

Microscopic mechanism of high-temperature superconductivity revealed by *ab initio* studies on hole-doped multilayer cuprates $\text{HgBa}_2\text{Ca}_2\text{Cu}_3\text{O}_8$ under pressure

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Triple-layer cuprate superconductor $\text{HgBa}_2\text{Ca}_2\text{Cu}_3\text{O}_8$ (Hg1223) keeps the record of the highest superconducting (SC) critical temperature $T_c \sim 134$ K among all the existing materials at ambient pressure. T_c further increases under pressure up to $T_c \sim 160$ K, thereby holding the key to the realistic and comprehensive understanding of the cuprate superconductors, which is one of the grand challenges in physics. However, the microscopic mechanism of high T_c of Hg1223 remains to be elucidated. Here, we perform first-principles calculations of Hg1223 both at ambient and applied pressures by solving *ab initio* Hamiltonians with an accurate variational solver supplemented by a neural network. The pressure dependence of the calculated *d*-wave SC order parameter and estimated T_c shows a structure supporting a T_c peak around 30 GPa in quantitative agreement with the reported experimental indications. The origin of the strong SC amplitude of Hg1223 at ambient pressure compared to other cuprates is identified as strong local Coulomb repulsion U attributed to poorer screening of the Coulomb interaction in multilayer compounds. The origin of the further increase in T_c under pressure is ascribed to a unique interplay of three elements, namely, increased electron hopping t , decreased U , and more importantly, strongly reduced nonlocal Coulomb repulsion V with increasing pressure. The origin of Cooper pairing is identified as the emergent local attraction counterintuitively arising from the strong bare local repulsion U for Hg1223. It explains why Hg1223 shows the highest T_c among other cuprates, while the pairing mechanism is common in other cuprates as well. The emergent attraction is interpreted from “attraction from reduced repulsion,” originating from the release of the fluctuating doubly occupied sites characterized from the “false vacuum” in the Mott insulator to the double-occupation-free *d*-wave SC states caused by the carrier doping. This instantaneous and local attraction is in marked contrast with the case of the conventional BCS SC mediated by bosonic glues. The local attraction is consistent with the electron fractionalization supported in experimental analyses. The coexistence of SC and antiferromagnetic orders is also demonstrated as a characteristic feature of the multilayer system with self-doping in the underdoped region. The quantitative and microscopic understanding of the high T_c in Hg1223 provides a new perspective on the origin of the cuprate superconductivity and offers a new route explicitly using this channel of the emergent attraction for future superconductivity research aiming at designing and optimizing SC materials.

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I. INTRODUCTION

Since the discovery of high-temperature superconductors (SCs) in copper oxides (cuprates) in 1986 [1], cuprate SCs continue to hold the record of the highest SC critical temperature (T_c) at ambient pressure. Although a tremendous amount of research has been conducted with significant advances, understanding the microscopic mechanisms behind materials dependencies of T_c and high- T_c superconductivity itself continues to be a central challenge in condensed

matter physics. Strong electron correlations that characterize the cuprate SCs accompanied by severe competition among intertwined different states and fluctuations require accurate many-body solutions.

Recently, by using state-of-the-art quantum many-body solvers, *ab initio* low-energy effective Hamiltonians for single-, double-, and infinite-layer cuprates have been solved, which successfully reproduced the rich material dependence of T_c [2]. More specifically, based on the effective Hamiltonians for CaCuO_2 with an infinite-layer structure, $\text{HgBa}_2\text{CuO}_4$ (Hg1201), $\text{Bi}_2\text{Sr}_2\text{CuO}_6$ (Bi2201) with a single-layer structure, and $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ (Bi2212) with a double-layer structure by using an accurate variational Monte Carlo (VMC) method [3,4] combined with a neural network algorithm [5] using only experimental crystal structures as inputs, the material and doping concentration dependencies of the SC order parameter and T_c have been quantitatively reproduced. The materials dependence has been understood primarily from the difference in an *ab initio* Hamiltonian parameter for the dimensionless

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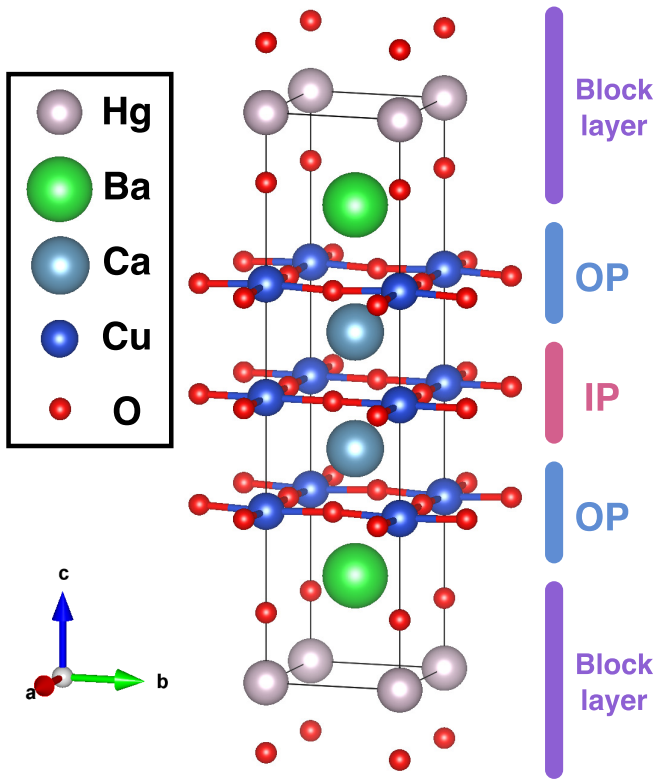


FIG. 1. Crystal structure of Hg1223 and directions of primitive vectors (*a*, *b*, *c*). The inner CuO₂ plane (IP) sandwiched by the two outer CuO₂ planes (OPs) is separated from the equivalent next three CuO₂ planes by the block layers, and the neighboring IP and OP are separated by interstitial Ca atoms. The crystal structure [6] is visualized from data in the Crystallography Open Database [7–9] using VESTA [10].

ratio of the local interaction U to the nearest-neighbor hopping amplitude $|t_1|$. The severe competition among the SC, antiferromagnetic (AF), and stripe phases is in agreement with the observations in the experiments as well.

However, significantly higher T_c was observed in multilayer, especially triple-layer cuprates, while the numerical analysis of the multilayer cuprates on the same level as Ref. [2] is so far missing because of the demanding computational cost ascribed to the large unit cell.

Among all the available materials at ambient pressure, the highest T_c was reported at $T_c \sim 134$ K in a triple-layer cuprate compound HgBa₂Ca₂Cu₃O₈ (Hg1223) [11–15] (see Fig. 1 for the crystal structure). When the pressure is applied, Hg1223 shows a further increased T_c forming domelike pressure dependence and reaches the peak at around 30 GPa, as high as $T_c \sim 160$ K, which is the highest record among the cuprates [13,15]. Therefore, without identifying the microscopic origin of such high- T_c values based on the *ab initio* parameter-free framework, the mechanism of the cuprate SC would not be ultimately understood.

In this study, we quantitatively elucidate why multilayer cuprates including triple-layer Hg1223 show the substantially higher T_c than the single-, double-, and infinite-layer compounds. We also discuss the origin of the observed domelike pressure dependence of T_c , which peaks around

30 GPa. The materials dependence of the single-, double-, and infinite-layer compounds was captured without paying serious attention to the difference in the off-site interactions because they are similar at ambient pressure [2]. Here, in contrast, we show that the pressure-induced reduction of off-site interactions is substantial and plays a major role in enhancing SC. We also clarify that, in the multilayer compounds, the induced self-doping brings about the difference in carrier concentration between the IP and OPs, which allows for the emergence of coexisting SC and AF order as was experimentally observed [16,17].

Based upon the successful and quantitative reproduction of the materials dependence of the SC for the cuprates, including the triple-layer Hg1223, we identify the realistic mechanism of SC: The Cooper pair is induced by the instantaneous and local attraction caused by the reduction of repulsion in the background of strong “vacuum fluctuation,” which is a unique property of the strongly correlated doped Mott insulator. This mechanism is radically different from the conventional BCS SC [18], where the retardation is essential to induce the boson-mediated weak-coupling SC to circumvent the unavoidable electron-electron Coulomb repulsion.

Our calculations are based on the *ab initio* low-energy effective Hamiltonians of Hg1223 derived in Ref. [19]. The pressure-dependent lattice constants and atomic positions used in deriving these Hamiltonians were obtained from the first-principles structural optimization performed in Ref. [19]. The crystal structure was parametrized by the lattice constants *a* and *c* and five internal structure parameters, from which the atomic coordinates at each pressure were determined. The relevant hopping parameters were obtained from the matrix elements between maximally localized Wannier orbitals. In the procedure to derive the effective Hamiltonians, Refs. [19,20] employed a state-of-the-art methodology of constrained GW (cGW) scheme [21–23], which completely removes the double counting problem known in the constrained RPA [24,25]. In addition, the self-interaction correction (SIC) together with the level renormalization feedback (LRFB) [26] are taken into account, in which the charge distribution among the atomic O2*p* and Cu3*d_{x²-y²}* orbitals, which strongly hybridize to form the antibonding and bonding orbitals, is appropriately treated in a self-consistent fashion. This procedure in total is called cGW-SIC + LRFB scheme. The antibonding orbital constituting the low-energy effective Hamiltonian contains a small portion of other atomic orbitals, such as hybridizing Cu3*d_{3z²-r²}* and O2*p_z* orbitals, which contribute to the interlayer hopping. The ground state of the derived effective Hamiltonian for the antibonding orbital on lattices is obtained in this paper by applying the VMC method [3,4] refined from the seminal work [27–30]. We further enhance the accuracy by combining the VMC method with neural network techniques [5,31,32].

This paper is organized as follows: In Sec. II, we briefly describe the *ab initio* low-energy effective Hamiltonian and the VMC method. In Sec. III, we present the ground states of Hg1223 at ambient pressure obtained by the VMC method. In Sec. IV, we present the pressure dependence of the SC order parameter and T_c in Hg1223. In Sec. V, we discuss the effective attraction emergently arising from the strong local repulsion as the origin of Cooper pairing of Hg1223

and compare with the cases of other cuprates with lower T_c on the basis of the successful and realistic reproductions of the experimental trend. Section VI is devoted to discussions. Finally, in Sec. VII, we summarize our findings and discuss their consequences. We also provide perspectives for future studies.

II. METHODS

A. *Ab initio* effective Hamiltonians

We solve the *ab initio* triple-layer effective Hamiltonian of Hg1223 for the antibonding orbital consisting of strongly hybridized $\text{Cu}3d_{x^2-y^2}$ and $\text{O}2p_\sigma$ orbitals as derived in Ref. [19], which has the form

$$\mathcal{H} = \mathcal{H}_t + \mathcal{H}_U + \mathcal{H}_V + \mathcal{H}_\mu, \quad (1)$$

with

$$\mathcal{H}_t = \sum_{I,J,\sigma} t_{IJ} c_{I,\sigma}^\dagger c_{J,\sigma}, \quad (2)$$

$$\mathcal{H}_U = \sum_I U_I n_{I,\uparrow} n_{I,\downarrow}, \quad (3)$$

$$\mathcal{H}_V = \frac{1}{2} \sum_{I \neq J} V_{IJ} n_I n_J, \quad (4)$$

$$\mathcal{H}_\mu = \sum_I \mu_I n_I. \quad (5)$$

Here, I and J are multiindices of site and layer, e.g., (i, α) and (j, β) , respectively; sites i and j correspond to positions $\mathbf{r}_i = (x_i, y_i)$ and $\mathbf{r}_j = (x_j, y_j)$ on an $L \times L$ square lattice, whereas α and β indicate the layer, namely, IP or OP. In Fig. 1, here and later, two OPs should be distinguished as OP1 and OP2, where the summations over α and β run over IP, OP1, and OP2. The operator $c_{I,\sigma}^\dagger$ ($c_{I,\sigma}$) creates (annihilates) an electron with spin σ and index I . The operator $n_{I,\sigma} = c_{I,\sigma}^\dagger c_{I,\sigma}$ counts the number of electrons with spin σ and index I , and the total number operator is given by $n_I = n_{I,\uparrow} + n_{I,\downarrow}$. The system contains one IP and two OPs as shown in Fig. 1, and the total number of sites is $N_s = N_l L^2$, with N_l being the number of layers ($N_l = 3$ for the triple-layer Hg1223). The hopping parameter is given by t_{IJ} and the strengths of the on-site and off-site Coulomb interactions are represented as U_I and V_{IJ} , respectively. Both hopping and off-site interaction parameters are functions of the relative coordinate vector $\mathbf{r}_i - \mathbf{r}_j$ between sites i and j because of the translational invariance of the crystal structure. The chemical potential of the IP μ_I relative to that of the OP controls the relative carrier densities under the condition of a fixed total number of electrons for a fixed size in the canonical ensemble and is a function of the total number of electrons $N_e = [(1 - \delta)N_s]$.

We consider hopping and interaction parameters that extend over long distances and assume that their values are nonzero within the range satisfying $r = |\mathbf{r}_i - \mathbf{r}_j| \leq 3\sqrt{2}$ between sites i and j . Here, the distance is counted in the length unit of the nearest Cu-Cu distance, both within the xy plane and along the interlayer z direction. Beyond this range, the parameters are negligibly small. The dominant hopping and interaction parameters employed in the present calculations are summarized in Table I, for which we employ

TABLE I. Selected parameters for the *ab initio* low-energy effective Hamiltonian of Hg1223 at 0, 30, and 60 GPa. We show the nearest-neighbor hopping parameter t_1 , the on-site Coulomb interaction U , and the nearest-neighbor off-site Coulomb interaction V_1 for the IP and the OP. All values are given in eV. Complete lists including other parameters are given in Tables II–IV.

IP	t_1	U	V_1
0 GPa	−0.4857	4.5184	0.9878
30 GPa	−0.5833	4.3615	0.8752
60 GPa	−0.6492	4.1751	0.6842
OP	t_1	U	V_1
0 GPa	−0.4849	4.5443	0.9785
30 GPa	−0.5554	4.4986	0.8056
60 GPa	−0.5812	4.2381	0.5832

the values from Ref. [20]. For other parameters, see complete lists in Tables II–IV in Appendix A. They include both intralayer and interlayer parameters. In most cases, the parameters for hopping and interaction are smaller than 10% of the largest nearest-neighbor hopping and on-site interaction, respectively, when $r = |\mathbf{r}_i - \mathbf{r}_j| > 2$. Although parameters with such small values contribute little to qualitative physical properties, we keep values at longer distances with $r > 2$ up to the 11th neighbor to perform more realistic simulations of actual systems and for quantitative comparisons with real compounds. The truncated hopping parameters employed in the VMC calculations accurately reproduce the cGW-SIC + LRFB band dispersion near the Fermi level. The resulting Fermi surfaces of the individual bands agree well with those obtained by the full parameter set of the cGW-SIC + LRFB calculations.

