Phonon-mediated superconducting transitions in layered cuprate superconductors

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A phonon-mediated *d*-wave BCS-like model is presented for a homologous series of layered cuprate superconductors. We show that the dependence of both the superconducting transition temperature T_c and oxygen isotope exponent α on the doping level, number of CuO_2 layers, pressure, and cation disorder in the model superconducting series $HgBa_2Ca_{n-1}Cu_nO_{2n+2+\delta}$ can be well reproduced within one single theoretical framework. We also find that the theoretical model accounts well for the superconducting transitions in other homologous series such as the bismuth- and thallium-based family. Our results suggest that the interlayer coupling plays an important role in the significant enhancement of T_c and in the systematic reduction of α in a layered homologous series.

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I. INTRODUCTION

A striking feature of the known copper-oxide superconductors is the presence of CuO_2 planes. The role of interlayer coupling between the CuO_2 planes was recognized[1] soon after the superconducting transition temperature T_c was discovered to be enhanced from 35 K in single CuO_2-layer compound $La_{2-x}Ba_xCuO_2$ to 90 K in two CuO_2-layer compound $YBa_2Cu_3O_{7-\delta}$. Interlayer effects were particularly emphasized in the superconductivity of homologous series of bismuth-, thallium-, and mercury-based superconductors.[2–8] Within each family of cuprates, the initial discovery of the T_c increase when increasing the number of CuO_2 layers (n) from $n=1$ to $n=3$ gave rise to the expectation that T_c may increase further when the structural cell has more CuO_2 layers. However, the later experiments showed that the trilayer material has the maximum T_c in each homologous series.[8,9] In monolayer or bilayer compounds, the hole content is uniform on the crystallographically equivalent CuO_2 planes. There exist two crystallographically inequivalent outer and inner CuO_2 planes in the multilayer compounds when $n \geqslant 3$. The former and latter are characterized by a fivefold pyramidal and a fourfold square oxygen coordination, respectively. Nonhomogeneous charge distribution of holes among the inequivalent CuO_2 planes was proposed to account for the observed T_c behavior in multilayer members,[9–11] which was supported by nuclear magnetic resonance (NMR) measurements[12–17] and the bond-valence-sums analysis.[18,19] A variety of theoretical models have been developed to include such charge imbalance in understanding the universal relation between T_c and the number of CuO_2 layers in layered superconductors.[20–25] The theoretical results support that an interlayer coupling is necessary to enhance T_c, although the charge imbalance suppresses T_c in multilayer compounds. Recent NMR studies[26,27] revealed the coexistence of the superconductivity and antiferromagnetism in five-layered compounds. The long-ranged 0-Josephson coupling was used to interpret such a coexistence with a rather high value of T_c.[28] These layered models provided a better understanding of the layering-structure effects without taking phonons into account.

The large T_c value and the appearance of antiferromagnetic order in the multilayerd cuprates seemingly cast a great doubt on the role of the electron-phonon interaction. The characteristic layering structures of the high-T_c compounds of homologous series suggest that special two-dimensional physics is important, whereas phonons in two dimensions are probably no different from those in three. However, strong electron-phonon coupling was just found in four-layered mercury-based compound with $T_c=123$ K by Raman scattering.[29] The estimated large electron-phonon interaction parameter λ can even lead to a very high T_c in this material if taken literally in the BCS theory. A significant contribution of the electron-phonon interaction to the superconductivity was also observed in other mercury-based compounds by x-ray-absorption spectroscopic studies.[30] The question of a possible connection between the electron-phonon coupling and the remarkably high-T_c of layered mercury-based superconductors remains open. Measurements of angle-resolved photoemission spectroscopy (ARPES)[31–34] and tunneling[35] provide further evidence for phonons to be a relevant player in the basic physics of high-T_c superconductivity. The observed sizeable isotope effect,[36] either in the materials with relatively low T_c values or when a compound is doped far away from the optimal doping level, offers key experimental evidence for the electron-phonon interaction in high-T_c cuprates. Within a homologous family, the oxygen isotope exponent α decreases systematically with increasing the number of CuO_2 layers in a way that is opposite to T_c in optimally doped compounds.[37] These elaborate experiments strongly suggest that one should include phonons in the theory of high-T_c superconductivity. It is crucial to examine the validity of previous theoretical interpretations when a phonon-containing model is presented.

Besides the CuO_2-layer effect, the homologous series of copper-oxides provides an attractive opportunity to study the effects of hole doping, isotope substitution, external pressure, and cation disorder. It is well known that there is a universal parabolic behavior of T_c with doping for any member in a

homologous series.[38,39] The dramatic enhancement of T_c was usually generated by external pressure in layered cuprates, specifically in mercury-based family.[40–44] The cation disorder was found to suppress T_c in many cuprates.[45–48] The comprehensive understanding of these superconducting transition properties within one single theoretical framework is still challenging. Another crucial question is whether these interesting effects can be understood based on a phonon-mediated theory.

In this work we address these issues by studying rather rich superconducting transition features in a model homologous series of $HgBa_2Ca_{n-1}Cu_nO_{2n+2+\delta}$. We develop a phonon-mediated *d*-wave BCS-like model for layered superconductors. The systematic investigations of mercury-based superconducting series enable us to clarify some properties shared by different cuprates. We show that the theoretical model is successful in explaining the dependence of both the superconducting transition temperature T_c and oxygen isotope exponent α on the doping level, number of CuO_2 layers, pressure, and lattice distortion in cuprate superconductors. The interlayer coupling is found to play an important role in the significant enhancement of T_c and in the systematic reduction of α in a layered homologous series.

The outline of this paper is as follows: In Sec. II, we give the phonon-mediated *d*-wave BCS equation in the general case of any CuO_2 layers per unit cell. We choose $HgBa_2Ca_{n-1}Cu_nO_{2n+2+\delta}$ as an example to study the possibility of phonon-mediated superconductivity. Section III is devoted to the numerical results and analysis for the superconducting transition temperature as functions of the hole concentration and number of CuO_2 layers at atmosphere pressure. In Sec. IV, we give the general formalism for the oxygen isotope exponent. We predict the oxygen isotope effects and discuss the role of interlayer coupling on them. Section V is focused on the pressure dependence of both the superconducting transition temperature and isotope exponent. Three variables are proposed to account for the pressure effects. We use the framework to analyze presently available data, compare our results to experiments, and suggest further measurements. In Sec. VI, we show how the phonon frequency changes the superconducting transition temperature and isotope effect. The physical implication of this behavior to the observed lattice distortion is also discussed. In Sec. VII, we examine the validity of the theoretical model in other homologous series such as the Bi- and Tl-based family. Conclusions are presented in Sec. VIII.

