Superconductivity Studied by Solving *Ab Initio* Low-Energy Effective Hamiltonians for Carrier Doped CaCuO₂, Bi₂Sr₂CuO₆, Bi₂Sr₂CaCu₂O₈, and HgBa₂CuO₄

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Understanding the materials dependence together with the universal controlling parameter of superconductivity (SC) in copper oxide superconductors is one of the major challenges in condensed matter physics. Here, we numerically analyze SC by using *ab initio* low-energy effective Hamiltonians consisting of the antibonding combination of Cu $3d_{x^2-v^2}$ and O $2p_{\sigma}$ orbitals without adjustable parameters. We have performed the state-of-the-art variational Monte Carlo calculations for the four carrier doped cuprates with diverse experimental optimal SC critical temperature T_c^{opt} : CaCuO₂ ($T_c^{\text{opt}} \sim 110$ K), Bi₂Sr₂CuO₆ ($T_c^{\text{opt}} \sim 10-40$ K), $Bi_2Sr_2CaCu_2O_8$ ($T_c^{opt} \sim 85-100$ K), and $HgBa_2CuO_4$ ($T_c^{opt} \sim 90$ K). Materials and hole doping concentration (δ) dependencies of the SC order parameter F_{SC} and the competition with spin or charge order show essential and quantitative agreement with the available experiments on the four materials in the following points. (1) In a wide range $0.05 \le \delta \le 0.25$, the ground state is commonly the uniform SC state, which is severely competing with the charge or spin stripe and antiferromagnetic states. (2) F_{SC} at the optimum doping shows amplitude consistent with the superfluid density measured in the muon spin resonance and its dome structure found in δ dependence shows consistency with that of the SC gap in the tunneling and photoemission measurements. Based on the confirmed materials dependence, we further find insights into the universal SC mechanism. (I) $F_{\rm SC}$ increases with the ratio $U/|t_1|$ within the available realistic materials, indicating that $U/|t_1|$ is the principal component controlling the strength of the SC in the real materials dependence. Here, U and t_1 are the on-site Coulomb repulsion and the nearest neighbor hopping, respectively, in the ab initio Hamiltonians. (II) A universal scaling $T_c^{opt} \sim 0.16 |t_1| F_{SC}$ holds. (III) SC is enhanced and optimized if U is increased beyond the real available materials, and it is further enhanced when the off-site interaction is reduced, while the presence of the off-site interaction is important to make the SC ground state against other competing states. The present findings provide useful clues for the design of new SC materials with even higher T_c^{opt} .

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I. INTRODUCTION

The mechanism and origin of the large superconducting (SC) gap, high superfluid density, and high critical temperatures T_c observed in high- T_c superconductors, such as Subject Areas: Condensed Matter Physics, Strongly Correlated Materials, Superconductivity

copper oxides, remain a central challenge in condensed matter physics. In these copper oxides, the *d*-wave SC state is severely competing with other orders, such as spin and charge stripes or antiferromagnetic (AFM) states, and the observed T_c widely ranges from above 130 K to below 10 K. Understanding and reproducing these diverse phenomena without relying on adjustable parameters is hence desirable, especially when clarifying their origin. When *ab initio* calculations are able to reproduce systematic materials dependence quantitatively by solely relying on their crystal structures, it provides us with valuable insight into the universal mechanism behind and into the

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principal components for the enhancement of SC beyond existing materials.

Many studies have suggested severe competitions of the SC with charge or spin stripe and AFM states theoretically based on simplified Hubbard-like or t-J Hamiltonians as models of the cuprate superconductors [1–8]. A positive correlation between U and T_c or SC tendency was also pointed out by taking U as an adjustable parameter in the Hubbard-type Hamiltonians [9]. However, ab initio studies without adjustable parameters are few and it is not clear whether the diversity of the materials dependence can be accounted for. There still exists a limited number of *ab initio* studies: The phase diagram including the SC phase was reproduced by solving the *ab initio* Hamiltonian for a particular case of Hg compound [10]. Ab initio Hamiltonians were derived by Nilsson *et al.* for several cuprate compounds, which reported an empirical observation without solving the Hamiltonians that the experimental optimal T_c is generally higher for larger U/|t| in their parameters [11,12]. The relation of the charge transfer energy to T_c was also pointed out [13,14]. Aside from the cuprates, there exist some ab initio studies on strong-coupling superconducting materials such as the iron-based superconductors [15], fullerene [16], and nickelates [17] to discuss the superconducting properties. However, to our knowledge, there exist no systematic studies on the SC properties by solving solely ab initio Hamiltonians without adjustable parameters with the help of accurate manybody solvers to reveal the origin of the diverse materials dependence, especially for the challenging cuprates. Unless reproducing the materials dependent properties quantitatively, the universal mechanism would also not be able to be identified confidently either.

In this paper we show properties of typical cuprate superconductors calculated by solving the ab initio Hamiltonians of four families of materials, namely, carrier doped CaCuO₂, HgBa₂CuO₄ (abbreviated as Hg1201 hereafter), Bi₂Sr₂CuO₆ (Bi2201 hereafter), and Bi₂Sr₂CaCu₂O₈ (Bi2212 hereafter) [12], by applying a state-of-the-art quantum many-body solver based on the variational Monte Carlo (VMC) algorithm [18,19], including the combination with neural network [20,21] elaborated from earlier variational algorithms [22-25]. It is experimentally known that the optimum critical temperature T_c^{opt} is realized at around $\delta = 0.1-0.15$ [26,27] for doped CaCuO₂ $(T_c^{\text{opt}} \sim 110 \text{ K})$ and Hg1201 $(T_c^{\text{opt}} \sim 90 \text{ K})$, and at around $\delta = 0.15 - 0.25$ [28-30] for doped Bi2212 ($T_c^{\text{opt}} \sim$ 85–100 K) and Bi2201 ($T_c^{\text{opt}} \sim 10$ –40 K). We elucidate that similarity and diversity among the four families, especially the amplitude of the SC order parameters and T_c^{opt} in the experiments, are accounted for by using the present ab initio results, which provides us with insights into the materials dependence and the universal mechanism.

We emphasize that our *ab initio* analyses contain essential differences from most of the Hubbard models studies. One important difference is the presence of the realistic off-site interactions. We will clarify that this crucially stabilizes the charge uniform SC state without clear stripe long-range order.

The dominance of the SC for all the four families is successfully demonstrated. In addition, the δ dependence of $F_{\rm SC}$ has a dome structure with the peak at $\delta \sim 0.05-0.1$ consistently with the experimental indications. On the other hand, the dome peak of T_c appears at larger $\delta > 0.1$ in the experiment. This shift from the dome peak of $F_{\rm SC}$ is understood from the decreasing renormalization factor with decreasing doping, which does not affect $F_{\rm SC}$ but T_c .

Although T_c^{opt} has a variety among these four families, we show universally that (I) the higher SC order parameter F_{SC} at the optimal doping basically results from a larger ratio $U/|t_1|$, where U is the on-site repulsive Coulomb interaction and t_1 is the nearest neighbor hopping in our *ab initio* parameters of single-band effective Hamiltonian for the antibonding (AB) orbital of strongly hybridized Cu $3d_{x^2-y^2}$ and O $2p_{\sigma}$ orbitals. Furthermore, we show that (II) T_c^{opt} is determined by the scaling $T_c^{\text{opt}} \sim 0.16|t_1|F_{\text{SC}}$.

The δ dependence of the local energy suggests a universal superconducting mechanism: Though the bare interaction is strongly repulsive, the Mottness converts it to the strong effective attraction required for the Cooper pairing.

Despite monotonic increase of $F_{\rm SC}$ with $U/|t_1|$ within the existing materials, we further show that (III) larger $U/|t_1|$ beyond the *ab initio* values makes the peak of $F_{\rm SC}$ followed by the reduction. We find that $F_{\rm SC}$ can be roughly 30% more enhanced than the *ab initio* case when U is 20% increased beyond the *ab initio* value by preserving the transfer and other off-site interaction parameters. We also show that $F_{\rm SC}$ is further enhanced to as much as the double of the existing material by the additional reduction of the off-site Coulomb interaction. These searches beyond the *ab initio* parameters for the existing materials offer a guide for future experimental materials design.

This paper is organized as follows. Section II presents the methods and computational details. First, the effective Hamiltonians studied in this paper are summarized in Sec. II A. Then, we give the numerical methods in Sec. II B and define the physical quantities in Sec. II C. We present in Sec. III the results for each of the four families of compounds. Based on the obtained *ab initio* results, in Sec. IV, we further explore the direction to enhance and optimize the SC order parameter by controlling the effective interaction parameters beyond the *ab initio* values, to gain insights into the future materials design. In Sec. V, we discuss our analyses. Summary and conclusion are given in Sec. VI.

II. METHODS

A. Ab initio effective Hamiltonian

Within this paper, we solve the *ab initio* single-band effective Hamiltonians for CaCuO₂, Hg1201, Bi2201, and Bi2212 as derived in Ref. [12]. It should be noted that the single band is constructed from the AB orbital of strongly hybridized Cu $3d_{x^2-y^2}$ and O $2p_{\sigma}$ orbitals and not from the atomic single orbital of Cu $3d_{x^2-y^2}$. This is justified by very large hybridization gap of the AB and bonding (B) or nonbonding (NB) orbitals. See Appendix D of Ref. [12] and the last paragraph of Sec. V in this paper. Here the transfer and interaction parameters are derived at values close to the experimental optimum hole concentration (at 10% doping for CaCuO₂ and Hg1201 and at 20% for the two Bi compounds). This choice is appropriate in this paper, because properties at optimum hole concentration are the central subject. The effective Hamiltonians have the form

 $\mathcal{H}=\mathcal{H}_{\mathrm{kin}}+\mathcal{H}_{U}+\mathcal{H}_{V},$

(1)

with

$$egin{aligned} \mathcal{H}_{\mathrm{kin}} &= \sum_{i,j,\sigma} t_{ij} c_{i\sigma}^{\dagger} c_{j\sigma}, \ \mathcal{H}_{U} &= \sum_{i} U n_{i\uparrow} n_{i\downarrow}, \ \mathcal{H}_{V} &= rac{1}{2} \sum_{i
eq j} V_{ij} n_{i} n_{j}. \end{aligned}$$

Here *i*, *j* are the unit cell indices of the maximally localized Wannier function [31,32] and $c_{i\sigma}^{\dagger}(c_{i\sigma})$ is the corresponding creation (annihilation) operator of an electron of spin σ at the site *i*. The number operator is given by $n_i = \sum_{\sigma} n_{i\sigma}$ and $n_{i\sigma} = c_{i\sigma}^{\dagger} c_{i\sigma}$. Hopping amplitudes t_{ij} in the kinetic energy \mathcal{H}_{kin} depend on the relative coordinate vector $\mathbf{r}_i - \mathbf{r}_j$ by assuming the translational invariance of the crystal structure. The direct effective Coulomb interaction given by \mathcal{H}_{II} is scaled by the on-site interaction U, and off-site interaction \mathcal{H}_V is the sum over the combination of the site *i* and j, which is proportional to V_{ij} . Leading values for all of the four materials are listed in Table I. For longer-ranged part of t_{ij} and V_{ij} , see Sec. S1 A in Supplemental Material (SM) [33]. Note that the Hamiltonian parameters for Hg1201 in Ref. [12], which we employ, are improved from Ref. [34]. It results in slightly different physical quantities on the quantitative level in the present solution in comparison to Ref. [10].

