**Ab initio** low-energy effective Hamiltonians for high-temperature superconducting cuprates Bi$_2$Sr$_2$CuO$_6$, Bi$_2$Sr$_2$CaCu$_2$O$_8$ and CaCuO$_2$

Jean-Baptiste Moréé, Motoaki Hirayama, Michael Thobias, Youhei Yamaji, and Masatoshi Imada

1 Waseda Research Institute for Science and Engineering, Waseda University, 3-4-1, Okubo, Shinjuku, Tokyo 169-8555, Japan
2 Department of Applied Physics, University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-8565, Japan
3 RIKEN Center for Emergent Matter Science, Wako, Saitama 351-0198, Japan
4 JST, PRESTO, Hongo, Bunkyo-ku, Tokyo 113-8565, Japan
5 Center for Green Research on Energy and Environmental Materials, National Institute for Materials Science, Namiki, Tsukuba-shi, Ibaraki, 305-0044, Japan
6 Toyota Physical and Chemical Research Institute, 41-1, Yokomichi, Nagakute, Aichi 480-1192, Japan

We derive low-energy effective Hamiltonians (LEH) for several high-temperature superconducting (SC) copper oxides. We focus on Bi$_2$Sr$_2$CuO$_6$ (Bi2201, $N_f = 1$, $T_c \sim 10$ K), Bi$_2$Sr$_2$CaCu$_2$O$_8$ (Bi2212, $N_f = 2$, $T_c \sim 84$ K) and CaCuO$_2$ (Ca11, $N_f = \infty$, $T_c \sim 110$ K), where $N_f$ is the number of laminated CuO$_2$ planes between neighboring block layers, and $T_c$ is the experimental optimal SC transition temperature. We apply the latest methodology of the multiscale ab initio scheme for correlated electron systems (MACE) [Hirayama et al, Phys. Rev. B 99, 245155 (2019)]. The LEH for HgBa$_2$CuO$_4$ (Hg1201, $N_f = 1$, $T_c \sim 90$ K) is also derived to benchmark the consistency with the literature. We mainly discuss the single-orbital LEH consisting of the antibonding (AB) combination of Cu3$d_{x^2−y^2}$ and O2$p_x$ orbitals in the CuO$_2$ plane, by which the experimental phase diagram of Hg1201 was reproduced [Ohgoe et al, Phys. Rev. B 101, 045124 (2020)]. The derived LEHs provide reliable starting points to understand the difference in $T_c$ and SC mechanism, when they are solved by accurate quantum many-body solvers. We further discuss the following features: (1) The onsite effective interaction $U \sim 3.6−4.5$ eV increases monotonically with $T_c$, which is consistent with the expectation of increasing SC order parameter $\Delta_{SC}$ with increasing $U$. (2) Inspection of the three-orbital LEH consisting of the AB and two counterpart bonding (B) orbitals suggests that the single-orbital LEH looks sufficient to describe low-energy physics. (3) For $N_f \geq 2$, off-site interactions between neighboring CuO$_2$ layers ($\sim 0.7$ eV) and within a CuO$_2$ layer ($\sim 1.0$ eV) are comparable. We discuss their contributions to both $\Delta_{SC}$ and the stability of the SC state.

I. INTRODUCTION

Unconventional superconductivity (SC) occurs in cuprates [1] with the transition temperature $T_c$ reaching the maximal value $T_{c}^{exp} \sim 138$ K at ambient pressure for HgBa$_2$Ca$_2$Cu$_3$O$_{8}$ [2], and even higher values ($\gtrsim 150$ K) for Hg-based compounds under pressure [3, 4]. It is empirically observed that $T_{c}^{exp}$ has a correlation with $N_f$, the number of CuO$_2$ layers sandwiched by the neighboring block layers; in the Bi-based cuprates Bi$_2$Sr$_2$Ca$_{N_f−1}$Cu$_{N_f}$O$_{2N_f+4}$: $T_{c}^{exp} < 10$ K for $N_f = 1$ (Bi2201) [5, 6], up to $\sim 40$ K under optimal substitution [7]; $T_{c}^{exp} \sim 84$ K for $N_f = 2$ (Bi2212) [8−11]; $T_{c}^{exp} \sim 110$ K for $N_f = 3$ (Bi223) [12] and HgBa$_2$Ca$_2$Cu$_3$O$_8$ also belongs to $N_f = 3$. Namely, $T_{c}^{exp}$ increases progressively with $N_f$ and this trend is also satisfied for carrier doped CaCuO$_2$, which is interpreted as $N_f = \infty$, and reaches $T_{c}^{exp} \sim 110$ K [13]. However, the microscopic mechanism that causes this trend is not well understood.

If appropriate LEHs that correctly describe low-energy physics could be derived systematically for compounds that have different $N_f$ and the relevant material dependent parameters are revealed, it would provide hints to the origin of the difference in $T_{c}^{exp}$ and eventually the SC mechanism in cuprates.

Historically, single-orbital Hubbard models with adjustable parameters of onsite Coulomb repulsion $U$ and the hopping $t$ have been extensively studied to understand the cuprate superconductors. This single orbital is expected to be an antibonding orbital originating from strongly hybridized atomic Cu $3d_{x^2−y^2}$ and O $2p_z$ orbitals, which is centered on each Cu atom and denoted as AB hereafter. However, it is not trivial whether the degrees of freedom beyond the antibonding orbital AB play only minor roles in understanding physics of SC. The band structure derived from density functional theory (DFT) suggests atomic Cu $3d_{x^2−y^2}$, $3d_{3z^2−r^2}$ and O $2p_z$ orbitals, abbreviated as $x$, $z$ and $p$ orbitals hereafter, are located relatively close to the Fermi level and potentially contribute to low-energy physics. For instance, charge transfer energy $\Delta E_{x}^{exp}$ between atomic $x$ and $p$ orbitals smaller than the onsite Coulomb energy for $x$ was claimed to lead to essential insufficiency of the single-band Hubbard-type model [14] including the negative correlation of $\Delta E_{x}^{exp}$ to $T_{c}^{exp}$ [15]. The positive correlation between $T_{c}^{exp}$ and $\Delta E_{xz}$ defined between $x$ and $z$ orbitals was also addressed [16]. An inapplicability of the AB Hubbard model to overdoped cuprates was also claimed from earlier X-ray absorption spectra [17, 18]. Further-
more, theoretical studies of the Hubbard model [19–30] showed the increasing superconducting order for larger $|U/t_1|$ in the superconducting solution of the simple Hubbard model at finite hole doping, where $U$ is the on-site interaction and $t_1$ denotes the nearest neighbour hopping within a CuO$_2$ layer. However, this solution becomes an excited state for large $|U/t_1|$ and the true ground state is dominated by severely competing charge-ordered (CO) states contrary to the widely spread SC phase found in the doping concentration dependence of the experiments [19, 23–30].

Nonetheless, a recent study [31] of the ab initio single-orbital AB Hamiltonian [32] for Hg1201 ($T^c_{\text{exp}} \sim 90$ K [33]) derived by MACE, without adjustable parameters and beyond the simple Hubbard model, was able to reproduce the experimental phase diagram at zero temperature, including the dominant SC phase under hole doping. Off-site interaction parameters beyond the Hubbard model have turned out to be crucially important: Although they reduce the long-range $d$-wave superconducting order parameter $\Delta_{\text{SC}} = \sqrt{P_{dd}}$ monitored by the $d$-wave Cooper pair correlation function $P_{dd}$, they allow the stabilization of the SC state over the competing CO states, because the CO is more severely damaged, so that the SC ground state is successfully predicted. This supports that the ab initio single-orbital AB Hamiltonian offers a promising framework for the in-depth understanding of the SC mechanisms in the cuprates, provided that the ab initio LEH is carefully derived beyond the Hubbard picture. In this paper, we extend the work along this line and derive the ab initio Hamiltonians systematically for several compounds that have different $N_t$ to gain insights into the substantial dependence of $T^c_{\text{exp}}$ on $N_t$.

In the early stage of the derivation of the ab initio LEH for the cuprates within the MACE [34], the hopping parameters (one-particle part) were at the level of the local density approximation (LDA) or generalized gradient approximation (GGA), and the effective interactions (two-particle part) were at the level of the constrained version (cRPA) of the random phase approximation (RPA) [35, 36]. At this level, the $T^c_{\text{exp}}$ dependence of the LEH parameters was studied [37–42], without the recent improvement of the MACE by the constrained GW (cGW) method [43–46], self-interaction correction (SIC) [44] and level renormalization feedback (LRFB) [32]. Recent LDA/GGA+cRPA studies [39–41] at this level reported that the high-$T_c$ SC is favored by a higher value of $|U/t_1|$. However, they studied the Hamiltonian without considering the off-site interaction and did not consider the competition with the CO. For instance, in Ref. 39, the cRPA value of $U$ for TlBa$_2$CuO$_5$ (Tl1201, $N_t = 1$, $T^c_{\text{exp}} \sim 50$ K) is $\sim 40\%$ lower than their estimate for Hg1201 ($N_t = 1$, $T^c_{\text{exp}} \sim 90$ K) and Ca11 ($N_t = \infty$, $T^c_{\text{exp}} \sim 110$ K). In addition, their value of $U$ for Hg1201 is $\sim 2.9$ eV, which is substantially underestimated with respect to $\sim 3.8$ eV in Ref. 46. It is known that an insufficient treatment of the disentanglement procedure [47] can cause the underestimate. Still, the enhanced SC for larger $|U/t_1|$ is consistent with the Hubbard model study mentioned above [19, 21, 23–30].

In this paper, we apply the state-of-the-art methodology of the MACE [32], by using the RESPACK code [48]. We use the pseudopotential (PP) and plane wave formalisms, to reduce the computational cost compared to the all-electron (AE) implementation. This allows us to treat compounds with more atoms in the unit cell such as Bi2201 and Bi2212 even with the improved MACE scheme mentioned above [32]. It enables to derive LEHs for different $N_t$ and to study the systematic dependence of the LEH parameters on Hg1201 ($N_t = 1$), Bi2201 ($N_t = 1$), Bi2212 ($N_t = 2$) and Ca11 ($N_t = \infty$), which helps reaching our present goal to understand the microscopic origin of strongly increasing $T_c$. Our comparison of the LEH for Hg1201 ($N_t = 1$) with the literature using the AE implementation in Refs. 32 and 46 is useful to establish the accuracy and reliability of our PP framework.

