## **Variation of the superconducting transition temperature of hole-doped copper oxides**

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The experimentally observed difference of the superconducting critical temperature  $T_c$  of hole-doped cuprates is studied by using an extended interlayer coupling model for layered *d*-wave superconductors. We show that the change of the maximum  $T_c$  from series to series is determined by the next-nearest-neighboring hopping  $t'$ , while the difference of the maximum  $T_c$  among the compounds in a homogeneous series is controlled by the interlayer pairing strength. Our results also provide helpful guidelines in the search for new high- $T_c$ superconductors.

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The nature of high-temperature superconductors is a challenging problem in condensed-matter physics. A common feature of copper-oxide superconductors is the presence of  $CuO<sub>2</sub>$  plane. It has been observed that the superconducting critical temperature  $T_c$  varies parabolically with the hole concentration  $n_H$  in CuO<sub>2</sub> plane with a maximum  $T_c^{max}$  at an optimal doping level.<sup>1,2</sup> Furthermore, in the homogeneous series compounds  $A_mB_2Ca_{n-1}Cu_nO_{2n+y+\delta}$  (*A* = Bi, Tl, or Hg,  $B =$ Sr or Ba,  $m=2$  or 1,  $y=4$ , 3, or 2),  $T_c^{max}$  initially increases with the number of  $CuO<sub>2</sub>$  layers  $(n)$  per unit cell, maximizes when  $n=3$ , and then decreases with further increasing  $n$ ,<sup>3</sup> as shown in Fig. 1. However,  $T_c^{max}$  attainable is different from series to series, e.g.,  $35$  K in  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ (Ref. 4) and 97 K in HgBa<sub>2</sub>CuO<sub>4+ $\delta$ </sub>.<sup>5</sup> An obvious question is what is the crucial parameter that governs the  $T_c^{max}$  of each family.

Among various parameters proposed, the Madelung potential at the apical oxygen relative to that at the planar  $\alpha$  oxygens<sup>6</sup> was found to correlate with  $T_c^{max}$  rather well, pointing to the primary importance of the apical oxygens for the electronic structure relevant to superconductivity. Further investigations<sup>7,8</sup> revealed that the effect of the apical oxygens on high- $T_c$  superconductivity in reality translates into a correlation between  $T_c^{max}$  and the next-nearest-neighbor hopping parameter  $t'$  in the  $t$ - $t'$ -*J* model with  $t$  and  $J$  being the nearest-neighbor hopping parameter and antiferromagnetic interaction, respectively. In these approaches,  $t'$  was considered as a single parameter reflecting the main difference among various cuprates. If we consider the homologous series, the universality of such a correlation would be seriously questioned. For example, the bilayer and trilayer  $TI_2$ -based and Hg-based compounds have almost same  $t'$  (Ref. 8), but their  $T_c^{max}$ 's are significantly different.

Our goal in this work is to extract and identify which parameters govern the  $T_c$  behaviors in hole-doped cuprates. We apply an interlayer coupling model to  $CuO<sub>2</sub>$  layer systems and then calculate  $T_c$  based on the Bardeen-Cooper-Schrieffer (BCS) gap equation with *d*-wave symmetry. Our results suggest that the difference of  $T_c^{max}$  from family to family is the result of different next-nearest-neighbor hop-

ping  $t'$ , while the difference of  $T_c^{max}$  between the compounds in a homologous family is controlled by the interlayer coupling strength  $T<sub>I</sub>$ .

The effective layered Hamiltonian we consider is

$$
H = \sum_{lk\sigma} (\varepsilon_k - \mu) c_{k\sigma}^{\dagger l} c_{k\sigma}^l - \sum_{lkk'} V_{kk'} c_{k\uparrow}^{\dagger l} c_{-k\downarrow}^{\dagger l} c_{-k'\downarrow}^l c_{k'\uparrow}^l
$$
  
+ 
$$
\sum_{\langle ll'\rangle} \sum_k T_j(k) c_{k\uparrow}^{\dagger l} c_{-k\downarrow}^{\dagger l} c_{-k\downarrow}^l c_{k\uparrow}^l,
$$
 (1)

where  $\varepsilon_k$  is the quasiparticle dispersion,  $\mu$  is the chemical potential,  $c_{k\sigma}^{\dagger l}$  is a quasiparticle creation operator pertaining to the layer (*l*) with two-dimensional wave vector *k* and spin  $\sigma$ . The summation over  $ll'$  runs over the layer indices of the unit cell. The intralayer interaction  $V_{kk}$  is assumed to be independent of *l*. The interlayer tunneling is parametrized by  $T_J(k) = T_J(\cos k_x - \cos k_y)^4$  (Ref. 9).