II. THEORETICAL APPROACH

Let us start with the model Hamiltonian including the basic in-plane pairing term and a weak interlayer tunneling coupling term:

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

where $c^{\dagger l}_{k\sigma}$ is a quasiparticle creation operator on layer l with spin projection σ and wave-vector k, ε_k is the quasiparticle dispersion, μ is the chemical potential, the summation over ll' runs over the layer indices of the unit cell, and the pairing potential $V_{kk'}$ is assumed to be independent of l, originating from some of the proposed mechanisms, which we do not attempt at specifying. The interlayer tunneling is parameterized by $V_{\perp}(k) = V_{\perp} g^4(k)$, with $g(k) = \cos k_x - \cos k_y$ and $V_{\perp}$ being the interlayer tunneling strength.[49]

By characterizing the superconducting gap by the order parameter $b^l_k = \langle c^l_{k\uparrow} c^l_{-k\downarrow} \rangle$, we have the equation for the gap function Δ^l_k based on BCS theory,

$$\Delta^l_k = -\sum_{k'} V_{kk'} b^l_{k'} + V_{\perp}(k)(b^{l+1}_k + b^{l-1}_k), \quad (2)$$

where $b^l_k = \Delta^l_k \chi^l_k$ and the generalized pair susceptibility is $\chi^l_k = (2E^l_k)^{-1} \tanh(\beta E^l_k/2)$ with the quasiparticle spectrum $E^l_k = \sqrt{(\varepsilon_k - \mu)^2 + |\Delta^l_k|^2}$ and $\beta = (k_B T)^{-1}$.

The spatial dependence of the gap is taken as[5] $\Delta^l_k = \Delta^{\pm}_k e^{\pm i\alpha l}$. The general solution of the homologous part is $\Delta^l_k = \Delta^+_k e^{i\upsilon l} + \Delta^-_k e^{-i\upsilon l}$. Because the gap vanishes on the layer ends $l=0$ and $n+1$, the natural boundary conditions for the gap are $\Delta^0_k = \Delta^{n+1}_k \equiv 0$. The wave vector of the oscillating gap can be determined by

$$\begin{pmatrix} 1 & 1 \\ e^{i\upsilon l} & e^{-i\upsilon l} \end{pmatrix} \begin{pmatrix} \Delta^+_k \\ \Delta^-_k \end{pmatrix} = 0.$$

The vanishing determinant of the matrix has a nontrivial solution only when $\upsilon = \xi\pi/(n+1)$ with ξ being an integer, so $\Delta^+_k = -\Delta^-_k \equiv \Delta_k$. The solution of the spatial dependence of the gap is then given by $\Delta^l_k = 2i\Delta_k \sin[l\pi\xi/(n+1)]$. The solution with the lowest energy is nodeless inside the CuO_2 layers which leads to $\xi = 1$ for the superconducting state. The spatial dependence of the gap can be expressed by $\Delta^l_k = 2i\Delta_k \sin[l\pi/(n+1)]$.

Around T_c, we can approximate $\chi^l_k \simeq (2E_k)^{-1} \tanh(\beta_c E_k/2) \equiv \chi_k$. Substituting χ^l_k and Δ^l_k into Eq. (2), we have a simple Δ_k equation

$$\Delta_k + \sum_{k'} V_{kk'} \chi_{k'} \Delta_{k'} = f(n) V_{\perp}(k) \chi_k \Delta_k, \quad (3)$$

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

Considering a phonon-mediated interaction, we may take $V_{kk'} = -Vg(k)g(k')$; $|\varepsilon_k - \mu|$ or $|\varepsilon_{k'} - \mu| < \omega_0$ where $V > 0$ is the in-plane pairing interaction strength, and ω_0 is the cutoff of the phonon frequency. Assuming no cutoff for the interlayer pairing tunneling process, one can rewrite the gap equation (3) by

$$1 = \sum_k \frac{V g^2(k) \chi_k}{1 - f(n) V_{\perp}(k) \chi_k} \theta(\omega_0 - |\varepsilon_k - \mu|), \quad (4)$$

where the gap function $\Delta_k = \Delta g(k)/[1 - f(n) V_{\perp}(k) \chi_k]$, and the step function $\theta(x)$ takes care of the condition $|\varepsilon_k - \mu|$ or $|\varepsilon_{k'} - \mu| < \omega_0$. The T_c can be obtained by solving Eq. (4) with $\Delta = 0$. The constraint condition for the hole concentration n_H in CuO_2 plane in conjunction with μ is given by

$$n_H = \frac{1}{2} - \frac{1}{2}\sum_k \tanh\left(\frac{|\varepsilon_k - \mu|}{2k_B T_c}\right). \quad (5)$$

Although the interlayer tunneling has anisotropic *s*-wave symmetry and thus represents interlayer *s*-wave pairing, and the function $g(k)$ has *d*-wave symmetry,[4] the solution of the gap equation in reality has pure *d*-wave symmetry, which has been revealed by the quasiparticle tunneling spectra of multilayer cuprates.[50] The validity of the *d*-wave BCS formalism in describing the superconducting state of cuprates has been supported by recent measurements of ARPES[51] and transport properties.[52,53]

Once knowing the dispersion ε_k and the values of ω_0, V, and/or $V_\perp$, one can obtain the T_c behavior from Eqs. (4) and (5). We chose a typical $\omega_0=0.060$ eV. The energy scale lies within the kink range of (0.050–0.080 eV) from ARPES.[31–34] Far-infrared measurements[54] have established that the phonon modes scarcely change amongst the cuprates irrespective of their T_c changes. An unchanged mean boson energy of 52 ± 8 meV for all dopings was recently detected by tunneling.[35] Thus, the choice of $\omega_0=0.060$ eV should capture the reasonable energy scale of the phonon cutoff frequency for cuprate superconductors.