The chemical potential difference $\mu = \mu_{\text{OP}} - \mu_{\text{IP}}$ between the IP and the OP is determined in a way such that μ reproduces the carrier density in each layer obtained by GW calculations. For the details of the determination of μ , see Appendix B. The final hole doping δ dependencies of μ at 0, 30, and 60 GPa are given by

$$\mu_{0 \text{ GPa}} = 5.192 - 9.78\delta, \quad (6)$$

$$\mu_{30 \text{ GPa}} = 5.087 - 11.2\delta, \quad (7)$$

$$\mu_{60 \text{ GPa}} = 2.990 - 6.36\delta, \quad (8)$$

respectively. At ambient pressure, δ_{IP} is always smaller than δ_{OP} . With increasing pressure and doping δ , the chemical potential difference μ becomes smaller, and the carrier densities of the IP and the OP tend to be closer to each other, as one can see later in Figs. 2 and 7.

B. Numerical methods

We employ an elaborated VMC method, which is available as an open source software [3,4,33] for calculating the ground-state properties of Hg1223. The variational wave function is given by

$$|\psi\rangle = \mathcal{P}_G \mathcal{P}_J \mathcal{P}_{\text{dh}} |\phi_{\text{pair}}\rangle, \quad (9)$$

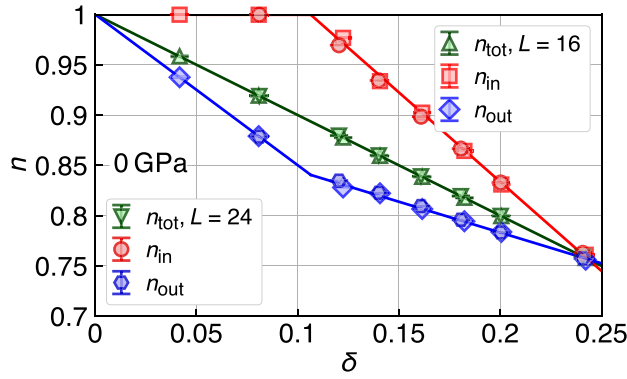


FIG. 2. Inner- and outer-layer hole densities as a function of total hole density δ for the *ab initio* Hamiltonian of Hg1223 at ambient pressure. n_{tot} is the averaged electron density per Cu site, whereas n_{in} and n_{out} are the averaged electron densities per site in the inner and outer planes, respectively. The IP always has lower hole density. The plots for $L = 16$ and 24 show negligible size dependence, suggesting that the $L = \infty$ limit is nearly reached.

where the Gutzwiller factor $\mathcal{P}_G$ [34], the charge Jastrow correlation factor $\mathcal{P}_J$ [35,36], and the doublon-holon correlation factor $\mathcal{P}_{\text{dh}}$ [37] are defined by

$$\mathcal{P}_G = \exp \left(\sum_I g_I n_{I,\uparrow} n_{I,\downarrow} \right), \quad (10)$$

$$\mathcal{P}_J = \exp \left(\sum_{I < J} v_{IJ} n_I n_J \right), \quad (11)$$

$$\mathcal{P}_{\text{dh}} = \exp \left[\sum_{t=1,2} \sum_{m=0}^4 \sum_{l=d,h} \sum_I \gamma_I^{(t,m,l)} \xi_I^{(m,l)} \right], \quad (12)$$

respectively. Here, the doublon is a site occupied by both up- and down-spin electrons and the holon represents an empty site. Variational parameters are g_I , v_{IJ} , and $\gamma_I^{(t,m,l)}$. We assume that $g_I = g_{(i,\alpha)}$ does not depend on the position $\mathbf{r}_i$, but can take different values depending on the layer α . On the other hand, $v_{IJ} = v_{(i,\alpha)(j,\beta)}$ is chosen to be a function of the relative position $|\mathbf{r}_i - \mathbf{r}_j|$ and can vary for different layer pairs (α, β) . The many-body operator $\xi_I^{(m,l)}$ is diagonal in the real space representation and takes 1 if a doublon ($l = d$) [holon ($l = h$)] exists at index I and m holons (doublons) at t th nearest neighbors surround it; otherwise, $\xi_I^{(m,l)}$ is 0. The parameter $\gamma_I^{(t,m,l)} = \gamma_{(i,\alpha)}^{(t,m,l)}$ does not depend on the position $\mathbf{r}_i$, but can take different values depending on the layer α and t th nearest neighbors. The pair-product wave function [3,38,39] is given by

$$|\phi_{\text{pair}}\rangle = \left(\sum_{I,J=1}^{N_s} \sum_{\sigma,\sigma'} f_{IJ}^{\sigma\sigma'} c_{I,\sigma}^\dagger c_{J,\sigma'}^\dagger \right)^{N_c/2} |0\rangle, \quad (13)$$

where $f_{IJ}^{\sigma\sigma'}$ are variational parameters and are assumed to have a sublattice structure with translational invariance.

The variational wave function can further be improved by applying additional correlation factors to the aforementioned wave function, and the corresponding energy becomes closer to the exact values by performing a zero-variance

extrapolation. We typically consider the wave function with the restricted Boltzmann machine (RBM) correlation factor [5,31,32], which has the form

$$|\psi_{\text{RBM}}\rangle = \mathcal{P}_G \mathcal{P}_J \mathcal{P}_{\text{dh}} \mathcal{P}_{\text{RBM}} |\phi_{\text{pair}}\rangle. \quad (14)$$

Here, the RBM correlation factor is chosen as

$$\mathcal{P}_{\text{RBM}} = \exp \left(\sum_{I,\sigma} a_{I\sigma} n_{I,\sigma} \right) \prod_J \cosh \left(b_J + \sum_{I,\sigma} W_{IJ\sigma} n_{I,\sigma} \right), \quad (15)$$

with $a_{I\sigma}$, b_J , and $W_{IJ\sigma}$ being variational parameters. Increasing the ratio of the number of hidden parameters N_h to the number of sites, namely, $\alpha_{\text{RBM}} = N_h/N_s$, improves the accuracy of the variational wave function. In practice, we apply the RBM correlation factor on top of the optimized wave function $|\psi\rangle = \mathcal{P}_G \mathcal{P}_J \mathcal{P}_{\text{dh}} |\phi_{\text{pair}}\rangle$, although the RBM correlation factor itself can simulate the effects coming from $\mathcal{P}_G$ and $\mathcal{P}_J$. Another way of improving the wave function is to consider the Lanczos method [40,41]. We choose the appropriate factors α_n in the wave function

$$|\psi_M\rangle = \left(1 + \sum_{n=1}^M \alpha_n \mathcal{H}^n \right) |\psi\rangle \quad (16)$$

so that its energy is minimized. We apply the Lanczos method up to the first step ($M = 1$) [42,43] in this study because of the high computational cost, which increases exponentially in M .

We assume antiperiodic-periodic boundary conditions and calculate ground-state candidate states of lattices with $N_s = 3L^2$ sites. The system sizes that we have studied range from $L = 16$ to $L = 28$, where the system contains 2352 sites at $L = 28$. We first calculate ground-state candidate states and then compare the energy per site $E/N_s = \langle H \rangle / N_s$ of each state after the variance extrapolation [44–46] to the zero limit with the help of results obtained after Lanczos and RBM operations if necessary. The variance is evaluated by $(\delta E)^2 = (\langle H^2 \rangle - \langle H \rangle^2) / \langle H \rangle^2$ and satisfies $\delta E = 0$ for true eigenstates. We determine the ground state by choosing the one with the lowest energy among the candidate states. Such ground-state candidate states include the SC, metallic, staggered AF, and C4S8 stripe [47,48] states as well as the coexistence of the SC and AF. See the detailed procedure in Appendix C. Since the Lanczos procedure is generally considered to have negligible effects on analyzed physical quantities such as correlations and order parameters [42,49–51], we present order parameters without applying the Lanczos method hereafter.

For important quantities such as SC and AF order parameters, we take the size extrapolation to the thermodynamic limit. In some cases below, however, we only show the results for $24 \times 24 \times 3$ lattice for Hg1223 with an explicit remark to save the demanding computational time for the size extrapolation, when the size dependence is small.

C. Physical quantities

We calculate the spin and charge structure factors to distinguish whether the state exhibits magnetic and charge orders. The spin structure factor in each layer α (= IP, OP) is

defined by

$$S_s^\alpha(\mathbf{q}) = \frac{1}{L^2} \sum_{i,j} \langle S_{i,\alpha} \cdot S_{j,\alpha} \rangle e^{i\mathbf{q} \cdot (\mathbf{r}_i - \mathbf{r}_j)}, \quad (17)$$

where $S_i = (S_i^x, S_i^y, S_i^z) = \sum_{\tau,\tau'} c_{i,\tau}^\dagger \boldsymbol{\sigma}_{\tau,\tau'} c_{i,\tau} / 2$ is the spin-1/2 operator and $\boldsymbol{\sigma}_{\tau,\tau'}$ is the Pauli matrix. Similarly, the charge structure factor in each layer is defined by

$$S_c^\alpha(\mathbf{q}) = \frac{1}{L^2} \sum_{i,j} \langle n_{i,\alpha} n_{j,\alpha} \rangle e^{i\mathbf{q} \cdot (\mathbf{r}_i - \mathbf{r}_j)}. \quad (18)$$

The system exhibits long-range order when $S_s^\alpha(\mathbf{Q})/L^2$ or $S_c^\alpha(\mathbf{Q})/L^2$ at the Bragg peak $\mathbf{Q}$ is extrapolated to a nonzero value in the thermodynamic limit ($L \rightarrow \infty$). The order parameter in the thermodynamic limit is evaluated by

$$m_{s,c}^\alpha = \sqrt{\lim_{L \rightarrow \infty} \frac{S_{s,c}^\alpha(\mathbf{q})}{L^2}}. \quad (19)$$

The SC states in the present study are of d -wave type, and the corresponding SC correlation function in each layer α is given by

$$P_d^\alpha(\mathbf{r}) = \frac{1}{L^2} \sum_{\mathbf{r}_i} \langle \Delta_d^{\alpha\dagger}(\mathbf{r}_i) \Delta_d^\alpha(\mathbf{r}_i + \mathbf{r}) + \text{H.c.} \rangle, \quad (20)$$

where $\Delta_d^\alpha(\mathbf{r}_i)$ is the order parameter with the form

$$\Delta_d^\alpha(\mathbf{r}_i) = \frac{1}{\sqrt{2}} \sum_{\mathbf{r}} f_d(\mathbf{r}) (c_{\mathbf{r}_i,\alpha,\uparrow} c_{\mathbf{r}_i+\mathbf{r},\alpha,\downarrow} - c_{\mathbf{r}_i,\alpha,\downarrow} c_{\mathbf{r}_i+\mathbf{r},\alpha,\uparrow}). \quad (21)$$

The form factor at $\mathbf{r} = (x, y)$ that reflects the $d_{x^2-y^2}$ symmetry is given by

$$f_d(\mathbf{r}) = \delta_{y,0}(\delta_{x,1} + \delta_{x,-1}) - \delta_{x,0}(\delta_{y,1} + \delta_{y,-1}). \quad (22)$$

For SC states in large systems, the SC correlation function nearly saturates for sufficiently long distances. This long-distance correlation is proportional to the square of the SC order parameter $F_{SC}^\alpha(L)$ for a finite system in layer α . To practically estimate this value, we average the long-distance part of $P_d^\alpha(\mathbf{r})$ and evaluate a quantity, which is given by

$$[F_{SC}^\alpha(L)]^2 = \bar{P}_d^\alpha(L) = \frac{1}{N_{\text{far}}} \sum_{|\mathbf{r}| > \sqrt{2}L/4} P_d^\alpha(\mathbf{r}). \quad (23)$$

Here, position $\mathbf{r} = (x, y)$ runs over all sites within a region $(-L/2, L/2)^2$ and N_{far} is the number of lattice points satisfying $|\mathbf{r}| \in (\sqrt{2}L/4, \sqrt{2}L/2)$. Then, the SC order parameter in the thermodynamic limit $F_{SC}^{\alpha,\infty}$ for layer α is evaluated by

$$\bar{P}_d^{\alpha,\infty} = \lim_{L \rightarrow \infty} \bar{P}_d^\alpha(L), \quad (24)$$

$$F_{SC}^{\alpha,\infty} = \lim_{L \rightarrow \infty} F_{SC}^\alpha(L) = \sqrt{\bar{P}_d^{\alpha,\infty}}. \quad (25)$$

The linear $1/L$ scaling is assumed during the extrapolation [52] because quasiparticle excitations of the d -wave SC state at nodal points exhibit the Dirac-type linear dispersion.

All the physical quantities including the energy are estimated by the standard Monte Carlo sampling. Error bars for a fixed system size are estimated by the statistical errors of each quantity. In the procedure of size extrapolations, we also estimate the error bars coming from the extrapolation.

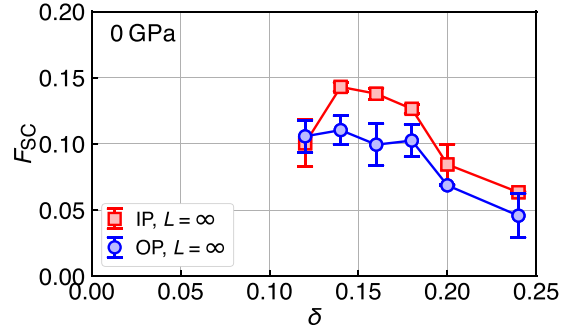


FIG. 3. Doping concentration dependence of the SC order parameter F_{SC} defined in Eq. (25) at ambient pressure. The red and blue lines show F_{SC} for the IP and OP, respectively, after size extrapolation to the thermodynamic limit $L \rightarrow \infty$. The error bars are due to the size extrapolation error.

III. RESULTS AT AMBIENT PRESSURE

In this section, we show the solutions for the ground state of the effective Hamiltonian for Hg1223 at ambient pressure and discuss SC properties competing with AF and stripe states. A characteristic feature of the multilayer compounds, not present in the single-, double-, and infinite-layer compounds, is self-doping, in which the carrier concentration is self-optimized and varies among layers. Figure 2 shows how the inner- and outer-layer hole densities, δ_{IP} and δ_{OP} , respectively, evolve as a function of averaged hole density per layer $\delta = (\delta_{IP} + 2\delta_{OP})/3$ for the *ab initio* Hamiltonian of Hg1223 at ambient pressure. From Eq. (6), the IP always has a lower chemical potential of electrons (higher chemical potential of holes) in the realistic carrier density region, so that the IP has a lower hole density. This is consistent with the experimental indications of the multilayer compounds [16,17].

A. Doping concentration dependence of SC order

We show the hole density (δ) dependence of the SC order parameter F_{SC} in the thermodynamic limit at the IP and OP (two OPs show the same behavior) in Fig. 3. Hereafter, for F_{SC}^∞ obtained after the extrapolation to the thermodynamic limit, we drop ∞ as F_{SC} . (The procedure of obtaining F_{SC} from the extrapolation of the SC correlation P_d to the thermodynamic limit is described in Appendixes D and E.) The dominance of the d -wave SC order demonstrates the consistency with the experimental indications. The optimum value of $F_{SC} \sim 0.14$ at $\delta = 0.14 - 0.16$ is found in the IP and exceeds the maximum value ~ 0.13 of the single-, double-, and infinite-layer compounds reported for Bi2212 [2]. The origin of the highest optimum F_{SC} among the cuprates studied here and in Ref. [2] is mainly attributed to the highest value of $U/|t_1|$ due to the poor screening. The optimum hole density is roughly consistent with Cu valence measurement [15,53] and NMR data [54] by considering its temperature dependence.