From four different materials, we learn that the realistic range of available *ab initio* Hamiltonian parameters is estimated to be $6 \leq U/|t_1| \leq 10$, $0.2 \leq |t_2/t_1| \leq 0.3$, $1.2 \leq |V_1/t_1| \leq 2.0$, $0.5 \leq |V_2/t_1| \leq 1.2$, etc. (see also Table IV). In this paper, we investigate whether the

TABLE I. *Ab initio* single-band effective Hamiltonian for CaCuO₂, Hg1201, Bi2212, and Bi2201 taken from Ref. [12]. *U* is the on-site interaction. The *n*th neighbor hopping amplitude and Coulomb interaction are denoted as t_n and V_n , respectively. Interlayer hoppings and interactions are neglected here. All values are given in eV.

	t_1	t_2	<i>t</i> ₃	t_4	<i>t</i> ₅
CaCuO ₂	-0.521	0.132	-0.047	0.008	0.000
Hg1201	-0.544	0.111	-0.043	0.010	0.000
Bi2212	-0.452	0.135	-0.053	-0.001	0.007
Bi2201	-0.527	0.140	-0.042	0.009	-0.007
	U	V_1	V_2	V_3	V_4
CaCuO ₂	4.221	0.969	0.539	0.380	0.316
Hg1201	3.999	1.002	0.596	0.448	0.389
Bi2212	4.226	0.915	0.518	0.366	0.312
Bi2201	4.393	1.030	0.602	0.450	0.395

diversity of the SC properties can be understood within this range of parameters.

We note that the effective Hamiltonian parameters in Eq. (1) are restricted to a single CuO₂ layer. However, in the case of multilayer cuprates $CaCuO_2$ and Bi2212, the distance between CuO₂ layers is comparable to the cell parameter along the x direction, so that interlayer coupling parameters (given in Table VI in Appendix H for Bi2212) also exist in the effective Hamiltonian [12], and its amplitude $V_n^l \lesssim 0.6$ eV is comparable to that of the intralayer off-site interaction $V_n \lesssim 0.9$ eV. This interlayer coupling is ignored in Eq. (1), but potentially plays a role in the SC properties. This role is actually examined in Sec. III C 1 in the case of Bi2212, which ensures that the SC order parameter F_{SC}^{∞} (defined later in Sec. II C) and physical quantities are essentially not affected by the interlayer coupling within the present case of CaCuO₂ and Bi2212 as we discuss in Sec. III C 1. Thus, we restrict to Eq. (1) even in the case of CaCuO₂ and Bi2212. We employ this "single-layer approximation" for all the four materials throughout this paper unless otherwise noted.

B. Numerical methods

We solve the Hamiltonian in Eq. (1) by applying the many-variable variational Monte Carlo (mVMC) method [18,19] with a trial wave function of the form $|\psi\rangle = \mathcal{P}^{G}\mathcal{P}^{J}\mathcal{P}^{dh}|\phi^{pair}\rangle$. Here we consider the Gutzwiller factor $\mathcal{P}^{G} = \exp(-g\sum_{i}n_{i\uparrow}n_{i\downarrow})$ [35], the Jastrow correlation factor $P^{J} = \exp(\sum_{i < j} \alpha_{ij}n_{i}n_{j})$ [36,37], the doublonholon correlation factor $\mathcal{P}^{dh} = \exp[-\sum_{m=0}^{4} \sum_{\ell=1,2} \alpha_{m}^{(\ell)} \times \sum_{i} \xi_{i(m)}^{(\ell)}]$ [38], and a generalized pairing wave function of the form $|\phi\rangle^{pair} = (\sum_{i\sigma,j\sigma'} f_{i\sigma,j\sigma'} c_{i\sigma}^{\dagger} c_{j\sigma'}^{\dagger})^{N_{c}/2} |0\rangle$. The variational parameters are g, α_{ij} , $\alpha_{m}^{(\ell)}$, and $f_{i\sigma,j\sigma'}$.

We also supplement the mVMC technique with the restricted Boltzmann machine (RBM) [20] and the first-order

Lanczos step to improve the wave function and also to take the zero limit of the variance extrapolation to improve the estimate by following the variance extrapolation method [39–41] and by using the simple mVMC, mVMC + Lanczos, and mVMC + RBM results (see Appendixes A and B and Ref. [42] for the detailed procedure).

Competing states with spin or charge order or strong fluctuations can be studied by imposing the mean-field order at the initial trial wave function [19]. The correlated metallic state without any symmetry breaking can also be studied by using the ground-state wave function of the noninteracting system as an initial state. These initial states are then relaxed to lower the energy by optimizing the variational parameters. If the competing states exist, the optimization leads to multiple locally stable solutions. The true ground state is determined by comparing the total energy after taking the variance extrapolation described in Appendix B and if possible after careful size extrapolation to see the thermodynamic limit.

The computational details are the following. For all numerical solutions of finite-size lattices subsequently presented in this paper, we assumed the antiperiodicperiodic boundary condition on a $N = L \times L$ square lattice of length L, where N is the number of sites on the singlelayer system by ignoring the interlayer coupling except for Bi2212. For Bi2212, we examine the two-layer system to examine the effect of interlayer coupling, because a unit cell of Bi2212 contains two layers and the interlayer coupling could play roles in the SC. Within a layer, hoppings and interactions were taken into account up to t_9 and V_9 , i.e., up to the 2D distance $\mathbf{R} = (3,3)$ in the unit of the Cu-Cu distance, while contributions smaller than 0.001 eV were ignored. Unless explicitly mentioned this is applied throughout the whole paper. We concentrate solely on hole doped cases, where the hole doping is given via $\delta = 1 - N_e/N$ and N_e is the number of electrons in the system.

C. Physical quantities

The physical quantities discussed in this paper are defined as follows: The total energy per site $E/N = \langle \mathcal{H} \rangle / N$ is calculated after the variance extrapolation as it is summarized in Appendix B. To see whether the state has spin or charge order, we compute the spin structure factor,

$$S_s(\boldsymbol{q}) = \frac{1}{N} \sum_{i,j} \langle \boldsymbol{S}_i \cdot \boldsymbol{S}_j \rangle \mathrm{e}^{\mathrm{i}\boldsymbol{q}(\boldsymbol{r}_i - \boldsymbol{r}_j)}, \qquad (2)$$

and the charge structure factor,

$$S_c(\boldsymbol{q}) = \frac{1}{N} \sum_{i,j} \langle n_i n_j \rangle \mathrm{e}^{\mathrm{i}\boldsymbol{q}(\boldsymbol{r}_i - \boldsymbol{r}_j)}, \qquad (3)$$

where $S_i = (S_i^x, S_i^y, S_i^z) = \sum_{\sigma,\sigma'} c_{i\sigma}^{\dagger} \sigma_{\sigma\sigma'} c_{i\sigma'}/2$ is the spin-1/2 operator and $\sigma_{\sigma\sigma'}$ is the Pauli matrix $[=(\sigma_{\sigma\sigma'}^x, \sigma_{\sigma\sigma'}^y, \sigma_{\sigma\sigma'}^z)]$.

The long-range order is determined whether $S_s(q)/N$ or $S_c(q)/N$ remains after taking the limit $N \to \infty$. The SC long-range order is measured by the *d*-wave SC correlation function:

$$P_d(\mathbf{r}) = \frac{1}{2N} \sum_{\mathbf{r}_i} \langle \Delta_d^{\dagger}(\mathbf{r}_i) \Delta_d(\mathbf{r}_i + \mathbf{r}) + \text{H.c.} \rangle.$$
(4)

Here $\Delta_d^{\dagger}(\boldsymbol{r}_i)$ describes the order parameter of the form

$$\Delta_d(\mathbf{r}_i) = \frac{1}{\sqrt{2}} \sum_{\mathbf{r}} f_d(\mathbf{r}) (c_{\mathbf{r}_i \uparrow} c_{\mathbf{r}_i + \mathbf{r} \downarrow} - c_{\mathbf{r}_i \downarrow} c_{\mathbf{r}_i + \mathbf{r} \uparrow}), \quad (5)$$

where the $d_{x^2-y^2}$ -wave symmetry is included via the form factor:

$$f_d(\mathbf{r}) = \delta_{r_y,0}(\delta_{r_x,1} + \delta_{r_x,-1}) - \delta_{r_x,0}(\delta_{r_y,1} + \delta_{r_y,-1}).$$
(6)

Then, we deduce $P_d(\mathbf{r})$ over the long-range part as

$$\bar{P}_d(L) = \frac{1}{M} \sum_{\sqrt{2L/4} < |\boldsymbol{r}|} P_d(\boldsymbol{r}), \tag{7}$$

where $\mathbf{r} = (r_x, r_y)$ includes all sites within the square $(-L/2, L/2)^2$ and *M* is the number of lattice points satisfying $\sqrt{2}L/4 < |\mathbf{r}| < \sqrt{2}L/2$. We define the SC order parameter in the thermodynamic limit $F_{\text{SC}}^{\text{sc}}$ by

$$\begin{split} F^{\infty}_{\rm SC} &= \lim_{L \to \infty} F_{\rm SC}(L) = \sqrt{\bar{P}^{\infty}_d}, \\ F_{\rm SC}(L) &\equiv \sqrt{\bar{P}_d(L)}, \\ \bar{P}^{\infty}_d &= \lim_{L \to \infty} \bar{P}_d(L). \end{split} \tag{8}$$

III. AB INITIO RESULTS

In this section we present our calculated *ab initio* results on the four families of compounds. We first analyze the results for doped CaCuO₂ in detail and then compare it with the result of doped Hg1201. Doped Bi2201 and Bi2212 suffer from the experimental uncertainty of the apical oxygen position due to the supermodulation of the crystal structure. Since it generates a variance of the Hamiltonian parameters if one assumes the translational symmetry of the Hamiltonian parameters, we show the properties by indicating this range. All of the four materials show dominance of the *d*-wave SC in the doping concentration dependence and the calculated results reproduce the experimental materials dependence of the strength of SC, which makes it possible to extract the universal properties and systematic trends as well.



FIG. 1. SC correlation function $P_d(r)$ for CaCuO₂ at different hole dopings. (a) $\delta = 0.028$, (b) $\delta = 0.045$, (c) $\delta = 0.087$, (d) $\delta = 0.101$, (e) $\delta = 0.125$ (here L = 28 instead of L = 30), (f) $\delta = 0.167$, (g) $\delta = 0.208$, (h) $\delta = 0.247$. In each case, we show $P_d(r)$ for the square lattice size L = 24, 30, 36. For the same distance r we employ the largest value of correlation. We perform the same procedure in later figures. Inset of each panel: size extrapolation of $\bar{P}_d(L)$ to the thermodynamic limit \bar{P}_d^{∞} , whose numerical value is listed in Table II. The gray line shows the linear approximation. Statistical errors originating from the Monte Carlo sampling are smaller than the symbol size.

A. Doped CaCuO₂

Here, we present the results of our calculation for the doped $CaCuO_2$ in the following order.

- (1) δ dependence of SC properties; in particular, \bar{P}_d^{∞} and the *d*-wave SC order parameter F_{SC}^{∞} . (See the definitions in Sec. II C.)
- (2) Competition between the SC state and other competing states including stripe states.

1. Properties of superconducting phase

First, we discuss the r dependence of the pairing correlation $P_d(r)$ for $L \times L$ lattice and its long-ranged part $\bar{P}_d(L)$: Figure 1 shows $P_d(r)$ and $\bar{P}_d(L)$ for several choices of square lattices with the linear size L from 24 to 36 and hole doping δ from 0.028 (2.8%) to 0.247 (24.7%). For each value of δ , we observe that \bar{P}_d does not significantly depend on L, suggesting the existence of a strong SC longrange order in the thermodynamic limit in this ground-state candidate. This is indeed confirmed by a size extrapolation, i.e., plot of $\bar{P}_d(L)$ as a function of 1/L to estimate \bar{P}_d^{∞} in the limit $L \to \infty$ via linear regression, as shown in the insets of Figs. 1(a)–1(h). The linear 1/L scaling was employed in Ref. [10] and is expected to work because of Dirac-type linear dispersion for the quasiparticle excitation of the d-wave superconductor at the nodal points. Here, we have shown the data calculated from the transfer and interaction parameters in the Hamiltonian fixed at 10% hole doping for simplicity as we addressed above. However, we can test its robustness by taking δ dependent transfer and interaction parameters. The result is shown in Appendix C and the difference is small.