For the dispersion we use $\varepsilon_k = t'_{\text{eff}} \cos k_x \cos k_y + t''_{\text{eff}}(\cos 2k_x + \cos 2k_y)$ with t'_{eff} and t''_{eff} being the effective next-nearest-neighbor and next-next-nearest-neighbor hoppings, respectively. Such a dispersion can reproduce the bandwidth and Fermi-surface shape of many cuprate superconductors.[55,56] According to the t-t'-J model, $t'_{\text{eff}}=J+2t'$ and $t''_{\text{eff}}=J/4$, where J is the antiferromagnetic interaction. Experiments and calculations give a $J=0.128$ eV which does not depend significantly on the compounds.[57] Values of $J=0.128$ eV and $t''_{\text{eff}}=0.032$ eV are hence expected to be a generally good representation for all Cu-O materials. For a homologous series, the t'_{eff}, t''_{eff}, and V values is assumed to be the same. Their T_c difference for the materials having different CuO_2 layers is only controlled by the interlayer tunneling strength $V_\perp$. The value of $V_\perp$ can be determined by using the experimental values of the maximum superconducting transition temperature $T_c^{\max}$ for the monolayer and bilayer compounds in each homogeneous series.

There is only a single CuO_2 layer in the monolayer system ($V_\perp=0$). The $T_c^{\max}$ value solely depends on the choice of t'_{eff} and V. The driving force of high-T_c superconductivity is believed to come mainly from the almost same CuO_2 plane. One can reasonably assume a compound-independent V for these materials. Thus, the variation of $T_c^{\max}$ among different families is due to the change in t'_{eff}. It has been established[8,58,59] that a larger t'_{eff} or t' is always in favor of a higher $T_c^{\max}$. By using $t''_{\text{eff}}=0.032$ eV and changing t'_{eff} from -0.032 to 0.0914 eV, Eq. (4) bears the experimentally observed $T_c^{\max}$ ranging from 30 to 97 K of various optimally doped monolayer cuprates when taking $V=0.037\,62$ eV. $HgBa_2CuO_{4+\delta}$ has the highest $T_c^{\max}=97$ K amongst the monolayer cuprates reported so far and thus has the largest t'_{eff}. Other optimally doped monolayer compounds with a $T_c^{\max}<97$ K should have a smaller t' or t'_{eff}. Therefore, we can estimate the relative t'_{eff} by using the experimentally observed $T_c^{\max}$ for an individual material.

In choosing the dispersion ε_k, the quasiparticle band is considered to be the same for any member within a homologous series. The universality of the low energy exitation of the Bi-based family has been revealed from ARPES measurements.[60–63] That is, the quasiparticle band is rather flat near $(\pi,0)$, while it is quite dispersive and defines a clear Fermi crossing along the $(0,0)$-(π,π) direction. The dispersion used here has the overall features as observed. Although there is a small change of the electronic states due to bilayer band splitting,[62,64] the splitting is suppressed by strong many-body effects.[21] It has also been found[62] that the bilayer splitting is much smaller in the superconduting state than that in the normal state. We then assume safely that the bilayer splitting has no significant influence on the bulk superconducting properties studied, although it may be necessary for interpreting anisotropic transport properties in the normal state. To clarify its origin, more experimental studies on the multilayer cuprates are needed.

III. SUPERCONDUCTING TRANSITION TEMPERATURE

We first examine how the phonon-mediated BCS-like model describes the T_c behavior for layered cuprates. We chose $HgBa_2Ca_{n-1}Cu_nO_{2n+2+\delta}$ as an example because T_c is highest both at ambient condition and under high pressure in any member of this homologous series and it strongly depends on the number of CuO_2 layers. The structure of all these compounds is relatively simple, and furthermore, T_c can be changed by doping level over a relatively wide regime. Thus, this homologous series is a very good candidate for studies.

For mercury-based compounds, we take $t'_{\text{eff}}=0.0914$ eV, $t''_{\text{eff}}=0.032$ eV, and $V=0.037\,62$ eV.[8] Such a choice can bear the experimentally observed T_c versus n_H behavior of $HgBa_2CuO_{4+\delta}$ with a maximum T_c of 97 K at the optimal doping based on a *d*-wave BCS-like model without taking phonons into account.[8] Meanwhile, the $V_\perp/V$ ratio was estimated to be 0.0332 by taking the experimental values of T_c of 97 and 127 K for the monolayer and bilayer mercury-based compounds, respectively. In order to keep the consistency of the theory, we use the same dispersion ε_k, V, and $V_\perp$ in the calculations.

Figure 1 shows the calculated T_c in the homologous series of $HgBa_2Ca_{n-1}Cu_nO_{2n+2+\delta}$ as a function of the hole concentration n_H. The obtained T_c of all these materials varies parabolically with the doping level. This is a good agreement between the theory and experiments in the monolayer, bilayer, and trilayer compounds.[38,39] These results support the negligible phonon contribution to interlayer coupling process. Increasing the number of CuO_2 layers shifts upwards the whole T_c versus n_H curves. For the larger members, the relation between T_c and n_H becomes almost undistinguishable. All these features are consistent with those obtained from a *d*-wave BCS-like model.[8]

Figure 2 shows the CuO_2-layer dependence of the calculated maximum T_c in the homogeneous series of the optimally doped $HgBa_2Ca_{n-1}Cu_nO_{2n+2+\delta}$. The experimental data points[11] are also plotted for comparison. As can be seen, T_c initially increases with increasing the number CuO_2 layers

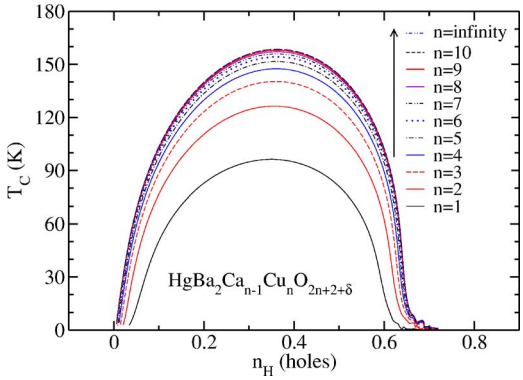

FIG. 1. (Color online) The superconducting transition temperature T_c versus the hole concentration n_H in the homologous series of $HgBa_2Ca_{n-1}Cu_nO_{2n+2+\delta}$.

and then saturates as $n \rightarrow \infty$. This behavior is in good agreement with those obtained from previous theoretical studies.[5,8,11] The upper limit of T_c for the infinite layer compound is 161.6 K, which is very close to the reported value of 162.1 K from the previous theoretical consideration.[8]