We analyze a three-orbital LEH, called the ABB LEH below, consisting of the antibonding AB orbital as well as two counterpart bonding orbitals denoted by B. By examining the ABB LEH, we demonstrate that the single-orbital AB Hamiltonian looks enough, by showing that the lower Hubbard band (LHB) from the AB manifold is located above the upper Hubbard band (UHB) from the B manifold and both are nearly separated. In this AB Hamiltonian, we find that the monotonic increase in experimental $T^c_{\text{exp}}$ with ab initio $U$ is caused by the progressive reduction of the cRPA screening of the Coulomb interaction. It is consistent with the previous speculation for the dependence of the SC order on $U$, but the present quantitative estimates will allow detailed clarification about the severe competition with the charge ordering/phase separation. Also, we discuss the possible role of interlayer (i.e., between different CuO$_2$ layers) interaction to the stability of CO for $N_t \geq 2$ (Bi2212 and Ca11).

This paper is organized as follows: Section II describes our method and computational details. The first part of the LEH derivation starting from the Kohn-Sham (LDA or GGA) level and improving it to the GW level supplemented with LRFB correction [32] is outlined in Sec. III. In Sec. IV, we start from the GW+LRFB electronic structure to derive AB LEH. In Sec. V, we analyze material dependence of the derived LEH parameters, and their effect on $\Delta_{\text{SC}}$ and stability of SC state. Appendices A, B and C show details of the preprocessing summarized in Sec. III. Appendix D demonstrates the validity of the single-orbital AB Hamiltonian derived in Sec. IV. In Appendix E, we propose an approximation (not used in this paper, but useful for future studies) to reduce the computational cost of the MACE for compounds with large number of bands as the cuprates with $N_t \geq 2$, without loss of accuracy.
II. METHODS AND COMPUTATIONAL DETAILS

Effective LEHs in the present paper have the form

\[
\hat{H} = \sum_{\langle i,\mathbf{R},\sigma \rangle} \sum_{\langle j,\mathbf{R}',\sigma' \rangle} t_{ij}^{\sigma\sigma'} (\mathbf{R} - \mathbf{R}') c_{i\sigma\mathbf{R}} \hat{c}_{j\sigma'\mathbf{R}'} + \sum_{\langle i,\mathbf{R} \rangle} \sum_{\langle j,\mathbf{R}',\sigma' \rangle} U_{ij}^{\sigma\sigma'} (\mathbf{R} - \mathbf{R}') c_{i\sigma\mathbf{R}} \hat{c}_{j\sigma'\mathbf{R}'} \hat{c}_{j\sigma'\mathbf{R}'} c_{i\sigma\mathbf{R}},
\]

where \( \mathbf{R} \) is the coordinate of the unit cell in the space \([abc]\) expanded in the \((a,b,c)\) frame in Fig. 1. The indices \( i,j \) denote the orbitals within the unit cell, and \( \sigma, \sigma' \) denote the spin indices. By using these notations, \( c_{i\sigma\mathbf{R}} \) and \( \hat{c}_{i\sigma\mathbf{R}} \) are respectively the creation and annihilation operators in the spin-orbital coordinate \((i, \sigma)\) at \( \mathbf{R} \), and \( t_{ij}^{\sigma\sigma'} (\mathbf{R} - \mathbf{R}') \) and \( U_{ij}^{\sigma\sigma'} (\mathbf{R} - \mathbf{R}') \) are respectively the hopping and direct interaction parameters between spin-orbitals \((i, \sigma)\) at \( \mathbf{R} \) and \((j, \sigma')\) at \( \mathbf{R}' \), which satisfy translational symmetry. Other interaction parameters (Hund, exchange and pair hopping parameters, and parameters beyond two-body interaction) are assumed to play minor roles and ignored. If \( \mathbf{R} = \mathbf{R}_0 = [000] \) and \( i = j \), we abbreviate \( U_{ii}^{\sigma\sigma'} (\mathbf{R}_0) \) as the onsite interaction \( U \) for the AB Hamiltonian, and \( U_l \) for multi-orbital Hamiltonians.

In the case of the AB Hamiltonian, there is only one AB orbital per unit cell for Hg1201, Bi2201 and Ca11 \((i = j = 1)\), but two for Bi2212 \((i, j = 1, 2\), as in Fig. 1). For a comparison of energy scales in the series of the cuprates, we separate \( U_{ij} (\mathbf{R}) \) into four different categories of parameters, denoted as onsite, intralayer, interlayer ("ii") and distant interlayer ("dd") parameters. A few typical interaction parameters are represented schematically in Fig. 1. Intralayer, off-site interaction parameters are \( V_{nn} = U_{11} (\mathbf{R}_n) \), where \( \mathbf{R}_n \) gives the position of the \( n^{th} \) nearest neighbour orbital within the CuO2 layer (we have \( \mathbf{R}_1 = [100], \mathbf{R}_2 = [110], \mathbf{R}_3 = [200], \mathbf{R}_4 = [210], \mathbf{R}_5 = [220] \) and \( \mathbf{R}_6 = [300] \)). Interlayer interaction parameters are defined only for Bi2212 and Ca11, as \( V_n^d = U_{12} (\mathbf{R}_n) \) for Bi2212 and \( V_n^d = U_{11} (\mathbf{R}_n + c) \) for Ca11, where \( c = [001] \) is defined as in Fig. 1. Distant interlayer parameters for Hg1201, Bi2201 and Bi2212 are defined as those between different CuO2 layers separated by a block layer, that is, \( V_n^d = U_{11} (\mathbf{R}_n + c) \) for Hg1201 and Bi2201 and \( V_n^d = U_{11} (\mathbf{R}_n + 2c) \) for Bi2212. For Ca11, there are no block layers; instead of \( V_n^d \), we define the second interlayer parameters as \( V_n^{dd} = U_{11} (\mathbf{R}_n + 2c) \).

The one-particle part \( t_{ij} (\mathbf{R}) \) is classified into intralayer \((t_{ii})\), interlayer \((t_{dd})\) and distant interlayer \((t_{dd}^d)\) hopping parameters, which are defined similarly.

Distant interlayer parameters are usually neglected in the AB Hamiltonian, even within the most recent MACE methodology [32, 46]. Neglecting \( t_{dd}^d \) parameters is justified by their small amplitude \((\lesssim 0.01 \text{ eV for Bi2201 and Bi2212})\). Although \( V_n^d \) parameters are small

\( (V_n^d/U \lesssim 8\% \text{ in this paper}) \), they are not always practically negligible. It is worth noting that the distant interlayer Coulomb interactions lead to the screening of the intralayer interactions due to the dielectric or metallic responses from the distant layers. It may be possible to include the screening effect from layers outside the 2D Hamiltonian by using the dimensional downfolding procedure [49], which we do not consider here. In the following, we will focus on intralayer and interlayer parameters; in the analysis of the LEHs, the maximal values of \( V_n^d \)
parameters for Hg1201, Bi2201 and Bi2212, and $V_{\alpha}^{2l}$ parameters for Ca11, will be compared.

We compute the effective parameters $t_{ij}(\mathbf{R})$ and $U_{ij}(\mathbf{R})$ by using our new implementation of the MACE scheme [52, 53] within the RESPACK code [48]. We first compute the electronic structure at the Kohn-Sham (KS) level. Then, the medium-energy ($M$) space, composed of 3$d$-like bands from Cu and 2$p$-like bands from O, is improved at the $GW$+LRFB level, by following the methodology in Ref. 32; other bands are left at the KS level. We then use the resulting $GW$+LRFB electronic structure as a starting point to derive the AB Hamiltonian at cRPA and $cGW$ levels; the cRPA allows to remove the double counting in the screening in the two-particle part, whereas the c$GW$ allows to remove the exchange and correlation double counting term in the one-particle part.

Now, we give computational details. Structural data is taken from [33, 50, 51]. DFT calculations are done with QUANTUM ESPRESSO [52, 53] and optimized norm-conserving Vanderbilt pseudopotentials (PPs)\footnote{We used the PPs $X\_ONCV\_PBE\_1.0$ upf ($X$ = Bi, Sr, Ca, Hg, Ba, Cu and O) from the http://www.quantum-espresso.org distribution.} using the GGA-PBE functional [55]. For Ca11 and Hg1201, we regenerate the PPs by using the ONCVPSP code [56], and switch the functional to the Perdew-Zunger LDA [57] to remove the double counting in the screening in the two-particle part, whereas the c$GW$ allows to remove the exchange and correlation double counting term in the one-particle part.

The Kohn-Sham (KS) electronic structure is composed of 20 bands for Cu and 2$p$-like bands from O, is improved at the $GW$+LRFB level, by following the methodology in Ref. 32; other bands are left at the KS level. We then use the resulting $GW$+LRFB electronic structure as a starting point to derive the AB Hamiltonian at cRPA and $cGW$ levels; the cRPA allows to remove the double counting in the screening in the two-particle part, whereas the c$GW$ allows to remove the exchange and correlation double counting term in the one-particle part.

Now, we give computational details. Structural data is taken from [33, 50, 51]. DFT calculations are done with QUANTUM ESPRESSO [52, 53] and optimized norm-conserving Vanderbilt pseudopotentials (PPs)\footnote{We used the PPs $X\_ONCV\_PBE\_1.0$ upf ($X$ = Bi, Sr, Ca, Hg, Ba, Cu and O) from the http://www.quantum-espresso.org distribution.} using the GGA-PBE functional [55]. For Ca11 and Hg1201, we regenerate the PPs by using the ONCVPSP code [56], and switch the functional to the Perdew-Zunger LDA [57] to remove the double counting in the screening in the two-particle part, whereas the c$GW$ allows to remove the exchange and correlation double counting term in the one-particle part.

III. PREPROCESSING OF INITIAL ELECTRONIC STRUCTURE AT KS LEVEL TO PROCEED TO GW+LRFB LEVEL

Here, we discuss the preprocessing of the starting electronic structure. Before the improvement of the $M$ space from the KS level to the $GW$+LRFB level, we discuss a technical preprocessing treatment to eliminate electron pockets of $p$-like bands originating from the BiO block layer created as an artifact of the KS level calculation for Bi2201 and Bi2212. This treatment is necessary to make fast convergence to the accurate LEH.