We assume that the superconducting gap is characterized by the nonvanishing order parameter  $b_k^l = \langle c_{k\uparrow}^l c_{-k\downarrow}^l \rangle$ . Based on the BCS theory, the gap function  $\Delta_k^l$  satisfies the following equation:



FIG. 1. Dependence of the critical temperature  $T_c^{max}$  at optimal doping as a function of the number of  $CuO<sub>2</sub>$  layers  $(n)$  of various homogeneous series.

$$
\Delta_k^l = \sum_{k'} V_{kk'} b_{k'}^l + T_j(k)(b_k^{l+1} + b_k^{l-1}),\tag{2}
$$

where  $b_k^l = \Delta_k^l \chi_k^l$  and the generalized pair susceptibility is  $\chi_k^l = (2E_k^l)^{-1}$ tanh $(\beta E_k^l/2)$  with the quasiparticle spectrum  $E_k^l = \sqrt{(\varepsilon_k - \mu)^2 + |\Delta_k^l|^2}.$ 

The spatial dependence of the gap takes the form<sup>10</sup>  $\Delta_k^l$  $=$   $\Delta_k^{\pm}e^{\pm i\alpha l}$ . Then the general solution of the homogeneous part is

$$
\Delta_k^l = \Delta_k^+ e^{i\alpha l} + \Delta_k^- e^{-i\alpha l}.
$$
 (3)

Considering the fact that the gap vanishes on the layer ends  $l=0$  and  $n+1$ , the natural boundary conditions for the gap are  $\Delta_k^0 = \Delta_k^{n+1} \equiv 0$ . The wave vector of the oscillating gap is then determined by

$$
\begin{pmatrix} 1 & 1 \ e^{i\alpha l} & e^{-i\alpha l} \end{pmatrix} \begin{pmatrix} \Delta_k^+ \\ \Delta_k^- \end{pmatrix} = 0.
$$

The vanishing determinant of the matrix provides a nontrivial solution only when  $\alpha = \xi \pi/(n+1)$  with  $\xi$  being an integer. Thus we obtain  $\Delta_k^+ = -\Delta_k^- = \Delta_k$ . The solution of spatial dependence of the gap is then given by

$$
\Delta_k^l = 2i \Delta_k \sin\left(\frac{l\pi\xi}{n+1}\right). \tag{4}
$$

The solution with the lowest energy is nodeless inside the *n* CuO<sub>2</sub> layers which leads to  $\xi=1$  for the superconducting state. Thus, the spatial dependence of the gap has the form

$$
\Delta_k^l = 2i \Delta_k \sin\left(\frac{l\pi}{n+1}\right). \tag{5}
$$

Around critical temperature  $T_c$ , we can take  $\chi_k^l$  in a  $\text{simple}$  form:  $\chi_k^l \approx (2E_k)^{-1} \tanh(\beta E_k/2) \equiv \chi_k$  with  $E_k$  $=\sqrt{(\varepsilon_k - \mu)^2 + |\Delta_k|^2}$ . In this case, Eq. (2) can be rewritten as

$$
\Delta_k^l = \sum_{k'} V_{kk'} \chi_{k'} \Delta_{k'}^l + T_J(k) (\chi_k \Delta_k^{l+1} + \chi_k \Delta_k^{l-1}). \tag{6}
$$