Amongst the optimally doped multilayer compounds, only the theoretical T_c value in the trilayer material is close to the experimental value. There is a large difference between the theory and experiments for the compounds having more CuO_2 layers. Within the present theoretical framework, the outer and inner CuO_2 planes are considered to be equivalent in all multilayer systems. This treatment is reasonable for the trilayer systems in which the difference of the hole content between the outer and inner planes is almost negligible and is nearly the same at the optimal doping from the NMR measurements.[13,14,16,17] That is why the theoretical results for the $n=3$ member agree well with experiments. For other multilayer compounds, nonhomogeneous charge distribution among the inequivalent CuO_2 planes becomes more pronounced.[12–19] Therefore, one should include the charge imbalance in the theory in order to account for the experimentally observed T_c reduction in multilayer systems. Constant efforts have led to a better understanding of this behavior.[20–25] Interestingly, Angilella and Pucci[23] presented a nice analysis of the effect of nonuniform hole distribution by using a similar BCS equation. A shift from the uniform distribution was found to make either inner or outer planes prevail. The appearance of the upper limit of T_c in uniform charge-distributed multilayer materials offers a great hope of the possible enhancement of T_c as high as ~160 K if one can fabricate homogeneous copper-oxide multilayers by atomic layer-by-layer growth.

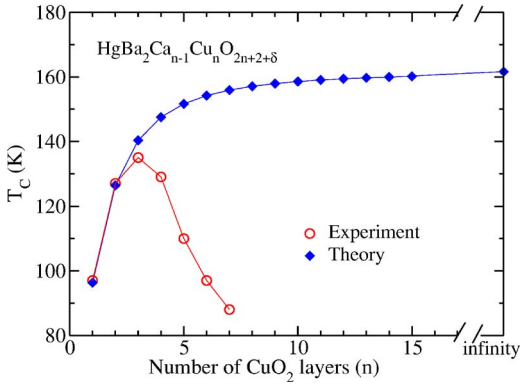

FIG. 2. (Color online) Variation of the superconducting transition temperature T_c with the number of CuO_2 layers in the optimally doped $HgBa_2Ca_{n-1}Cu_nO_{2n+2+\delta}$. The experimental data points were taken from Ref. 11.

It is worth emphasizing that the true comparison between the theories and experiments crucially depends on the sufficient accuracy of the experimental information provided. For an individual multilayer mercury-based compound, the experimentally reported T_c value has been generally believed to be its maximum value at the optimal doping. NMR is also considered as a sensitive probe to identify the doping level by measuring the temperature dependence of Knight shift K_{ab} or K_c. It has been accepted[65,66] that $K_{ab,c}$ in underdoped (overdoped) compounds decreases (increases) with decreasing temperature when approaching the T_c, whereas $K_{ab,c}$ in the optimally doped compounds are almost temperature independent at high temperatures near 300 K. Knight shift is therefore the fingerprint of the doping character. A continuous reduction of $K_{ab,c}$ with decreasing temperature down to T_c was generally observed in the $n=3$,[16] $n=4$,[16] and $n=5$ (Ref. 26) members with $T_c=133$, 123, and 108 K, respectively, signalling that all these multilayer compounds are underdoped. Furthermore, the Hall number per CuO_2 plane was estimated to be 0.12 for the $n=5$ member with $T_c=108$ K.[67] This value is relatively small compared to many other optimally doped cuprates.[68,69] The signature of the optimal doping in cuprate superconductors can be also identified by the temperature dependence of the Hall coefficient R_H at high-magnetic fields.[68] There has been no such identification in multilayer mercury-based compounds. It remains unclear whether the experimental reported T_c's are the maximum values of the multilayer mercury-based compounds with the optimal doping.

IV. OXYGEN ISOTOPE EFFECT

We now study how the c-axis layering structure affects the isotope effect in mercury-based superconductors. We focus ourselves on the analysis in the monolayer, bilayer, and trilayer materials. This consideration is based on the following experimental facts: (i) Most experimental data over an entire doping regime are available only for the compounds having $n \leqslant 3$; (ii) Oxygen doping of the member $n \geqslant 4$ does not induce any noticeable decrease of T_c and there is no convincing evidence for supporting the optimal doping; (iii) an overdoped material for $n \geqslant 4$ is extremely hard to be synthesized and only strongly reducing or highly oxidizing conditions promote appreciable T_c variations; and (iv) the structures of the members with $n \geqslant 4$ always contain multiple intergrowths of the lower and/or higher members of the homologous series.

For phonon-mediated pairing, ω_0 can be assumed to vary with the isotope mass M as $M^{-1/2}$. By differentiating Eq. (4) with respect to both T_c and ω_0, we can express α as

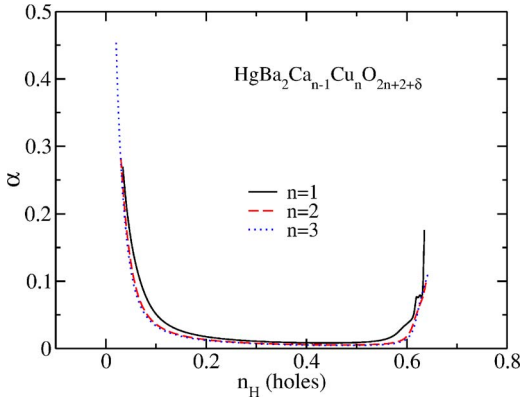

FIG. 3. (Color online) The oxygen isotope exponent α versus the hole concentration n_H in $HgBa_2Ca_{n-1}Cu_nO_{2n+2+\delta}$ $(n=1,2,3)$.

$$\alpha = \frac{1}{2}\omega_0 T_c \tanh\left(\frac{\omega_0}{2T_c}\right)\frac{I_1}{I_2},$$

$$I_1 = \sum_k \frac{g^2(k)\delta(\omega_0 - |\varepsilon_k - \mu|)}{2\omega_0 - A(\omega_0)},$$

$$I_2 = \sum_k \frac{g^2(k)B\theta(\omega_0 - |\varepsilon_k - \mu|)}{[2|\varepsilon_k - \mu| - A(|\varepsilon_k - \mu|)]^2}, \tag{6}$$

where $A(x)=f(n)V_\perp(k)\tanh(x/2T_c)$ and $B=|\varepsilon_k-\mu|^2\mathrm{sech}^2(|\varepsilon_k-\mu|/2T_c)$.

