## 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  $A_m B_2 Ca_{n-1} Cu_n O_{2n+y+\delta}$  (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 La<sub>2-x</sub>Sr<sub>x</sub>CuO<sub>4</sub> (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 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 Tl<sub>2</sub>-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 hopping t', while the difference of  $T_c^{max}$  between the compounds in a homologous family is controlled by the interlayer coupling strength  $T_J$ .

The effective layered Hamiltonian we consider is

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

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.

$$\Delta_{k}^{l} = \sum_{k'} V_{kk'} b_{k'}^{l} + T_{J}(k) (b_{k}^{l+1} + b_{k}^{l-1}), \qquad (2)$$

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  $\Delta_k^+ = -\Delta_k^- \equiv \Delta_k$ . 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 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

$$\Delta_{k}^{l} = \sum_{k'} V_{kk'} \chi_{k'} \Delta_{k'}^{l} + T_{J}(k) (\chi_{k} \Delta_{k}^{l+1} + \chi_{k} \Delta_{k}^{l-1}).$$
(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:

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

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

For monolayer insulator La<sub>2</sub>CuO<sub>4</sub>, 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  $T_J$ .

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' 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  $\cos k_{y} \cos k_{y}$ 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  $d_{\text{Cu-O}(a)}$  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  $V_{\rm Cu}$  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  $d_{\rm Cu-O(a)}$ . Band-structure calculations<sup>8</sup> revealed that t' increases with  $d_{\rm Cu-O(a)}$  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.<sup>29</sup> 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<sub>2</sub>CuO<sub>4+δ</sub> 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=0.037 62 eV from curves of  $T_c^{max}$  versus t'. Equation (9) yields t'=-0.0183 eV for the optimally doped HgBa<sub>2</sub>CuO<sub>4+δ</sub>. For other optimally doped monolayer compounds with  $T_c^{max} < 97$  K, t' should be smaller than -0.0183 eV because of their shorter  $d_{Cu-O(a)}$ . 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  $d_{Cu-O(a)}$  between the copper and apical oxygen atoms, the distance  $d_{Cu-O(p)}$  between the copper and in-plane oxygen atoms, and the calculated values of the bond valence sums of copper  $V_{Cu}$  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)	$d_{\text{Cu-O}(a)}$ (Å)	$d_{\operatorname{Cu-O}(p)}$ (Å)	V <sub>Cu</sub>	$\Delta V_M$ (eV)
La <sub>1.85</sub> Sr <sub>0.15</sub> CuO <sub>4</sub>	35	2.4124	1.8896	2.539	49.620
$Bi_2Sr_{1.61}La_{0.39}CuO_{6+\delta}$	36	2.461	1.901	2.437	48.437
TlBa <sub>1.2</sub> La <sub>1.8</sub> CuO <sub>5</sub>	52	2.500	1.9240	2.280	48.409
Tl <sub>2</sub> Ba <sub>2</sub> CuO <sub>6</sub>	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>2*n*+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.<sup>3</sup> 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 TI-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.<sup>10,31-33</sup>

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  $2^{28,34-36}$  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 studies36 on the Hg-based series revealed that the highest  $T_c^{max}$  corresponds to the smallest difference in  $n_H$  between two types of  $CuO_2$  planes. When the number of  $CuO_2$  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- $T_c$  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.14,15 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	1	2	3	4	5	œ	$T_J/V$
$\overline{\text{Bi}_2\text{Sr}_2\text{Ca}_{n-1}\text{Cu}_n\text{O}_{2n+4+\delta}}$	36 (36)	90 (90)	115.5 (110)	127.8	134.7	150.7	0.1945
$TlBa_2Ca_{n-1}Cu_nO_{2n+3+\delta}$	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
$HgBa_2Ca_{n-1}Cu_nO_{2n+2+\delta}$	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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