Substitution of Eq.  $(5)$  into Eq.  $(6)$  yields a simple  $\Delta_k$  equation

$$
\Delta_k - \sum_{k'} V_{kk'} \chi_{k'} \Delta_{k'} = f(n) T_J(k) \chi_k \Delta_k, \qquad (7)
$$

where  $f(n) = 2 \cos[\pi/(n+1)].$ 

To account for the experimental observed *d*-wave gap, we assume a *d*-wave pairing potential

$$
V_{kk'} = Vg(k)g(k'), g(k) = \cos k_x - \cos k_y.
$$
 (8)

The gap magnitude is thus  $\Delta_k = \Delta_{0}g(k)$  and the parameter  $\Delta_0$  is determined by the following self-consistent equation:

$$
1 = \frac{1}{2N} \sum_{k} \frac{Vg^{2}(k) + f(n)T_{J}(k)}{E_{k}} \tanh\left(\frac{\beta E_{k}}{2}\right). \tag{9}
$$



FIG. 2. (Color) The critical temperature  $T_c$  vs the hole concentration  $n_H$  for various next-nearest-neighbor hopping parameters  $t'$ with the interaction strength  $V=0.038$  eV (a) and for various *V* with  $t' = -0.02$  eV (b) in monolayer cuprates.

The value of  $T_c$  in layered *d*-wave superconductors is then obtained by solving Eq. (9) at  $\Delta_0=0$ .

In order to self-consistently calculate  $T_c$  for a given  $\mu$  in conjunction with the equation determining  $n_H$ , we need an explicit form of  $\varepsilon_k$ . It has been established<sup>11–15</sup> that the quasiparticle excitation spectrum of cuprates can be well described by the  $t$ - $t'$ - $J$  model. Within the framework of the *t*-*t'*-*J* model, the dispersion  $\varepsilon_k$  is given by<sup>13,15</sup>

$$
\varepsilon_k = (J + 2t')\cos k_x \cos k_y + \frac{J}{4}(\cos 2k_x + \cos 2k_y). \tag{10}
$$

For monolayer insulator  $La_2CuO_4$ , experiments<sup>16</sup> and theoretical calculations<sup>17</sup> give a  $J=0.128$  eV. There are small variations of *J* among various Cu-O insulators<sup>18</sup> but we expect a value of  $J=0.128$  eV is a generally good representation for all Cu-O materials. Then one can determine  $T_c$  as a function of  $n_H$  based on Eqs. (9) and (10) once having knowledge of  $t'$ , *V*, or/and  $T<sub>J</sub>$ .

First we consider the variation of  $T_c$  in monolayer (*n*  $=1$ ) hole-doped cuprates. Figures 2(a) and 2(b) show the calculated  $T_c$  in monolayer superconductors as a function of  $n_H$  in some interested parameter range of  $t'$  and *V*. As shown,  $T_c$  initially increases with increasing  $n_H$ , takes a maximum around an optimal doping level  $n_H^{opt}$ , and then decreases with further increasing  $n_H$ . This parabolic relation between  $T_c$  and  $n_H$  agrees with general experimental observations in monolayer cuprates.<sup>1,2</sup> We notice that  $T_c^{max}$  systematically changes with  $t'$ , but it monotonically increases with *V*, as one expects. The difference between these two parameters is that  $n_H^{opt}$  depends significantly on  $t'$ , while it scarcely changes for different values of *V*. These results indicate that the parameters controlling  $T_c^{max}$  would be either  $t'$ or *V* or both of them.

In Fig. 3 we plotted the  $t'$  dependence of both  $T_c^{max}$  and  $n_H^{opt}$  for monolayer cuprates. As *t*<sup>*'*</sup> increases,  $T_c^{max}$  increases and then decreases through a maximum for all *V* studied. Note that the enhancement of  $T_c^{max}$  from 25 to 100 K occurs



FIG. 3. The calculated maximum critical temperature  $T_c^{max}$  (a) and the optimal hole concentration  $n_H^{opt}$  (b) as a function of the next-nearest-neighbor hopping parameter  $t'$  (or  $J+2t'$ ) for various interaction strengths *V* in monolayer cuprates.