The IP and OP exhibit a similar doping dependence of the SC dome structure. In fact, although the Hamiltonian parameters are similar between the IP and OP at ambient pressure except for the chemical potential, F_{SC} shows the maximum at $\delta_{IP} = 0.06$ for the IP while at $\delta_{OP} = 0.12$ for the OP if we plot as a function of the hole density in each layer as is seen in

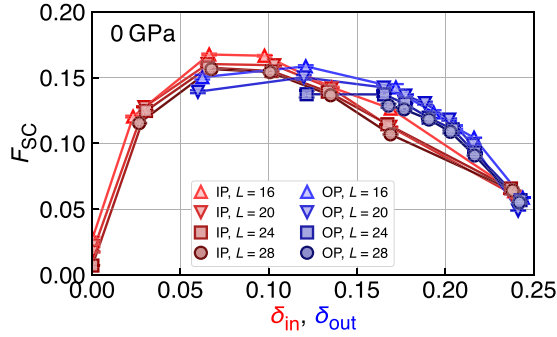


FIG. 4. Size-dependent SC order parameters F_{SC} for the IP and OP at ambient pressure as a function of hole density δ_{IP} and δ_{OP} in each layer for several choices of system sizes. In this figure, F_{SC} is defined as the converged values of $\sqrt{P_d}$ at long distances before the size extrapolation.

Fig. 4. This shift between the IP and OP cannot be explained if the IP and OP are independent. On the other hand, the shift can be caused by the synchronization between the IP and OP as one sees as a function of the averaged density δ in Fig. 3 suggesting a substantial interlayer proximity effect, which may also enhance the amplitude of F_{SC} at the IP relative to the OP because the IP is sandwiched by the two OPs. To assess the role of parameter inhomogeneity at 0 GPa, we also set the OP parameters equal to the IP parameters and find nearly unchanged doping dependence of the SC order parameters, indicating that the inhomogeneity has little effect on them. We emphasize that this interlayer proximity effect occurs near ambient pressure, where the Hamiltonian parameters for the IP and OP do not differ significantly. Under pressure, however, the parameter difference between the two layers becomes more pronounced. Indeed, at 60 GPa, the off-site Coulomb interactions in the IP become substantially stronger than those in the OP (see Table I); therefore, the interlayer proximity effect is not strong enough to make the perfect synchronization.

B. Coexistence of AF and SC

The carrier density in the IP is much closer to the half filling than that in the OP, which tends to stabilize AF order in the IP, while the OP stabilizes SC order. Therefore, if the interlayer coupling is switched off by keeping the self-doped hole density, one would expect the segregation and independent survival of the SC order in the OP and the AF order in the IP in an appropriate range of the averaged hole density, because the microscopic coexistence is not seen in the numerical result of the single-, double-, and infinite-layer cuprates [2]. However, this segregation is not a microscopic coexistence, but an interlayer phase separation. If a small but nonzero tunneling is introduced between the segregated IP and OP, as in the realistic situation of the multilayer cuprates, the proximity may lead to the coexistence in both planes. Here, we discuss such a possibility of the microscopic and homogeneous coexistence of AF and SC orders at ambient pressure.

To see a typical example, we focus on the case of 8% doping on average. When we use the *ab initio* Hamiltonian

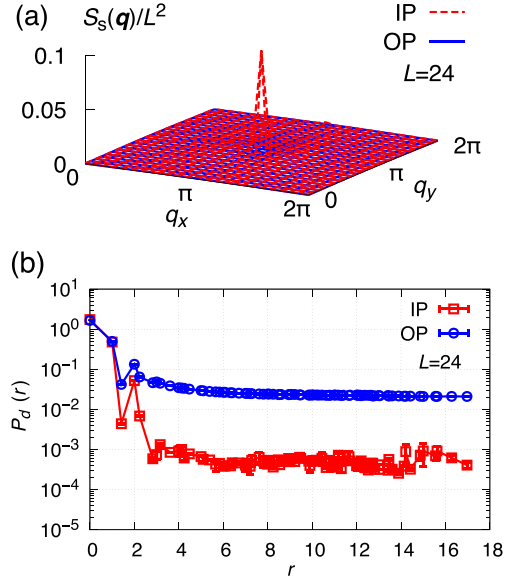


FIG. 5. Spin structure factors defined in Eq. (17) [panel (a)] and SC correlation defined in Eq. (20) [panel (b)] of the IP and OP for 24×24 lattice. The chemical potential difference between the IP and OP is $\mu = 3.2$, which is decreased from the value $\mu = 4.41$ for the Hg1223 *ab initio* Hamiltonian at ambient pressure estimated from the GW calculation. The spin structure shows a prominent peak at $\mathbf{Q} = (\pi, \pi)$ for the IP, which may grow to the Bragg peak in the thermodynamic limit, whereas it shows no such peak for the OP. Both the IP and OP exhibit the SC correlation saturated at long distances. Because the AF correlation tends to degrade the SC correlation, the saturated value of the SC correlation is larger for the OP than for the IP.

for Hg1223 at ambient pressure, the IP becomes nearly half filled by the self-doping and exhibits the AF order. However, in the underdoped region, the GW calculation tends to be less reliable due to the strong correlation effect of the precursor to the Mott insulator in the real compounds ignored in the GW calculation, which implies that the approach to the Mott insulator in the IP will be slower in reality in accordance with the experimental indications. To mimic this effect, we study the effect of μ reduced from the GW adjusted value, $\mu = 4.41$ to $\mu = 3.2$, which makes the difference in δ_{IP} and δ_{OP} smaller to $\delta_{IP} = 0.07$ and $\delta_{OP} = 0.085$ instead of $\delta_{IP} = 0$ and $\delta_{OP} = 0.12$ for $\mu = 4.41$. (For the detailed analyses regarding the dependence on the chemical potential difference between the IP and OPs, please see Appendix F.)

Figure 5 shows the SC and AF correlations in the OP and IP for a 24×24 lattice. The size extrapolation to the thermodynamic limit in Fig. 6 indeed supports the coexistence. (See also Fig. 24. The coexisting state has the lowest energy below the uniform SC and uniform AF states. We have not studied the comparison with possible long-period stripe states, which is left for future studies.) The result supports the coexistence at the inner layer in a certain range of parameters by the mechanism of proximity from the robust SC order in the outer layer that lowers the energy from a uniform non-SC AF state. The coexistence of the AF and SC in multilayer cuprates is overall consistent with experiments [16,17,55].

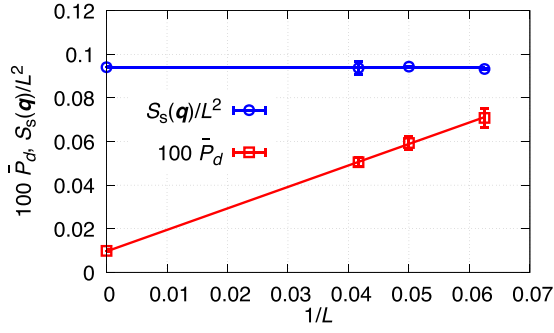


FIG. 6. Size extrapolation of $S_s(\mathbf{q})/L^2$ and $\bar{P}_d$ defined in Eqs. (17) and (23), respectively, for the IP. At chemical potential $\mu = 3.2$ for 8% doping concentration, the AF order defined in Eq. (19) is $m_s = 0.3067(6)$ while the SC order defined in Eq. (25) is $F_{SC} = 0.0099(3)$, supporting the coexistence of AF and SC orders.

IV. RESULTS UNDER PRESSURE

In this section, we show the solutions for the ground state under pressure, especially the pressure dependence of the SC order parameter and T_c .

A. Pressure dependence of SC order

Figure 7 illustrates the δ dependence of $n_{IP} = 1 - \delta_{IP}$ and $n_{OP} = 1 - \delta_{OP}$ while Fig. 8 shows the SC order parameter in the thermodynamic limit, F_{SC} in the IP and OP at 30 and 60 GPa, which confirms the dominance of the SC order for $\delta > 0.12$ even under pressure. For detailed data for finite sizes

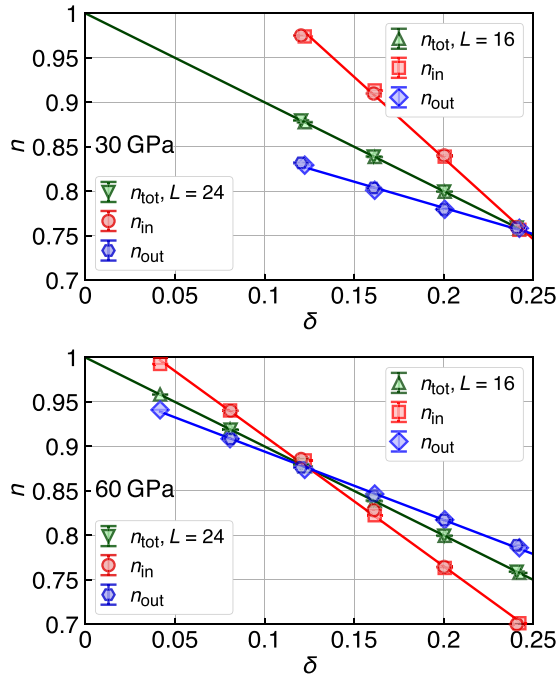


FIG. 7. Inner- and outer-layer hole densities as a function of total hole density δ for the *ab initio* Hamiltonian of Hg1223 at 30 GPa [panel (a)] and 60 GPa [panel (b)]. The comparison between the results at 16×16 and 24×24 lattices demonstrates that the size dependence is negligible.

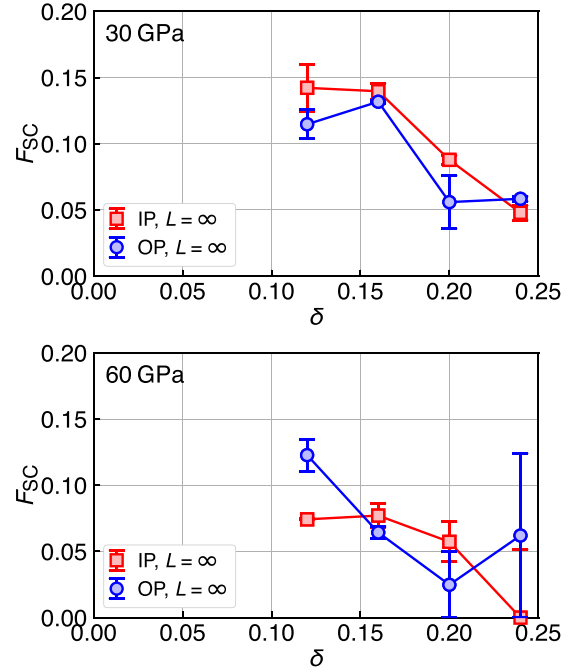


FIG. 8. Doping concentration dependence of the SC order parameter F_{SC} at 30 GPa [panel (a)] and 60 GPa [panel (b)] after the size extrapolation to the thermodynamic limit. The red and blue lines are for the IP and OP, respectively.

and their size extrapolations, see Appendixes D and E. Both at 30 and 60 GPa, F_{SC} is comparable between the IP and OP while in most cases, the IP shows slightly larger F_{SC} . In Fig. 9, we plot F_{SC} for all the pressures at the hole density $\delta = 0.16$, which is the optimal value at ambient pressure. Since F_{SC} is always larger for the IP than for the OP at $\delta = 0.16$, we plot only the values for the IP. The SC order parameter F_{SC} (black circles) remains nearly constant from 0 to 30 GPa and decreases from 30 to 60 GPa. We analyze the origin of this trend below. In the inset of Fig. 9, the pressure dependence of the Hamiltonian parameters $|t_1|$, $U/|t_1|$ and $V_1/|t_1|$ at the IP are plotted by taking the data from Tables II–IV. From 0 to 60 GPa, $|t_1|$ increases, while $U/|t_1|$ and $V_1/|t_1|$ decrease significantly. In particular, $V_1/|t_1|$ decreases to less than a half.

We plot F_{SC} at $\delta = 0.16$ in Fig. 10 for four choices of pressures of Hg1223 at the corresponding values of $U/|t_1|$. We also plot F_{SC} for the infinite-, single-, and double-layer compounds, CaCuO₂, Hg1201, Bi2201, and Bi2212 at the optimal doping and ambient pressure taken from Ref. [2] together. The data for Hg1223 under pressure clearly deviate from the universal trend for the single-, double-, and infinite-layer compounds at ambient pressure grouped by the shaded belt. This implies that F_{SC} is not solely determined by $U/|t_1|$ under pressure. In fact, although the off-site Coulomb interaction is similar among various cuprates at ambient pressure (for instance, the nearest-neighbor Coulomb interaction $V_1 \sim 1$ eV; see Table V), it is decreased substantially under pressure (for instance, $V_1 \sim 0.88$ and 0.68 eV at 30 and 60 GPa, respectively) (see Appendix A for details). This reduction is along the same line as the reduction of U under pressure (U for Hg1223 is ~ 4.52 , 4.36 , and 4.18 eV at 0, 30, and 60 GPa, respectively), which is ascribed to the increased screening

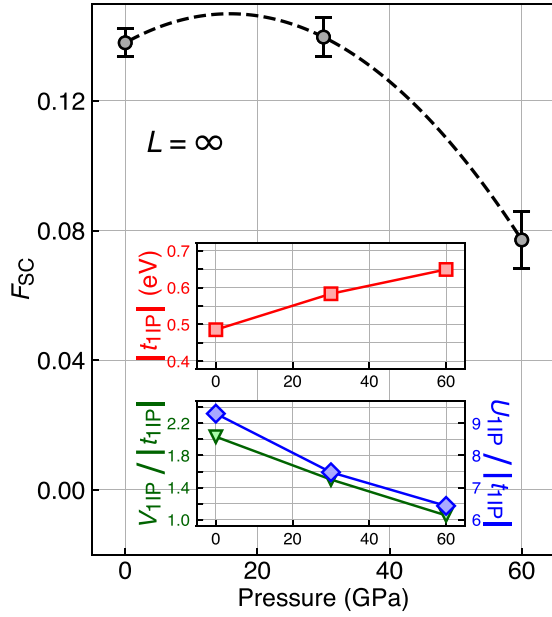


FIG. 9. Pressure dependence of the SC order parameter (F_{SC}) in the thermodynamic limit after the size extrapolation for the IP at $\delta = 0.16$ of Hg1223. The black dashed curve is a quadratic fitting. Inset: Pressure dependence of the nearest-neighbor hopping parameter ($|t_{IP}|$) and the ratio of the on-site Coulomb interaction parameter to the nearest-neighbor hopping parameter ($U_{IP}/|t_{IP}|$) as well as the ratio of the nearest-neighbor Coulomb interaction parameter to the nearest-neighbor hopping parameter ($V_{IP}/|t_{IP}|$), taken from Ref. [19] and Table I. In this plot, all the hopping and Coulomb interaction parameters are presented for the IP, where the larger SC order is observed at $\delta = 0.16$. Error bars are estimated from the size extrapolation errors.

from the block layers and apical oxygens caused by the shrunk lattice constant under pressure. It shares a common feature observed before in quasi-two-dimensional (2D) systems [56], where the reduction of Coulomb interaction caused by the off-plane screening is similar irrespective of the mutual distance (namely, regardless of whether $U, V_1, V_2, \dots$) (and hence the reduction is relatively more significant for larger mutual distance).

The large enhancement of F_{SC} by the reduction of the off-site interaction was actually pointed out in Ref. [2], and the present result is consistent with it. The gradual pressure dependence of F_{SC} for Hg1223 instead of the steeper reduction in the general trend at ambient pressure is now understood by the interplay of reduced U and V_i (and reduced V_i with $i > 2$ as well), where the reduction of F_{SC} by reduced $U/|t_1|$ is partially compensated by a large reduction of the off-site interactions.