Figure 3 shows the relation between the α and hole concentration n_H in $HgBa_2Ca_{n-1}Cu_nO_{2n+2+\delta}$ $(n=1,2,3)$. Here the ambient-pressure T_c values of 127 and 135 K for the bilayer and trilayer compounds, respectively, are used to determine their $V_\perp$ values in order to reproduce experiments. The calculated α versus n_H curves have many interesting features. The obtained α is positive in all these layered materials. It has a minimum on the overdoped side and becomes large for the highly underdoped or highly overdoped samples. We see that α decreases monotonically with doping on the underdoped side, reaching a minimum on the overdoped side after passing through the optimal doping level, and then increases gradually when the material is highly overdoped. This is the generic picture observed experimentally in many copper-oxide superconductors.[36,37,70,71] Increasing the number of CuO_2 layers moves the whole α versus n_H curves downwards. There have been no reports of the isotope effect studies in mercury-based compounds. A good agreement between theory and experiments has been reached in the sister system $Bi_2Sr_2CaCu_2O_{8+\delta}$ over a whole doping range.[37] Thus, the present theoretical model does catch the essential features of the oxygen isotope effect.

At optimal doping, the calculated α decreases systematically with an increasing number of CuO_2 layers in each homologous family. This behavior is in qualitative agreement with the experimental results observed in bismuth-based superconductors.[37] We have found that the calculated α strongly depends on the number of CuO_2 layers in the homologous series of the low-T_c groups. However, the CuO_2 layer effect on α becomes weak in high-T_c mercury-based series. The systematic variation of T_c among various copper-oxide materials is believed to be dominated by the hopping parameter t'_{eff}.[8,58,72] Similar to the T_c behavior, the α difference between different superconducting series is also related to the band structures. Within a homologous series, the band structure used is assumed to be the same. Both the T_c and α difference among the materials having different CuO_2 layer(s) within the unit cell is controlled by the interlayer coupling between the adjacent CuO_2 layers which is naturally responsible for the CuO_2 layer dependence of the isotope effect. The interlayer coupling has a tendency to enhance the T_c value but to suppress the α value.

V. PRESSURE EFFECT

Next we study the pressure effect in mercury-based superconductors. It has been found[73,74] that both n_H and V are two intrinsic pressure variables. Within the phonon-mediated BCS-like model, we need to include ω_0 as another pressure-sensitive variable. Assuming that these variables change with pressure through a term given by $1-v(P)/v(0)$, we write

$$y_i(P) = y_i(0)\left[1 + \frac{d\ln y_i}{dP}B_0\left(1 - \frac{v(P)}{v(0)}\right)\right], \tag{7}$$

where $y_1=V$, $y_2=n_H$, and $y_3=\omega_0$ over the range studied. The pressure dependence of the relative volume can be well described by the first-order Murnaghan equation of state $v(P)/v(0)=(1+B'_0P/B_0)^{-1/B'_0}$, where $v(0)$ is the cell volume at ambient condition, $B_0(\equiv 1/\kappa_v)$ is the bulk modulus, $\kappa_v \equiv -d\ln v/dP$ is the volume compressibility, and B'_0 is the pressure derivative of B_0.

For the cutoff phonon frequency $\omega_0(P)$, we may take the pressure dependence of the frequency ω_{B1g} of the B_{1g} Cu-O bond-buckling phonon since it has been ascribed as a bosonic mode influencing high-T_c superconductivity.[32] The information on $\omega_{B1g}(P)$ can be obtained by high-pressure Raman scattering measurements. There has been no measurement of actual ω_{B1g} dependence on pressure for Hg-based cuprates. High-pressure Raman spectra for Hg-based compounds[75] showed that the Raman frequency ω of the apical oxygen A_{1g} mode increases with pressure at nearly the same rate as the normalized frequency below 5 GPa for Hg1201 and Hg1223, i.e., $d\ln\omega/dP=7.5\times10^{-3}$ GPa^{-1}. A similar result was also obtained theoretically for the A_{1g} mode in Hg1201.[76] Considering the fact that the magnitude of $d\ln\omega/dP$ remains roughly the same for the B_{1g} and A_{1g} mode,[77] we may have $d\ln\omega_0/dP=7.5\times10^{-3}$ GPa^{-1} for the optimally doped Hg-based materials.

For structural parameters, we use the measured bulk moduli B_0 of 65.4, 91.6, and 83.8 GPa and their derivatives B'_0=4.53, 5.30, 5.80 for Hg1201, Hg1212, and Hg1223, respectively.[78] Measurements of the Hall effect under high pressure are recognized to be an effective method of providing an accurate value of the pressure-induced change of the hole content. In general, the Hall coefficient R_H decreases with pressure in high-T_c cuprates.[69,79] Recent studies[79] revealed that the Hall coefficient in multilayered cuprates decreases with pressure with a relatively low rate $d\ln R_H/dP$

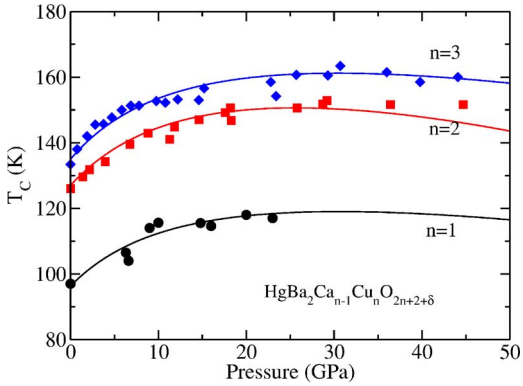

FIG. 4. (Color online) Pressure dependence of the superconducting transition temperature T_c in the optimally doped $HgBa_2Ca_{n-1}Cu_nO_{2n+2+\delta}$ $(n=1,2,3)$. Theoretical results are plotted by the curves. Circles, squares, and diamonds denote the experimental data points taken from Ref. 40 for the monolayer, bilayer, and trilayer compounds, respectively.

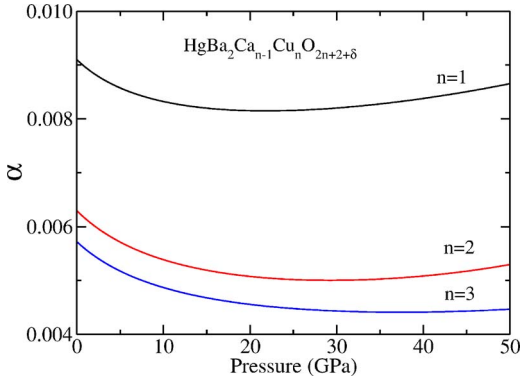

FIG. 5. (Color online) Calculated oxygen isotope exponent α as a function of pressure in the optimally doped $HgBa_2Ca_{n-1}Cu_nO_{2n+2+\delta}$ $(n=1,2,3)$.