over a rather wide parameter space. Such a huge enhancement completely covers the variation of  $T_c^{max}$  among the monolayer high- $T_c$  superconductors. The occurrence of the maximum implies that the enhancement of  $T_c^{max}$  due to the increase in  $t'$  is limited. Since  $J+2t'$  is the coefficient of the cos  $k_x \cos k_y$  term in Eq. (10), the effect of  $J + 2t'$  on  $T_c^{max}$  is obviously the same as that of  $t'$ . That means that  $T_c^{max}$  increases with increasing the coefficient of the  $\cos k_x \cos k_y$ term in quasiparticle dispersion, saturates, and then decreases with the further increase of this coefficient. This nonmonotonic  $T_c^{max}$  dependence is consistent with those reported previously within the framework of the tight-binding approximation.<sup>19,20</sup> We also notice that  $n_H^{opt}$  behaves in a similar manner with *t'* as  $T_c^{max}$ . For  $J+2t' > 0$ ,  $n_H^{opt}$  decreases with increasing *t'*. Although  $T_c^{max}$  depends on *V*,  $n_H^{opt}$  is nearly independent of *V* over a wide range of *t'*.

To trace the clue to the change of  $T_c^{max}$  among monolayer cuprates, we list in Table I the experimental results of  $T_c^{max}$ (Refs. 2, 4, 5, 21, and 22) the distance  $d_{Cu-O(a)}$  between the copper and apical oxygen atoms, and the distance  $d_{\text{Cu-O}(p)}$ between the copper and in-plane oxygen atoms taken from the works in Refs. 5 and 6, the calculated values of bond

valence sums  $(BVS)$  of copper  $V_{Cu}$  and the difference in the Madelung site potential for a hole between the copper and the in-plane oxygen  $\Delta V_M$ . To get effective BVS of copper, we follow the method proposed by Brown.<sup>23</sup> The results of  $\Delta V_M$  based on the structural data are taken from the works in Refs. 6 and 24. Here we observe one important experimental fact:  $T_c^{max}$  increases systematically with enlarging  $d_{Cu-O(a)}$ . Band-structure calculations<sup>8</sup> revealed that  $t'$  increases with  $d_{\text{Cu-O}(a)}$  for the monolayer cuprates reported so far. Thus the increase of  $T_c^{max}$  with increasing  $t'$  should capture the basic physics of the monolayer cuprates.

It has been proposed<sup>25,26</sup> that  $V_{Cu}$  and  $\Delta V_M$  are two essential factors governing  $T_c$  and represent an essentially equivalent physical content. Materials with larger  $T_c^{max}$  tend to have a smaller  $V_{Cu}$  (Ref. 25) or  $\Delta V_M$  (Ref. 26). Since the variation of  $V_{Cu}$  or  $\Delta V_M$  reflects the corresponding change of  $n_H$  (Refs. 26–28) the increase of the calculated  $T_c^{max}$  with decreasing  $n_H^{opt}$  for a wide  $t'$  range is obviously consistent with the experimental data shown in Table I. This  $n_H^{opt}$  dependence of  $T_c^{max}$  is also consistent with the muon spin resonance measurements.29 On the other hand, the fact that the change of  $T_c^{max}$  with *V* is almost independent of  $n_H^{opt}$  [Fig.  $3(b)$  rules out the possibility of *V* being a dominant factor in governing the change in  $T_c^{max}$ . The present results lead us to conclude that the increase of  $T_c^{max}$  with  $d_{\text{Cu-O}(a)}$  among the monolayer cuprates is a result of the increase in *t'*. One prediction is that  $T_c^{max}$  decreases with further increasing  $t'$ after a saturation. Thus, materials with a relatively long  $d_{\text{Cu-O}(a)}$  bond length would not always expect to have a high  $T_c^{max}$ .

The values of  $t'$  were determined in a self-consistent way as follows. From Fig.  $3(a)$  we learned that there exists a maximum for a given *V*. Among the monolayer cuprates discovered so far,  $HgBa_2CuO_{4+\delta}$  possesses the highest  $T_c^{max}$  of 97 K. Assuming this is the highest value in all monolayer cuprates, we derived a value of  $V=0.03762$  eV from curves of  $T_c^{max}$  versus *t'*. Equation (9) yields  $t' = -0.0183$  eV for the optimally doped  $HgBa_2CuO_{4+\delta}$ . For other optimally doped monolayer compounds with  $T_c^{max}$  < 97 K, *t'* should be smaller than  $-0.0183$  eV because of their shorter  $d_{\text{Cu-O}(a)}$ . The relative  $t'$  is then obtained by using the experimentally observed  $T_c^{max}$ .