To corroborate our understanding of the effect of the reduced off-site interaction under pressure, we calculated for a hypothetical system of modified single-layer Hg1201, where we reduced only the off-site interactions ($V_i, i \geq 1$) from the *ab initio* values of Hg1201 with the factor $\xi = 0.686$ as ξV_i at nearly 15% doping concentration ($\delta = 0.146$). Here, this factor ξ mimics $V_i/|t_1|$ for pressurized Hg1223 that has $U/|t_1|$ corresponding to Hg1201. The result is plotted as a red open square in Fig. 10, which indeed fits the trend of Hg1223

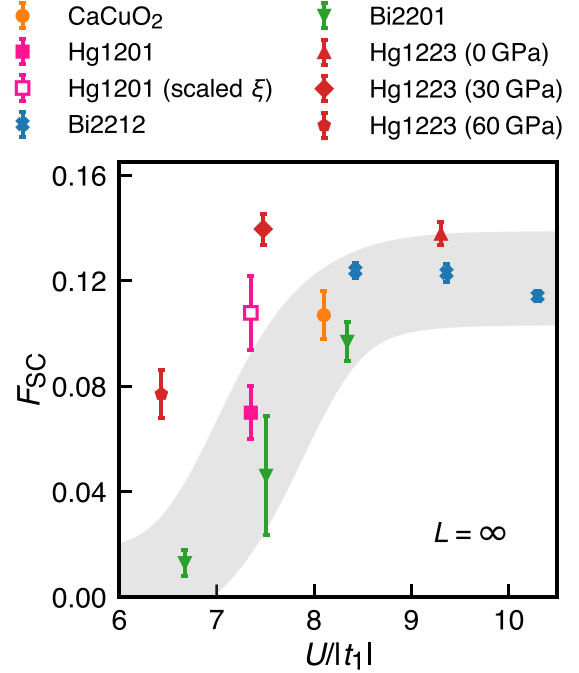


FIG. 10. Comparison of the SC order parameter in $U/|t_1|$ dependence among cuprate SCs in the thermodynamic limit after the size extrapolation. For Hg1223, the abscissa is the ratio $U_{IP}/|t_{IP}|$ at the IP. The data for CaCuO₂, Hg1201, Bi2212, and Bi2201 at $\delta \sim 0.15$ are taken from Ref. [2]. The plot for “Hg1201 (scaled ξ)” (open magenta square) is obtained by calculating the ground state for the Hg1201 single-layer Hamiltonian (filled magenta square) with reduced off-site interaction V_i by a factor $\xi = 0.686$.

under pressure. In the multilayer systems, there seems to be an additional cooperative effect from the OP and IP through the proximity to further enhance the SC.

This suggests the importance of controlling the off-site interaction in material design, as is already pointed out in hypothetical parameter search [2]. The proposal in Ref. [2] indeed becomes a reality here as the pressure effect, which works to reduce the effective interaction in the relative ratio to the values for ambient pressure more efficiently for the off-site part than for the on-site one, which helps enhance the superconductivity in Hg1223. If U could be retained under pressure simultaneously with reduced off-site interaction, F_{SC} could be much more enhanced, as we will discuss below.

B. Pressure dependence of T_c

It was proposed that T_c at the optimum hole density can be estimated by using the *ab initio* result in the scaling form

$$T_c \sim 0.16|t_1|F_{SC} \quad (26)$$

for the single-, double-, and infinite-layer compounds at ambient pressure [2]. Since F_{SC} of Hg1223 under pressure does not follow the common trend of cuprates at ambient pressure and the triple-layer structure contains nonequivalent IP and OP, it is highly nontrivial whether this scaling form is also valid in the triple-layer Hg1223 under pressure. To test the validity, in Fig. 11, we plot the pressure dependence of T_c estimated by using Eq. (26) together with t_1 and the calculated

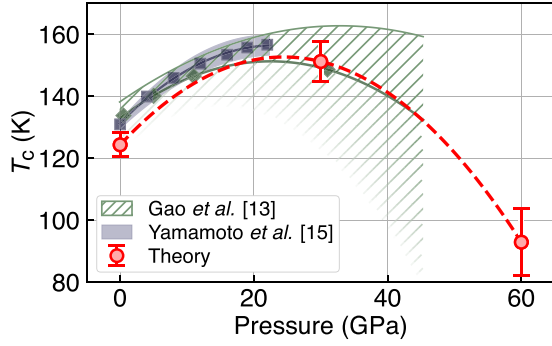


FIG. 11. Comparison of the pressure dependence of T_c between theory and experiment for Hg1223. We estimate the theoretical T_c (red circles) at $\delta = 0.16$ by using Eq. (26) for the IP. The red dashed curve is a guide to the eye obtained by a quadratic fit of the theoretical T_c . Experimental T_c contains uncertainty. The upper bound of the uncertainty is the onset temperature (onset T_c) at which the resistivity ρ starts deviating downward. The lower bound should, in principle, be the temperature at which the resistivity becomes zero. The trajectory of the inflection point defined by $d^2\rho/dT^2 = 0$ is widely used as another metric of T_c . We show two examples of experimental reports. The gray shaded belt depicts the experimental T_c at $\sim 16\%$ doping concentration reported in Ref. [15], with the upper and lower bounds being the onset and zero-resistivity temperature, respectively. The black curve with squares shows the inflection-point trajectory. The greenish hatched belt is from Ref. [13], where the upper bound is the onset and the lower bound drawn as the green thick curve with diamonds represents the inflection-point trajectory. In Ref. [13], the zero resistivity was not reached even at the reported lowest temperature ~ 80 K. Therefore, the true T_c could be lower than the lower bound of the greenish hatched zone by considering possible nonuniformity and inhomogeneity of pressure in terms of hydrostatic nature above 30 GPa (see Sec. IV B). The theoretical T_c shows a peak at around 30 GPa, consistently with the experimental T_c .

F_{SC} at the IP responsible for the higher T_c and compare it with the experimental T_c reported in Refs. [13,15,57]. The experimental and numerical values of T_c agree fairly well at 0 and 30 GPa and they are maximized around 30 GPa, which supports the validity of the scaling form (26) under pressure for the triple-layer compound despite the large deviation of F_{SC} from the trend in the single-, double-, and infinite-layer compounds at ambient pressure.

Note that beyond 30 GPa, T_c in the experimental report is largely uncertain, because only one report exists [13], where only the onset temperature for the superconductivity is available, and the zero resistivity was not reached at any temperature, implying the inhomogeneity in samples or possible nonuniformity of hydrostatic pressure. The origin of the T_c peak structure in Hg1223 under pressure is now understood from the scaling form (26): With increasing pressure, the SC order parameter F_{SC} is nearly constant up to 30 GPa and then decreases above 30 GPa, whereas the nearest-neighbor hopping parameter $|t_1|$ monotonically increases. To keep the plateau of F_{SC} , the reduction of off-site Coulomb repulsion plays an important role. Then, the pressure dependence of T_c exhibits a maximum in the intermediate pressure region near 30 GPa, as shown in Fig. 11, because of the interplay between F_{SC} and $|t_1|$.

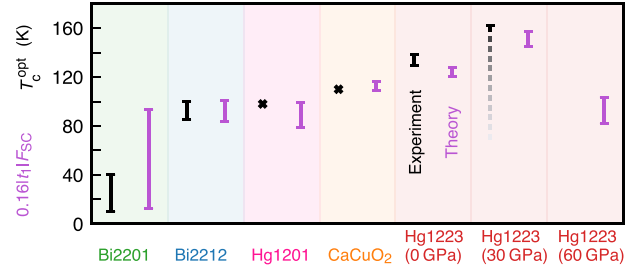


FIG. 12. Materials dependence of SC critical temperature T_c : Comparison between the present *ab initio* predictions and experimental observations for triple-layer Hg1223 at ambient and applied pressures is plotted together with single-, double-, and infinite-layer compounds [2]. The black bar in the left row for each case indicates experimental optimum T_c . Theoretical estimates are depicted in each right row based on the *ab initio* calculations with the help of Eq. (26) using calculated F_{SC} after the size extrapolation to the thermodynamic limit. The dashed line for Hg1223 (30 GPa) represents an ambiguity in experimental T_c (see Secs. IV B and VI for more details). No experimental value of T_c has been reported for Hg1223 at 60 GPa.

In Fig. 12, we show the comparison between the experimental T_c and the present estimates for CaCuO₂, Hg1201, Bi2201, Bi2212, and Hg1223 at ambient pressure as well as Hg1223 under pressure. The *ab initio* estimates essentially reproduce all the experimental indications. In particular, it captures the experimental trend of the higher T_c of Hg1223 in comparison to others.

V. EFFECTIVE ATTRACTION

To understand the mechanism of superconductivity, it is crucially important to uncover the origin of the effective attraction that causes the Cooper pairing. In this section, we report a detailed analysis obtained from the *ab initio* calculations, which should be relevant to both Hg1223 and other cuprate compounds.

In general, effective static interaction of particles can be estimated from the energy required to add a particle to the system. Namely, the effective interaction energy between the added particle and the particles already existing in the system is measured by the shift of the chemical potential from an N -particle system $\mu(N)$ to that of an $(N+1)$ -particle system $\mu(N+1)$; in other words, $\mu(N+1) - \mu(N) \sim \partial\mu/\partial n$. Since $\mu = \frac{\partial E}{\partial n}$, with n and E being the particle density and the energy density, respectively, the effective interaction is given by $\frac{1}{2} \frac{\partial^2 E}{\partial n^2}$. Partial interaction energy can also be defined by decomposing the total energy density E into the sum of partial energies by following the proposal in Ref. [2]. Here, by decomposing Eq. (1) into Eqs. (2)–(5), we focus on the local interaction, $\frac{1}{2} \frac{\partial^2 E_U}{\partial \delta^2}$, where $E_U = \langle \mathcal{H}_U \rangle$ denotes the ground-state average of the local interaction.

To estimate the effective interaction at $\delta = \delta_m$ practically, we fit the on-site Coulomb energy $E_U(\delta)$ by the quadratic function

$$E_U(\delta) = c_0 + c_1(\delta - \delta_m) + c_2(\delta - \delta_m)^2, \quad (27)$$

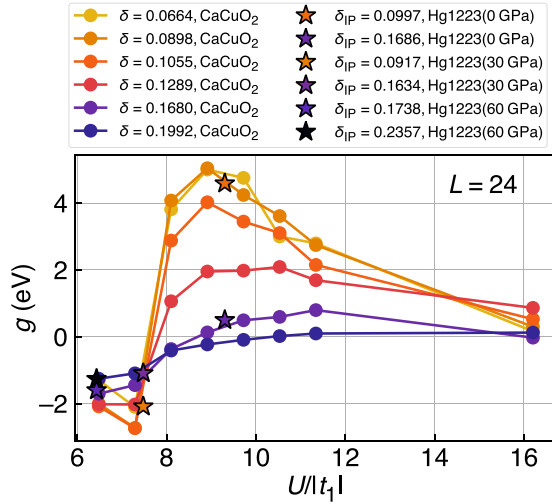


FIG. 13. Coulomb interaction dependence of the estimated attraction g at each doping. All g 's are extracted from data for $L = 24$. The symbol color distinguishes the doping concentration (δ). For Hg1223, we estimate g from the on-site Coulomb energy E_U of three contiguous doping concentrations in the IP, as shown in Fig. 25(b). The corresponding doping concentration δ_{IP} is given by the average of the three contiguous doping concentrations.

where c_0 , c_1 , and c_2 are the fitting parameters. The coefficient c_2 is the effective interaction, and we rewrite it in terms of the effective attraction $g \equiv -c_2$ hereafter. We typically consider E_U at five discrete and adjacent hole densities, say, $\delta_1 < \delta_2 < \delta_3 < \delta_4 < \delta_5$, and assign the quadratic fitting parameter c_2 around $\delta_m = \delta_3$ as the interaction strength.

We show $U/|t_1|$ dependence of the effective attraction g and the SC order parameter F_{SC} for a number of choices of δ in Figs. 13 and 14, respectively, for Hg1223 at 0, 30, and 60 GPa. In this analysis, E_U is further decomposed into the IP and OP contributions, and we choose the curvature of either

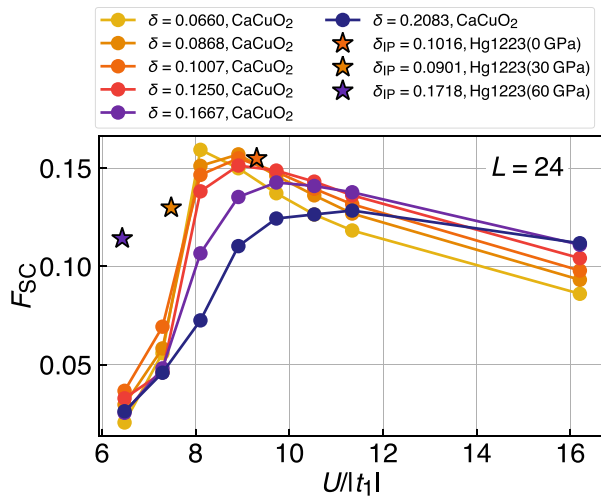


FIG. 14. Coulomb interaction dependence of the SC order parameter F_{SC} at each doping. For Hg1223, we show F_{SC} for a finite-size system ($L = 24$) at the optimal doping concentration ($\delta = 0.16$). The corresponding parameters, $U/|t_1|$ and δ_{IP} , are for the IP.

the IP or OP that shows the stronger SC order parameter. As references, we also show g and F_{SC} calculated for hypothetical Hamiltonians modified from that of the hole-doped CaCuO₂ only by changing the U value by hand from the *ab initio* value $U = 4.22$ eV ($U/|t_1| = 8.1$).

In the case of CaCuO₂ and its derivatives, the effective attraction g shown in Fig. 13 is enhanced in underdoped regions and is maximized at the interaction $U/|t_1| \sim 9$ in the realistic range $8 \leq U/|t_1| \leq 12$. The attraction is gradually reduced with increasing doping. The attraction g for the triple-layer cuprate, Hg1223, in overall trend, agrees well with that of CaCuO₂ at the corresponding values of $U/|t_1|$ and δ . This means that the effective attraction generated by the on-site repulsive Coulomb interaction emerges in the same way in all the single-, double-, triple-, and infinite-layer cuprates.

The SC order parameter F_{SC} is shown in Fig. 14 for the Hamiltonians for CaCuO₂, and its derivatives, which also shows the maxima at the interaction $U/|t_1| \sim 9$, just as in the case of g . Furthermore, the peak structure is asymmetric around the peak in both g and F_{SC} : The tail is steeper for the weak-coupling side $U/|t_1| < 9$, whereas it is broader for the strong-coupling side $U/|t_1| > 9$. The SC order parameter of the triple-layer cuprate, Hg1223, follows the trend of CaCuO₂, although F_{SC} is higher at higher pressure thanks to the reduced off-site Coulomb interaction in Hg1223, as is discussed in Sec. IV A. The overall similar behavior between g and F_{SC} in $U/|t_1|$ and δ dependencies with a prominent peak around $U/|t_1| \sim 9$ and asymmetric $U/|t_1|$ dependence around the peak strongly supports that the local attraction g is indeed the origin of the SC order.