$\approx -3\%--7\%$ GPa^{-1}. There are no reports of such measurements in Hg-based cuprates. First-principles study showed that the application of pressure up to 5 GPa adds almost same 0.025 holes for all Hg-based compounds,[80] approximately yielding a $d\ln n_H/dP$ of 0.03 GPa^{-1} at the optimal doping.[81] This value corresponds to a value of $d\ln R_H/dP \sim -5\%$ GPa^{-1} for Hg-based compounds, consistent with the measurements in other homologous series,[79] according to the relation $d\ln R_H/dP=-1/B_0-d\ln n_H/dP$. At the moment, we take $d\ln n_H/dP=0.03$ GPa^{-1} in the calculations. The pressure coefficient of the pairing interaction strength V was found to obey a simple formula $d\ln V/dP=\gamma/B_0$ in high-T_c cuprates with γ being a material-dependent parameter.[18,19] We thus have all the parameters required to obtain the high-pressure behavior of T_c.

Figure 4 shows the calculated T_c as a function of pressure up to 50 GPa for the optimally doped $HgBa_2Ca_{n-1}Cu_nO_{2n+2+\delta}$ $(n=1,2,3)$ by taking γ=1.7, 2.0, and 1.9, respectively. As pressure is increased, T_c increases initially until passing saturation at a critical pressure P_c; at even higher pressures T_c decreases slightly. We notice that our theoretical model reproduces the saturated T_c's and the P_c's of these materials well. The theoretical results are in very good agreement with those experimentally obtained by Gao *et al.*[40] This is the first effort of the application of the phonon-mediated *d*-wave BCS-like model to the pressure effect on T_c in cuprate superconductors. Good agreement between theory and experiment supports V, n_H, and ω_0 as three intrinsic pressure variables. It is worth noting that three similar parameters that appear in the McMillan formula were found to be responsible for the pressure effect in conventional superconductors.[82,83] It has been found[84–87] that the pressure dependence of T_c along the *c* axis is very small in the optimally doped $YBa_2Cu_3O_{7-\delta}$. This experimental observation indicates that $V_\perp$ should not be treated as a pressure-dependent parameter. Thus, enhancing T_c by pressure is not controlled by interlayer coupling. We therefore attribute the superconductivity primarily to intraplanar pairing interactions. The same conclusion was previously drawn by Schilling[88] from the observation of the T_c increase related to the reduction in the area of the CuO_2 planes.

It has been shown[74] that there exists a correlation between the critical pressure P_c and the difference between the superconducting transition temperatures ΔT_c at the ambient pressure and critical pressure in the optimally doped cuprates. ΔT_c increases under P_c at the rate of near 1 K/GPa, signaling that increasing pressure to a critical level would drive T_c to a saturation value in an optimally doped high-T_c superconductor. This ubiquitous behavior may serve as an indicator of a hallmark of hole-doped high-T_c cuprates, as pointed out by Schirber *et al.*[89]

The significant T_c increase with pressure points to the possibility that one may obtain the further enhancement of T_c at ambient condition in similar compressed structures through the preparation of epitaxial films deposited on substrates having smaller lattice parameters than those of the optimally doped Hg-based compounds. This idea is based on the uniaxial pressure effect on T_c within the *ab* plane and along the *c* axis.[84–87] It has been proven true by Locquet *et al.*,[90] who reported a doubling T_c of 49 K in a slightly underdoped $La_{1.9}Sr_{0.1}CuO_4$ thin film from its bulk value of 25 K by tuning epitaxial strain. The dramatic T_c behavior was suggested to come from strain-induced modification of either the pairing interaction[91] or the band structure.[92] An epitaxially compressive strain-induced metal-insulator transition above room temperature was also observed in maximum colossal magnetoresistance manganite thin films.[93]

We now turn to the analysis of the high-pressure behaviors of the oxygen isotope effect. Based on the parameters determined above, we can compute the variation of α with pressure by using Eqs. (4)–(7). In Fig. 5, we plotted the pressure dependence of α up to 50 GPa in the optimally doped $HgBa_2Ca_{n-1}Cu_nO_{2n+2+\delta}$ $(n=1,2,3)$. We find that α first decreases with increasing pressure in a way similar to $d\ln T_c/dP$, reaching a minimum at some pressure level near the critical pressure at which T_c is maximized, after that it slightly increases with pressure. Such a slight increase in α is reasonable on physical grounds, since both the phonon frequency and hole content would have a tendency to enhance α when the applied pressure is extremely high. A systematic decrease in α with the number of CuO_2 layers is observed

over the entire pressure range that was studied. This indicates again that the interlayer coupling plays an important role in determining superconducting properties in multilayer cuprates. For Hg-based materials, measurements of the isotope effect under high pressure would be of interest, especially the examination of the enhancement of α at high pressures.

It should be mentioned that the high-pressure behaviors of T_c and α are not sensitive to the choice of $d \ln \omega_0/dP$ but depend strongly on the $d \ln n_H/dP$ values. Since there is no measurement of Hall coefficient dependence on pressure for Hg-based cuprates, we took the theoretical $d \ln n_H/dP$ values from first-principles calculations. These values agree nicely with the early phenomenological estimations for these materials.[94] Such a choice of $d \ln n_H/dP$ is believed to be physically plausible. The hole concentration can be expressed as $n_H=N/v$, where N is the number of carriers within a unit cell having a volume of v. Thus, n_H must change under pressure even though N remains unchanged. Taking the partial derivative of n_H with respective of pressure yields $d \ln n_H/dP=1/B_0$. We then have $d \ln n_H/dP=0.011 \sim 0.015$ when the number of carriers N keeps a constant. In fact, there are some effects such as pressure-induced charge transfer or pressure-induced delocalization. These effects should contribute to the N change under pressure, yielding a larger $d \ln n_H/dP$, accordingly.