TABLE I. Summary of the experimental results of the critical temperature  $T_c^{max}$  at optimal doping, the distance  $d_{\text{Cu-O}(a)}$  between the copper and apical oxygen atoms, the distance  $d_{\text{Cu-O}(p)}$  between the copper and in-plane oxygen atoms, and the calculated values of the bond valence sums of copper  $V_{\text{Cu}}$  and the difference in the Madelung site potentials  $\Delta V_M$  for a hole between the in-plane oxygen and copper atoms in some typical monolayer cuprates.

Cuprates	$T_c^{max}$ (K)	$d_{\text{Cu-O}(a)}$ (A)	$d_{\text{Cu-O}(p)}$ (A)	$V_{\rm Cu}$	$\Delta V_M$ (eV)
$La1.85Sr0.15CuO4$	35	2.4124	1.8896	2.539	49.620
$Bi_2Sr_{1.61}La_{0.39}CuO_{6.6}$	36	2.461	1.901	2.437	48.437
$TIBa1$ <sub>2</sub> La <sub>18</sub> CuO <sub>5</sub>	52	2.500	1.9240	2.280	48.409
$Tl_2Ba_2CuO_6$	90	2.714	1.9330	2.135	47.081
$HgBa_2CuO_{4+\delta}$	97	2.780	1.9375	2.091	46.81



FIG. 4. The calculated critical temperature  $T_c$  vs the hole concentration  $n_H$  in HgBa<sub>2</sub>Ca<sub>n-1</sub>Cu<sub>n</sub>O<sub>2n+2+ $\delta$ </sub> as a function of the number of  $CuO<sub>2</sub>$  layers.

Next we consider  $n$ , the number of  $CuO<sub>2</sub>$  layers, dependence of  $T_c^{max}$  in the layered homogeneous series. In general,  $T_c^{max}$  initially increases with *n*, maximizes at  $n=3$ , and then decreases with further increasing  $n^3$ . To calculate  $T_c$  for multilayers, we use the same dispersion  $\varepsilon_k$  and *V* as obtained from the monolayer. The interlayer tunneling strength  $T_j$  is determined by using the experimental values of  $T_c^{max}$  for monolayer and bilayer compounds in the same homogeneous series. As an example, in Fig. 4, we show curves of calculated  $T_c$  versus  $n_H$  as a function of layer number *n* in the Hg-based series. The theoretical curves exhibit the generic parabolic behavior. Previously, the relation between  $T_c$  and  $n_H$  has been well established for the monolayer, bilayer, and trilayer Hg-based superconductors.<sup>30</sup> Compared to the available experimental data, the agreement is excellent.

The calculated  $T_c^{max}$  in four typical homogeneous series are summarized in Table II. The experimental results are also listed for comparison. As can be seen,  $T_c^{max}$  initially increases with increasing *n* and then saturates as  $n \rightarrow \infty$ . This behavior is in good agreement with those obtained from both the interlayer mechanism<sup>10,31</sup> and Ginzburg-Landau theory.<sup>32,33</sup> The upper limit of  $T_c^{max}$  for infinite layer compound is in the range of 139.4–164.6 K. The highest  $T_c^{max}$  of 164.6 K is found in the Tl-based series. Our results for *n*  $=$  3 agree with experiments very well. The predictions made here for  $T_c^{max}$  of the trilayer compound are the best ones compared to previous theories. $10,31-33$ 