Fig. 2 shows the KS (GGA) band structures for Bi2201 and Bi2212 on the panels (a) and (c). The $M$ space is composed of 23 bands for Bi2201 and 34 bands for Bi2212. The AB-like bands cross the Fermi level, creating the hole-like Fermi surface (FS) centered around $X = (\pi, \pi)$ typically observed in experiment for cuprates, including Bi2201 with partial La-substitution of Sr [61, 62] or Pb-substitution of Bi [63, 64], and Bi2212 [64, 65]. However, $p$-like bands coming from the hybridization of 6$p$ orbitals from Bi and 2$p$ orbitals from O (dubbed hereafter as Bi $p$-bands) cross the Fermi level as well, creating the electron pocket centered around $D = (\pi, 0)$, for both compounds. In the case of Bi2212, this is consistent with local density functional (LDF) full-potential linearized augmented plane wave (FLAPW) calculations in the literature [66, 67]. However, such electron pockets are not observed in the experimental FS for Bi2212 [64, 65] or substituted Bi2201 [61–64]. We attribute the appearance of the electron pockets to the insufficient accuracy of the GGA approximation. If we keep these unphysical pockets, they would create unphysical screening at both the RPA and cRPA levels in the $GW$ calculation and LEH derivation.

To remove this unphysical effect, we preprocess the electronic structure already at the KS level, in order to realize the realistic Bi $p$-bands. First, for Bi2201, we perform a structural optimization of the relative positions of Bi and O atoms in the BiO layer and apical O atoms (details are given in Appendix A). This yields the KS(opt) band structure on the panel (a) of Fig. 2. We next take into account the experimental condition of the superconductors with hole doping, where Bi is partially substituted. Here we simulate it within the PP, by using the implementation of the virtual crystal approximation (VCA) [68] in QUANTUM ESPRESSO [52, 53]. We interpolate the PPs for Bi and Pb by using proportions of 1/$\delta$ in the KS and KS(opt) is now absent in the KS(opt,$\delta$)). This yields the KS(opt) band structure on the panel (a) of Fig. 2. We next take into account the experimental condition of the superconductors with hole doping, where Bi is partially substituted. Here we simulate it within the PP, by using the implementation of the virtual crystal approximation (VCA) [68] in QUANTUM ESPRESSO [52, 53]. We inter
FIG. 2. Band structures of Hg1201, Bi2201, Bi2212 and Ca11 at the KS level (all bands) and GW+LRFB level (improvement of bands within M space). For Bi compounds, we show the intermediary KS(opt) and KS(opt,δ) band structures for Bi2201, and KS(δ) band structure for Bi2212. High-symmetry points are defined in Cartesian coordinates, in units of $2\pi/|a|$, $2\pi/|b|$ and $2\pi/|c|$, as $\Gamma = [0.0 0.0 0.0]$, $D = [0.5 0.0 0.0]$, $Z = [1.0 0.0 0.0]$ and $X = [0.5 0.5 0.0]$. For Ca11, we redefine $Z = [0.0 0.0 0.5]$, and we define $R = [0.5 0.0 0.5]$ and $A = [0.5 0.5 0.5]$.

that the hole doping $\delta = 0.2$ has been chosen, where $\delta$ is controlled by the value of $x$ and $\delta = 0.2$ corresponds to $x = 0.1$. This is a realistic choice and physically justified, because the optimal hole doping is $\delta \sim 0.19$ for substituted Bi2201 [7] and $\sim 0.27$ for Bi2212 [69, 70].

As for Hg1201 and Ca11, the KS (LDA) band structures are shown in panels (e) and (f) of Fig. 2. The M space is composed of 17 and 11 bands, respectively. Although we could perform the treatment similar to the Bi compounds, we do not need to do so, because unphysical electron pockets do not show up.

Then, we preprocess the M space at the GW+LRFB level, following the methodology in Ref. 32. For Hg1201, Bi2201 and Ca11, the M space is disentangled [47] from other bands crossing the AB-like band ($s$-bands from Hg and Ba near X point for Hg1201, Bi $p$-bands near X point for Bi2201, and interstitial $s$-like band from Ca near A point for Ca11). For Hg1201 and Bi2201, we use the inner window (between $-7$ eV and the Fermi energy) to preserve the band dispersion of the $s$-bands and $p$-bands. The resultant disentangled M space is preprocessed at the GW level; the GW self-energy is calculated as a function of frequency, then evaluated and linearized around KS eigenvalues as in [32, 46] to construct the quasiparticle GW electronic structure (which includes bands outside the M space, left at the KS level). We next improve the GW electronic structure at the GW+LRFB level as follows. We restart from the GW electronic structure and derive a three-orbital LEH ($xp$ Hamiltonian) at the cRPA level and $cGW$ level with the SIC ($cGW$-SIC) [44]; the details are given in Appendix B. We solve this $xp$ Hamiltonian with the mVMC code [23, 71, 72] to deduce the LRFB correction $\Delta\mu$ of the charge transfer en-

\[ \text{For this part, the computational cost may be reduced by using the approximation proposed in Appendix E.} \]
energy $\Delta E_{zp}$ between the $x$ and $p$ levels; details are given in Appendix C. We use $\Delta \mu$ to improve the GW self-energy as in Ref. 32; then, we evaluate and linearize this self-energy around KS eigenvalues, and deduce the quasiparticle $GW+\text{LRFB}$ electronic structure for the M space; the corresponding band structures are shown on the panels (b), (d), (e) and (f) of Fig. 2. We note that, for Hg1201, the $GW+\text{LRFB}$ band structure is in good agreement with Fig. 7 in Ref. 32. The preprocessed M space, together with other bands left at the KS level, constitutes the $GW+\text{LRFB}$ electronic structure, which is used as a starting point for the derivation of the AB Hamiltonian.

IV. SINGLE-ORBITAL AB HAMILTONIAN

We now proceed to the main process to derive the AB Hamiltonian for all four aforementioned compounds. We start from the preprocessed electronic structure at the $GW+\text{LRFB}$ level and construct the maximally localized Wannier (MLW) orbitals with the AB character: We set the initial guesses as one $x$ atomic orbital centered on each Cu atom in the unit cell (one orbital for Hg1201, Bi2201 and Ca11, two orbitals for Bi2212). The outer window consists in the M space, from which the lowest bands are excluded (4, 7, 10 and 2 lowest bands for Hg1201, Bi2201, Bi2212 and Ca11, respectively) to avoid catching bonding character. We minimize the spillage functional [74] on this outer window to extract the AB-like band which spans the AB subspace, then minimize the spread functional [74] on the AB subspace to deduce the AB MLW orbital. We then disentangle [47] other bands within the outer window from the AB band; these disentangled bands, together with the AB band, the unmodified bands excluded from the outer window, and other bands left at the KS level, are denoted as the “$GW+\text{LRFB(AB)}$” electronic structure. As an illustration, for Bi2201, we show the comparison between $GW+\text{LRFB}$ and $GW+\text{LRFB(AB)}$ band structures (restricted to M space) in the panel (a) of Fig. 4 of Appendix D.

Then, we start from the $GW+\text{LRFB(AB)}$ electronic structure, and compute the two-particle part at the cRPA level and corresponding one-particle part at the cGW level, denoted as $cGW+\text{LRFB(AB)}$. We do not include the SIC, since it is only useful for multi-orbital Hamiltonians with nondegenerate energy levels.

a. Validity of the AB Hamiltonian — Before showing results for the AB Hamiltonian, we discuss why it is legitimate to restrict the present study to the single-orbital picture, without the need to include other degrees of freedom in the LEH. In Appendix D, we extend the AB Hamiltonian to the ABB Hamiltonian, which includes the AB orbital plus two B orbitals. We analyze the competition between (i) the average onsite Coulomb repulsion on AB and B manifolds, and (ii) the charge transfer energy between AB and B manifolds. We show that (i) is weaker than or barely equal to (ii). As a consequence, the upper Hubbard band from the B manifold remains well below the Fermi level, and it is nearly separated from the lower Hubbard band from AB orbital. That is, the B manifold does not essentially contribute to low-energy physics, and may be excluded safely from the LEH.

b. Benchmark (Hg1201) — For the AB Hamiltonian, results are summarized in Table I. Here, we detail the irreducible intralayer and interlayer parameters up to $n = 6$; the complete list of parameters may be obtained from the authors upon request. For the one-particle part, corresponding band structures are shown in Fig. 3. Also, important quantities and ratios between effective parameters are shown in Tables I and II. First, we restrict to Hg1201, and compare our result with the AE result [32]. The two-particle part is close for both calculations: The difference in $U$ does not exceed $\sim 0.18$ eV (less than 5%), and the difference in
Two-particle part (cRPA) (eV)

|                | $U$ | $V_1$ | $V_2$ | $V_3$ | $V_4$ | $V_5$ |
|----------------|-----|-------|-------|-------|-------|-------|
| Hg1201 (AE)    | 3.846 | 0.834 | 0.460 | 0.318 | 0.271 | 0.209 | 0.233 |
| Hg1201         | 4.029 | 0.900 | 0.520 | 0.373 | 0.329 | 0.265 | 0.146 |
| Bi2201         | 3.617 | 0.845 | 0.488 | 0.333 | 0.342 | 0.298 | 0.156 |
| Bi2212         | 4.270 | 0.881 | 0.508 | 0.373 | 0.328 | 0.276 | 0.148 |
| Ca11           | 4.482 | 1.044 | 0.554 | 0.356 | 0.288 | 0.201 | 0.118 |
| $V_0^i$        | $V_1^i$ | $V_2^i$ | $V_3^i$ | $V_4^i$ | $V_5^i$ |
| Bi2212         | 0.629 | 0.461 | 0.377 | 0.307 | 0.283 | 0.246 | 0.132 |
| Ca11           | 0.749 | 0.521 | 0.396 | 0.289 | 0.247 | 0.184 | 0.108 |

One-particle part (cGW+LRFB(AB)) (eV)

|                | $d_0$ | $t_1$ | $t_2$ | $t_3$ | $t_4$ | $t_5$ | $t_6$ |
|----------------|-------|-------|-------|-------|-------|-------|-------|
| Hg1201 (AE)    |       |       |       |       |       |       |       |
| Hg1201         | -0.509 | 0.127 | -0.077 | 0.018 | 0.004 | -0.004 |       |
| Bi2201         |       |       |       |       |       |       |       |
| Bi2201         | -0.494 | 0.112 | -0.055 | 0.018 | 0.002 | -0.002 |       |
| Bi2212         | -0.507 | 0.174 | -0.071 | 0.003 | -0.016 | 0.007 |       |
| Bi2212         | -0.077 | -0.433 | 0.147 | -0.048 | -0.004 | -0.003 | -0.001 |
| Ca11           |       |       |       |       |       |       |       |
| Ca11           | -0.059 | -0.521 | 0.120 | -0.029 | 0.008 | -0.007 | -0.001 |       |

TABLE I. Effective Hamiltonian parameters for the AB Hamiltonians of Hg1201, Bi2201, Bi2212 and Ca11. For Hg1201, we give the all-electron (AE) result from Ref. 32, for comparison. Interlayer hopping parameters $t_n^i$ other than for $n=0$ are not shown (their amplitude is $\sim 0.01$ eV). We also give other useful quantities in the lower panel: We list $T_c^{\exp}$ for the convenience of readers and give the onsite bare interaction $v$, screening ratios $R = U/v$ and $R_0 = V_0^i/U$ (where $v_0^i$ is the bare interaction corresponding to $V_0^i$), and ratio $V_0^i/U$. We also give $V_0^d/U$ for Hg1201, Bi2201 and Bi2212 and $V_0^d/U$ for Ca11, where $V_0^d$ and $V_0^{d'}$ are the strongest among $V_n$ and $V_n^d$, respectively.