After taking the size extrapolation to the thermodynamic limit, we show the δ dependence of the order parameter F_{SC}^{∞} calculated from Eq. (8) and $\bar{P}_d^{\infty} = \lim_{L \to \infty} \bar{P}_d(L)$ in Fig. 2 and the numerical values in Table II. This shows a rapid increase of F_{SC}^{∞} from 0 at $\delta = 0$ up to $\delta \sim 0.05$ as a function of δ followed by a plateau around $0.05 \le \delta \le 0.1$ and



FIG. 2. The SC order parameter $F_{\rm SC}$ as a function of δ for doped CaCuO₂. The gray filled circles show the values of $F_{\rm SC}(L)$ at L = 24 square lattice calculated from $\bar{P}_d(L)$ shown in Fig. 1 by using $F_{\rm SC}(L) = \sqrt{\bar{P}_d(L)}$. The red squares are the sizeextrapolated values $F_{\rm SC}^{\infty}$ calculated from \bar{P}_d^{∞} . Inset shows the corresponding $\bar{P}_d(L)$ at L = 24 and \bar{P}_d^{∞} .

TABLE II. Size-extrapolated SC correlation function \bar{P}_d^{∞} and order parameter $F_{\rm SC}$ for doped CaCuO₂ for several choices of doping δ . The fitting error is of the order of $\sim 10^{-4}$ for \bar{P}_d^{∞} and $\sim 10^{-3}$ for $F_{\rm SC}^{\infty}$.

δ	$ar{P}^\infty_d$	F_{SC}^{∞}
0.028	0.0139(6)	0.118(2)
0.045	0.0188(2)	0.137(1)
0.087	0.0177(8)	0.133(3)
0.101	0.0174(16)	0.132(6)
0.125	0.0136(9)	0.116(4)
0.167	0.0078(2)	0.088(1)
0.208	0.0039(2)	0.062(1)
0.247	0.0016(2)	0.040(3)

monotonic decrease with further increasing δ above around 0.1 in the thermodynamic limit.

The dome structure ubiquitously observed for T_c in the cuprates is qualitatively similar to the δ dependence in F_{SC}^{∞} , but the peak for F_{SC}^{∞} is located at somewhat lower $\delta \sim 0.05$ than the case of experimental T_c , where the optimum δ is observed to be $\delta \sim 0.12$ [43]. We will discuss this discrepancy in Sec. V. However, the monotonic decrease of F_{SC}^{∞} with increasing δ for $\delta \geq 0.1$ is consistent with the universal trend of δ dependence of the SC gap identified from the angle-resolved photoemission spectra (ARPES) and the scanning tunneling microscope (STM) of the cuprates in general [44,45], though the SC gap in the experimental estimate contains an ambiguity associated with the contribution from the pseudogap.

In the mean-field picture, the SC gap is the product of the order parameter F_{SC}^{∞} and the effective attractive interaction. If we consider the experimentally observed maximum gap ~50 meV [44,45] and $F_{SC}^{\infty} \sim 0.13$, the characteristic scale of attractive interaction is as large as ~0.4 eV. This imposes a constraint on theories for the SC mechanism.

The sharp increase of F_{SC}^{∞} between $\delta = 0$ and 0.05 and the subsequent dome structure are similar to the earlier study by a VMC method and a cluster dynamical meanfield study [9,46] for the Hubbard model, where rapidly increasing F_{SC} from 0 at $\delta = 0$ already reaches $F_{SC} \sim 0.1$ at $\delta = 0.03$ in the present notation. In case of the Hubbard model, however, it was argued that the ground state is actually not SC but stripe-ordered states [4–6].

2. Competition of SC, stripe, and AFM states

Now, we analyze the competition between the SC and other states. The energies of the SC state and other states at L = 24 are given in Fig. 3. We see that the SC state has the lowest energy in the region from $\delta = 0.05$ to $\delta = 0.25$, indicating that the SC phase is dominant in the ground state of doped CaCuO₂. The SC ground state, however, is severely competing with the C4S8, and C3S3-like stripe states, and AFM-type state within the energy difference of



FIG. 3. (a) Variance extrapolated energies of doped CaCuO₂ for various ground-state candidates, SC, charge or spin stripe states C3S3 and C4S8, and AFM state as a function of hole doping δ on a L = 24 square lattice. All energies are subtracted by the function $\mathcal{F}(\delta) = -12.76470 \times \delta + 6.44626$ for better visibility. (b) Energy difference ΔE for the variance extrapolated data from (a).

5–10 meV. Here, CmSn denotes the charge- and spinordered stripe state with the periodicity of *m* lattice spacing for the charge modulation and the period *n* for the spin order. Spin and charge real-space patterns and structure factors are explicitly illustrated in Secs. S2–S4 of SM [33] for the C4S8, C3S3-like, and AFM-like excited states. In the region studied here, we find only C4S8 and C3S3 as candidates of the competing stripe order, which has similarity to an earlier study of the simple Hubbard model with the next nearest neighbor transfer in the range $0.2 < |t_2/t_1| < 0.3$ and $0.05 < \delta < 0.25$ [6] and an *ab initio* study [10]. Note that C4S8 at $\delta = 1/8$ and C3S3 at $\delta = 1/6$ have a particular commensurability energy gain. In Fig. 3(b), a dip exists in the AFM states at $\delta = 0.167$. At the moment, the origin of the dip is not clear.

Figures 14 and 15 in Appendix D show size dependence of spin and charge structure factors. Although the demanding computation cost does not allow larger system calculation, the available size dependence supports that the AFM states up to $\delta = 0.16$ and the C4S8 stripe state at finite doping around $\delta \sim 0.12$ do have the long-range order as one can see in Figs. 14(a) and 15, respectively. However, although the initial trial states are ordered mean-field states, long-ranged order seems absent or is very weak after the optimization in VMC calculations for the C3S3 stripe states and seems replaced by well-developed short-ranged correlations at $\delta \neq 0$. This is the reason why we add "-like" for On the comparison between SC and stripe or AFM states, similar severe competitions were reported in Hubbard models. However, the present *ab initio* results have a crucial difference, where the charge uniform SC phase is the dominant ground state, while the ground states of Hubbard models irrespective of the presence or absence of t_2 mostly have the stripe long-range order. The reason for this difference originates from the presence of realistic off-site interaction in the *ab initio* case as was already pointed out in Ref. [10]. We discuss this point in comparison to the Hubbard models in Appendix E.

B. Doped Hg1201 compared with CaCuO₂

Now, we present our results in the case of Hg1201, and compare it to CaCuO₂. In this comparison, we find (i) the positive correlation between F_{SC}^{∞} and $U/|t_1|$ and (ii) the relation $T_c^{\text{opt}} \sim 0.16|t_1|F_{SC}^{\infty}$ from the comparison with the experimental T_c .

The pairing correlation for Hg1201 Hamiltonian and the size extrapolation are shown in Fig. 4 for $\delta = 0.146$, indicating the existence of the SC long-range order. The size of the order parameter is $F_{SC}^{\infty} \sim 0.09$ as compared with ~ 0.116 for doped CaCuO₂ at $\delta \sim 0.12$, respectively, which are both close to each optimal concentration. The difference in F_{SC}^{∞} between Ca and Hg compounds can be compared with the difference in $U/|t_1| = 8.10$ for CaCuO₂ and 7.35 for Hg1201. As we discuss later, F_{SC}^{∞} rapidly increases with $U/|t_1|$ if we monitor the effect of $U/|t_1|$ beyond the



FIG. 4. SC correlation function $P_d(r)$ for Hg1201 at $\delta = 0.146$, which is close to the experimental optimal doping, at the square lattice size L = 24, 28, 30, 32, and 36. Error bars are smaller than the symbol size. Inset: size extrapolation of P_d to the thermodynamic limit $L \rightarrow \infty$. Because of relatively scattered data we employ the average of the two biggest lattice sizes (L = 32, 36) for the size extrapolation $L \rightarrow \infty$. In fact, the value at the largest sizes is consistent with the systematic δ dependence observed near $\delta = 0.146$ after the size extrapolation (not shown). The error bar at 1/L = 0 is estimated as the biggest difference to F_{SC}^{∞} from the given data.

ab initio value around $U/|t_1| = 7-8$. Therefore, F_{SC}^{∞} amplifies the increase in $U/|t_1|$ while effects of other parameters are minor: Namely, F_{SC}^{∞} should have a functional form $F_{SC}^{\infty}(U/t_1, V_i/t_1, t_i/t_1)$ with $1 \le i \le 9$ in general, but V_i/t_1 and t_i/t_1 dependencies are weaker as compared to dependence on $U/|t_1|$ in the realistic parameter range. See Appendix F for the example of $|t_2/t_1|$ dependence. See also Fig. 9 for the V_1/t_1 dependence. In both cases, the change in F_{SC} at the optimum doping is at most 10% in the realistic parameter range. In fact, in the comparison of Bi2212, Bi2201, CaCuO₂, and Hg1201, $|t_2/t_1|$ is ~0.30, 0.27, 0.25, and 0.20, respectively, which does not have systematic correlation with T_c^{opt} . Appendix F shows tiny anticorrelation of $|t_2/t_1|$ and F_{SC} , but is practically negligible at the optimal doping. After careful examination of other parameters as well, the difference of $F_{\rm SC}^{\infty}$ in these four compounds studied is concluded to be ascribed to the difference in $U/|t_1|$.

The materials dependent F_{SC}^{∞} may also be compared with the difference in $T_c^{opt} \sim 110$ and 90 K for CaCuO₂ and Hg1201, respectively, because T_c may be proportional to the order parameter F_{SC}^{∞} . Since T_c has the dimension of energy and should also be scaled by the overall characteristic energy scale t_1 , T_c may be proportional to $|t_1|F_{SC}^{\infty}$. In fact, the ratio of $T_c^{opt}/|t_1|F_{SC}^{\infty}$ as a nondimensional quantity is ~0.16(1) at the optimal doping $\delta \sim 0.12$ for CaCuO₂ and ~0.16(2) at the optimal point $\delta \sim 0.15$ for Hg1201, as we show in Table III, supporting the hypothesis that T_c^{opt} is universally given from the relation

$$T_c^{\text{opt}} \sim 0.16 |t_1| F_{\text{SC}}^{\infty} \tag{9}$$

at the optimal doping.