VI. PHONON SOFTENING

In principle, a softening of the phonon frequencies could be expected due to a reduction of the force constants. The change of the force constants is usually caused by the change of the bonds or by the separation of the CuO_2 planes by the difference in the cation ionic radii. It has been shown[95,96] that the B_{1g} phonon frequency decreases with increasing cation ionic radius in many cuprates. The B_{1g} Cu-O bond buckling can therefore serve as a measure of the local lattice distortion. Such an effect is distinct from the well known effects such as doping and number of CuO_2 layers. Besides modifying phonon frequencies, the degree of lattice distortion is also strongly coupled to the electronic band structure of the saddle points, and in particular to their extended character.[97] Thus, the lattice distortion results in both the changes of phonon frequency and electronic band structure which combine to change T_c in cuprate superconductors.

We have investigated the phonon frequency dependence of T_c in the optimally doped $HgBa_2Ca_{n-1}Cu_nO_{2n+2+\delta}$ ($n=1,2,3$). The theoretical results are presented in Fig. 6. A systematic reduction of T_c is seen with the phonon frequency softening in all these materials within the reasonable ω_0 range. When $\omega_0 < 0.060$ eV, the T_c reduction is significant. The phonon softening effect is more pronounced in the monolayer material, but it becomes weaker with the increasing number of CuO_2 layers. The interlayer coupling competes with the phonon softening to suppress the T_c reduction. This behavior is physically acceptable because the cation substitution is always produced in the block layer(s) and the CuO_2 planes in the multilayer system are not greatly affected. However, one expects a significant influence on the B_{1g} phonons in the octahedral monolayer. Our theoretical

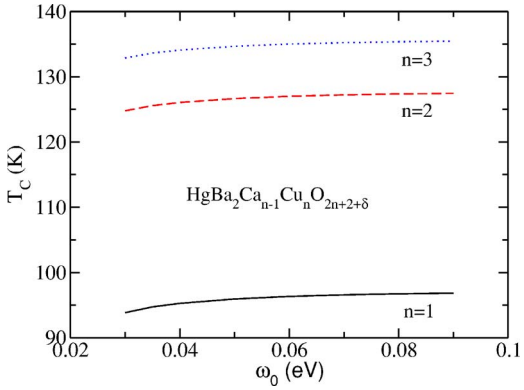

FIG. 6. (Color online) Cutoff frequency ω_0 dependence of both the superconducting transition temperature T_c in the optimally doped $HgBa_2Ca_{n-1}Cu_nO_{2n+2+\delta}$ ($n=1,2,3$).

results are consistent with the observations where T_c was found to reduce dramatically with increasing cation disorder in monolayer La- and Bi-based systems[45–47] but it is not significantly affected in bilayer systems.[48]

The reduction of T_c through the phonon softening is further supported by the isotope effect. Figure 7 shows the phonon frequency dependence of the oxygen isotope exponent α in the optimally doped $HgBa_2Ca_{n-1}Cu_nO_{2n+2+\delta}$ ($n=1,2,3$). It is seen that α increases with the phonon frequency softening in a way opposite to the T_c reduction. The smaller the phonon frequency, the larger the α value. It is interesting to notice that α also systematically decreases when the number of CuO_2 layers is increased over the entire ω_0 range studied.

The Fermi surface topology in high-T_c cuprates in the antinodal region is believed to be mainly affected by a change of t'_{eff}. The variation of t'_{eff} can account for the variation of T_c among various optimally doped cuprates.[8,58,72] Recently, Chen and Su (Ref. 98) emphasized the importance of t'_{eff} in the rare-earth ionic size effect on T_c of the optimally doped $RBa_2Cu_3O_{7-\delta}$ superconductors. Experimentally, T_c was observed to increase systematically with the increasing the rare-earth ionic radius ($r_{R^{3+}}$).[99,100] Increasing $r_{R^{3+}}$ leads to the increase of t'_{eff} (Ref. 98) and the softening of B_{1g} phonon frequencies.[95] Although the B_{1g} phonon softening tends to

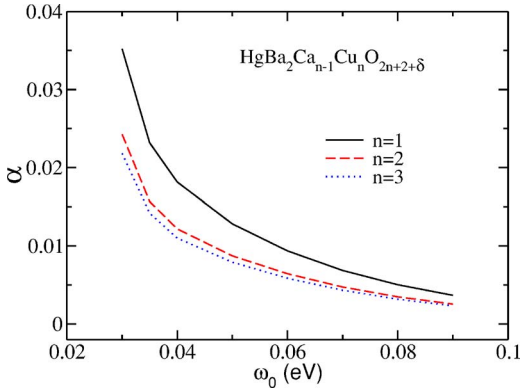

FIG. 7. (Color online) Cutoff frequency ω_0 dependence of both the oxygen isotope exponent α in the optimally doped $HgBa_2Ca_{n-1}Cu_nO_{2n+2+\delta}$ ($n=1,2,3$).

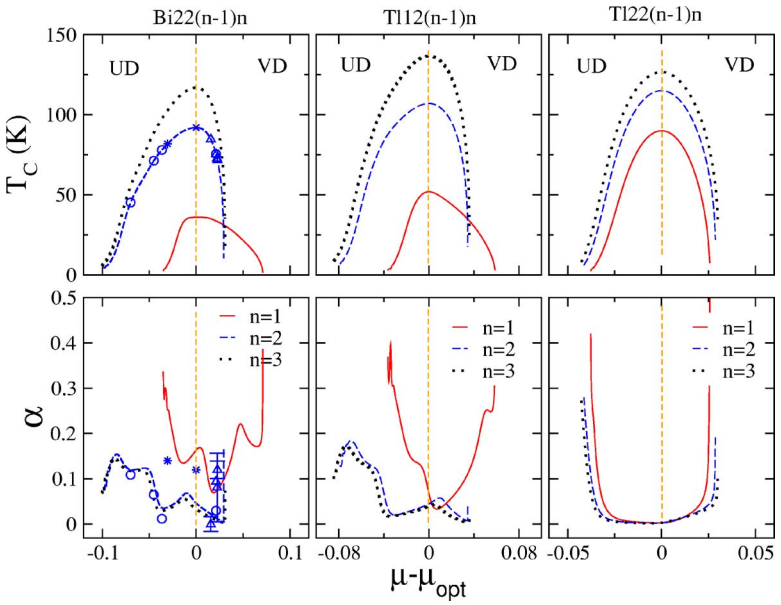

FIG. 8. (Color online) The superconducting transition temperature T_c and oxygen isotope exponent α versus the chemical potential μ relative to the optimal doping in layered cuprate superconductors $Bi_2Sr_2Ca_{n-1}Cu_nO_{2n+4+\delta}$ [Bi22$(n-1)n$], $TlBa_2Ca_{n-1}Cu_nO_{2n+3+\delta}$ [Tl12$(n-1)n$], and $Tl_2Ba_2Ca_{n-1}Cu_nO_{2n+4+\delta}$ [Tl22$(n-1)n$]. Our theoretical results are plotted by the curves. The experimental data for Bi2212 are denoted by triangles from Ref. 71, circles from Ref. 70, and stars from Ref. 37. The vertical dashed lines at $\mu=\mu_{opt}$ denote the optimal doping level. UD and VD represent the underdoped and overdoped sides, respectively.

decrease T_c as seen in Fig. 6, the enhancement of t'_{eff} is in favor of the increase of T_c. As a result, the experimentally observed increase of T_c with $r_{R^{3+}}$ is dominated by the electronic states through the enhancement of t'_{eff} in $RBa_2Cu_3O_{7-\delta}$.