$V_n$ does not exceed $\sim 0.08$ eV. As for the one-particle part, the difference in hoppings does not exceed $\sim 0.02$ eV, although the value of $|t_3|$ is underestimated. The difference in values of $|U/t_1|$ is only $\sim 8\%$ and values of $|t_2/t_1|$, $V_1/U$ and $V_2/V_1$ are well reproduced. Thus, the overall agreement is deemed acceptable.

c. *Two-particle part* — Second, we discuss material dependence of the two-particle part. The overall trends are summarized as follows. (1) $U$ increases progressively with $N_f$ and $T_c^{\exp}$, (2) long-range $V_n$ decays faster when $N_f$ and $T_c^{\exp}$ increases, and (3) important interlayer parameters appear for $N_f \geq 2$.

(1) $U$ increases progressively from $\sim 3.6$ eV for Bi2201 ($N_f = 1$, $T_c^{\exp} < 10$ K) to $\sim 4.5$ eV for Ca11 ($N_f = \infty$, $T_c^{\exp} \sim 110$ K). As a consequence, $U$ increases monotonically with $T_c^{\exp}$, in agreement with other studies [39, 40], but the values are substantially larger in the present estimate. Here, several important points are found.

(1.i) The progressive increase in $U$ is mainly caused by the progressive decrease in the onsite cRPA screening (increase in $R = U/v$): $R \sim 0.26$ for Bi2201 ($T_c^{\exp} < 10$ K), $0.28$ for Bi2212 ($T_c^{\exp} \sim 84$ K), $0.29$ for Hg1201 ($T_c^{\exp} \sim 90$ K), and $0.32$ for Ca11 ($T_c^{\exp} \sim 110$ K). Interestingly, the progressive increase in $R$ with $T_c^{\exp}$ still holds within three-orbital Hamiltonians (ratio $R_x$ in Table IV of Appendix B, and $R_{AB}$ in Table VI of Appendix D), which suggests the cRPA screening channel between AB-like band and B-like bands is not responsible for the increase in $R$. Instead, two factors (1.ii) and (1.iii) detailed below seem to account for the increase in $R$.

(1.ii) The first factor likely to be effective to the cRPA screening is caused by progressive substitution of apical O atoms by apical Cu atoms when $N_f$ increases: For each Cu atom, the two apical O atoms for $N_f = 1$ are progressively replaced by apical Cu atoms when $N_f$ increases, as seen in Fig. 1. This explains the progressive increase in $R$ for Bi2201, Bi2212 and Ca11. Indeed, for a given Cu atom, the apical O atoms harbor only non-correlated orbitals, and thus, fully participate in the short-range cRPA screening. On the other hand, apical Cu atoms harbor both non-correlated orbitals and correlated AB MLW orbitals: The former participate in the short-range cRPA screening but not the latter. As a consequence, replacing apical O with apical Cu weakens the short-range cRPA screening, which increases $R$.

(1.iii) The second factor: Number of bands contributing to the screening in the target bands. At fixed $N_f$, when the number of atoms in the unit cell increases, the band structure becomes denser and the screening is increased. In our case, concerning Bi2201 and Hg1201, $R$ is smaller for Bi2201 ($N_f = 1$, $T_c^{\exp} < 10$ K) than for Hg1201 ($N_f = 1$, $T_c^{\exp} \sim 90$ K), because there are more atoms in the block layer for Bi2201. Concerning Bi2212 ($N_f = 2$, $T_c^{\exp} \sim 84$ K) and Hg1201 ($N_f = 1$, $T_c^{\exp} \sim 90$ K), $R$ is similar: There are more atoms in the block layer in Bi2212 compared to Hg1201, but this is compensated by the replacement of apical O atoms by apical Cu atoms in Bi2212 mentioned in (1.ii).

(1.iv) Besides cRPA screening, the onsite bare interaction $v$ contributes to increase the value of $U$ for Bi2212. Indeed, $v$ is similar for Bi2201, Hg1201 and Ca11 ($\sim 14$ eV) but higher for Bi2212 ($\sim 15$ eV). The AB MLW orbital is more localized for Bi2212, which we interpret as a consequence of the buckling of Cu–O–Cu bonds within CuO$_2$ layers (as seen in Fig. 1): Although the cell parameter along $a$ is the same for Bi2201 and Bi2212 ($\sim 3.81$ Å), in-layer O atoms are slightly distorted out of the CuO$_2$ plane in Bi2212 because of GdFeO$_2$-type tilting of CuO$_6$ octahedron [75], which increases the distance between Cu and in-layer O atoms. This contributes to increasing the localization of MLW orbitals. The tilting also contributes to decrease the...
hopping amplitude $|t_1|$ as well as the total bandwidth, which is indeed the case of Bi2212 and is discussed in d.(1) below.

(2) Very roughly speaking, there exists a trend that the decay of intralayer interactions $V_n$ with increasing distance (increasing $n$) is faster when $N_c$ and $T_{\text{exp}}$ increase. Indeed, $V_3/V_1 = 0.45$ for Bi2201, 0.42 for Bi2212 and Hg1201, and 0.34 for Ca11. Also, $V_6/U = 0.043$ for Bi2201, $0.034$ for Bi2212, $0.036$ for Hg1201, and 0.026 for Ca11. This trend is originated from the same mechanism of the screening from dense bands as (1-iii).

(3) Interlayer parameters are important for Bi2212 and Ca11 ($N_c \geq 2$). Indeed, $V_3/U = 0.15$ for Bi2212 and 0.17 for Ca11, and $V_3/V_1$ takes intermediate values between $V_1$ and $V_2$. The larger value of $V_3/U$ for Ca11 compared to Bi2212 comes from the weaker short-range eRPA screening, as discussed above. The latter screening is encoded within $R_0^d = V_0^d/v_0^d$ (where $v_0^d$ is the bare interaction corresponding to $V_0^d$), which is 0.16 for Bi2212 but 0.20 for Ca11.

d. One-particle part — Now, we comment on the one-particle part at the cGW+LRFB(AB) level for each compound (Table I; corresponding band structures are shown in Fig. 3). Here as well, we point out several findings:

(1) As for $|t_1|$, it is similar for Bi2201, Hg1201 and Ca1 ($\sim 0.50$ eV), but smaller for Bi2212 ($\sim 0.43$ eV) due to the stronger localization of AB MLW orbitals, which was discussed previously. Importantly, whereas $U$ increases monotonically with $N_c$ and $T_{\text{exp}}$ as in Refs. 39 and 40, $|U/t_1|$ has the highest value for Bi2212, which looks against the “larger $T_{\text{exp}}$, larger $|U/t_1|”$ trend. The origin of larger $|U/t_1|$ in Bi2212 is the relatively smaller value of $|t_1|$. In fact, $T_c$ is expected to be determined not only by $|U/t_1|$ but the characteristic energy scale $|t_1|$ itself. Therefore, lower $T_c$ despite larger $|U/t_1|$ can be naturally understood from nearly 15% smaller $|t_1|$ for Bi2212 than Hg1201 and Ca11.

(2) As for long-range intralayer hoppings, there exists a rough trend that the spatial range of non-negligible hopping decreases when $N_c$ and $T_{\text{exp}}$ increase, concomitantly with the faster decay of $V_n$ at increasing $n$. This tendency is not consistent with the comparison on the LDA level [76], where, for instance, $|t_2/t_1|$ is $\sim 0.37$ for Hg1201 and $\sim 0.27$ for Bi2201, while they are respectively 0.23 and 0.34 in the present estimate. This opposite trend can be naturally understood by the self-energy effect taken into account by refined MACE procedure with the cGW+LRFB treatment beyond the LDA level. Indeed, $|t_2/t_1|$ is $\sim 0.34$ for Bi2201 and Bi2212 as compared to $\sim 0.23$ for Hg1201 and Ca11. Also, $|t_3/t_1| = 0.14$ for Bi2201, 0.11 for Bi2212 and Hg1201, and 0.06 for Ca11. It is an intriguing future issue whether smaller hopping of the long-range part helps the SC.

(3) As for interlayer hoppings, they become non-negligible for Bi2212 and Ca11: $|t_0/t_1| \sim 0.18$ for Bi2212 and $\sim 0.11$ for Ca11. We note that $|t_0|$ is larger for Bi2212. This can be explained as follows: For Bi2212, compared to Ca11, the distance between O atoms in adjacent CuO$_2$ layers is smaller due to the buckling of Cu–O–Cu bonds, which increases the overlap between a given AB Wannier orbital and that in the next layer as hinted by the shape of the isosurfaces for Bi2212 and Ca11 in Fig. 1.