In the Uemura plot [47], it was observed from the muon spin resonance (μ SR) measurement that T_c^{opt} is proportional to the ratio between the superfluid density n_s , here interpreted as $F_{\rm SC}^{\infty}/\sqrt{2}$, and the effective mass m^* . Since the mass enhancement from the bare band mass m_0 , namely, m^*/m_0 at the optimal hole density may be similar in the cuprates, T_c^{opt} is indeed expected to be roughly proportional to $|t_1| F_{\rm SC}^{\infty}$ according to the Uemura plot, because the inverse band mass is essentially determined by the dominant transfer t_1 . In addition, n_s estimated from the relaxation rate $\sigma \sim 2 \ \mu s^{-1}$ from the μSR measurement for the cuprates with $T_c \sim 80-100$ K corresponds to $n_s m_0/m^* \sim 4 \times 10^{21} \text{ cm}^{-3}$. If we cut out the volume including one Cu atom with the c axis length ~ 6 Å as in Ref. [47] irrespective of the unit cell volume to compare with F_{SC}^{∞} defined as the value per Cu atom, this corresponds to $F_{\rm SC}^{\infty} \sim 0.10$ by considering the definition $F_{\rm SC}^{\infty} = \sqrt{2n_s}$ and $m^*/m_0 \sim 5$ assumed in Ref. [47]. Then it is also quantitatively consistent with the present result of $F_{\rm SC}^{\infty} \sim$ 0.10 at the optimum doping. These indicate that our results

TABLE III. Comparison of long-ranged SC correlation \bar{P}_d^{∞} and the order parameter F_{SC}^{∞} with $U/|t_1|$ as well as comparison between $|t_1|F_{SC}^{\infty}$ and T_c^{opt} for doped CaCuO₂ at $\delta = 0.125$, and Hg1201 at $\delta = 0.146$ (these values of δ are chosen in accordance with those closest to experimental optimal values, $\delta \sim 0.12$ and 0.15, respectively, and to allow the numerical size extrapolation to be easier). Note that the ratio $T_c^{\text{opt}}/(|t_1|F_{SC}^{\infty})$ is given as a nondimensional quantity by using 1 eV = 1.16×10^4 K. The parentheses in the last digit indicate the error bar.

	$U/ t_1 $	\bar{P}_d^∞	F_{SC}^{∞}	$ t_1 F_{\mathrm{SC}}^{\infty}$ (eV)	T_c^{opt} (K)	$T_c^{\text{opt}}/(t_1 F_{\text{SC}}^\infty)$
CaCuO ₂	8.10	0.0136(9)	0.116(4)	0.060(2)	110	0.16(1)
Hg1201	7.35	0.008(2)	0.09(1)	0.048(5)	90	0.16(2)

indeed capture the realistic situation more or less quantitatively. We will show in Sec. V that F_{SC}^{∞} also agrees with the estimate from the angle resolved photoemission spectra in case of Bi2212 and Bi2201.

C. Doped Bi2201 and Bi2212

In this subsection, we discuss ab initio results for Bi2212 and Bi2201 and compare them to each other. Unfortunately, in these two compounds, an uncertainty exists in the experimental crystal parameters that causes the uncertainty in the effective Hamiltonian parameters as well. In particular, the distance d_{Oap}^z between an apical oxygen and the nearest Cu atom is not fully precisely determined and the available experimental data have considerable variations [48–53]. This uncertainty is also related to the structural distortion and long-period modulation of the CuO₂ plane arising from the effect from the BiO layer [54,55] as we discuss in Sec. V. Recent ab initio studies have clarified that this uncertainty leads to a possible variety of effective Hamiltonian parameters, especially owing to the variation of the apical oxygen position [12].

In principle, the structural optimization in *ab initio* calculations is desired to predict the stable atomic position. However, such an optimization in strongly correlated electron systems is at the moment not necessarily accurate enough and we leave this task for future studies. Instead, in this paper, we admit a range of Hamiltonian parameters and discuss the consequence.

As analyzed in Ref. [12], the apical oxygen position sensitively affects the effective Hamiltonian parameters, primarily the value of U. For Bi2212, the value $U \sim 4.2$ eV in Table I is intermediate and the uncertainty range is between 4.0 and 4.7 eV for U by considering that d_{Oap}^z may range from 2.25 to 2.45 Å. On the other hand, the value $U \sim 4.4$ eV for Bi2201 in Table I is the upper bound and the uncertainty ranges from $U \sim 4.4$ to 3.5 eV by considering that d_{Oap}^z may range from $d_{Oap}^z = 2.6$ to 2.45 Å. We first present in Secs. III C 1 and III C 2 the results obtained from the parameters shown in Table I, namely, U = 4.2 eV for Bi2212 and U = 4.4 eV for Bi2201 and then discuss in Sec. III C 3 the possible range of SC properties originating from this uncertainty later.

1. Bi2212

We begin with the results for Bi2212. Figure 5 shows $P_d(r)$ and $\bar{P}_d(L)$ at $\delta = 0.167$ for L from 16 to 36 by switching off the interlayer transfers and interactions. Namely, we first show the results obtained by solving the single-layer Hamiltonian despite the actual two-layer unit cell of Bi2212. The case of δ dependent Hamiltonian is also shown in Appendix C, where the difference is small. Similarly to CaCuO₂, we identify a SC ground state with large \bar{P}_d and it does not change significantly by increasing L, where the size extrapolation gives $P_d^{\infty} \sim 0.0151$, as shown in the inset of Fig. 5, which corresponds to a SC order parameter $F_{\rm SC}^{\infty} \sim 0.12$. This relatively strong value of $F_{\rm SC}^{\infty}$ is understandable, because the $U/|t_1|$ ratio has a value of $U/|t_1| \sim 9.4$, which is the strongest among all four considered compounds. Again the enhanced F_{SC}^{∞} originates from the larger $U/|t_1|$ in accordance with the observation in the comparison of Hg1201 and CaCuO₂ in Sec. III B. This large F_{SC}^{∞} is also consistent with the high T_c (up to ~100 K) [30]. We will discuss more intricate aspect in Sec. V.

The competition with other phases is seen in δ dependence of the total energy shown in Fig. 19 in Appendix I. Similarly to CaCuO₂, the SC state is the ground state in most of the doping concentration, while it is severely competing with spin- or charge-ordered states.



FIG. 5. SC correlation function $P_d(r)$ for Bi2212 at $\delta = 0.167$ for the square lattice size L = 16, 24, 30, 36. Inset: size extrapolation of \bar{P}_d to the thermodynamic limit. The gray line shows the linear extrapolation to $1/L \rightarrow 0$ by using the values for L = 16, 24, 30, 36. Error bars are smaller than the symbol size.

Now, we extend the calculation by switching on the interlayer terms and solve the two-layer Hamiltonian obtained in Ref. [12] to examine the effects of interlayer coupling. For the calculations we take two identical layers (in terms of intralayer parameters for t_{ii} , V_i , and U, where iand *j* are intralayer combination), coupled by the interlayer terms listed in Table VI (Appendix H). The interlayer contributions are restricted to the leading interlayer hopping term of size $t_0^l = -0.098$ eV and nearest and next nearest neighbor interlayer interaction of size $V_0^l =$ 0.643 eV and $V_1^l = 0.463$ eV. See Ref. [12] for more details. A comparison between the single- and two-layer cases of $P_d(r)$ for L = 16 and $\delta = 0.167$ is shown in Fig. 6. We see that the long-range average of $P_d(\mathbf{r})$ in the twolayer case is close to that of $P_d(\mathbf{r})$ in the single layer case, which demonstrates that $P_d(\mathbf{r})$ is not significantly affected by the interlayer Coulomb interaction and hopping parameters. Indeed, for the single layer we found a long-range average of the SC correlation function of $\bar{P}_d^{\text{single}} = 0.0225$, while for the two-layer compound the average is $\bar{P}_d^{\text{two}} =$ 0.0213. The corresponding values of the SC order parameter are $F_{SC}^{\text{single},\infty} = 0.150$ and $F_{SC}^{\text{two},\infty} = 0.146$, which differ by only ~2.8%. The essentially same behavior between the single- and two-layer cases may not depend on the system size in accordance with the result of a two-layer Hubbard model at the optimum doping [56]. This similarity may be attributed to (i) the relatively small leading interlayer hopping parameter of $t_0^l = -0.098 \text{ eV}$ and also (ii) the robustness of the SC solution against the interlayer Coulomb interaction parameter, because the pairing occurs essentially only within a layer.

2. Bi2201

In the case of Bi2201, we again solve the single-layer Hamiltonian. The competition with other phases is seen in δ dependence of the total energy shown in Fig. 20 in



FIG. 6. SC correlation function $P_d(r)$ for Bi2212 at $\delta = 0.167$ in the cases of the single- and the explicit two-layer calculations. In the single-layer case a 16×16 square lattice was considered, which corresponds to a $16 \times 16 \times 2$ lattice in the two-layer case. Error bars are smaller than the symbol size.



FIG. 7. SC correlation function $P_d(r)$ for Bi2201 at $\delta = 0.167$ at the square lattice sizes L = 24, 30, 36. Inset: size extrapolation of \bar{P}_d to the thermodynamic limit. The gray line shows the linear extrapolation to $1/L \rightarrow 0$ by using the values for L = 24, 30, 36. Error bars are smaller than the symbol size.

Appendix I. Figure 7 shows $P_d(r)$ and its extrapolation to the thermodynamic limit, which suggests the stable longranged SC order. Again, the obtained value $F_{SC}^{\infty} \sim 0.10$ is consistent with the rule that larger $U/|t_1|$ leads to larger F_{SC}^{∞} because $U/|t_1|$ is the second largest among the four materials in the estimate shown in Table I. The smaller $U/|t_1|$ relative to Bi2212 leads to weaker SC. However, on a more quantitative aspect, we need to be careful about the uncertainty of the Hamiltonian parameter. We will discuss this issue below.

3. Effect of structural uncertainty on possible variation of SC properties

Since we have the uncertainty of the Hamiltonian parameters particularly for the interaction as we discussed above, we here monitor the effects of modifying the effective interactions for Bi2212 and Bi2201, which well represent the effect of variant apical oxygen position as shown in Appendix C of Ref. [12]. Namely, Table I with preserved transfer parameters fixed at each *ab initio* value, together with interaction scaling, represents most of the effect of the apical oxygen shift and we scale the Hamiltonian Eq. (1) such that

$$\mathcal{H}(\alpha,\xi) = H_{\rm kin} + \alpha H_U + \xi H_V, \tag{10}$$

where $\alpha = \xi = 1$ represents the *ab initio* case given in Table I, α scales the on-site Coulomb interaction term H_U , and ξ scales the remaining off-site interactions V_i . Since the apical oxygen shift alters the interaction parameters in the way $\alpha \sim \xi$ [12], we examine the dependence on $\alpha = \xi$ below.

Figures 8(a) and 8(b) show the size scaling and $\alpha = \xi$ dependence of P_d^{∞} for Bi2212 and Bi2201 at $\delta = 0.167$, which is close to the experimental optimum doping. Further details are given in Sec. S5 of SM [33]. Figure 8(c) shows the corresponding $\alpha = \xi$ dependence of the SC order



FIG. 8. Variation of SC order arising from uncertainty of the apical oxygen position via $\alpha = \xi$ scaling for Bi2212 and Bi2212. The realistic range is $0.95 \le \alpha = \xi \le 1.1$ for Bi2212 and $0.8 \le \alpha = \xi \le 1.0$ for Bi2201. Size extrapolation of \bar{P}_d for (a) Bi2212 at $\delta = 0.167$ and $\alpha \in \{0.8, 0.9, 1.0, 1.1, 1.2\}$, (b) Bi2201 at $\delta = 0.167$ and $\alpha \in \{0.8, 0.9, 1.0\}$. (c) F_{SC}^{∞} as a function of $\alpha = \xi$ for Bi2212 (orange) and Bi2201 (blue).

parameter F_{SC}^{∞} . For Bi2212, by taking the realistic uncertainty range $4.0 \le U \le 4.7 \text{ eV}$ (corresponding to $0.95 \le \alpha = \xi \le 1.1$) obtained from $2.25 \le d_{\text{Oap}}^z \le 2.45$ Å, we find the range of $0.011 \le P_d^\infty \le 0.015$, namely, $0.10 \le F_{\text{SC}}^\infty \le 0.12$. On the other hand, the uncertainty for Bi2201 obtained from $2.45 \le d_{\text{Oap}}^z \le 2.6$ Å leads to $3.5 \le U \le 4.4$ eV (corresponding to $0.8 \le \alpha = \xi \le 1.0$), which results in $0.00017 \le P_d^\infty \le 0.0094$ ($0.013 \le F_{\text{SC}}^\infty \le 0.10$). The lower bound for Bi2201 causes a fatal damage to the SC order.