However, there exist two important different trends between g and F_{SC} . (1) One is the hole density dependence: g increases strongly with decreasing hole density, while F_{SC} shows much smaller δ dependence. (2) The other is that F_{SC} shows a clear deviation for Hg1223 under pressure from the trend of modified CaCuO₂, namely, substantially enhanced F_{SC} for Hg1223 at 30 and 60 GPa than the modified CaCuO₂ is observed, as is already ascribed to the effect of off-site interaction in Sec. IV A. On the other hand, g does not show such an enhancement for Hg1223 under pressure. These discrepancies give us insights into the nature of superconductivity.

On point (1), we note that the attractive interaction induces the SC order, but they are not the same. In fact, it is known experimentally that the SC gap monotonically increases with decreasing hole concentration δ even in the underdoped region, while T_c has a dome structure and decreases to zero toward the Mott insulator [58]. This detachment has the same origin as the difference between g and F_{SC} . Namely, at least in the underdoped region, F_{SC} must decrease with the carrier density [59] and must vanish at the transition to the insulator, while g may remain nonzero. Therefore, F_{SC} is expected to be better represented by g multiplied by δ at least in the underdoped region. Based on this observation, we plot $(g + A)\delta$ in Fig. 15, where A is 2. The offset A is considered because even for noninteracting fermions, where the local interaction vanishes as well, $c_2 = -g$ must be positive because of the Pauli exclusion principle, and the origin of the local interaction should be located at a negative g , which makes the effective attraction $g + A$ with the positive offset A . The value of A can be inferred from the value of c_2 at small U . Figure 15 shows

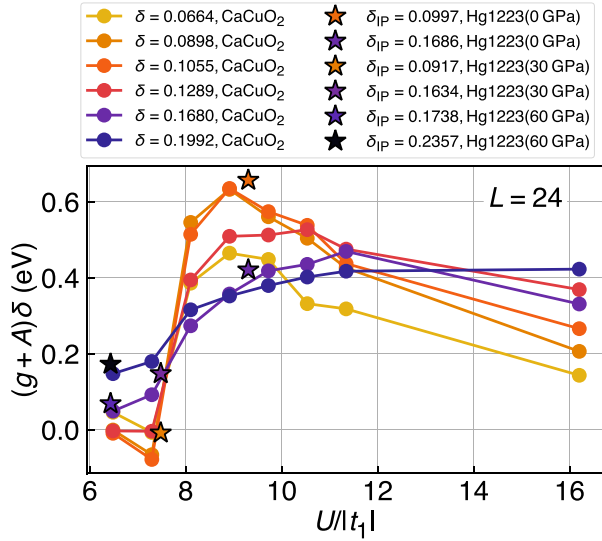


FIG. 15. Product of the attraction $g + A$ and doping δ as a function of the Coulomb interaction for $L = 24$. The constant A is chosen as 2.

a better correlation with F_{SC} than Fig. 13 on the point that the large enhancement in the undoped region is suppressed around the relevant region $U/|t_1| \sim 8-10$.

Let us discuss point (2). We have addressed that F_{SC} is dominantly determined by g at ambient pressure. However, it is likely that off-site effective interactions play a certain role, which may show up as the off-site interaction dependence of A ; namely, A is similar at ambient pressure because the off-site interactions are similar (for instance, $V_1 \sim 1$ eV throughout the cuprates), while A should increase substantially under pressure due to reduced off-site interaction consistently with the finding in Sec. IV A. This well resolves point (2). It implies that

$$F_{SC} \sim \frac{1}{8} \frac{g^*}{|t_1|} \delta \quad (28)$$

is roughly satisfied in the realistic region, where the effective total attraction $g^* = g + A$, after including the effect of off-site interaction in A , can generally be expressed by the dimensionless scaling function G as $g^* = |t_1| G(\frac{t_2}{|t_1|}, \frac{t_3}{|t_1|}, \dots, \frac{U}{|t_1|}, \frac{V_1}{|t_1|}, \dots)$. The calculated results indicate that G most dominantly depends on $U/|t_1|$ and secondarily on $V_1/|t_1|$ as F_{SC} .

The finding of the close correlation between F_{SC} and the local and instantaneous emergent attraction g looks to support an SC mechanism entirely different from the conventional one mediated by some bosonic glue, such as the dynamical spin or charge fluctuation scenarios. In fact, it introduces an unexplored perspective as we will discuss below. However, we also make a remark on the relation to the spin-fluctuation scenario. The spin fluctuation can be measured by the dynamical spin structure factor

$$S_s(\mathbf{q}, \omega) = \frac{1}{L^2} \int dt \sum_{i,j} \langle S_i(t=0) \cdot S_j(t) \rangle e^{i\mathbf{q} \cdot (\mathbf{r}_i - \mathbf{r}_j) + i\omega t}, \quad (29)$$

while the local Coulomb energy E_U can be rewritten by using the local moment $\langle m^2 \rangle = \langle S^2 \rangle = \frac{3}{4} \langle (n_{i,\uparrow} - n_{i,\downarrow})^2 \rangle = \frac{3}{4} \langle n_i \rangle - 2 \langle n_{i,\uparrow} n_{i,\downarrow} \rangle$ as

$$E_U = U \left[\frac{N_e}{2N_s} - \frac{2}{3} \int d\omega d\mathbf{q} S_s(\mathbf{q}, \omega) \right]. \quad (30)$$

Therefore, the local attraction measures the energy-momentum integrated amplitude of the dynamical spin correlation as

$$g = \frac{U}{3} \frac{\partial^2}{\partial \delta^2} \int d\omega d\mathbf{q} S_s(\mathbf{q}, \omega). \quad (31)$$

Since $S_s(\mathbf{q}, \omega)$ is extended not only to the so-called spin-fluctuation region $\mathbf{q} \sim (\pi, \pi)$ and paramagnon energy range or superexchange range $\omega < J \sim 4t_1^2/U$ but also to a much wider range in doped Mott insulators, it makes a very broad structure of $S_s(\mathbf{q}, \omega)$. In the weak-coupling SC materials, the attraction is limited to the characteristic energy range of glue around the Fermi surface, such as the Debye frequency for phonon-mediated SC, while in the present case, a much wider range of energy comparable even to the Mott gap is involved, where the picture of instantaneous attraction becomes relevant. In strongly correlated systems, it is often *not* appropriate to employ the analysis localized in momentum-energy space; rather, it is better to start from a spatially and temporally local picture. See Appendix G for further analyses of attraction.

VI. DISCUSSION

Here, we discuss why the local emergent attraction arises counterintuitively from the originally strong repulsion term E_U . The strong Coulomb repulsion suppresses the double occupation of electrons and induces the AF Mott insulating state at half filling. The Mott insulator is regarded as the “vacuum” of the charge carrier protected by the charge gap. Nevertheless, the averaged double occupation $D = \langle n_{i\uparrow} n_{i\downarrow} \rangle$ and the corresponding exciton density (the density of the bound doublon and holon) do not vanish unless U becomes infinite. Keeping in mind this common property of the Mott insulator, and the fact that the emergent attraction generated by the U term shows similar behavior among all the cuprates including Hg1223, we analyze an example of CaCuO₂ to elucidate the origin of the attraction.

We show in Fig. 16 $U/|t_1|$ dependence of E_U in the case of the modified CaCuO₂ Hamiltonians. There exist peaks around $U/|t_1| \sim 6.5$ with inflection points around $U/|t_1| \sim 8-9$. Even the simple Hubbard model in the metastable d -wave SC state shows a qualitatively common trend, as is shown in Fig. 26 in Appendix G. The nonzero E_U in the limit $\delta \rightarrow 0$ may be interpreted as the “vacuum-polarization” energy, and the Mott insulator at $\delta = 0$ is regarded as the “false vacuum,” particularly in the intermediate $U/|t_1|$ because of large quantum fluctuations.

This vacuum-polarization structure is in contrast to the band insulator, where the charge density does not fluctuate from 0 or 2 per site, depending on the unoccupied or occupied band, respectively, and the exciton density is strictly zero in the ground state. In the present case of the Mott insulator, the vacuum polarization (quantum fluctuation) generates a

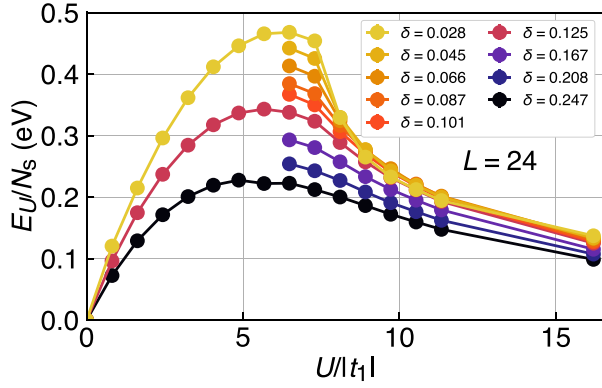


FIG. 16. On-site Coulomb interaction energy per site ($E_U/N_s = U/D$) as a function of $U/|t_1|$ for hole-doped CaCuO_2 at $L = 24$. The peak in E_U/N_s appears at $U/|t_1| \sim 6.5$ for any $\delta \in (0, 0.25)$.

nonzero density of doublon-holon pair (exciton) causing high zero-point energy of E_U .

The introduction of charge carriers induces the collapse of the Mott insulator (collapse of the gap) accompanied by the release of the zero-point energy by rapidly reduced doublon-holon pair density and hence E_U , as one sees in Fig. 16 most prominently in the intermediate $U/|t_1| \sim 5$ –9. This nonlinear reduction of the local energy from the “false vacuum” with negative curvature results in the relative local attraction playing the role of the origin of the Cooper pairing. This mechanism may be called “attraction by reduced repulsion.” In the cuprates, E_U peaks around $U/|t_1| \sim 6.5$, and the inflection point around $U/|t_1| \sim 8$ –9 in the Mott insulator is indeed the parameter region of the largest attraction $g + A$ and the strongest SC order parameter F_{SC} , as is illustrated in Figs. 14 and 15. The universality of this trend is discussed in Appendix G.

The d -wave SC state changes the sign of the order parameter across the nodal line in the Brillouin zone and prohibits the double occupation of electrons on the same site by symmetry. Therefore, it has a large energy gain if the order develops by reducing E_U , which largely contributes to stabilizing the d -wave SC state against AF, stripe, and Fermi liquid metal, because these competing states allow the double occupation. By the energy gain, the SC order grows, which is accelerated by the nonlinear reduction of E_U with the negative curvature of δ dependence generating emergent attraction synergetically in a self-consistent fashion.

A complementary view for the conversion from the repulsion to the attraction in the nodal d -wave SC state is known by considering the scattering process arising from the U term containing $U c_{k\uparrow}^\dagger c_{-k\downarrow}^\dagger c_{-q\downarrow} c_{q\uparrow}$, with $k \sim (\pi, 0)$ and $q \sim (0, \pi)$ [60]. This generates the momentum transfer (π, π) , which is enhanced if the AF fluctuation is large, while in the mean-field decoupling $\langle c_{k\uparrow}^\dagger c_{-k\downarrow}^\dagger \rangle \langle c_{-q\downarrow} c_{q\uparrow} \rangle$, the d -wave phase change between $(\pi, 0)$ and $(0, \pi)$ converts the repulsion $U > 0$ into attraction.

The tight connection between the emergence of attraction and the role of d -wave SC is clearly seen in Fig. 17, which shows a sharp contrast of the δ dependence of E_U between the normal metal and the SC state for CaCuO_2 . Only the d -wave SC state shows the negative curvature and thus the

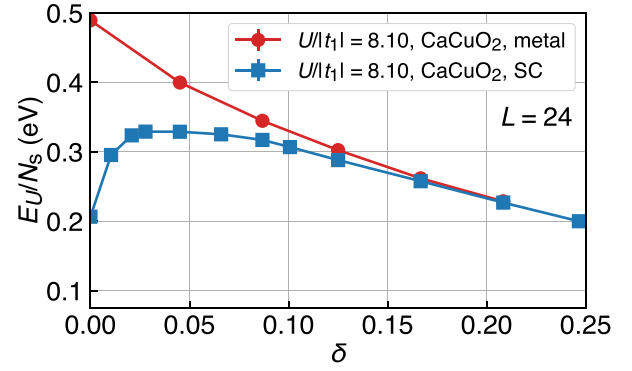


FIG. 17. Doping concentration (δ) dependence of the on-site Coulomb interaction energy E_U for metallic and SC states in CaCuO_2 with $L = 24$.

emergent attraction. Note that the normal metal compared here is different from the metal at temperatures above T_c with the pseudogap.

As shown in Figs. 11 and 12, the theoretically estimated T_c exhibits a peak at ~ 30 GPa upon increasing pressure, behaving consistently with the experimental T_c . On the other hand, the estimated T_c is slightly lower for stronger pressure. One of the possible origins is the inhomogeneity of applied pressure in the experiments, which unsettles the definition of the experimental T_c : For stronger pressure, the experimentally observed onset temperatures ($T_{c,\text{onset}}$) available so far are much higher than the other $T_{c,\text{mid}}$ determined by the inflection point of the temperature derivative of the resistivity.

Among a huge number of studies, let us pick up some work, which pays attention to the relation between T_c and materials parameters from general viewpoints on the cuprates, which might have relations to the present work. The importance of the apical oxygen position to control T_c was empirically pointed out in the cuprates in general in Ref. [61]. Also empirically, parameters of the crystal structure controlling T_c were discussed [62,63]. Several experimental studies on the cuprates reported the relation between T_c and the character of dopant atoms and disorder [64,65]. The optimum value for the highest T_c was discussed in the $U/|t_1|$ dependence [66,67]. Features of the crystal structure to optimize the SC were discussed with the help of machine learning [68]. Further studies on the relation between the above work and the present results await future analyses.

On the other hand, many studies pointed out relations between T_c and other physical quantities such as SC energy gaps and AF spin-fluctuation energies. For instance, the Uemura plot focused on the proportionality of T_c and the SC carrier density measured by the μ SR penetration depth in more comprehensive studies on the materials, not only on the cuprates [69]. This essentially shares the common viewpoint with Eq. (26). It was pointed out that the scaling $T_c \propto F_{\text{SC}}$ can be understood from the scaling of the Berezinskii-Kosterlitz-Thouless transition temperature in a weakly interacting 2D Bose gas, $T_{\text{BKT}} \propto n_b$ [70], by interpreting the superfluid density F_{SC} as the boson density n_b in the Bose-Einstein condensation (BEC) picture. However, it does not tell us how F_{SC} is determined theoretically, which the

present work has elucidated for the cuprates, especially with the pressure dependence of Hg1223 by the *ab initio* studies. In fact, Eq. (28) unveils the origin of the SC caused by the instantaneous, local, and emergent attraction. We note that F_{SC} is quantitatively consistent with the value reported in Ref. [71], as discussed in Ref. [2].

There exist several studies on the multilayer cuprates. Differentiation in roles of the IP and OP was theoretically emphasized in realizing the high T_c of Hg1223 [72,73]. While the difference in the hole density they presumed is qualitatively similar to the present result, and the interlayer differentiation plays a role through the proximity, the main origin of the higher T_c is found to be ascribed to high $U/|t_1|$ in the present work with quantitative agreement with the experiments.