V. DISCUSSION

In Sec. IV, we discussed the origins of the differences in the AB Hamiltonians for different compounds. Material dependence of the LEH parameters clarified in the present paper will contribute to understanding of material dependence of the SC on the microscopic level and hence to understanding of the universal mechanism of SC when they are solved by reliable quantum many-body solvers in future. However, even before solving them, one can gain insight into the materials dependence of the experimentally observed SC phases from the derived AB Hamiltonians. We first discuss two points: (1) amplitude of the SC order parameter $\Delta_{\text{SC}}$, and (2) competition of the SC state with charge inhomogeneous states at low temperatures.

| Starting point | Compound | $T_{\text{exp}}$ (K) | $|U/t_1|$ | $|t_1|$ (eV) | $|t_2/t_1|$ | $|t_3/t_1|$ | $|V_1/U|$ | $|V_2/V_1|$ | $|V_3/V_1|$ |
|---------------|----------|---------------------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|
| (a) Ref. 46   | GW       | La$_2$CuO$_4$       | $\sim 45$ | 10.36     | 0.48      | 0.15      | 0.21      | 0.25      | 0.56      |
| Ref. 46      | GW       | Hg1201              | $\sim 90$ | 9.49      | 0.46      | 0.26      | 0.16      | 0.22      | 0.54      |
| Ref. 32      | GW+LRFB  | Hg1201              | $\sim 90$ | 7.56      | 0.51      | 0.25      | 0.15      | 0.22      | 0.39      |
| (b) This work | GW+LRFB  | Hg1201              | $\sim 90$ | 8.16      | 0.49      | 0.23      | 0.11      | 0.22      | 0.42      |
|              | GW+LRFB  | Bi2201              | $\sim 10$ | 7.13      | 0.51      | 0.34      | 0.14      | 0.23      | 0.45      |
|              | GW+LRFB  | Bi2212              | $\sim 84$ | 9.86      | 0.43      | 0.34      | 0.11      | 0.18      | 0.42      | 0.15      |
|              | GW+LRFB  | Ca11                | $\sim 110$ | 8.60      | 0.52      | 0.23      | 0.06      | 0.11      | 0.23      | 0.34      | 0.17      |

TABLE II. Summary of important ratios in the AB Hamiltonian at the cRPA and cGW+LRFB(AB) levels. We also give the starting point (electronic structure before construction of AB Wannier orbital, disentanglement and calculation of effective parameters), together with the values of $T_{\text{exp}}$ (in K) and $|t_1|$ (in eV). In Ref. 46, the GW electronic structure is used as a starting point instead of the GW+LRFB one; the framework of the rest of the calculation is similar.
On (1), $\Delta_{SC}$ is given by the long-range value of the pair-pair correlation function $P_{dd}$, as $\Delta_{SC} = \sqrt{P_{dd}(r \to \infty)}$. According to Refs. 31 and 28, $\Delta_{SC}$ strongly increases with $|U/t_1|$. This simple trend is in good agreement with the correlation between higher $T^\text{exp}_c$ and larger $U$ in our results and the previous cRPA studies [39, 40].

On the other hand, there exists exceptions such as La$_2$CuO$_4$ (La201), where $U$ is large but $T_c$ is low [32, 46]. It was argued before that too large $|U/t_1|$ leads rather to the CO or charge inhomogeneous states than the SC [29, 32, 46, 77].

Besides $U$, off-site intralayer interactions $V_n$ are important as well. For Hg1201, $|V_3/U|$ and $|V_3/V_1|$ were shown [31] to have a significant effect on SC: Starting from the “only $U$” case, $V_1$ strongly reduces the value of $P_{dd}$ (from $\sim 5 \times 10^{-2}$ to $\sim 2 \times 10^{-3}$), but $V_3$ partly compensates this reduction ($P_{dd}$ increases to $\sim 7 \times 10^{-3}$). Despite the overall reduction of $P_{dd}$ and thus $\Delta_{SC}$ due to $V_n$ parameters, the latter more destabilizes the competing CO state, and relatively favor the SC ground state.

In our results, $|V_3/U|$ is similar for all compounds, whereas $|V_3/V_1|$ decreases with $N_t$ and $T^\text{exp}_c$. This is counterintuitive since a lower value of $|V_3/V_1|$ is not expected to favor a SC ground state or high value of $\Delta_{SC}$ [31]. However, for $N_t \geq 2$, the effect of $V_3$ may be enhanced by off-site interlayer interactions. Indeed, the intralayer effect of $V_3$ is to destabilize the CO state by causing frustration in the charge ordering. In the case of $N_t \geq 2$, the interlayer interaction becomes comparable or larger than $V_3$. It is plausible that $V_n^d$ parameters enhance the frustration effect on top of $V_3$, and would contribute to further destabilization of the CO state. This is an intriguing issue to be examined in the future.

In future studies, two important questions should be clarified: (1) Do $V_n^d$ parameters affect the value of the SC order parameter $\Delta_{SC}$? (2) Do $V_n^d$ parameters contribute to further destabilization of CO states? The AB Hamiltonians presented in this paper may be used as a basis to investigate these questions.

Finally, we discuss the role of $|t_2/t_1|$. This is larger for Bi2201 and Bi2212 ($\sim 0.34$) compared to Hg1201 and Ca11 ($\sim 0.23$). The latter is close to and consistent with the AE result for Hg1201 [32, 46] ($|t_2/t_1| \sim 0.25 - 0.26$; the LRFB does not alter the value of $|t_2/t_1|$). Within the simple framework of the Hubbard model with only $t_1$ and $t_2$ in the one-particle part [29], the stripe CO ground state becomes more extended in the doping concentration dependence and more stable compared to the SC state with increasing $|t_2/t_1|$, which suggests that the SC state is favored by a small $|t_2/t_1|$ in the competition. This is in agreement with the smaller $|t_2/t_1|$ for higher-$T_c$ compounds Hg1201 and Ca11. Though a rough correlation between $|t_2/t_1|$ and $N_t$ can be seen, the correlation between $|t_2/t_1|$ and $T^\text{exp}_c$ is less clear. In fact, the comparison of ab initio Hamiltonians [46] for Hg1201 and La201 show a lower value of $|t_2/t_1|$ for La201, despite a lower $T^\text{exp}_c$. It is desirable to clarify the role of the large $|t_2/t_1| \sim 0.34$ for Bi2201 and Bi2212 on the stability of SC in later studies, by using the present ab initio Hamiltonians as the base.

**SUMMARY**

We have derived and compared LEHs for Bi2201, Bi2212 and Ca11 to gain insights into the mechanism of cuprate superconductivity from the differences of the Hamiltonian parameters of the three compounds that show diverse superconducting transition temperatures. We have also derived the LEH for Hg1201 to compare with Ref. 32. The Hamiltonians are also derived by aiming at serving for future studies to obtain detailed physical properties by accurate quantum many-body solvers.

We have employed the following steps for the derivation: (1) Preprocessed quasiparticle electronic structure is used as a starting point for the derivation of the LEH. Namely, we refined the band structure outside the target low-energy bands near the Fermi level of Bi2201 and Bi2212, which leads to the consistency with the spectroscopic experiments, and improved the exchange and correlation from the KS level to the GW+LRFB level. (2) By using the result of (1), the single-band AB LEHs are derived on the cGW+LRFB level.

In addition to (2), three-orbital ABB LEHs were obtained, which showed that B orbitals have few effect on the low-energy physics except for the indirect cRPA-type screening effect to renormalize the effective interaction within the AB subspace, which justifies the focus to the single-orbital AB Hamiltonian.

We then compared the dependence of effective parameters on compounds within the single-band AB Hamiltonian. The main findings are the following: When the number of layers in the unit cell, $N_t$ increases in this series of multi-layer compounds, $U$ increases whereas the decay of off-site interactions $V_n$ and hoppings $t_n$ is faster. In addition, non-negligible interlayer parameters emerge for $N_t \geq 2$.

Our result on the systematic dependence of the ab initio LEH parameters for four studied compounds leads us to infer that larger $U$ favors larger SC order parameter $\Delta_{SC}$ and hence higher $T_c$, whereas interlayer interaction parameters for high-$T^\text{exp}_c$ compounds with $N_t \geq 2$ could amplify the destabilization of competing CO states caused by intralayer off-site interactions. The derived ab initio LEHs provide a reliable starting point to investigate these hypotheses by solving them by a reliable quantum many-body solver.

**ACKNOWLEDGEMENTS**

We thank Kazuma Nakamura for useful discussions. This work was supported by MEXT as “Program
for Promoting Researches on the Supercomputer Fugaku” (Basic Science for Emergence and Functionality in Quantum Matter —Innovative Strongly-Correlated Electron Science by Integration of “Fugaku” and Frontier Experiments—, JPMXP1020200104) and used computational resources of supercomputer Fugaku provided by the RIKEN Center for Computational Science (Project ID: hp200132, hp210163, and hp220166). Part of the computation was done using the facilities of the Supercomputer Center, the Institute for Solid State Physics, the University of Tokyo. We also acknowledge the financial support of JSPS Kakenhi Grant No. 16H06345. Fig. 1 was drawn by using software VESTA [85].

Appendices

A. STRUCTURAL OPTIMIZATION FOR BI2201

Here, we give computational details and results about the structural optimization for Bi2201 in Sec. III. The optimization is performed by using VASP [78–80]. We use the projector augmented wave (PAW) [81, 82] potential sets recommended by VASP. We take $12 \times 12 \times 6$ $k$-mesh and set the kinetic-energy cutoff to 500 eV. We optimize relative positions of Bi and O atoms within the block layer, and apical O atoms; we do not modify relative positions of Cu, in-layer O and Sr atoms and cell parameters, reported in Ref. 50. Modifications of the relative positions are given in Table III for information. As discussed in Sec. III, this treatment reduces the electron pocket around the D point, but is not sufficient to remove it completely. We have checked that this result is not changed if we optimize the relative positions of Sr atoms in addition.

| Atom          | $x$      | $y$      | $z$      |
|---------------|----------|----------|----------|
| O(apical,A)   | $-0.0160$| $-0.0160$| $0.0841$ |
| O(apical,B)   | $0.0160$ | $0.0160$ | $-0.0841$|
| Bi(A)         | $0.0037$ | $0.0037$ | $0.0367$ |
| Bi(B)         | $-0.0037$| $-0.0037$| $-0.0367$|
| O(block,A)    | $-0.0703$| $-0.0703$| $0.0000$ |
| O(block,B)    | $0.0703$ | $0.0703$ | $0.0000$ |

TABLE III. Variations of relative positions of apical O atoms, Bi atoms, and O atoms within the BiO block layer, between experimental [50] and optimized crystal structures, in Cartesian coordinates (unit is cell parameter [Å] = 3.8097 Å). A and B indicate atoms above and below the CuO$_2$ plane within the unit cell, respectively.

B. THREE-ORBITAL HAMILTONIAN FOR LRFB

Here, we derive the three-orbital Hamiltonian which is used in Sec. III for the calculation of the LRFB correction. This Hamiltonian is denoted as $x_p$ in this paper; it is equivalent to the three-orbital Hamiltonian in Ref. 46, and is sometimes denoted as $d_{pp}$ Hamiltonian in the literature. We start from the GW electronic structure, consider the whole M space as the outer window, and construct three MLW orbitals. We set one $x$ atomic orbital centered on each Cu atom and two $p$ atomic orbitals centered on in-plane O atoms as initial guesses for the construction of the MLW orbitals. We then disentangle other bands within M space from the $x$ $p$ subspace; these disentangled bands, together with the $x$ $p$ subspace and bands outside M space (left at the KS level), constitute the GW($x$ $p$) electronic structure, which is used as a starting point to derive the effective interaction at the cRPA level and one-particle part at the cGW-SIC level. Table IV shows effective parameters for the $x$ $p$ Hamiltonian.

