noted that the maximum intrinsic $T_c(II)(P)$ in type-II planes exceeds the maximum $T_c(P)$ of 120 K in the IL compound.[70] There is also a kink in $T_c(i)(P)\sim P$ curves. The calculated pressure level of the kink occurrence is at 10.5 GPa, which is just the experimentally observed value of 10.5 GPa.[7] We found that the experimental data obtained by Tristan Jover *et al.*[7] lie between the two intrinsic $T_c(i)(P)\sim P$ curves when pres-

sure is above 10.5 GPa. Therefore the type-II CuO_2 plane should become the dominant conducting layer under higher pressure.

In the above calculation, we found that the intrinsic $T_c(II)$ of the type-II CuO_2 plane in Tl-2234 can increase strongly from 56.6 to 152.8 K under the application of pressure, while a modest increase of $T_c(II)$ occurs from 126.7 to 131.9 K in Tl-2223. This is because the type-II CuO_2 plane in Tl-2234 is highly underdoped due to the depletion of intrinsic hole concentration compared to that in Tl-2223. Therefore, mixed-phase samples of Tl-2223 containing some superconducting Tl-2234 phase should have a higher T_c than the pure Tl-2223 phase. This behavior has been confirmed in the pressure effects on T_c in Hg-1223 and Hg-1234 by Ihara *et al.*,[82] who reported the highest onset T_c of 156 K at 25 GPa for a Hg-1223 and Hg-1234 mixed-phase sample and 140 K at 13 GPa for a Hg-1223 phase sample.

IV. SUMMARY AND CONCLUSION

We have presented a simple pressure-induced charge transfer model for layered cuprate superconductors based on the sheet-charge theory. The model, which utilized the assumption that each CuO_2 plane has its intrinsic hole concentration, critical temperature, and pressure derivative of T_c, can distinctly predict the pressure effect on the intrinsic $T_c(i)$ among the crystallographically inequivalent CuO_2 layers. The effect of interlayer coupling between the adjacent CuO_2 layers on the superconducting transition temperature has been investigated using the generalized Lawrence-Doniach theory based on the hypothesis that the distinct intrinsic critical temperature corresponding to each order parameter ψ_i is probably different for the two types of CuO_2 planes. We studied the dependence of transition temperature on the type and number of CuO_2 layers in bilayer thallium-based superconductors both at ambient pressure and under high pressure. Our major results and predictions are the following.

(1) The transition temperature T_c increases with interlayer coupling λ. At any λ, the larger the difference between the intrinsic $T_c(II)$ and $T_c(I)$, the stronger the increase of T_c with the number of CuO_2 layers.

(2) T_c is an increasing function of n, reaching 95% of the maximum possible value $T_n(\infty)$ by $n=5$. The upper limit of T_c for the bilayer thallium-based cuprates is 146 K.

(3) For the optimally doped single, double, and triple CuO_2 sheets thallium-based superconductors, the predicted saturation values of $T_c(I)$ of the type-I CuO_2 plane of 91.1, 119.6, and 133.9 K are obtained when $P=2.3$, 2.9, and 6.0 GPa, respectively. For the underdoped Tl-2234 compound with $T_c=113$ K, the calculated $T_c(I)$ of 118.5 K is obtained at $P=6.0$ GPa. These predictions are in good agreement with experiments.

(4) The intrinsic $T_c(i)$ of the type-I and type-II CuO_2 planes follow from two different parabolas which cross at high pressure in multilayer compounds Tl-2223 and Tl-2234. The results indicate that at low pressures T_c is the intrinsic $T_c(i)$ of the type-I CuO_2 plane, and at the relatively high pressures the intrinsic effect of the type-II plane dominates.

(5) Under the application of pressure, the intrinsic $T_c(II)$ of the type-II CuO_2 plane in Tl-2234 increases strongly compared with a modest increase of $T_c(II)$ in Tl-2223. We suggest that the strong pressure effect on the intrinsic $T_c(II)$ in the type-II CuO_2 plane results from its underdoped nature.

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