A recent study [74] on Hg1223 at ambient pressure, based on the atomic orbital basis combined with the dynamical mean-field framework similarly to the choice of Ref. [75], instead of the bonding-antibonding basis of strongly hybridized $Cu3d_{x^2-y^2}$ and $O2p_\sigma$ orbitals employed in the present work, proposed a different view. Reference [74] obtained similar optimum SC order parameter at ambient pressure between Hg1201 and Hg1223 although the experimental optimum T_c is substantially different (~ 98 K for Hg1201 and ~ 134 K for Hg1223), while the present work correctly captures this experimental trend of difference.

The advantage of the present work solving the lattice Hamiltonian is that this approach is able to treat the spatial and temporal quantum fluctuations on equal footing, which also allows treating the severe competitions of different orders and fluctuations as well beyond mean-field approximations. From the present *ab initio* work, detailed microscopic origins of dependencies on materials, hole density, and pressure have been elucidated by one-to-one correspondence to the *ab initio* parameters of the effective Hamiltonians, which makes the microscopic origins and mechanisms of the materials dependence clear and intuitively transparent. It also helps in understanding the physical origin of the empirical relations suggested in the literature discussed in the last paragraphs.

We now briefly mention guiding principles for realizing higher T_c than the available materials along the line of the mechanism of the cuprate SC. Since the T_c formula [Eq. (26)] widely holds, the strategy to be employed is to design (1) a wider bandwidth (consequently larger $|t_1|$) and (2) larger F_{SC} simultaneously. We learned in the present study that F_{SC} becomes optimized at (2a) $U/|t_1| \sim 9$, while (2b) smaller $V_1/|t_1|$ enhances F_{SC} . In real materials, it is not easy to satisfy (1), (2a), and (2b) simultaneously, because, for instance, the hydrostatic pressure makes the bandwidth wider while $U/|t_1|$ and $V_1/|t_1|$ (and also $V_2/|t_1|$, $V_3/|t_1| \dots$) smaller, which have opposite effects on F_{SC} . A possible direction is to search for the materials that have relatively small V_1/U , V_2/U , ... within the range of stable SC against the stripe (note that very small V_i generally is subject to making the stripe state more stable) by keeping $U/|t_1| \sim 9$ and large t_1 . Alternatively, if the condition $U/|t_1| > 9$ is satisfied together with not high off-site interactions, pressure is expected to work cooperatively to raise T_c . Smaller V_1/U , V_2/U , ... and larger t_1 could be prepared simultaneously by pressure as the present Hg1223 study demonstrated. Another possibility is to design a surface or

interface with an insulating substrate that has a high dielectric constant to screen the off-site interaction.

VII. SUMMARY, CONCLUSIONS, AND OUTLOOK

We have elucidated why the triple-layer Hg1223 shows the highest T_c as compared to the single-, double-, and infinite-layer cuprates at ambient pressure and a further increase in T_c under pressure, together with the mechanism of SC. This has been achieved by examining the ground states of the *ab initio* low-energy effective Hamiltonians of Hg1223. To explore the pressure dependence of the superconductivity, we have investigated the systems under pressure ranging from 0 to 60 GPa. By using the VMC method, we have calculated the *d*-wave SC order parameter F_{SC} in the thermodynamic limit and estimated T_c by following formula (26) and have found the stable and uniform *d*-wave SC phase in the ground state in the hole concentration region $0.1 < \delta < 0.25$.

Hg1223 at ambient pressure is found to be in a slightly strong-coupling region above the optimal value of $U/|t_1| \sim 9$, which is the primary reason for high T_c in comparison to single-, double-, and infinite-layer compounds studied in Ref. [2], where a high $U/|t_1|$ value (because of large U) originates from poorer screening. The pressure drives it into the weak-coupling region over the optimum $U/|t_1|$, which contributes to keeping F_{SC} in the plateau region around the peak at weak pressure $P < 30$ GPa, while it works to sharply decrease F_{SC} at $P > 30$ GPa. However, it is partially compensated by the largely decreasing off-site interaction, which works to enhance F_{SC} . Then, this interplay eventually makes a broad peak structure in the pressure dependence of F_{SC} .

As for T_c , the pressure makes the increase in $|t_1|$, which contributes to enhancing T_c . In addition, the pressure makes a reduction of $U/|t_1|$ as well, contributing to weak enhancement of T_c cooperatively in the region of $U/|t_1| > 9$ at low pressures < 30 GPa. On the other hand, higher pressure above 30 GPa corresponding to decreasing $U/|t_1|$ below 8 causes a substantial reduction of T_c due to the decay in F_{SC} , which becomes the dominant factor that controls T_c . This interplay is perfectly consistent with the experimental dome structure. It demonstrates that one effective route to enhance the superconductivity is to reduce the off-site interaction within the moderate and not too weak range to avoid the stripe phases. The determination of the boundary between the SC and stripe phases requires careful analysis in the case of weaker off-site interaction and will be left for a subject of future work.

The T_c formula at the optimal carrier density, $T_c \sim 0.16|t_1|F_{SC}$ established in the single-, double-, and infinite-layer cuprates [2] is shown to hold also in the triple-layer cuprates and to reproduce the essential feature of the pressure dependence, namely, the peak around 30 GPa.

We have also found the tendency that, in the underdoped region ($\delta \lesssim 0.1$), the filling in the IP is much closer to half filling than that in the OP. This stabilizes the AF order in the IP, simultaneously retaining a weak SC order as the proximity from the OP SC and enables the coexistence of the SC and AF.

The effective instantaneous attraction for Hg1223 that induces the Cooper pair is found and its origin is identified as the mechanism of "attraction by reduced repulsion" emerging

in the originally strongly repulsive on-site interaction term, which is common to the single-, double-, and infinite-layer cuprates. This mechanism is very general, as is also observed in the SC phase of the Hubbard model. The Cooper pair formed by the instantaneous attraction is in contrast with the weak-coupling BCS SC caused by the retarded interaction with a large coherence length. The SC at the optimum doping of the cuprates does not belong to the BEC regime either, because the attraction is relatively weaker than the kinetic energy scale (namely, effective Fermi energy expected from a large Fermi surface) and the preformed pair is not well developed above T_c . In fact, F_{SC} and T_c at the optimum doping rather increase with the increased attraction in the realistic cuprates as Eq. (28) suggests, instead of the reduction expected in the typical BEC regime.

In the underdoped region toward the Mott insulator, Eq. (26) shares similarity with the Berezinskii-Kosterlitz-Thouless (BKT) transition of a weakly interacting 2D Bose gas in the proportionality of T_c to the superfluid density F_{SC} , which is interpreted as the “boson density.” However, Eq. (28) indicates that the “boson density” is controlled in a nontrivial fashion by the emergent attractive interaction and the hole density in the present case. Despite the high electron density in the underdoped region, at which one can naively expect a large Fermi surface in metals, the cuprates do not show a diverging electron effective mass by keeping the high density of coherent electrons as a route to the Mott insulator. Instead, the incoherent part becomes dominating toward the Mott insulator, and the effective “boson” emerges as Eq. (28), if one can ignore the randomness and inhomogeneity in real materials.

A stringent test of the emergence of instantaneous attraction is to experimentally measure Eq. (31), for instance, by neutron scattering. Energy-momentum-resolved data will unveil the dynamical origin of the attraction.

The emergent attraction may cause the fractionalization of electrons [2,76], which is now supported by various experimental and theoretical studies [76–82]. To reach a further complete understanding of the superconductivity, it is desired to uncover the nature of the fractionalized hidden fermion splintered from the original electron and examine whether the weakly bound exciton proposed in the literature [59,76] is involved in the fractionalization.

There exist other strong-coupling unconventional SCs such as the iron-based SCs [83] and nickel oxide SCs [84,85]. It is an intriguing future subject to clarify whether these families have the common aspect to the cuprates along the line we reported in this paper. The relations to heavy-electron SC in f -electron systems and $4d$ -electron SC are also intriguing in a more general perspective of the fractionalization. Applying the present framework to other trilayer cuprates, such as Bi2223 [86] and compounds containing even more layers, would also be intriguing to examine the broader applicability of the proposed SC mechanism, which is left for future studies. Although their experimentally observed T_c values are lower and their treatment is beyond the scope of the present work because of the higher computational cost, multilayer compounds with four or more layers would also be interesting future targets.

In terms of the materials design strategy for higher T_c , one can propose to seek materials that allow “attraction by reduced

repulsion” from the “false vacuum,” as well as tuning toward optimum on-site repulsion, weaker intersite interaction, and larger bandwidth. Our findings will stimulate research on designing and optimizing SCs toward a higher SC critical temperature by explicitly utilizing the mechanism of the emergence of the attraction from the repulsion.

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DATA AVAILABILITY

The data that support the findings of this article are available upon request.

APPENDIX A: *AB INITIO* EFFECTIVE HAMILTONIANS UNDER PRESSURE

We show the parameters for the *ab initio* low-energy effective Hamiltonian of Hg1223 at 30 and 60 GPa in Tables II–IV. As in the case at ambient pressure, we use the hopping and interaction parameters that extend over long distances satisfying $r \leq 3\sqrt{2}$. In Table V, we list the core parameters t_1 , U , and V_1 of Hg1223, and compare them with already studied four hole-doped cuprate compounds CaCuO₂, Bi2201, Bi2212, and Hg1201 at ambient pressure and at optimum doping.

The on-site effective Coulomb interaction for the present single-band Hamiltonian has been improved by taking into account the energy-level correction between the $Cu d_{x^2-y^2}$ and $O 2p_\sigma$ atomic orbitals in the downfolding procedure to satisfy the charge consistency, namely, LRFB as proposed in Refs. [20,26]. By this LRFB, the strength of the on-site Coulomb interaction is commonly rescaled by 0.91–0.97 [19] and the rescaling of other Hamiltonian parameters is small, as was elucidated in Ref. [19]. In our VMC calculation, we use the effective Hamiltonian with this modified on-site Coulomb interaction. The original on-site Coulomb interactions U_{IP} and U_{OP} are multiplied by 0.95. In this article, the symbol U is used for the on-site Coulomb interaction after the LRFB correction. Tables II–IV contain the on-site Coulomb interactions after the correction by the LRFB.

APPENDIX B: ESTIMATION OF CHEMICAL POTENTIAL DIFFERENCE BETWEEN THE IP AND OPS

The carrier densities in the IP and OPs during the VMC calculations should be adjusted to reproduce those obtained

TABLE II. Parameters for the *ab initio* low-energy effective Hamiltonian of Hg1223 at 0 GPa. We show the hopping parameter t_l (including the chemical potential μ) and the Coulomb interaction V_l (including the on-site one U) within the same IP (IP-IP), those within the same OP (OP-OP), those between the IP and OP (IP-OP), and those between the different OPs (OP-OP') up to a distance $r = 2\sqrt{2}$. The chemical potential difference between the IP and the OP is given by μ , corresponding to the parametrizations, $\mu_{\text{IP}} = t_{0,\text{IP-IP}} = 0$ and $\mu_{\text{OP}} = t_{0,\text{OP-OP}} = \mu$ in Eq. (5). The hopping parameters t_0 with IP-OP and OP-OP' are on-site parameters connecting the different planes. The interaction parameters V_0 with IP-IP and OP-OP are the on-site Coulomb interaction parameters U_{IP} and U_{OP} , respectively. These Coulomb interactions in this table are the values after the LRFB correction [20,26]. The interaction parameters V_0 with IP-OP and OP-OP' are on-site Coulomb interaction parameters connecting the different planes. All values are given in eV.

index l of hopping t_l	distance r	IP-IP	OP-OP	IP-OP	OP-OP'
0	0	0	μ	-0.0619	+0.0219
1	1	-0.4857	-0.4849	+0.0067	-0.0057
2	$\sqrt{2}$	+0.1326	+0.1300	+0.0107	+0.0019
3	2	-0.0503	-0.0444	-0.0042	-0.0012
4	$\sqrt{5}$	+0.0036	+0.0004	+0.0001	-0.0018
5	$2\sqrt{2}$	+0.0024	+0.0050	+0.0014	+0.0027
6	3	-0.0108	-0.0113	-0.0005	-0.0015
7	$\sqrt{10}$	-0.0023	-0.0003	+0.0003	-0.0005
8	$\sqrt{13}$	+0.0017	+0.0025	-0.0011	+0.0016
9	4	-0.0039	-0.0039	-0.0017	+0.0016
10	$\sqrt{17}$	+0.0003	+0.0007	+0.0003	+0.0007
11	$3\sqrt{2}$	+0.0049	+0.0014	+0.0000	-0.0002
index l of interaction V_l	distance r	IP-IP	OP-OP	IP-OP	OP-OP'
0	0	4.5184	4.5443	0.7987	0.3953
1	1	0.9878	0.9785	0.5621	0.3392
2	$\sqrt{2}$	0.5631	0.5579	0.4375	0.2999
3	2	0.3901	0.3854	0.3318	0.2500
4	$\sqrt{5}$	0.3287	0.3257	0.2930	0.2315
5	$2\sqrt{2}$	0.2495	0.2485	0.2317	0.1943
6	3	0.2448	0.2437	0.2293	0.1938
7	$\sqrt{10}$	0.2299	0.2291	0.2163	0.1847
8	$\sqrt{13}$	0.1994	0.1991	0.1894	0.1661
9	4	0.1054	0.1054	0.1006	0.0879
10	$\sqrt{17}$	0.1012	0.1012	0.0967	0.0848
11	$3\sqrt{2}$	0.1740	0.1741	0.1678	0.1494

by GW calculations along the same idea employed for multi-orbital systems as was established in the procedure of LRFB [26]. This can be achieved by controlling the chemical potential difference between the IP and OP under the condition of a fixed total number of electrons for each system size in the canonical ensemble. Namely, this adjustment is accomplished by performing the VMC calculation of the *ab initio* effective Hamiltonian (1) by shifting the chemical potential difference μ until the inner and outer carrier densities meet the GW result, for instance, at $\delta = 0$ and 0.2 [87]. When the GW result is not available, we estimated it from the interpolation of the nearby available results of generalized gradient

TABLE III. Parameters for the *ab initio* low-energy effective Hamiltonian of Hg1223 at 30 GPa. The notations of the parameters are the same as in Table II.

index l of hopping t_l	distance r	IP-IP	OP-OP	IP-OP	OP-OP'
0	0	0	μ	-0.0678	+0.0021
1	1	-0.5833	-0.5554	+0.0031	-0.0019
2	$\sqrt{2}$	+0.1425	+0.1373	+0.0149	-0.0011
3	2	-0.0523	-0.0528	-0.0033	+0.0039
4	$\sqrt{5}$	+0.0038	+0.0066	-0.0019	-0.0016
5	$2\sqrt{2}$	+0.0023	+0.0009	+0.0033	+0.0012
6	3	-0.0051	-0.0080	-0.0023	+0.0003
7	$\sqrt{10}$	-0.0003	-0.0028	+0.0003	-0.0014
8	$\sqrt{13}$	-0.0016	+0.0013	+0.0002	+0.0013
9	4	-0.0010	-0.0006	-0.0018	-0.0010
10	$\sqrt{17}$	+0.0009	-0.0024	+0.0008	+0.0018
11	$3\sqrt{2}$	+0.0015	+0.0014	+0.0010	-0.0001
index l of interaction V_l	distance r	IP-IP	OP-OP	IP-OP	OP-OP'
0	0	4.3615	4.4986	0.7573	0.3696
1	1	0.8752	0.8056	0.4939	0.2957
2	$\sqrt{2}$	0.4683	0.4430	0.3651	0.2502
3	2	0.3115	0.3014	0.2663	0.2045
4	$\sqrt{5}$	0.2573	0.2574	0.2327	0.1850
5	$2\sqrt{2}$	0.1917	0.1959	0.1806	0.1534
6	3	0.1860	0.1912	0.1778	0.1519
7	$\sqrt{10}$	0.1750	0.1801	0.1674	0.1449
8	$\sqrt{13}$	0.1498	0.1544	0.1452	0.1290
9	4	0.0790	0.0818	0.0772	0.0683
10	$\sqrt{17}$	0.0763	0.0789	0.0741	0.0659
11	$3\sqrt{2}$	0.1298	0.1342	0.1270	0.1153

approximation (GGA) combined with its correction estimated from the difference between the GGA and GW charge densities. The carrier concentrations of the IP and OP thus obtained are shown in Figs. 2 and 7 and the corresponding chemical potential difference μ is given by Eqs. (6)–(8). The physical origin of positive μ seems to come from the larger positive charge surrounding the OP than the inner plane caused by the ions including the block layer, which is not taken into account by the VMC calculations. It requires the positive μ correction. Under pressure, a closer distance between the IP and OPs makes their chemical potential difference weaker, as is expected.