(1) We benchmark our result (the upper panel of Table IV) with respect to the AE implementation [32, 46] (the middle panel of Table IV). For Hg1201, our results compare to those of Ref. 46. Also, for Ca11, we derived the $x$ $p$ Hamiltonian by using the AE implementation, for comparison.

(1.i) The onsite bare interaction is different for the $x$ orbital. Typically, $v_x \sim 29$ eV in the AE result and $\sim 25$ eV in our result. This is due to the PPs used in our calculations; we have checked that the difference in $v_x$ may be corrected by regenerating the PPs with reduced cutoff radii, but at the expense of increasing the cutoff energy for wavefunctions from 100 Ry to at least 200 Ry. Nonetheless, effective interaction amplitudes are in good agreement; the difference is less than 5%. Also, the soft PPs allow computational tractability for compounds with a larger number of atoms in the unit cell, such as Bi2212.

(1.ii) The one-particle part is in good agreement, except the charge transfer energy $\Delta E_{xp}$ which is a bit underestimated in our new implementation: The difference is $\sim 10\%$ for Ca11 and $\sim 19\%$ for Hg1201. For Ca11, the difference in $\Delta E_{xp}$ does not exceed 10%, which remains acceptable. For Hg1201, although the difference is a bit larger, the final AB Hamiltonian is well reproduced, so that we deem it acceptable as well. This discrepancy is partly due to the difference in GW occupation numbers, which are more coherent in the PP result with respect to the AE result. These occupation numbers are used to calculate the SIC, as $t_i^{SIC} = -U_i n_i / 2 [44]$, so that, before applying the renormalization factor [43], $\Delta E_{xp}$ is modified by $\Delta E_{xp} = t_x^{SIC} - t_p^{SIC}$ (which is...
negative in practice). For $U_x = 8.51$ eV and $U_p = 5.35$ eV (our values for Hg1201), a modification $n_x \rightarrow n_x - \delta$ and $n_p \rightarrow n_p + \delta/2$ with $\delta$ as small as 0.04 electrons (corresponding to the difference between our result and the AE result) leads to an increase in $\Delta E_{xp}$ as large as 0.12 eV, which partly explains the larger $\Delta E_{xp}$ for the AE result.

(2) We quickly compare the $xp$ Hamiltonians for different compounds. (2.i) We discuss the effective interaction. $U_x$ and $R_x$ increase progressively with $N_e$ and $T_{\text{exp}}$, which is also the case for the AB Hamiltonian (Sec. IV). (2.ii) We compare $\Delta E_{xp}$ with $T_{\text{exp}}$. Interestingly, the “larger $T_{\text{exp}}$, smaller $\Delta E_{xp}$” trend (discussed before) is not valid here: $\Delta E_{xp}$ is roughly identical for Bi2201 and Bi2212 despite the large difference in $T_{\text{exp}}$. For Ca11 and Hg1201, $\Delta E_{xp}$ decreases with respect to Bi2201 and Bi2212, which is consistent with the increase in $T_{\text{exp}}$ for these compounds. Still, $\Delta E_{xp}$ is the lowest for Hg1201 despite the larger $T_{\text{exp}}$ for Ca11.

(2.i) and (2.ii) suggest that material dependence of $\Delta E_{xp}$ does not explain the differences in $T_{\text{exp}}$. The material dependence of $\Delta E_{xp}$ does not seem to be responsible either for the variations in the cRPA screening for the AB Hamiltonian. Instead, the latter variations may be caused by replacement of apical O with apical Cu and structural difference in the block layer, as discussed in Sec. IV.

### C. LRFB CORRECTION OF THE GW ELECTRONIC STRUCTURE

Here, we detail the LRFB correction [32] of the GW electronic structure. We take the three-orbital $xp$ Hamiltonian in Table IV. We solve this $xp$ Hamiltonian with the mVSCF code [23, 71, 72] as described in Ref. 32, to deduce the LRFB correction $\Delta \mu$ of the charge transfer energy $\Delta E_{xp}$ between the $x$ and $p$ levels.

Importantly, for Hg1201 [32] and Ca11, the ground state of the $xp$ Hamiltonian with $N_e = 5.0$ (half filling) has antiferromagnetic order, whereas the GW electronic structure is paramagnetic. As stated in Ref. 32, the difference in the character of the ground state introduces the correction from the exchange splitting (which is not taken into account at the GW level). In order to obtain a comparable correction for Bi2201 and Bi2212, we must obtain a ground state with similar character (that is, antiferromagnetic). However, the $xp$ Hamiltonian for Bi2201 and Bi2212 considers a total number of electrons $N_e = 4.8$. This poses a practical problem: The antiferromagnetic state is not the ground state anymore at hole doping $\delta = 0.2$, so that we cannot obtain a correction for Bi2201 and Bi2212 which is comparable to that for Hg1201 and Ca11.

In order to avoid this, we consider the following refinement: (1) For Bi2201 and Bi2212, we renormalize the total number of electrons in $xp$ subspace from $N_e = N_f \times 4.8$ to the undoped value $N_e = N_e \times 5.0$, and recalculate the GW occupation numbers and SIC [44]. Results are shown in Table IV (lower panel). $\Delta E_{xp}$ is reduced by $\sim 14\%$ for both compounds, due to the change in the SIC; other effective parameters are not changed. (2) In addition, we neglect interlayer parameters for Bi2212 and Ca11 in the $xp$ Hamiltonian, and consider only parameters within a given CuO$_2$ plane.

We obtain the LRFB correction $\Delta \mu$ of the energy difference between the $x$ and $p$ levels to reproduce the GW occupations, which is $\Delta \mu = 1.30$ eV for Bi2201, 1.35 eV for Bi2212, 1.10 eV for Hg1201 and 1.50 eV for Ca11. This correction is used to improve the GW electronic structure at the GW+LRFB level; details are given in

| $T_{\text{exp}}$ (K) | $N_e$ | $n_x$ | $n_p$ | $\Delta E_{xp}$ (eV) | $\mu$ (eV) | $U_x$ | $U_p$ | $v_x$ | $v_p$ | $R_x$ | $R_p$ | $\xi$ |
|----------------------|------|-------|-------|---------------------|----------|------|------|------|------|------|------|-----|
| Hg1201              | 90   | 5.0   | 1.474 | 1.763               | 2.03     | 1.32 | 0.80 | 8.51 | 5.35 | 25.19 | 17.03 | 0.398 |
| Bi2201              | 10   | 4.8   | 1.324 | 1.738               | 2.95     | 1.35 | 0.71 | 7.79 | 5.68 | 24.82 | 17.48 | 0.314 |
| Bi2212              | 84   | 4.8   | 1.318 | 1.741               | 3.05     | 1.34 | 0.87 | 8.70 | 5.82 | 25.58 | 17.66 | 0.340 |
| Ca11                | 110  | 5.0   | 1.457 | 1.773               | 2.36     | 1.36 | 0.77 | 9.72 | 6.30 | 25.35 | 17.58 | 0.383 |
| Hg1201 (AE)         | 90   | 5.0   | 1.437 | 1.781               | 2.41     | 1.26 | 0.75 | 8.84 | 5.31 | 28.82 | 17.11 | 0.307 |
| Ca11 (AE)           | 110  | 5.0   | 1.433(0) | 1.783(5)        | 2.62     | 1.29 | 0.74 | 9.33 | 6.16 | 28.99 | 17.84 | 0.322 |
| Bi2201              | 10   | 5.0   | 1.458 | 1.771               | 2.55     | 1.35 | 0.71 | 7.79 | 5.68 | 24.82 | 17.48 | 0.314 |
| Bi2212              | 84   | 5.0   | 1.452 | 1.774               | 2.62     | 1.34 | 0.87 | 8.70 | 5.82 | 25.58 | 17.66 | 0.340 |

TABLE IV. Three-orbital $xp$ Hamiltonian. We give the total number of electrons within $xp$ subspace ($N_e$), GW occupation numbers for $x$ and $p$ orbitals ($n_x$ and $n_p$), charge transfer energy between the $x$ and $p$ levels ($\Delta E_{xp}$), amplitudes of hoppings between neighbouring $x$ and $p$ orbitals ($|t_{xp}|$) and two $p$ orbitals within the unit cell ($|t_{pp}|$), intra-orbital effective interaction ($U_x$ and $U_p$) and bare interaction ($v_x$ and $v_p$) for $x$ and $p$ orbitals. We also give the intra-orbital screening ratio $R_i = U_i/v_i$, and the SIC $\xi = -U_i n_x n_p / 2 + U_p n_p / 2$. We also remind the values of $T_{\text{exp}}$ (in K). Upper panel shows the $xp$ Hamiltonian with $N_e$ corresponding to the Kohn-Sham and GW band structures. Middle panel shows the $xp$ Hamiltonian obtained by using the AE implementation [46] for Hg1201 and Ca11, for comparison. Lower panel shows the $xp$ Hamiltonian for the LRFB correction of Bi2201 and Bi2212. $N_e$ is renormalized to the undoped value, and $n_x$, $n_p$ and $\xi$ are recalculated accordingly (other parameters are not changed).
D. THREE-ORBITAL HAMILTONIAN FOR COMPARISON WITH AB HAMILTONIAN

Here, prior to the derivation of the single-orbital AB Hamiltonian in Sec. IV, we show why it is legitimate to restrict the LEH to the single-orbital picture. To do so, we extend the AB Hamiltonian to a three-orbital Hamiltonian, which includes B orbitals in addition. We detail the case of Bi2201, then give the final three-orbital Hamiltonian for all compounds.

a. Nonsuitability of the xz Hamiltonian to justify restriction to the AB Hamiltonian — In Appendix B, we derived the xz Hamiltonian by starting from the GW electronic structure. Here, we quickly discuss the xz Hamiltonian from the GW+LRFB electronic structure (same starting point as the AB Hamiltonian). For Bi2201, we show the occupation numbers of Wannier orbitals and intra-orbital bare interaction in the row (b) of Table V. The band dispersion and partial densities of states are shown in the panels (b) and (c) of Fig. 4. These results reveal that the xz Hamiltonian is not comparable to the AB Hamiltonian. In fact, the x orbital in the xz Hamiltonian has different character from the AB orbital in the AB Hamiltonian. In the xz Hamiltonian, both x and p orbitals have partial density of states in both AB and B bands, due to the strong x/p hybridization. The signature of this mixing is that p orbitals are not full (n_p ∼ 1.7 instead of 2.0). In particular, the x orbital (n_p ∼ 1.4) does not have the purely AB character, so that it is not comparable to the AB orbital.