Table IV summarizes the size-extrapolated \bar{P}_{d}^{∞} and corresponding F_{SC}^{∞} for the two Bi compounds when we use the Hamiltonian parameters listed in Table I and when we admit the uncertainty range of the interaction parameters for Bi2212 and Bi2201. The scaling Eq. (9) proposed for CaCuO₂ and Hg1201 is also valid in the Bi compounds, and the experimental T_{c}^{opt} is within the inferred range. We conclude that its materials dependence is well captured for the four studied materials (see also Fig. 11 in Sec. VI).

We realize that the fragility and diversity of T_c^{opt} experimentally observed in the range $10 < T_c^{\text{opt}} < 40 \text{ K}$ for Bi2201 is accounted for by the range of actual apical oxygen position. This range may be caused by the type of dopant atoms, impurities, and the spatial inhomogeneity caused by the supermodulation, which may depend on samples and the amplitude of the modulation. In fact, it was observed that the d_{Oap}^z periodically varies as much as 6% in accordance with the supermodulation for Bi2212 and a comparable modulation may exist for Bi2201 as well, which can be the origin of the experimental uncertainty [54,55]. The basic origin of this diversity and relatively low T_c among the four families of compounds is attributed to relatively small $U/|t_1|$ in the lower uncertainty range, at which the SC order becomes sensitively damaged by a slight decrease of $U/|t_1|$. We discuss more general aspects of the interaction dependence in Sec. IV. Even when we admit the uncertainty range, the general trend about the weaker SC of Bi2201 than those of Bi2212 is well explained by this ab initio result. It can also safely be addressed that Bi2212 has one of the strongest SC and T_c^{opt} among the four families comparably to $CaCuO_2$.

The effects of apical oxygen position on Hamiltonian parameters are discussed in Sec. V and in Appendix L.

IV. RESULTS BEYOND AB INITIO: INTERACTION DEPENDENCE OF SUPERCONDUCTING ORDER

We now study SC properties beyond the *ab initio* results. *Ab initio* results in the previous section successfully reproduce the experimental trend and have revealed that $U/|t_1|$ is the principally important Hamiltonian parameter to control the SC order parameter. Therefore, it is intriguing to examine the optimum Hamiltonian parameters to maximize the order parameter and hence the optimum T_c beyond the existing materials for the purpose of materials design to seek for higher T_c superconductors. We present $U/|t_1|$ dependence of F_{SC} as well as dependence on off-site interaction when tuning the interaction parameters artificially away from the *ab initio* value while keeping transfer parameters fixed at *ab initio* values. We here take an example of CaCuO₂ Hamiltonian at $\delta = 0.167$ and monitor the effect of α and ξ dependencies defined in Eq. (10).

TABLE IV. Comparison of long-ranged SC correlation \bar{P}_d^{∞} and the order parameter F_{SC}^{∞} with $U/|t_1|$ for Bi2212 and Bi2201 at $\delta = 0.167$, which is chosen in accordance with the experimental optimal values. The range of values represents the uncertainty range arising from the uncertainty of the apical oxygen position. This estimate helps the inference of the correct apical oxygen position (see text).

	U	$U/ t_1 $	$ar{P}^\infty_d$	$F_{\rm SC}^{\infty}$	$0.16 t_1 F_{\rm SC}^{\infty}$ (K)	T_c^{opt} (K)
Bi2212	4.0-4.7	8.8-10.4	0.011-0.015	0.10-0.12	83-101	85-100
Bi2201	3.5-4.4	6.7-8.4	0.00017-0.0094	0.013-0.096	12–93	10-40

We examine three types of scaling to go beyond the *ab initio* Hamiltonian: (i) scale only the on-site Coulomb interaction by α with fixed $\xi = 1.0$ [$\mathcal{H}(\alpha, 1.0)$], (ii) scale the full interaction part equally by using $\alpha = \xi$ [namely, $\mathcal{H}(\alpha, \alpha)$], and (iii) fix $\alpha = 1.2$ and scale the off-site Coulomb interaction uniformly via ξ by employing $\mathcal{H}(1.2, \xi)$, by considering the fact that (ii) shows the maximum SC order at $\alpha = \xi = 1.2$. For the cases (i) and (ii) we chose scaling values α ranging from 0.6 up to 4.0, while for the third case the range from 0.0 to 2.0 was chosen. Since the size dependence is not appreciable, we study L = 24 lattice. The SC order parameter F_{SC} are shown in Fig. 9(a) at $\delta = 0.167$ hole doping.

The results show that the SC order parameter can be enhanced with the amount of around 30% from the *ab initio* value when the interaction parameter is tuned to $\alpha \sim 1.2$ for (i) and around 20% at $\alpha = \xi \sim 1.2$ for (ii), which may allow T_c^{opt} as much as ~130–140 K, when compared to the *ab initio* results for CaCuO₂.

In the tuning (iii), we find that the order parameter further increases up to $F_{SC}^{\infty} \sim 0.22$ by decreasing ξ , which is twice as large as the *ab initio* case. However, we keep in mind that on-site and off-site interactions cannot



FIG. 9. (a) $F_{\rm SC}$ over scaling α or ξ (effective scaling of $U/|t_1|$ and V_i/U) of the SC state on the L = 24 square lattice and $\delta = 0.167$ hole doping. The inset is an enlarged plot around the peak $0.9 < \alpha, \xi < 1.5$. For further details, see the main text. (b) Variance extrapolated energy difference ΔE between the SC and C4S8 as a function of ξ in the case $\mathcal{H}(\alpha = 1.2, \xi)$.

independently be controlled in the usual experimental conditions. The present result offers a guide to enhance the SC in designing artificial structure and metamaterials including surface and interface, where quicker screening of the off-site interaction is desirable by keeping the on-site interaction at the optimal value (in this case $\alpha = 1.2$).

Another limitation to be considered is the competition with the stripe and AFM order. As far as we restrict the onsite interaction within the *ab initio* range, the SC energy is always lower than that of the stripe state, as one sees in Fig. 9(b). Here, we show the competition with the C4S8 because it is established that the most severe competitor is the C4S8 state. However, the decreasing of energy difference, such as at ($\alpha = 1.2$, $\xi = 0$), where the difference is ≤ 2 meV, may result in the thermal destruction of the SC order. For $\xi \geq 0.2$ the SC is still a stable ground state, while at small ξ , F_{SC} becomes nearly twice of the *ab initio* value for the doped CaCuO₂.

The order parameter decreases when $U/|t_1|$ is too large beyond the realistic range of the cuprates studied in this paper, as we see in Fig. 9(a). This reduction was already pointed out on the level of the Hubbard model [9]. The reduction at large $U/|t_1|$ is studied in Appendix J, which shows nontrivial power-law dependence of F_{SC} on $U/|t_1|$. The reduction itself may be easily understood on a qualitative level from the suppression of charge fluctuation with increasing $U/|t_1|$, which also suppresses the quantum entanglement caused by the suppression of both spin singlet fluctuation and dynamical exciton generation, as was reported in the literature [9,57]. It was pointed out that the enhanced quantum entanglement can be achieved by the fractionalization of electrons [57,58], which may be maximized at the optimum $U/|t_1|$.

V. DISCUSSION

By assuming the same ratio $T_c/(|t_1|F_{SC}^{\infty}) \sim 0.16$ with CaCuO₂ and Hg1201, we can infer the range of T_c arising from the uncertainty of the apical oxygen position and resultant uncertainty of $U/|t_1|$ listed in Table IV. The range of inferred T_c listed in Table IV for Bi2212 is consistent with the experimentally observed range of T_c^{opt} within the error bars as seen in Fig. 11. This suggests that the sample dependence of T_c^{opt} may be accounted for by the sample dependence of the apical oxygen position.

We note that the order parameter $F_{SC}^{\infty} \sim 0.10$ obtained here for Bi2212 and Bi2201 also shows consistency with the result obtained by using the machine learning of the angle-resolved photoemission spectroscopy data for Bi2212 and Bi2201, respectively, at the optimum doping [59], which gave $\langle c_{k\uparrow} c_{-k\downarrow} \rangle \sim 0.065$ at the antinodal point for Bi2212 and the momentum averaged value ~0.063 for Bi2201 at the optimum doping. These are translated commonly to $F_{SC}^{\infty} \sim 0.09$ in the present definition of F_{SC}^{∞} because of the relation $F_{SC}^{\infty} = \sqrt{2} \langle c_{k\uparrow} c_{-k\downarrow} \rangle$. In the case of Bi2201, the estimated $0.16|t_1|F_{SC}^{\infty}$ is also listed in Table IV. The comparison with the sample dependence of experimental T_c suggests that the true apical oxygen position is distributed near the lower bound $d_{Oap}^z \sim 2.45$ Å, if it is spatially uniform. Alternatively, if the supermodulation exists, the lower bound of d_{Oap}^z in the modulation may be close to 2.45 Å, because it governs the SC order as the bottleneck. It is desired to test this inference by precise and simultaneous measurements of the relation between T_c and d_{Oap}^z for Bi2212 and Bi2201.

The order parameter $F_{\rm SC}^{\infty}$ increases with decreasing hole doping for $\delta > 0.05$, as we find in Fig. 2, which follows the same trend as the SC gap as we discussed, but is slightly different from the dome structure known for T_c in the cuprates, where the peak of the dome is located at higher δ . Complete and quantitative understanding of this different trend is not the scope of this paper and is left for future studies. However, the origin of this difference can be inferred to be attributed to the increasing damping and incoherence of electrons in the underdoped region toward the metal-insulator transition as was analyzed before [44,45], which is represented as the enhanced self-energy of the normal electrons toward the Mott insulator. In the experimental conditions, the atomic doping or substitution introduces atomically spatial inhomogeneity, which is ignored in the present study and may cause the localization of carriers, which could also be the origin of slower increase of T_c upon doping.

In Appendix K, we show a qualitative difference of the momentum distribution between the optimal and underdoped hole concentrations, which suggests a signature of the increased damping at lower carrier concentration within the present *ab initio* study. We also discuss in Appendix K the subtlety and complexity in the underdoped region due to the pseudogap formation, involved in several quantities, which is not the scope of this paper.

The δ dependence of the energy decomposed to the kinetic, on-site and off-site interaction energies is analyzed in Appendix G. It should be noted that the on-site interaction energy $E_U = \langle \mathcal{H}_U \rangle$ has a convex curvature as a function of δ , which contributes to the effective attraction of electrons despite the original strong repulsion U. In fact, the $U/|t_1|$ dependence of the local attraction is qualitatively consistent with F_{SC} in Fig. 9. See Appendix G for more details. The local effective attraction may cause the Cooper pairing as well as the stripe formation. It was pointed out that the convex curve of the local energy generates bistable excitations, one in the underdoped side and the other in the overdoped side, inducing the electron fractionalization and the enhanced quantum entanglement through the quantum tunneling of the two excitations [57,58]. This line of further research is an important future subject.

The importance of the apical oxygen position has been pointed out from various viewpoints [60–63]. In this paper, we have elucidated the crucial role of controlling U in

general in the single-band description, which quantitatively explains the variation of T_c^{opt} and its uncertainty in the Bi compounds. In addition, the modulation of the SC gap with the modulation of the apical oxygen position in Bi2212 has indicated that the longer d_{Oap}^z induces the smaller SC gap [55]. This is consistent with the trend of F_{SC}^{∞} , which decreases when α is increased in the realistic range as shown in Fig. 8. This indicates that Bi2212 is located already slightly above the peak in the α dependence of $F_{\rm SC}^{\infty}$, which corresponds to $\alpha = 1.2$ for doped CaCuO₂ shown in Fig. 9. The present observation is also in accordance with the effect observed by laser irradiation aiming at the displacement of the apical oxygen position [64]. The control of d_{Oap}^z if possible in a spatially uniform fashion may help to optimize the SC in which the disturbance and pair breaking by the randomness caused by the inhomogeneous supermodulation in the case of the Bi compounds could be avoided.