APPENDIX C: PROCEDURE OF ENERGY-VARIANCE EXTRAPOLATION OF THE ENERGY

In strongly correlated electron systems, the ground-state candidate states described by the variational wave functions often energetically compete with one another. To fairly determine the true ground state, it is desirable to estimate the eigenenergy of each state by extrapolating the energy to the zero-variance limit. The variance $(\delta E)^2 = (\langle H^2 \rangle - \langle H \rangle^2) / \langle H \rangle^2$ can be systematically reduced by improving the quality of the wave functions. For this purpose, one can increase the number of variational parameters, such as those

TABLE IV. Parameters for the *ab initio* low-energy effective Hamiltonian of Hg1223 at 60 GPa. The notations of the parameters are the same as in Table II.

index l of hopping t_l	distance r	IP-IP	OP-OP	IP-OP	OP-OP'
0	0	0	μ	-0.0739	+0.0194
1	1	-0.6492	-0.5812	+0.0024	-0.0049
2	$\sqrt{2}$	+0.1509	+0.1274	+0.0141	+0.0004
3	2	-0.0578	-0.0406	-0.0060	+0.0019
4	$\sqrt{5}$	+0.0013	+0.0018	-0.0028	-0.0016
5	$2\sqrt{2}$	+0.0028	-0.0004	+0.0037	+0.0020
6	3	-0.0069	-0.0096	+0.0003	+0.0028
7	$\sqrt{10}$	+0.0009	-0.0014	-0.0007	+0.0011
8	$\sqrt{13}$	-0.0007	+0.0019	+0.0007	-0.0008
9	4	-0.0034	+0.0002	-0.0019	-0.0024
10	$\sqrt{17}$	+0.0010	-0.0001	-0.0000	-0.0004
11	$3\sqrt{2}$	+0.0005	+0.0015	+0.0002	-0.0024

index l of interaction V_l	distance r	IP-IP	OP-OP	IP-OP	OP-OP'
0	0	4.1751	4.2381	0.6922	0.3058
1	1	0.6842	0.5832	0.3593	0.2068
2	$\sqrt{2}$	0.3156	0.2878	0.2393	0.1566
3	2	0.1859	0.1743	0.1540	0.1110
4	$\sqrt{5}$	0.1419	0.1429	0.1267	0.0967
5	$2\sqrt{2}$	0.0908	0.0970	0.0850	0.0682
6	3	0.0870	0.0921	0.0826	0.0675
7	$\sqrt{10}$	0.0783	0.0840	0.0747	0.0613
8	$\sqrt{13}$	0.0598	0.0655	0.0581	0.0488
9	4	0.0331	0.0359	0.0321	0.0275
10	$\sqrt{17}$	0.0309	0.0337	0.0298	0.0256
11	$3\sqrt{2}$	0.0453	0.0505	0.0444	0.0376

in the RBM wave functions, and apply the power-Lanczos method to the optimized wave functions. The variance usually gets smaller as we improve the wave functions through this prescribed order. When the variational state is sufficiently close to the ground state, the energy difference between the ground state and the variational wave function becomes

TABLE V. Selected important parameters for the *ab initio* low-energy effective Hamiltonians of the IP of Hg1223 at 0, 30, and 60 GPa and those of CaCuO₂, HgBa₂CuO₄ (abbreviated as Hg1201), Bi₂Sr₂CaCu₂O₈ (Bi2212), and Bi₂Sr₂CuO₆ (Bi2201) at ambient pressure. We show the nearest-neighbor hopping parameter t_1 , the on-site Coulomb interaction U , and the nearest-neighbor off-site Coulomb interaction V_1 . All values are given in eV.

	t_1	U	V_1
Hg1223, IP, 0 GPa	-0.4857	4.5184	0.9878
Hg1223, IP, 30 GPa	-0.5833	4.3615	0.8752
Hg1223, IP, 60 GPa	-0.6492	4.1751	0.6842
CaCuO ₂	-0.521	4.221	0.969
Hg1201	-0.544	3.999	1.002
Bi2212	-0.452	4.226	0.915
Bi2201	-0.527	4.393	1.030

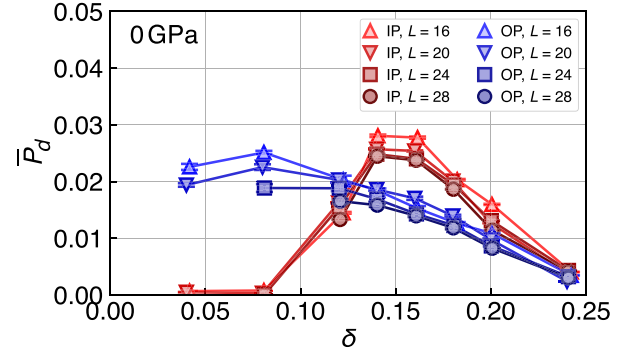


FIG. 18. Doping concentration dependence of the SC correlation averaged over long distances at 0 GPa. We observe dome structures both in the IP and OPs.

proportional to the energy variance [44]. The ground state is identified as the candidate state that achieves the lowest zero-variance energy after the linear extrapolation. This energy-variance extrapolation was shown to be useful and possess a wide versatility, as discussed in prior research [46]. Furthermore, the proportionality constant was shown to be nearly universal when the energy origin is set at the infinite temperature irrespective of the choices of the model Hamiltonian and the lattice structure [46]. Given its high degree of general applicability, this approach has been applied to various complex systems. For instance, the method has been applied to determine the ground states among competing stripe states of different periods, as well as SC and AF states, in various Hubbard-like models and *ab initio* Hamiltonians and demonstrated its usefulness [2,51,52]. For an example of the variance extrapolation, see Appendix F.

APPENDIX D: DETAILS OF THE SC CORRELATION

We show the doping concentration dependencies of the SC correlation $\bar{P}_d$ averaged over long distances in Figs. 18–20 with size dependence for calculated finite-size lattices. The characteristics of the dome structures between the IP and OP vary as a function of pressure.

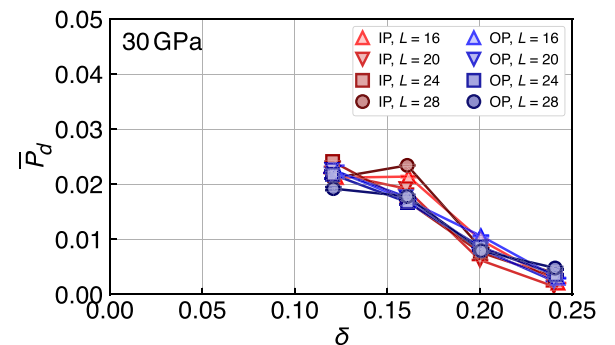


FIG. 19. Doping concentration dependence of the SC correlation averaged over long distances at 30 GPa.

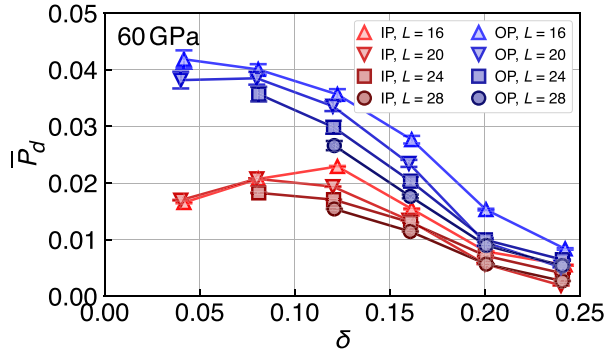


FIG. 20. Doping concentration dependence of the SC correlation averaged over long distances at 60 GPa.

APPENDIX E: PROCEDURE OF SIZE EXTRAPOLATION FOR THE SC CORRELATION FUNCTION

By using the SC correlations $\bar{P}_d$ averaged over long distances obtained in Appendix D, we extrapolate them to the thermodynamic limit. We summarize this size extrapolation in Figs. 21–23. Because the d -wave SC order possesses a gapless linear excitation, the size dependence of the SC correlation should be proportional to $1/L$, with L being the length of the lattice. All the data points are well fitted by this scaling formula. The SC order parameter F_{SC} in the thermodynamic limit is obtained from the square root as $F_{SC} = \sqrt{\bar{P}_d}$.

APPENDIX F: ANALYSIS OF COEXISTING AF AND SC ORDERS

Here, we show a realistic example of the coexistence. Since a state with the coexisting AF and SC orders is severely competing with the purely SC state, we show the variance extrapolation of energies discussed in Appendix C for the ground-state candidate states in Fig. 24. The data are for

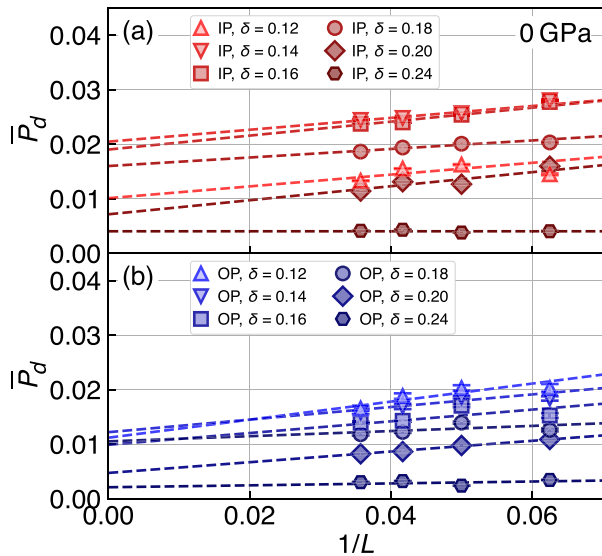


FIG. 21. Size extrapolation of the SC correlation averaged over long distances at 0 GPa. We show the extrapolation for the (a) IP and (b) OPs.

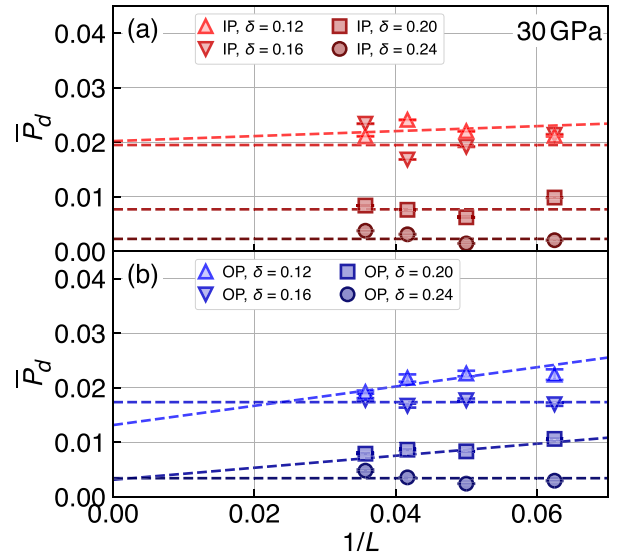


FIG. 22. Size extrapolation of the SC correlation averaged over long distances at 30 GPa. We show the extrapolation for the (a) IP and (b) OPs.

the system size $L = 16$ at 8% doping, with the chemical potential difference between the IP and OP being $\mu = 3.2$, which is smaller than the value corresponding to the *ab initio* Hamiltonian with the GW-based estimate of the LRFB. Here, the chemical potential shift μ is adjusted by hand so that the hole doping concentration in the IP becomes nonzero ($\delta_{IP} = 0.07$ in the present case) and should be regarded as a parameter search to explore the possibility of the coexistence in a physically relevant doped system. The IP is closer to half filling than the OP in this parameter region, and thus, the AF correlation is more developed in the IP. Ground-state candidate states include the state exhibiting SC order in both IP and OP, the state exhibiting AF order in both IP and OP,

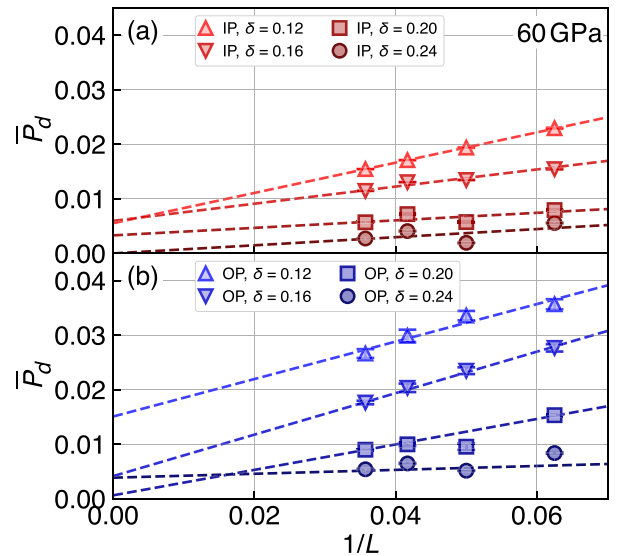


FIG. 23. Size extrapolation of the SC correlation averaged over long distances at 60 GPa. We show the extrapolation for the (a) IP and (b) OPs.

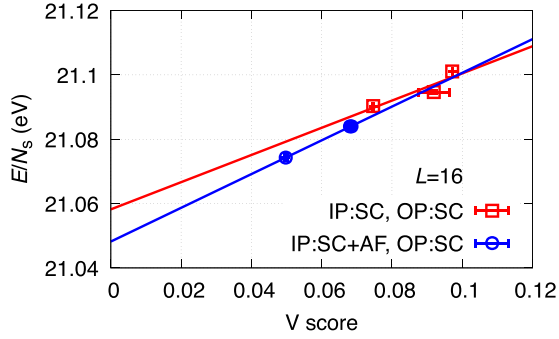


FIG. 24. Variance extrapolation of energies of ground-state candidate states. The abscissa is chosen as the V score [46], which is defined by $N_s(\langle H^2 \rangle - \langle H \rangle^2) / (\langle H \rangle - E_\infty)^2$, with E_∞ being the infinite-temperature energy. The states are obtained for $L = 16$ at 8% doping with the chemical potential difference $\mu = 3.2$. We show the energy of the pair-product wave function, that of the RBM wave function with $\alpha = 8$, and that after the one-step Lanczos method, which are aligned in the plots from the right to left in this order. At $\mu = 3.2$, the energy of the state exhibiting coexistence of SC and AF orders in the IP is lower than that of the state with inner-layer SC order. Note that the slope of the variance extrapolation is universal regardless of the details of the Hamiltonian, and the slope on a logarithmic scale in the present system [e.g., for the SC + AF state of the pair-product wave function, $\ln \frac{\langle H \rangle - E_0}{E_\infty - E_0} - \ln(\text{V score}) \sim \ln(0.0269) - \ln(0.0685) \sim -0.935$, with $\langle H \rangle = 16192.51$, $E_\infty = 17186.87$, and the extrapolated energy being $E_0 = 16165.02$] falls within the range of the slope fluctuations reported in previous studies [46].

and the state exhibiting SC order in the OP, but coexisting SC and AF orders in the IP. Although the variance extrapolation in Fig. 24 shows some uncertainty, the similar slopes of the two competing states support the stability of the extrapolation, and the ground state exhibits coexisting SC and AF orders over a certain parameter range.