Let us discuss the nature of the xz Hamiltonian in more details. The xz Hamiltonian is useful for the LRFB correction and improvement of the starting electronic structure beyond the quasiparticle GW approximation, for a more accurate derivation of the LEH. In the xz Hamiltonian, the p orbitals have some character within the AB-like band at the Fermi level (as revealed by the partial density of states and occupation number); this partial AB character is responsible for the role of p orbitals in the low-energy physics of the xz Hamiltonian. Still, it does not say anything about the importance of bands other than the AB-like band (namely, the two B-like bands) for the low-energy physics. As a result, it does not say anything about the necessity to include more than one band (and thus, one orbital) in the LEH.

b. AB/B transformation: From xz Hamiltonian to ABB Hamiltonian — In order to compare the AB and three-orbital Hamiltonians, we set the condition as follows: The AB orbital must be included in the three-orbital Hamiltonian. This can be achieved by considering the gauge degrees of freedom in the construction of Wannier orbitals. Once the spillage functional has been minimized [73] to extract the xz subspace (band dispersion in the panel (b) of Fig. 4), we still have to choose the unitary transformation U within this subspace, which yields the Wannier orbitals.

In the xz Hamiltonian, we chose U which minimizes the spread functional on the xz subspace. Now, we impose the following transformation, denoted as AB/B [83]

Starting from the xz subspace, we consider the AB band (which spans the AB subspace) and the two other bands (B subspace) separately. We project again the initial guess for the x orbital on the AB subspace, and the initial guesses for p orbitals on the B subspace. Then, we minimize the spread functional separately within each subspace; we obtain U on each subspace, yielding one MLW orbital in AB subspace and two MLW orbitals in B subspace. This Hamiltonian is denoted as ABB. For Bi2201, partial densities of states within the ABB Hamiltonian are shown in the panel (d) of Fig. 4: occupation numbers and intra-orbital bare interaction are shown in the row (d) of Table V. Although the total density of Kohn-Sham level (the difference is typically ∼ 1%), so that we may use them, as done in Appendix B. On the other hand, n_x and n_z are modified in the row (b), which makes the SIC wrong.

Furthermore, the modification of n_x and n_p between the rows (a) and (b) of Table V prevents us from performing the SIC in the derivation of the LEH. Indeed, the self-interaction is contained within the Hartree potential at the Kohn-Sham level, so that the SIC must consider n_x and n_p at the Kohn-Sham level [44]. Values of n_x and n_z at the GW level are close to those at the MLW orbitals and disentanglement. In the row (a), we show the occupation numbers of Wannier orbitals in AB subspace. This Hamiltonian is not comparable to the AB Hamiltonian, as shown in the section (c) of Table V. The band dispersion and partial densities of states are shown in the panels (b) and (c) of Fig. 4. These results reveal that the x orbital in the xz Hamiltonian is not comparable to the AB Hamiltonian. In fact, the x orbital in the xz Hamiltonian has different character from the AB orbital in the AB Hamiltonian. In the xz Hamiltonian, both x and p orbitals have partial density of states in both AB and B bands, due to the strong x/p hybridization. The signature of this mixing is that p orbitals are not full (n_p ∼ 1.7 instead of 2.0). In particular, the x orbital (n_p ∼ 1.4) does not have the purely AB character, so that it is not comparable to the AB orbital.

Let us discuss the nature of the xz Hamiltonian in more details. The xz Hamiltonian is useful for the LRFB correction and improvement of the starting electronic structure beyond the quasiparticle GW approximation, for a more accurate derivation of the LEH. In the xz Hamiltonian, the p orbitals have some character within the AB-like band at the Fermi level (as revealed by the partial density of states and occupation number); this partial AB character is responsible for the role of p orbitals in the low-energy physics of the xz Hamiltonian. Still, it does not say anything about the importance of bands other than the AB-like band (namely, the two B-like bands) for the low-energy physics. As a result, it does not say anything about the necessity to include more than one band (and thus, one orbital) in the LEH.

b. AB/B transformation: From xz Hamiltonian to ABB Hamiltonian — In order to compare the AB and three-orbital Hamiltonians, we set the condition as follows: The AB orbital must be included in the three-orbital Hamiltonian. This can be achieved by considering the gauge degrees of freedom in the construction of Wannier orbitals. Once the spillage functional has been minimized [73] to extract the xz subspace (band dispersion in the panel (b) of Fig. 4), we still have to choose the unitary transformation U within this subspace, which yields the Wannier orbitals.

In the xz Hamiltonian, we chose U which minimizes the spread functional on the xz subspace. Now, we impose the following transformation, denoted as AB/B [83]

Starting from the xz subspace, we consider the AB band (which spans the AB subspace) and the two other bands (B subspace) separately. We project again the initial guess for the x orbital on the AB subspace, and the initial guesses for p orbitals on the B subspace. Then, we minimize the spread functional separately within each subspace; we obtain U on each subspace, yielding one MLW orbital in AB subspace and two MLW orbitals in B subspace. This Hamiltonian is denoted as ABB. For Bi2201, partial densities of states within the ABB Hamiltonian are shown in the panel (d) of Fig. 4: occupation numbers and intra-orbital bare interaction are shown in the row (d) of Table V. Although the total density of
Still, this ABB Hamiltonian (the row (d) of Table V) is not completely suitable for comparison with the AB Hamiltonian (the row (c) of Table V). Indeed, the onsite bare interaction for the AB orbital is \( v_{AB} \sim 12.5 \) eV, which is underestimated with respect to the AB Hamiltonian (\( v_{AB} \sim 13.9 \) eV), so that the AB MLW orbitals in both Hamiltonians are not completely equivalent. This comes from the difference in the AB subspace, encoded within the band dispersion of the AB band shown in the panels (a) and (b) of Fig. 4, where the band dispersions are similar, but not identical. In order to solve the discrepancy in \( v_{AB} \), we propose the following scheme. Instead of starting from the \( G\overline{W}+\text{LRFB} \) electronic structure, we start from the \( G\overline{W}+\text{LRFB(AB)} \) electronic structure in the panel (a) of Fig. 4. Then, we set the initial guesses, minimize the spillage functional, extract the \( xp \) subspace, and apply the AB/B transformation to calculate MLW orbitals for the ABB Hamiltonian as described previously. Results are given in the row (e) of Table V. Now, \( v_{AB} \) is very close to the value for the AB Hamiltonian. This improvement comes from the fact that the AB subspace within the AB Hamiltonian is already disentangled from other bands within the \( G\overline{W}+\text{LRFB(AB)} \) electronic structure: The spillage minimization procedure is able to pinpoint and mimic this subspace. Now, the AB MLW orbitals in both AB and ABB Hamiltonians are completely equivalent.
c. Results for the ABB Hamiltonian — Now, we derive the ABB Hamiltonian for all compounds, by considering the following procedure. We start from the GW + LRFB(AB) electronic structure, calculate the AB and B MLW orbitals as described previously, and disentangle the rest of the M space from the ABB subspace. These disentangled bands, together with the ABB substructure, constitute the GW + LRFB(ABB) electronic structure. We start from the latter, and calculate the ABB Hamiltonian for all compounds, by considering the following procedure. We start from the AB subspace and other bands outside the M space (left at the KS level), constitute the GW + LRFB(ABB) electronic structure. We start from the latter, and calculate the two-particle part at the cRPA level and one-particle part at the cGW-SIC level. Importantly, the occupation numbers \( n_{AB} \) and \( n_B \) of MLW orbitals are strictly the same as at the KS level, so that it is possible to perform the SIC.

Results are shown in Table VI; we reproduce values of \( v_{AB} \) and \( U_{AB} \) from the AB Hamiltonian in Table I for comparison. First, \( U_{AB} \) is larger than for the AB Hamiltonian. This does not come from the AB MLW orbital, which is equivalent in both Hamiltonians since \( v_{AB} \) is nearly identical. The difference comes from the cRPA screening, which excludes the channel between AB and B orbitals within the ABB Hamiltonian, but includes it within the AB Hamiltonian. This screening channel contributes to the reduction of \( U_{AB} \) in the AB Hamiltonian, with respect to the ABB Hamiltonian.

Second, the charge transfer energy \( \Delta E_{ABB} \) between the AB and B orbitals is very large (\( \Delta E_{ABB} \sim 7.5 - 8.5 \) eV) compared to the \( xp \) Hamiltonian (\( \Delta E_{xp} \sim 2 - 3 \) eV in Appendix B). This is due to the different character of MLW orbitals, combined to the SIC. Indeed, the update of the onsite energies \( \epsilon_i \) due to the SIC is \( \epsilon_i^{SIC} = -U_i/2 \) [44]. For the \( xp \) Hamiltonian, we have \( t_x = -5.16 \) eV and \( t_p = -4.94 \) eV, so that the improvement of \( \Delta E_{xp} \) due to the SIC (before applying the renormalization factor [43]) is \( \epsilon_i^{SIC} = \epsilon_i^{SIC} - t_i^{SIC} \sim -0.2 \) eV, as seen in Table IV. On the other hand, for the ABB Hamiltonian, we have \( \epsilon_i^{SIC} = -1.65 \) eV and \( t_i^{SIC} = -4.72 \) eV. As a result, \( \epsilon_i^{SIC} = \epsilon_i^{SIC} - t_i^{SIC} \sim -3.1 \) eV, which explains the large value of \( \Delta E_{ABB} \) within the ABB LEH compared to \( \Delta E_{xp} \) within the \( xp \) LEH.

d. Exclusion of B manifold from the Hamiltonian — For the ABB Hamiltonian, the large \( \Delta E_{ABB} \) suggests that B orbitals are far below the Fermi level and play no role in low-energy physics except for the cRPA-type screening from the B orbitals, so that it is legitimate to exclude them from the LEH. Still, we must take into account the strong electronic correlation encoded within \( U_{AB} \) and \( U_B \) in order to verify this. Here, we propose the following criterion: It is safe to exclude the B manifold from the LEH if the upper Hubbard band from the B manifold (UHB) remains well below the Fermi level, and does not overlap strongly with the lower Hubbard band from the AB manifold (LHB). That is, the charge transfer energy must be larger than or similar to the intraorbital Coulomb repulsion, and the insulating phase of the mother compound should retain the purely AB-like Mott insulating character, without B → AB charge transfer insulating behaviour.