The effects of the displacement of the apical oxygen position on the Hamiltonian parameters were discussed in Ref. [12]. In Appendix L, we readdress this issue in relation to earlier work.

We have mainly focused on the quantities at optimal doping to clearly extract the diversity of the materials dependence, where the experimental subtlety due to the effects from extrinsic randomness as well as the complexity of physical quantities arising from the pseudogap formation in the underdoped region is irrelevant. The behavior of suppressed T_c , the SC carrier density n_s , and the coherent spectral weight arising from the pseudogap formation in the underdoped region are left for future studies. See Appendix K for more details.

In general, the *d*-wave SC correlation has a dip at (1, 1) distance. The origin is speculated as follows. The singlet pair between electrons at (0, 0) and (1, 0) sites dynamically interferes with the singlet pair between (1, 1) and (1, 0), because these two singlets are incompatible. The same is true for the singlets, which share (0, 1) site. This double interfere makes the SC correlation of the pair between (0, 0) and (1, 1) smaller very generally. However, at the moment, we do not know the origin of particularly large dip around $\delta = 1/8$, which is left for future study.

There exist studies proposing the possibility of the pair-density-wave (PDW) states [65]. However, in the present *ab initio* Hamiltonians, the PDW correlation remains small as is shown in Fig. S7 of SM [33], where peak is absent at nonzero momentum. If the stripe long-ranged order coexists in the SC ground state, the PDW order must be trivially accompanied. However, as one sees in Fig. 14(b) and in Fig. S6 [33], the stripe correlation exists but remains small and is scaled to zero in the thermodynamic limit.

In this paper, we have employed the single-band Hamiltonian for the AB orbital, because the hybridization gap between AB and B or NB orbitals is so large (~8–9 eV) that the B or NB bands are more or less completely filled and inactive [see Fig. 10(b) of Ref. [34] and Appendix D of Ref. [12]]. When one starts from the three bands constructed from Cu $3d_{x^2-y^2}$ and O $2p_{\sigma}$ atomic orbitals, the analysis would be more complicated. See Appendix M for this issue.

VI. SUMMARY AND CONCLUSIONS

We have studied the superconductivity in the *ab initio* Hamiltonians for CaCuO₂, Hg1201, Bi2201, and Bi2212 derived by using the experimental crystal structure in Ref. [12] without adjustable parameters. The dominance of SC order against severely competing stripe states and AFM state in a wide range of hole concentration is shown in the solutions for the ground state of all four materials obtained from the variational Monte Carlo calculations, which agrees with the experimental results.

The SC order parameter F_{SC}^{∞} at the optimal doping shows consistency with the superfluid density measured in the µSR and the machine learning analysis of the ARPES data for Bi2212 and Bi2201. F_{SC}^{∞} decreases with increasing doping for the doping concentration $\delta > 0.05$, showing a similarity to the SC gap reported in the STM and ARPES measurements. On the other hand, F_{SC}^{∞} quickly decreases to zero toward $\delta = 0$ for $\delta < 0.05$ forming a dome structure which has a similarity to experimental T_c , but the dome peak appears at slightly lower δ for the calculated F_{SC}^{∞} . This may be attributed to the reduced renormalization factor suggested by the broadened momentum distribution.

From the comparison of the four materials, we have revealed that $U/|t_1|$ is a crucial parameter to control the strength of the SC order; larger $U/|t_1|$ materials show larger SC order parameter $F_{\rm SC}^{\infty}$ in the realistic materials. This explains that T_c and the SC gap at the optimum doping are larger for CaCuO₂ than Hg1201, where T_c^{opt} is well scaled by $|t_1| F_{\text{SC}}^{\infty}$ as $T_c^{\text{opt}} \sim 0.16 |t_1| F_{\text{SC}}^{\infty}$. Though the experimental uncertainty in the crystal structure prohibits a quantitative comparison, F_{SC}^{∞} is also larger for Bi2212 than Bi2201 at least qualitatively, in agreement with the experimental indications. When we apply the same scaling $T_c^{\text{opt}} \sim$ $0.16|t_1|F_{\rm SC}^{\infty}$ to the two Bi compounds with the calculated order parameter, it also well explains the experimental sample dependence of T_c . The strong dependence of $F_{\rm SC}^{\infty}$ on $U/|t_1|$ for real materials is summarized in Fig. 10: In the range of $7.0 \le U/|t_1| \le 8.0$, F_{SC}^{∞} sharply increases and the calculated sensitive materials dependence of F_{SC}^{∞} is well captured within this range. This simply means that, except for Bi2212, most of the realistic materials we have studied are positioned in the weak-coupling side, where the SC order parameter rapidly increases with increasing $U/|t_1|$. The good scaling of T_c^{opt} by $0.16|t_1|F_{\text{SC}}^{\infty}$ is also summarized in Fig. 11, which indicates that the detailed difference of $U/|t_1|$ within the range of $7 < U/|t_1| < 9$ in the *ab initio* parameters reproduces the diverse materials dependence



FIG. 10. F_{SC}^{∞} as a function of $U/|t_1|$ for the four cuprate compounds at $\delta = 0.167$ plotted from the list in Tables III and IV.



FIG. 11. Experimental T_c^{opt} (black crosses or bars) in comparison to the $T_c = 0.16|t_1|F_{\text{SC}}^{\infty}$ scaling for each compound (purple bars). T_c is taken from Tables III and IV.

of T_c^{opt} . Since the larger variance for the theoretical prediction on Bi2201 is ascribed to the experimental uncertainty of the apical oxygen, it is desired to precisely determine the apical oxygen position in the experiments.

Based on the successful reproduction of the materials dependent properties, the underlying superconducting mechanism is identified by the effective local attraction emerging from the Mottness, which converts the original strong repulsion to the attraction.

The SC order parameter has the maximum above the *ab initio* values of $U/|t_1|$ at 20% larger value of $U/|t_1|$ with the enhancement of 20%–30%. If one can control on-site and off-site interaction independently, further optimization of the SC order parameter as much as the factor 2 larger value beyond the available compounds synthesized so far without falling into other competing states can be achieved as the theoretical maximum value in the present mechanism. By increasing $|t_1|$ as well as the whole parameter values

uniformly, T_c^{opt} should obviously increase accordingly. These offer a clue for the materials design in the future.

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APPENDIX A: LANCZOS METHOD AND RESTRICTED BOLTZMANN MACHINE PROCEDURE

1. Lanczos method

To further improve the accuracy of the VMC calculations or the variance extrapolation of the competing ground state candidates, we apply the Lanczos method [42]. To do so the optimized mVMC wave function $|\Psi\rangle$ is extended by

$$|\psi_n\rangle = \left(1 + \sum_{n=1}^M \alpha_n \mathcal{H}^n\right) |\Psi\rangle.$$
 (A1)

The factor in front of $|\Psi\rangle$ can be regarded as an additional projection operator with variational parameters α_n , which when chosen appropriately–further reduce the energy. Although one could systematically improve the wave function by increasing M, the computational cost increases exponentially with M, too. Hence we employ the Lanczos method only up to the first step (n = 1) within this work.

2. Restricted Boltzmann machine procedure

To further improve the mVMC wave function we apply a restricted Boltzmann machine as a variational wave function $|\Psi\rangle$, as first suggested in Ref. [66]. Here we follow the notations given in Refs. [20,42]. The variational wave function including RBM takes the following form:

$$|\Phi\rangle = \mathcal{P}^{\rm G} \mathcal{P}^{\rm J} \mathcal{P}^{\rm dh} \mathcal{N} |\phi^{\rm pair}\rangle, \tag{A2}$$

where $\ensuremath{\mathcal{N}}$ is the additional RBM correlator. The RBM correlator,

$$\mathcal{N} = \prod_{k} 2 \cosh\left(b_k + \sum_{i} W_{ik} \sigma_i\right) e^{\sum_{i} a_i \sigma_i}, \quad (A3)$$

introduces the additional variational parameters b_k (hidden layer), a_i (visible layer), and W_{ik} (network).

In practice we apply the additional RBM projection after the wave function is already optimized via mVMC calculations; i.e., the variational parameters of the optimized wave function $|\phi\rangle = \mathcal{P}^{G}\mathcal{P}^{J}\mathcal{P}^{dh}|\phi^{pair}\rangle$ are kept fixed during the RBM procedure.

The accuracy of the wave function depends on the number of hidden and visible parameters (N_h, N_v) [66]. Hence we can define a hidden variable density as $\alpha_{\text{RBM}} = N_h/N_v$ as measure for the accuracy. Note that the number of RBM variational parameters increases with α , too.

Within this work the RBM procedure was applied with $\alpha_{\text{RBM}} = 4$.

APPENDIX B: VARIANCE EXTRAPOLATION PROCEDURE

The total energy per site $E/N = \langle \mathcal{H} \rangle / N$ is calculated after the variance extrapolation. The true ground state as a function of the variance $(\delta E)^2 = (\langle \mathcal{H}^2 \rangle - \langle \mathcal{H} \rangle^2) / \langle \mathcal{H} \rangle^2$ is obtained by taking the limit $\delta E \to 0$, because the true eigenstate satisfies $\delta E = 0$ [39–41]. If several different states such as SC, spin- and charge-ordered as well as normal metallic states are competing, the ground state is determined from the lowest-energy state after the variance extrapolation. In practice, the variance extrapolation is a linear regression of the energies per site E/N (e.g., obtained from combinations of mVMC, Lanczos, or RBM) over the



FIG. 12. Variance extrapolation for the example of CaCuO₂ at $\delta = 0.167$ on a L = 24 lattice. The inset shows an enlarged plot around $(\delta E)^2 = 0$.



FIG. 13. Comparison of F_{SC} between that obtained by using δ dependent Hamiltonians (red) and the fixed Hamiltonian (black) as in the main text.

variance δE for a specific state, to approximate the zero variance value of the energy.

As an example, the variance extrapolation for $\delta = 0.167$ in CaCuO₂ is shown in Fig. 12. From the inset we confirm the lowest energy of the SC state with severely competing AFM and stripe states very close to it (the difference is around 5 meV). However, the overlap of the error bars is small and in comparison to AFM, C4S8, and C3S3 states, the SC state definitely has lower energy.

APPENDIX C: RESULTS OF δ DEPENDENT HAMILTONIAN

The hole density dependence of F_{SC} at L = 24 is compared between the case presented in the main text and that calculated by the δ dependent Hamiltonian given in Sec. S1 B of SM [33]. The results for CaCuO₂ in Fig. 13(a) and for Bi2212 in Fig. 13(b) show that the difference of the two Hamiltonians is small and the essential feature can be analyzed by taking the Hamiltonians analyzed in the main text.

APPENDIX D: THERMODYNAMIC LIMIT OF THE TRIAL WAVE FUNCTION AFTER OPTIMIZATION FOR CaCuO₂

Here we discuss the stability of the AFM, C3S3, and C4S8 states in the thermodynamic limit for CaCuO₂. To do so, each state is stabilized on different lattice sizes and the spin and charge structure factors $[S_s(q)]$ and $S_c(q)$] are calculated.

We plot $S_s(q)$ as a function of 1/L or $1/L^2$ depending on the cases of the presence or absence, respectively, of the AFM order by following the convention and perform a linear extrapolation. The result is shown in Fig. 14, where in Fig. 14(a) the peak height of the spin structure factor follows a linear trend with a nonzero offset, indicating a stable longranged AFM order. In Fig. 14(b), the peak of the spin structure is scaled to zero in the thermodynamic limit, which indicates the absence of the long-ranged AFM order in the SC state at this doping. We did not find the coexistence of the SC and AFM order at other doping either.