The present study shows that interlayer coupling is the driving force for the enhancement of  $T_c^{max}$  for multilayer systems. This does not conflict with the experimental fact that  $T_c^{max}$  decreases as  $n \ge 3$ . In fact, there exist fivefold (outer) and fourfold  $CuO<sub>2</sub>$  (inner) planes surrounded by pyramidal and square oxygens in the multilayer system. Investigations carried out by different experimental techniques and model calculations<sup>28,34–36</sup> showed that the distribution of charge carriers is nonhomogeneous among the  $CuO<sub>2</sub>$  sheets and the hole concentration in the outer  $CuO<sub>2</sub>$  plane is larger than that in the inner  $CuO<sub>2</sub>$  plane. BVS analyses<sup>28</sup> and NMR studies $36$  on the Hg-based series revealed that the highest  $T_c^{max}$  corresponds to the smallest difference in  $n_H$  between two types of  $CuO<sub>2</sub>$  planes. When the number of  $CuO<sub>2</sub>$  layer is larger than three, the reduction of  $T_c^{max}$  comes from the large difference in  $n_H$  between the outer and inner  $CuO<sub>2</sub>$ planes. For compounds with more than three  $CuO<sub>2</sub>$  planes, the enhancement of  $T_c^{max}$  seems possible at ambient pressure if one can adequately dope the inner planes.

Finally, we would like to comment on the possibility of applying Eq.  $(10)$  to the high- $T_c$  superconducting oxides. Angle-resolved photoemission  $(ARPES)$  experiments<sup>37</sup> reveal that there exists a flat region near  $(\pi,0)$  in many high-*Tc* compounds. Such an extended region of flat  $CuO<sub>2</sub>$ -derived bands seems a universal property of the holedoped cuprates. The hole dispersion relation of Eq.  $(10)$  derived from the  $t$ - $t'$ - $J$  model reproduces well the flat bands similar to those observed in ARPES experiments.<sup>37</sup> Dagotto and his co-workers<sup>12</sup> have shown that the effect of strong correlations can quantitatively account for such flat bands. It has been found<sup>12,38</sup> that the sign, doping, and temperature dependence of the Hall coefficient, thermopower, specific heat, magnetic susceptibility is in excellent quantitative agreement with experiments when the dispersion obtained from the *t*-*t'*-*J* model is used. Furthermore, the momentumdependent spectrum as a function of hole concentration has been calculated based on the  $t$ - $t'$ - $t''$ - $J$  model by using both the exact diagonalization and Monte Carlo method.<sup>14,15</sup> The calculated results explain the experimental data of ARPES for hole doped  $Bi_2Sr_2CaCu_2O_{8+\delta}$  from underdoped to overdoped regime. The results indicate that once the flat region around  $(\pi,0)$  has already formed, it remains almost unchanged upon additional doping. This means that the electronic structure is essentially the same in a high- $T_c$  material with different doping level. Therefore, the dispersion relation of Eq.  $(10)$  is essential in describing the low-energy physics of high-temperature superconductors.

In summary, we have investigated the observed  $T_c$  variation in hole-doped cuprates on the basis of an extended in-

TABLE II. The critical temperature  $T_c^{max}$  and the ratio of  $T_J/V$  in homogeneous copper-oxide series at optimal doping. The brackets are the experimental data taken from the works of Refs. 2–6, 21, 22, 28, and 33.

n				4		$\infty$	$T_I/V$
$\text{Bi}_2\text{Sr}_2\text{Ca}_{n-1}\text{Cu}_n\text{O}_{2n+4+\delta}$	36(36)	90 (90)	115.5(110)	127.8	134.7	150.7	0.1945
$TIBa2Can-1CunO2n+3+\delta$	52 (52)	107(107)	131.3 (133.5)	143.0 (127)	149.5	164.6	0.1930
$Tl_2Ba_2Ca_{n-1}Cu_nO_{2n+4+\delta}$	90 (90)	115(115)	125.2(125)	130.1 (116)	132.9	139.4	0.0906
$HgBa2Can-1CunO2n+2+\delta$	97 (97)	127 (127)	139.2 (135)	145.2 (129)	148.6 (110)	156.4	0.1135

terlayer coupling model. We demonstrate that the nextnearest-neighboring hopping *t'* dominates the variation of the maximum  $T_c$  from series to series and the interlayer coupling strength controls the difference of the maximum  $T_c$ among the compounds in a layered homogeneous series. These results also provide helpful guidelines in the search for new high- $T_c$  superconductors.

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