APPENDIX G: UNIVERSALITY OF ATTRACTION

Figure 25 shows the doping concentration (δ) dependence of the on-site Coulomb interaction energy E_U per site as a function of $U/|t_1|$ for CaCuO_2 [panel (a)] and Hg_{1223} [panel (b)]. For Hg_{1223} , the doping concentration dependencies are presented separately for the IP and OP. Below $U/|t_1| = 8$, the on-site Coulomb interaction energy for CaCuO_2 exhibits the concave upward δ dependence with the positive curvature, corresponding to nearly vanishing effective attraction and weak SC order. Above $U/|t_1| = 8$, the δ dependence of E_U turns concave downward, inducing the effective attraction that may stabilize superconductivity. When $U/|t_1|$ becomes very large ($U/|t_1| \gtrsim 16$), the on-site Coulomb interaction energy decays almost linearly in δ , suppressing the effective attraction.

The δ dependence of E_U in Hg_{1223} follows the same trend as that of CaCuO_2 . Near the optimal doping (16%), T_c is dominated by the SC order in IP. As the pressure increases, the parameter $U/|t_1|$ in IP decreases from 9.30 to 6.43 (see Tables II–IV). Reflecting this reduction in $U/|t_1|$, the δ_{IP} dependence of E_U of IP, as shown in Fig. 25(b), changes from

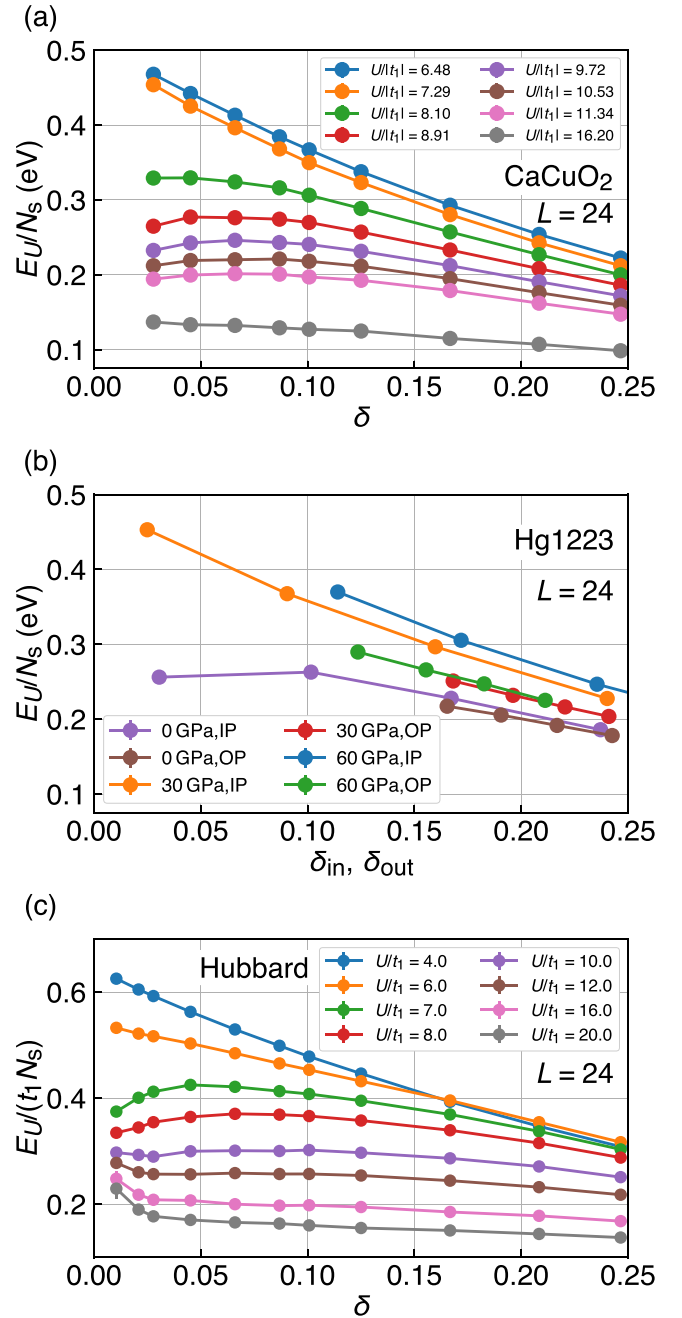


FIG. 25. Doping concentration (δ) dependence of the on-site Coulomb interaction energy E_U as a function of $U/|t_1|$ for CaCuO_2 [panel (a)], Hg_{1223} [panel (b)], and the simple Hubbard model [panel (c)] with the size $L = 24$. The energy E_U commonly shows the concave upward δ dependence for weaker $U/|t_1|$, whereas it exhibits the concave downward δ dependence indicating attractive interaction for stronger $U/|t_1|$. In the case of the simple Hubbard model, we also observe the concave upward δ dependence (namely, repulsive interaction) for very large U in the underdoped region.

concave downward to concave upward as the pressure increases. This observation suggests a reduction in the effective attraction due to pressure, consistent with the decrease in the SC order at high pressure (see Fig. 9).

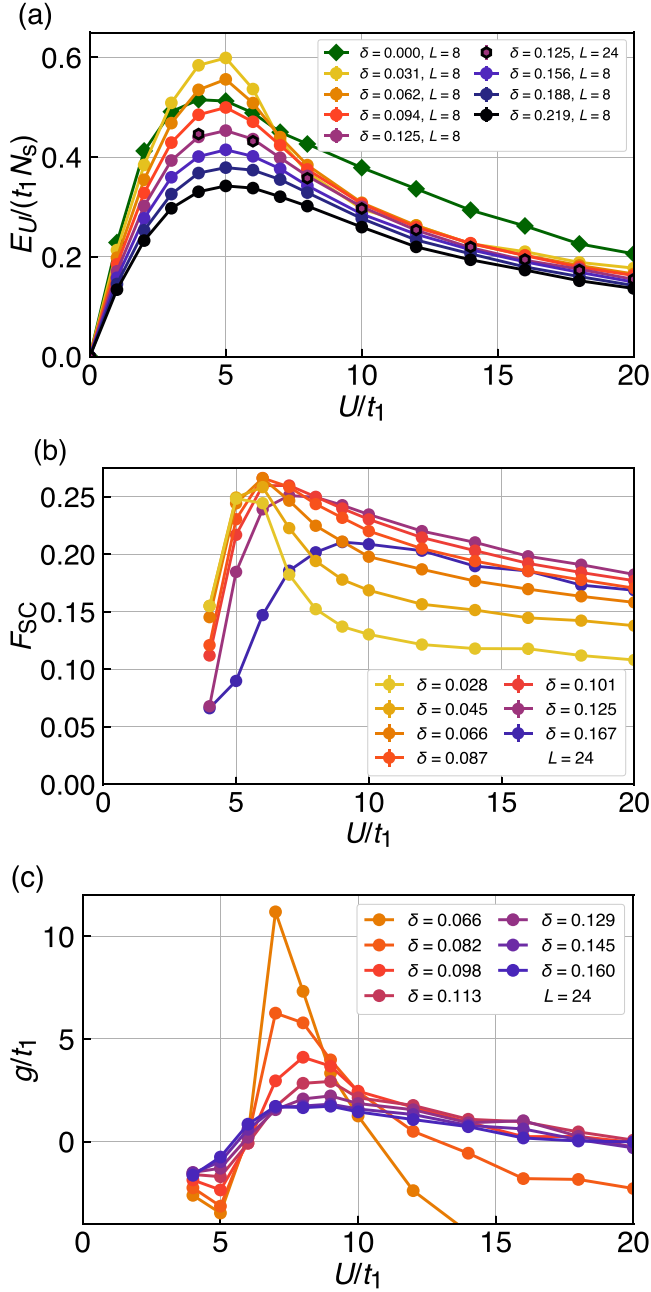


FIG. 26. On-site interaction (U) dependence of the on-site Coulomb interaction energy E_U [panel (a)], the SC order parameter F_{SC} [panel (b)], and the estimated attraction g [panel (c)] at various choices of δ for the simple Hubbard model on a square lattice. The data of the AF states are shown at half filling ($\delta = 0$), whereas those of the SC states are presented for nonzero doping concentrations ($\delta > 0$). The on-site Coulomb interaction energies, E_U 's per site, are nearly the same between system sizes $L = 8$ and $L = 24$ and they are close to the values in the thermodynamic limit. For F_{SC} and g , we plot selected data for $L = 24$. All the physical quantities exhibit a peak at $U \sim 5$ – 8 , which gradually increases upon doping for F_{SC} . The peak intensities of both F_{SC} and g diminish with increasing the doping concentration.

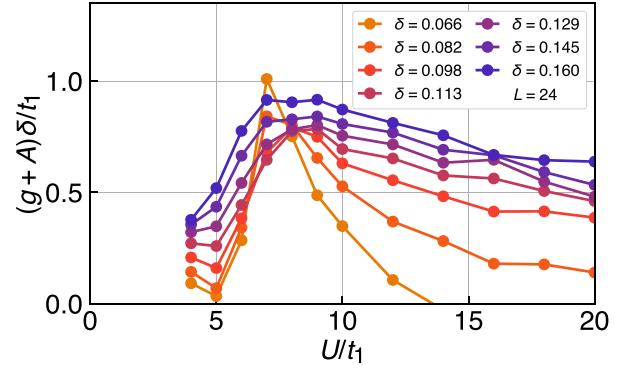


FIG. 27. On-site interaction (U) dependence of the product of the effective attraction g and the doping concentration δ for the simple Hubbard model on a square lattice with the size $L = 24$. Here, we choose the constant shift of g as $A = 4$, which corresponds to the minimum value of g for a weak interaction region, as shown in Fig. 26(c). The quantity $(g + A)\delta$ mostly reproduces the interaction dependence of F_{SC} in Fig. 26(b). For an underdoped region, the effective attraction from the on-site Coulomb interaction is relatively weaker under strong interaction, and instead, the effective attraction driven by the kinetic energy plays an important role, as shown in Fig. 28.

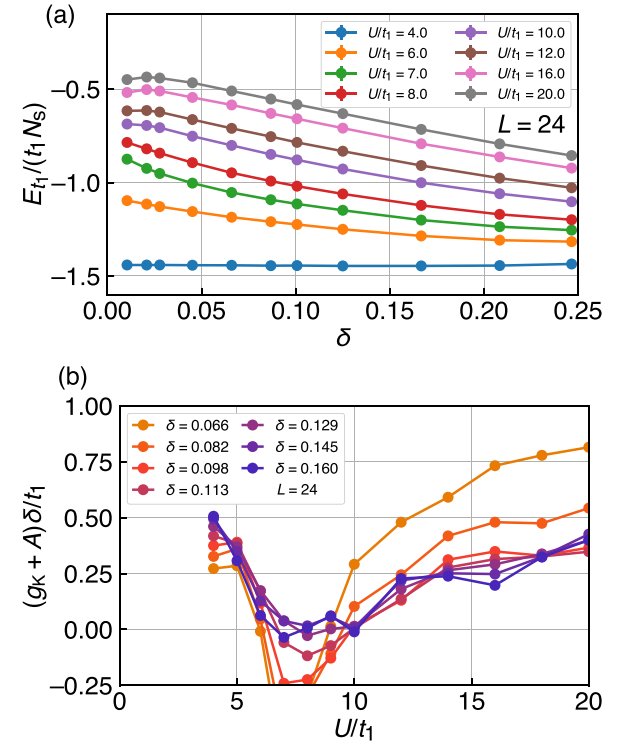


FIG. 28. Doping concentration (δ) dependence of the kinetic energy E_{t_1} as a function of U for the simple Hubbard model [panel (a)] with the size $L = 24$. On-site interaction, U dependence of the product of the doping concentration δ and the effective attraction g_K extracted from E_{t_1} [panel (b)]. Here, we choose the constant shift of g_K as $A = 4$ as in Fig. 27. The effective attraction driven by the kinetic energy emerges in an underdoped region when the Coulomb interaction is very strong.

To examine the universality of the pairing mechanism ascribed to the emergent local attraction generated by E_U , we have also calculated the local attraction g and F_{SC} for the simple Hubbard model, which has only the nearest-neighbor hopping t_1 and the local repulsion U in the metastable uniform d -wave SC state. (Note that the ground state has the charge inhomogeneous stripe order, and here, we focus on a metastable excited state that shows the SC order.) For the case of the simple Hubbard model, we take the energy unit by t_1 .

The similar trend in the δ dependence of E_U already appears in the simple Hubbard model without any further neighbor Coulomb interaction and hopping parameters [see Fig. 25(c)]. Because the *ab initio* Hamiltonians include substantial off-site Coulomb interaction parameters, the screening effects push the on-site Coulomb interaction that maximizes SC order to a stronger value than the Hubbard model. Indeed, as shown in Fig. 26, the Hubbard model exhibits the SC-order peaks at $U/|t_1| \sim 6$ –8, whereas the cuprate Hamiltonians display the SC-order peaks at $U/|t_1| \sim 8$ –10. Consistent with the interaction dependence of the SC order, the estimated effective attraction in the simple Hubbard model also peaks at $U/|t_1| \sim 7$ –8, which is slightly weaker than the peak interaction for the *ab initio* Hamiltonians. The peak of the SC order around $U/|t_1| \sim 6$ –8 is in overall agreement with earlier studies of the Hubbard model [67,88]. The asymmetric

peak structures for both F_{SC} and g in Fig. 26 with a long tail at large U are similar to the case of the cuprates in Figs. 13 and 14. The interaction that maximizes F_{SC} gradually increases in strength upon doping, in addition to the reduction of the peak value. This trend is also similar to the cuprates demonstrated in Fig. 14 and can commonly be understood from the screening by the doped carrier, which diminishes effective U . The core mechanism of this attraction in the cuprates is, therefore, likely to be already contained in the simple Hubbard model.

Note that the effective attraction also emerges in the kinetic energy in the strong-interaction limit, as was suggested in a previous study [88]. To examine this, we also investigate the energy of the SC state in the simple Hubbard model on a square lattice without any off-site Coulomb interactions and further-neighbor hoppings. When the on-site Coulomb interaction is very large ($U/|t_1| \sim 20$) and the system is underdoped (typically, $\delta < 0.03$), we find the concave downward doping dependence of the nearest-neighbor hopping energy [see Fig. 28(a)], consistently with the emergent attraction caused by the kinetic energy [see Fig. 28(b)]. However, we do not observe such behavior in the cases of *ab initio* low-energy effective Hamiltonians. It supports that the SC order in the real cuprate materials is mainly governed by the effective instantaneous attraction associated with the on-site Coulomb interaction energy rather than the kinetic energy.

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