The size scaling of the charge stripe states is shown in Fig. 15. Although the plot of the size dependence is not



FIG. 14. Size dependence of the spin structure factor $10^3 \times S_s(\mathbf{q}_{\text{max}})/N$ at (π, π) for (a) the AFM state after optimization at half filling (L = 12, 16, 24) and $\delta = 0.167$ (L = 12, 18, 24), and for (b) the SC state at $\delta = 0.167$ for different lattice sizes (L = 24, 30, 36).



FIG. 15. Size dependence of the charge and spin stripe state after the optimization of variational parameters for different lattice sizes and fillings. For C3S3 (red crosses) $\delta = 0.207$ and L = 12, 24 and for C4S8 (blue dot) $\delta = 0.125$ and L = 16, 24. (a) $10^3 \times S_s(\mathbf{q}_{\text{SDW}})/N$ vs 1/L. (b) $10^3 \times S_c(\mathbf{q}_{\text{CDW}})/N$ vs $1/L^2$. The dashed lines are only to guide the eyes. \mathbf{q}_{SDW} and \mathbf{q}_{CDW} are the wave numbers at the peak of S_s and S_c , respectively, for the spin density wave (SDW) and charge density wave (CDW) modulations, respectively.

sufficient due to very demanding computational cost for larger sizes, the trend suggests that only the C4S8 state shows a clear long-ranged order of spin and charge at around $\delta = 0.125$ in the thermodynamic limit. The C3S3 state seems to collapse to a paramagnetic state at the chosen doping of $\delta = 0.207$, at which C3S3 has relative stability. Hence we use the labeling "C3S3-like" instead of C3S3 in the main text. We do not go into details of the size dependence for the stripelike states, because they are in any case excited states.

APPENDIX E: COMPARISON TO HUBBARD MODEL STUDIES

Here we discuss the crucial difference of the *ab initio* results from the extensively studied Hubbard models

without the off-site interaction. In the Hubbard model studies, irrespective of the presence of the next nearest transfer t_2 or absence of it, in the hole doped region, a broad consensus seems to be formed, where the ground state has stripe type long-ranged charge order [3,5,6,67–72]. The charge or spin stripe states were also suggested to coexist with weak SC order in some cases [5,72], but other studies did not find the SC order [6,68,69,71].

In the Hubbard model studies, the numerical methods have a variety including the present VMC, density matrix renormalization group, constrained path quantum Monte Carlo, tensor network and density matrix embedding theory, which have their own advantages and disadvantages and they are complementary. When they can be compared with reliable solutions, the above Hubbard model calculations were benchmarked, which have shown comparable accuracy when compared between each state-of-the-art updated version. Even in quantum spin models, the level of accuracy of the above methods is roughly similar [21]. We also refer to recent thorough comparisons [73].

On the other hand, when realistic off-site interaction is included, the ground state is reported to be charge homogeneous superconducting state [10]. The off-site interaction substantially suppresses the SC order but the charge and spin stripe states are more damaged by the frustration introduced by the off-site interaction. The role of off-site interaction for the stabilization of the SC state relative to the stripe state was directly demonstrated in Ref. [10] by the comparison of *ab initio* result and that of the Hamiltonian obtained by switching off only the off-site interactions. We confirmed the similar behavior for the present Hamiltonians. The absence of developed stripe correlations in the SC ground state is seen in Fig. 14 herein and in Fig. S6 of SM [33].

APPENDIX F: *t*² **DEPENDENCE**

Here we show t_2 dependence of F_{SC} by starting from the *ab initio* Hamiltonian for hole doped CaCuO₂ with other Hamiltonian parameters fixed, where the effect of t_2 is



FIG. 16. SC correlation function $P_d(r)$ for modified t_2 from the CaCuO₂ Hamiltonian for L = 24 lattice.



FIG. 17. SC order parameter F_{SC} as a function of δ calculated for three choices of t_2 modified from CaCuO₂ Hamiltonian.

monitored beyond the *ab initio* value primarily within the realistic range of $|t_2/t_1|$ ($0.2 \le |t_2/t_1| \le 0.3$) in Figs. 16 and 17 for L = 24 lattice. F_{SC} slightly decreases with increasing t_2 , which is qualitatively consistent with a different approach [63]. However, the variation of F_{SC} is at most 10% near the optimum doping in the realistic range. Furthermore, F_{SC} has essentially no t_2 dependence in the range $0.0 \le |t_2/t_1| \le 0.2$. On the other hand, the period of the stripe order is known to sensitively depend on t_2 [5,6,68,69,71,72] in the ground states of Hubbard models and it may alter the superconductivity if it coexists, while the present charge uniform superconducting ground state is quite different.

APPENDIX G: ANALYSIS OF δ DEPENDENCE OF ENERGY

In Fig. 18 the total energy per site E_{tot} and the on-site Coulomb part E_U [see Figs. 18(a) and 18(c), respectively] are shown for doped CaCuO₂. Each energy contribution is subtracted by a linear function $F(\delta) = b_0 + b_1 \delta$ for better visibility, where b_0 and b_1 are listed in Table V [see gray lines in Figs. 18(a) and 18(c)] and are shown in Figs. 18(b) and 18(d). The subtracted energies are fitted by a quadratic function $\mathcal{P}(\delta) = c_0 + c_1 \delta + c_2 \delta^2$ to examine the curvature. Explicit values of the parameters are given in Table V.

The result shows that the total energy is concave as a function of δ of course, which is required from the thermodynamic stability, while only E_U exhibits convex behavior with $c_2 < 0$. Because the effective particle interaction is given by the δ^2 term, we find that the local quantity E_U is the origin of the attraction while the nonlocal energies contribute to $c_2 > 0$ in the total energy ensuring the thermodynamic stability. This supports the idea that the local strong correlation (repulsion) called Mottness turns to originate the effective attraction of the electrons, which is the underlying mechanism of both the Cooper pairing and charge segregation such as the stripes. The attraction is understood from the following Mottness. The local energy is retained high in the Mott insulator because of U.



FIG. 18. Energy per site as a function of δ in the SC state on a L = 24 square lattice. Total energy E_{total}/N (a) and $E_{\text{total}}/N - F(\delta)$ (b), as well as on-site Coulomb energy E_U/N (c) and $E_U/N - F(\delta)$ (d), are plotted. Here, a δ -linear function $F(\delta)$ defined below is subtracted in (b) and (d) for better visibility. Note that $F(\delta)$ is different depending on the type of the energy as seen in Table V. The gray line in the left-hand column indicates $F(\delta) = b_0 + b_1 \delta$, which is subtracted in (b) and (d). Right-hand column: energies after subtraction of $F(\delta)$ are fitted via a quadratic function $\mathcal{P}(\delta) = c_0 + c_1 \delta + c_2 \delta^2$. Fitting parameters are given in Table V.

However, if the carrier is doped, this is released by acquiring the itinerancy in a nonlinear fashion as a function of δ , which yields $c_2 < 0$ and the attraction. In accordance with the α dependence of F_{SC} , c_2 shows a similar behavior as $-c_2 = 1.47$, 2.24, 2.13, 1.85, and 1.53 at $\alpha = 1.0$, 1.1, 1.2, 1.3, and 1.4, respectively, with a peak around $\alpha \sim 1.1-1.2$.

TABLE V. Fitting parameters of linear correction function and quadratic fit function discussed in Fig. 18. See the main text for the explanation.

	b_0	b_1	c_0	c_1	c_2
$E_{\rm total}/N$	6.4995	-11.9723	0.0626	-2.5274	9.2307
E_U/N	0.3455	-0.5900	-0.0043	0.3651	-1.4749

TABLE VI. Effective interlayer Hamiltonian parameters (in eV) for Bi2212 at $\delta = 0.2$ taken from Ref. [12].

Bi2212						
V_0^l 0.643	V_1^l 0.463	$V_2^l \\ 0.368$	V_3^l 0.291	V_4^l 0.262	V_5^l 0.220	V_6^l 0.120
t_0^l -0.098	t_1^l -0.001	t_2^l 0.019	t_3^l -0.010	$\begin{array}{c}t_4^l\\0.007\end{array}$	$\begin{array}{c}t_5^l\\0.000\end{array}$	t_6^l -0.003

APPENDIX H: INTERLAYER CONTRIBUTIONS FOR Bi2212

The effective interlayer Hamiltonian parameters for Bi2212 are shown in Table VI, where V_n^l is the interlayer Coulomb interaction and t_n^l the interlayer hoppings. As defined in Ref. [12], the notation n = 0 represents the interaction or hopping between interlayer nearest neighbor Cu atoms (located one above the other), and $n \ge 1$ represents the interaction or hopping between a Cu atom and its (n + 1)th interlayer nearest neighbor (located above or below its intralayer *n*th nearest neighbor).

APPENDIX I: DOPING DEPENDENCE OF ENERGY FOR Bi2201 AND Bi2212

The energy per site as a function of δ and the competition of SC, stripe, and AFM states is shown in Fig. 19 for Bi2212 and Fig. 20 for Bi2201.

APPENDIX J: SCALING OF THE SC ORDER IN STRONG-COUPLING REGION

The SC order parameter F_{SC} in the strong-coupling region is plotted in Fig. 21 by monitoring *U* with other parameters of the Hamiltonian fixed at the *ab initio* values of CaCuO₂ for L = 24 lattice. F_{SC} is scaled as



FIG. 19. Variance extrapolated energies of Bi2212 for various ground-state candidates (SC, charge or spin stripe C3S3 and C4S8, and AFM states) as a function of hole doping δ on a L = 24 square lattice. (a) Total energies per site subtracted by $\mathcal{F}(\delta)$. All energies are subtracted by the function $\mathcal{F}(\delta) = -12.34985 \cdot \delta + 6.36953$. (b) Energy difference ΔE for the variance extrapolated data from (a).



FIG. 20. Variance extrapolated energies of Bi2201 for various ground-state candidates [SC, charge-spin stripe (C3S3 and C4S8), and AFM state] as a function of hole doping δ on a L = 24 square lattice. (a) Total energies per site subtracted by $\mathcal{F}(\delta)$. All energies are subtracted by the function $\mathcal{F}(\delta) = -15.38797 \cdot \delta + 7.79753$. (b) Energy difference ΔE for the variance extrapolated data from (a).

 $F_{\rm SC} \sim (|t_1|/U)^p$ with $p \sim 0.6$ irrespective of the hole density. As far as we know, there exists no theoretical argument in the literature including the role of the superexchange interaction $J \sim 4|t_1|^2/U$ to understand this scaling. The origin of this nontrivial power-law dependence imposes a severe constraint on the superconducting mechanism in the strong-coupling region and is left for future studies.



FIG. 21. SC order parameter in strong-coupling region for $\delta = 0.101$ and 0.167. The lattice size is L = 24. The dashed lines are fitting $F_{\text{SC}} \propto (|t_1|/U)^p$ with p = 0.58 for $\delta = 0.167$ and p = 0.60 for $\delta = 0.101$.

Overall δ and α dependences of F_{SC} are shown in Fig. 22. This shows that though the asymptotic behavior at large α is insensitive to δ , the optimal δ and α depend on each other. We note here α corresponds to $(1/8.1)U/|t_1|$.