It has been suggested[66] that the NMR oxygen linewidth associates with the lattice distortion: Tl- and Hg-based monolayer cuprates with large T_c and small linewidth, and La- and Bi-based monolayer cuprates with lower T_c and larger linewidth. The low-T_c monolayer cuprates are known to exhibit large structural distortion. Thus, the competition between the changes in phonon frequency and band structure due to the lattice distortion in low-T_c groups deserves further studies. For conventional superconductors, Garland *et al.*[101] showed that the effect of lattice disorder on T_c is controlled by the changes in the phonon frequency spectrum and electronic band density of states at the Fermi level. In this aspect, the disorder effect in superconducting materials seemingly has a common origin.

VII. OTHER MULTILAYER SYSTEM

Figure 8 shows the results of both the T_c and α for other typical layered families as a function of the chemical potential μ calculated from Eqs. (4) and (6). The calculated T_c of these layered materials varies parabolically with doping, having a maximum T_c when μ reaches a maximum in the density of states producing an optimal doping,[55] in agreement with experiments. Remarkably, the experimental fact that superconductivity occurs over a wide doping range in monolayer material but T_c drops rapidly for highly overdoped bilayer and trilayer materials can be reproduced from the present model as well.

The calculated α versus μ curves have the same character as in the mercury-based family. The obtained α is positive in all these layered materials. It has a minimum on the overdoped side and becomes large for the highly underdoped or highly overdoped samples. Like the La-based family in the low-T_c group,[102] the Bi- and Tl_1-based materials do not have a monotonic doping dependence of α. In high-T_c Tl_2- and Hg-based group, as the hole concentration increases, α first decreases from the underdoped side to the overdoped side and then increases for the highly overdoped samples. It has been suggested[72] that extended van Hove singularities play a more important role for the T_c behavior in the high-T_c group than the low-T_c group. This should also work well for the different isotope effects between the two groups.

Currently, the absence of the relevant information on many layered families makes the comparison of theory and experiment difficult. There are only reports of the isotope effect in the Bi2212 system with a wide doping level.[37,70,71] Bronemann *et al.*[70] obtained a positive α ranging between 0.03 and 0.109 with a minimum near 0.012 for the substituted compound $Bi_2Sr_2Ca_{1-x}Y_xCu_2O_{8+\delta}$. An increase in α with doping was observed by Pringle *et al.*[71] on overdoped $Bi_2Sr_2CaCu_2O_{8+\delta}$ samples. Recently, Chen *et al.*[37] obtained $\alpha=0.14$ and 0.12 for the underdoped and optimally doped materials having $T_c=81.8$ and 92.0 K, respectively. Taking the maximum T_c of 92 K for Bi2212, we can include experimental data points in Fig. 8. The measured α's are nicely situated around the theoretical curve for this material. The salient features of the α behavior are reproduced by our phenomenological model.

For optimally doped materials, the calculated α indeed decreases systematically with an increasing number of CuO_2 layers in each family. However, the CuO_2 layer effect on α becomes weak when passing from Bi-based to Hg-based series. For example, we obtain $\alpha=0.17$ for Bi2201, $\alpha=0.05$ for Bi2212, and $\alpha=0.03$ for Bi2223. The qualitative behavior of α is very similar to the experimental results.[37] For the Hg-based family we have $\alpha=8.6\times10^{-3}$, 6.2×10^{-3}, and 5.3×10^{-3} for monolayer, bilayer, and trilayer materials, respectively. The origin of such a difference is related to the band structures of these families. Within the same homologous series, the band structure used is assumed to be the same. Both the T_c and α difference among the materials having different CuO_2 layer(s) within the unit cell is only controlled

by the interlayer coupling, which is naturally responsible for the CuO_2 layer dependence of the isotope effect. There have been very few reports of isotope measurements in thallium- and mercury-based materials. The predictions made here for α await further experimental studies.

VIII. CONCLUSIONS

We have developed a phonon-mediated *d*-wave BCS-like model for layered copper-oxide superconductors. The homologous series of $HgBa_2Ca_{n-1}Cu_nO_{2n+2+\delta}$ has been chosen as a model system to investigate the superconducting transition features. We have shown that once the band structure and symmetry features are considered, this model can account well for the magnitudes of both the superconducting transition temperatures and oxygen isotope exponents as functions of doping level and number of CuO_2 layers. We have found that this theoretical model also reproduces the CuO_2 layer dependence of the pressure effect, isotope effect, and lattice distortion effect in typical Hg-based cuprates. Thus, all known distinct effects on T_c in cuprates have been interpreted within the present single theoretical framework. We also have found that the theoretical model works well for other homologous series such as the Bi-based and Tl-based families. We have concluded that the interlayer coupling plays an important role in the enhancement of T_c and in the systematic reduction of α in a layered homologous series. Some results of the calculations are certainly only in qualitative agreement with the experimental data, and a much better agreement could be obtained by adjusting the model parameters for each individual compound. However, the model is only intended to describe experimental trends rather than to reproduce all the data accurately. Many predictions have been made and hence await experimental examination. In spite of the simplicity and the limitations of the model, the results strongly suggest the role of the phonons should be included in any real microscopic mechanisms in explaining the superconducting behavior of cuprates. Very recently, we have demonstrated[103] that the variation of α amongst various optimally doped cuprates is controlled by the effective next-nearest-neighboring hopping. The oxygen isotope effect on the superconducting transition is also found to resemble the effect of pressure on the transition.

ACKNOWLEDGMENTS

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