APPENDIX K: COMPLEXITY IN UNDERDOPED REGION AND ANALYSIS ON MOMENTUM DISTRIBUTION

We show calculated momentum distribution defined by

$$n(\mathbf{k}) = \frac{1}{2N} \sum_{i,j,\sigma} \mathrm{e}^{\mathrm{i}\mathbf{k}(\mathbf{r}_i - \mathbf{r}_j)} \langle c_{i\sigma}^{\dagger} c_{j\sigma} \rangle, \qquad (\mathrm{K1})$$

because the jump or singularity of $n(\mathbf{k})$ at the Fermi level characteristic of metals measures the incoherence of the carrier. Here, we show $n(\mathbf{k})$ for the case of doped CaCuO₂ in Fig. 23 for the cases of $\delta = 0.028$ [Figs. 23(a) and 23(f)], 0.045 [Figs. 23(b) and 23(g)], 0.101 [Figs. 23(c) and 23(h)], 0.125 [Figs. 23(d) and 23(i)], and $\delta = 0.167$ [Figs. 23(e) and 23(j)] in the SC state on the L = 24square lattice. Although F_{SC}^{∞} is similar between $\delta = 0.028$ (or 0.045) and $\delta = 0.101$ (or 0.125), n(k) shows substantially more smooth and rounded shape for $\delta = 0.028$ and 0.04 than $\delta = 0.101$ and 0.125, demonstrating that the effect of the larger normal damping at smaller δ is responsible for this rounded behavior. The substantial increase in the damping with decreasing doping may be responsible for the suppression of T_c in the underdoped region irrespective of the high F_{SC}^{∞} . More quantitative studies will be presented elsewhere. In SM Fig. S9, we show the case of $\delta = 0.028$ and $\delta = 0.167$ on the L = 36square lattice to ensure that the size dependence is weak [33].

The strong damping is most prominent in the underdoped region around the antinodal points. This region is under the strong influence of the pseudogap, which makes the relation of the physical quantities nontrivial. The different behavior of T_c and F_{SC} is such an example. Deviation of the SC carrier density n_s and the weight of quasiparticle coherence peak at the antinodal point from $F_{\rm SC}$ and the SC gap $\Delta_{\rm SC}$ against the naive expectation may be another example. Although F_{SC} and the SC gap Δ_{SC} grow on top of the pseudogap as we revealed in the case of F_{SC} , n_s seems to be severely suppressed by the pseudogap around the antinodal point. This trend is indeed seen in the comparison of the muon penetration depth and the SC gap measurement [45,74–76], which causes difficulty in the comparison of our calculated result and experimental indications in the underdoped region. Such a complexity is expected to be small at the optimum doping region, while the prominent materials dependence is seen most prominently at the optimum doping. This is the reason why we focus on the materials dependence at the optimum doping.



FIG. 22. SC order parameter in the space of δ and $\alpha = (1/8.1)U/|t_1|$ modified from the *ab initio* CaCuO₂. Calculations were performed on L = 24 lattice.



FIG. 23. Momentum distribution n(k) in the Brillouin zone for doped CaCuO₂ on a L = 24 square lattice in the SC state. The doping concentration is $\delta = 0.028$ for (a),(f); 0.045 for (b),(g); 0.101 for (c),(h); 0.125 for (d),(i); and 0.167 for (e),(j). The lower panels [(f)–(j)] are contour plots.

APPENDIX L: EFFECT OF APICAL OXYGEN POSITION ON HAMILTONIAN PARAMETERS

We extend analysis on the effect of apical oxygen position examined in Ref. [12]. Studies on effects of the apical oxygen position represented by the distance d_{Oap}^z to Cu on effective Hamiltonian parameters are few [60,61,63]. In Table VII we show the Hamiltonian parameters of Bi2201 when the apical oxygen is artificially shifted.

As is already addressed in Ref. [12], U decreases with decreasing d_{Oap}^z because of increased screening from electrons at the apical oxygen, which is consistent with

TABLE VII.	Ab initio	single-band	effective	Hamiltonian	for
Bi2201 when	the apical of	oxygen is shi	fted. The	energy unit is	eV.

d_{Oap}^{z} (Å)	t_1	t_2	U	V_1	V_2
2.58 2.53	-0.527 -0.513	0.140 0.159	4.393 3.994	1.030 0.837	0.602 0.447
	$U/ t_1 $	$ t_2/t_1 $	$V_1 / t_1 $		
2.58 2.53	8.336 7.789	0.266 0.310	1.954 1.632		

the claim in Ref. [61]. In Ref. [60], $|t_2/t_1|$ increases with increasing d_{Oap}^z , for instance, in the comparison of Hg1201 and Bi2201. However, it shows opposite trend in Table VII and in the results in Ref. [12]. References [60,61] focused on the effect of orbitals whose energies are above the Fermi level such as Cu 4s and apical O $2p_z$ orbitals. The increasing d_{Oap}^z makes those levels lower because farther distance to the negative CuO_2 layer charge for the apical O $2p_{z}$ orbital and farther distance to the negatively charged apical oxygen for the Cu 4s orbital makes the Madelung potential lower. This lowering induces a larger hybridization with the AB orbital located around the Fermi level constructed from Cu $3d_{x^2-v^2}$ and in-plane O $2p_z$ orbital (our target band), which we call in-plane CuO_2 AB orbital. This increased hybridization especially enhances t_2 . However, this is not the whole story. The effective next nearest neighbor hopping t_2 in the single-band Hamiltonian is also altered by the effect from the bands below the Fermi level such as $3d_z$ orbital. Increasing d_{Oap}^z makes the lowering of the $3d_z$ level and hence causes the decrease in the hybridization with in-plane CuO₂ AB orbital which cancels the effect of the increased hybridization of the orbital above the Fermi level, as was pointed out in Ref. [63]. More precisely, the apical O $2p_z$ and Cu $3d_z$ orbitals are strongly hybridized and they form bonding and antibonding bands, and we need to consider all of these contributions, which are taken into account quantitatively in our calculations in the derivation of the effective Hamiltonian. As a consequence, the present estimate for t_2/t_1 has large difference from that in the estimates of Ref. [60] in some cases. For instance, in the case of Hg1201, $|t_2/t_1| \sim 0.36$ for Hg1201 in Ref. [60], while ~ 0.20 in the present study. Although Ref. [60] is based on complex approximations to estimate t_2/t_1 only by taking into account the contribution from the band above the Fermi level, recent standard way employs the maximally localized Wannier orbitals and its Hamiltonian matrix elements for the estimate of hopping, by considering all the bands contribution near the Fermi level. This is much simpler, more straightforward, and transparent for the estimate of the lattice fermion Hamiltonian parameters, which are used in Ref. [12] as the basis of our VMC calculations. In fact, recent estimates of $|t_2/t_1|$ for Hg1201 are 0.20 and 0.23 in Refs. [77,78] (in the revised manuscript), respectively, which are consistent with the present 0.20.

As we already mentioned in Appendix F, larger $|t_2/t_1|$ slightly but quantitatively suppresses F_{SC} in the realistic parameter range, which is opposite to the prediction in Ref. [60] but is consistent with Ref. [63]. Furthermore, and most importantly, too small dependence of optimal F_{SC} on $|t_2/t_1|$ clarified in this paper makes the role of $|t_2/t_1|$ on F_{SC} highly questionable. We find that the effect of the apical oxygen position on the superconductivity is primarily to control U.

APPENDIX M: COMPARISON TO APPROACH USING MULTIBAND HAMILTONIAN

There exists recent work based on the atomic orbitals containing Cu $3d_{x^2-y^2}$ and O $2p_{\sigma}$ orbitals [79–81]. Of course, multiorbital Hamiltonians should give essentially a similar answer if the derivation and the solving procedure are appropriate. On the other hand, the Hamiltonian becomes more complex with larger number of parameters, as it should be.

However, the antibonding band and nonbonding or bonding band are well separated with the hybridization gap (band center separation is ~ 9 eV and the direct gap is 5-6 eV for the cases we studied in this paper). See Appendix D of Ref. [12] for detailed analyses. In this circumstance, we can safely start from the picture of singleband Hamiltonian derived from the AB band of strongly hybridized Cu $3d_{x^2-y^2}$ and O $2p_{\sigma}$ orbitals only, because the B orbitals are more or less completely filled and inactive. See Appendix D of Ref. [12]. See also Fig. 10(b) of Ref. [34], where the completely filled B bands are verified for the Hg-based cuprate through all the relevant hole densities and this is universal in the curate superconductors. In the single AB band description, the B degrees of freedom are downfolded and give the renormalization to the AB orbital description. Since the AB-B hybridization gap is large, the perturbative downfolding procedure to renormalize and eliminate B degrees of freedom works well as a good approximation [12]. This is based on the multiscale ab initio scheme for correlated electrons (MACE) with refined GW approximation supplemented by the level renormalization feed back [34]. Except for the AB band, all the bands are either well below the Fermi level or above the Fermi level so that they can be perturbatively taken by the partial trace summation to give renormalizations to the AB degrees of freedom.

Of course, one can start from the three-band Hamiltonian where the charge transfer gap and covalency are relevant parameters. However, it should end up with this AB or B description after the basis transformation if one focuses on the low-energy physics in the realistic situation of the cuprates. The effect of the parameters of the charge transfer energy and the *d*-*p* covalency were taken into account in our downfolding procedure from the three-band to a single-band AB Hamiltonian in Ref. [12]. For instance, larger charge transfer gap results in poorer screening and larger correlation (*U*) as is confirmed in Ref. [12] [See the comparison of Table IV with Tables I and II in Ref. [12]]. Therefore, the three-band parameters are encoded in $U/|t_1|$ and other parameters in the AB single-band description indirectly and systematically in a complex manner.

There exist several recent analyses based on multiband Hamiltonian containing Cu $3d_{x^2-y^2}$ and O $2p_{\sigma}$ atomic orbitals for the cuprates or Hubbard-type models [79–81]. Three-band Hamiltonian constructed from the Cu $3d_{x^2-y^2}$

and O $2p_{\sigma}$ atomic orbitals derived and listed in Table IV of Ref. [12] shows rough consistency with the proposal by Ref. [79], in which the authors claim stronger superconducting order for smaller charge transfer gap ΔE_{xp} and larger d-p transfer t_{xp} in the notation of Ref. [12]. Naively, one would expect that smaller ΔE_{xp} makes stronger screening on the AB band and decreases effective U in the single-band picture, while larger t_{xp} directly leads to larger t_1 . Both result in smaller $U/|t_1|$, which appears to contradict the statement claimed in the present paper that larger $U/|t_1|$ leads to larger superconducting order parameter in the realistic parameter region. However, one needs to be careful about the parameter region employed in Ref. [79]. When one sees U_d (the direct on-site repulsion between atomic d orbital) dependence of the superconducting order parameter in Fig. 2 of Ref. [79], one clearly finds that the superconducting order parameter decreases from $U_d = 10$ to 18 or 14 in the energy scale of t_{pp} , transfer between neighboring Op orbital. When one compares the parameters with those in Table IV of Ref. [12], and compares with Tables I and II of Ref. [12], one notices that $U_d = 10$ in Ref. [79] already corresponds to the region around the optimum of $U/|t_1|$ in the present single-band description and further increase of U_d drives the system into the strong-coupling region with larger U, where the superconducting order decreases with increasing $U/|t_1|$, as one can see in our result in Fig. 9(a). This perfectly explains the U_d dependence between $U_d = 10$ and 18 in Ref. [79] as well as the $t_{pd} = t_{xp}$ and p level, ϵ_p dependences, because larger $t_{pd} = t_{xp}$ and smaller $\epsilon_p = \Delta E_{xp}$ both make smaller $U/|t_1|$, as was mentioned above, and leads to larger superconducting order parameter in the strong-coupling region, as is clarified in Fig. 9(a). It clarifies that the region studied in Ref. [79] is outside of the real materials dependence of the cuprates studied here. We further need more detailed studies of the correspondence between multiorbital and the present single-band description in real materials systematically, which is left for future studies.

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