We discuss in detail the latter point for the case of Bi2201, before discussing other compounds. We represent in Fig. 5 the band structure of Bi2201 at the cGW-SIC level, along with the onsite energy \( \epsilon_i \) for each manifold, the charge transfer energy \( \Delta E_{ABB} = \epsilon_{AB} - \epsilon_B \sim 8.2 \) eV, and other quantities defined below. In order to quantify the competition between intra-orbital Coulomb repulsion and charge transfer energy, a first possibility is to estimate the energy of the LHB \( E_{LHB} \) as \( \epsilon_{AB} - U_{AB}/2 \), and the energy of the UHB \( E_{UHB} \) as \( \epsilon_B + U_B/2 \). Equivalently, we define the dimensionless ratio

\[
\mathcal{L}_B^{(0)} = \frac{U_{AB} + U_B}{2\Delta E_{ABB}},
\]

so that \( \mathcal{L}_B^{(0)} \leq 1 \) if \( \epsilon_B + U_B/2 < \epsilon_{AB} - U_{AB}/2 \). For Bi2201, we obtain \( \epsilon_{AB} - U_{AB}/2 \sim -1.7 \) eV and \( \epsilon_B + U_B/2 \sim -5.5 \) eV, so that \( \mathcal{L}_B^{(0)} \sim 0.54 \) is well below 1.

However, the ratio \( \mathcal{L}_B^{(0)} \) in Eq. (2) is not appropriate enough as the criterion. For instance, it ignores the finite bandwidths \( W_{AB} \sim 4.2 \) eV and \( W_B \sim 4.1 \) eV of the AB and B manifolds at the cGW-SIC level (as seen in Fig. 5), which are comparable to \( U_{AB} \sim 4.1 \) eV, \( U_B \sim 4.7 \) eV and \( \Delta E_{ABB} \sim 8.2 \) eV. These finite bandwidths may be retained at least partially by the LHB and UHB when the LEH is solved, and should be taken into account in Eq. (2). Thus, we modify Eq. (2) as follows. We include the effect of \( W_{AB} = \epsilon_{AB} - \epsilon_B \) and \( W_B = \epsilon_B - \epsilon_B \), where \( \epsilon_{AB} \) (respectively, \( \epsilon_B \)) is the minimum (respectively, maximum) value of the band energy for each manifold, as defined in Fig. 5. To do so, we replace \( \Delta E_{ABB} \sim 8.2 \) eV by \( \Delta E_{min}^{AB} = \epsilon_{min}^{AB} - \epsilon_{max}^{B} \sim 5.0 \) eV. Equivalently, we assume that the minimum energy of the LHB is

\[
\epsilon_{min}^{AB} = \epsilon_{min}^{AB} - U_{AB}/2
\]

which is \( \sim -3.4 \) eV for Bi2201, and the maximum energy of the UHB is

\[
\epsilon_{max}^{B} = \epsilon_{max}^{B} + U_B/2
\]

which is \( \sim -4.0 \) eV for Bi2201. We redefine Eq. (2) as

\[
\mathcal{L}_B = \frac{U_{AB} + U_B}{2\Delta E_{min}^{AB}},
\]

and the criterion is satisfied if \( \mathcal{L}_B \lesssim 1 \) and \( \epsilon_{max}^{B} \) is well below the Fermi energy. Values of \( \mathcal{L}_B \) and \( \epsilon_{max}^{B} \) are shown in Table VI. We see that \( \epsilon_{max}^{B} \lesssim -4.0 \) eV for all compounds, which is well below the Fermi energy. In addition, \( \mathcal{L}_B \lesssim 1 \) for all compounds. Thus, LHB and UHB are nearly separated and are not strongly entangled. We note the fact that \( \mathcal{L}_B \) as defined in Eq. (5) is an upper bound, and the true value of \( \mathcal{L}_B \) in the practical resolution of the LEH may be lower because of the band narrowing of each UHB and LHB in comparison to \( W_B \) or \( W_{AB} \) due to the correlation effect as was observed.
E. REDUCTION OF THE COMPUTATIONAL COST OF THE GW CORRECTION OF M SPACE

Here, we propose an approximation to reduce the computational cost of the GW self-energy, without loss of accuracy. This approximation has not been used for calculations in this paper, but may be useful for future studies. One of the most difficult parts of the calculation is the GW correction of the M space: The computational cost of the GW self-energy for the N_M bands in the M space scales as N_M(N_M + 1)/2, which becomes challenging for cuprates with large N_M (e.g., N_M = 11 for Ca11 and 17 for Hg1201, but 23 for Bi2201 and 34 for Bi2212). For each wavevector in the irreducible Brillouin zone, we have to compute N_M diagonal elements and N_M(N_M − 1)/2 off-diagonal elements (ODEs) in the upper triangle; ODEs in the lower triangle may be deduced by Hermitianity. The large computational cost comes from ODEs.

However, in practice, ODEs have a sparse structure, especially for highly symmetric systems. Thus, it is desirable to anticipate which ODEs are negligible, and restrict the calculation of the GW self-energy to finite and important ODEs. To do so, the idea is to use as a guideline the ODEs of the Kohn-Sham exchange-correlation potential V^xc. First, matrix elements of V^xc are much cheaper to calculate than those of the GW self-energy. Second, we remark in practice that negligible (respectively, non-negligible) ODEs for the GW self-energy are also negligible (respectively, non-negligible) for V^xc: Although GW improves exchange and correlation beyond the Kohn-Sham level, the correction is mainly quantitative and does not change the overall structure of the matrix elements.

Thus, the procedure is the following. (1) Compute all N_M(N_M + 1)/2 matrix elements for V^xc; (2) determine ODEs whose amplitude is negligible, e.g., by using a cutoff energy ε; (3) assume these ODEs are negligible in the GW self-energy as well; (4) calculate the GW self-energy only for diagonal elements and non-negligible ODEs. We benchmarked this procedure in the case of Ca11; the cutoff energy ε = 0.05 eV allows to reproduce the GW band structure with excellent accuracy, while reducing the number of ODEs and computational cost by ~40%. Thus, the procedure can be useful for future studies of cuprates with large N_M, especially those for which N_M ≥ 2. The finite cutoff value ε may be determined for another cuprate with small N_M (e.g., Ca11). Then, this cutoff value may be considered for the cuprate with large N_M. Of course, the procedure may also be used for systems other than cuprates.

1. J. G. Bednorz and K. A. Müller. Possible high T_c superconductivity in the Ba-La-Cu-O system. Z. Phys. B, 64, 189, 1986.
2. P. Dai, B. C. Chakoumakos, G. F. Sun, K.W. Wong, Y. Xin, and D. F. Lu. Synthesis and neutron powder diffraction study of the superconductor HgBa_2Ca_2Cu_3O_8+δ by Ti substitution. Physica C: Superconductivity, 243(3):201–206, 1995.
3. M. Nuñez-Regueiro, J. L. Tholence, E. V. Antipov, J. J. Capponi, and M. Marezio. Pressure-induced enhancement of T_c above 150 K in Hg-1223. Science, 262(5130):97–99, 1993.
4. L. Gao, Y. Y. Xue, F. Chen, Q. Xiong, R. L. Meng, D. Ramirez, C. W. Chu, J. H. Eggert, and H. K. Mao. Superconductivity up to 164 K in HgBa_{2-x}Ca_xCu_{2}O_{8+δ} (x=1, 2, and 3) under quasi-hydrostatic pressures. Phys. Rev. B, 50:4260–4263, Aug 1994.
5. Hiroshi Maeda, Yoshiaki Tanaka, Masao Fukutomis, and Toshihisa Asano. A new high-Tc oxide superconductor without a rare earth element. Japanese Journal of Applied Physics, 27(1):209–210, Feb 1988.
[6] C. W. Chu, J. Bechtold, L. Gao, P. H. Hor, Z. J. Huang, R. L. Meng, Y. Y. Sun, Y. Q. Wang, and Y. Y. Xue. Superconductivity up to 114 K in the Bi-Al-Ca-Sr-Cu-O compound system without rare-earth elements. *Phys. Rev. Lett.*, 60:941–943, Mar 1988.

[7] Y. Arao, M. Tange, M. Yokoshima, H. Ikeda, and R. Yoshizaki. Optimization of the Bi-2201 superconductors with Pb and La co-doping. *Physica C: Superconductivity*, 426-431:351–354, 2005. Proceedings of the 17th International Symposium on Superconductivity (ISS 2004).

[8] R. M. Hazen, C. T. Previtt, R. J. Angel, N. L. Ross, L. W. Finger, C. G. Haddadiacos, D. R. Veblen, P. J. Heaney, P. H. Hor, R. L. Meng, Y. Y. Sun, Y. Q. Wang, Y. Y. Xue, Z. J. Huang, L. Gao, J. Bechtold, and C. W. Chu. Superconductivity in the high-$T_c$ Bi-Ca-Sr-Cu-O system: Phase identification. *Phys. Rev. Lett.*, 60:1174–1177, Mar 1988.

[9] M. A. Subramanian, C. C. Torardi, J. C. Calabrese, J. Gopalakrishnan, K. J. Morrissey, T. R. Askew, R. B. Flippin, U. Chowdhry, and A. W. Sleight. A new high-temperature superconductor: $\text{Bi}_2\text{Sr}_3\text{Ca}_n\text{Cu}_6\text{O}_{6+n+y}$. *Science*, 239(4845):1015–1017, 1988.

[10] J. M. Tarascon, W. R. McKinnon, P. Barboux, D. M. Hwang, B. G. Bagley, L. H. Greene, G. W. Hull, Y. LeFarge, N. Stoffel, and M. Giroud. Preparation, structure, and properties of the superconducting compound series $\text{Bi}_2\text{Sr}_2\text{Ca}_n\text{Cu}_{6-n}\text{O}_{y}$ with $n=1,2,$ and 3. *Phys. Rev. B*, 38:8885–8892, Nov 1988.

[11] J. L. Tallon, R. G. Buckley, P. W. Gilberd, M. R. Presland, I. W. M. Brown, M. E. Bowden, L. A. Christian, and R. Goguel. High-$T_c$ superconducting phases in the series $\text{Bi}_{2+\delta}\text{Sr}_{2n+4}\text{Ca}_n\text{Cu}_{6n+4}$.* Nature*, 356:6320–6326, Mar 2000.

[12] C. Michel, M. Hervieu, M. M. Borel, A. Grandin, F. Deslandes, J. Provost, and B. Raveau. Superconductivity in the Bi-Sr-Ca-O system. *Z. Phys. B*, 68:421, 1987.

[13] M. Azuma, Z. Hiroi, M. Takano, Y. Bando, and Y. Takeda. Superconductivity at 110 K in the infinite-layer compound $(\text{Sr}_{1-x}\text{Ca}_x)_{1-y}\text{Cu}_2\text{O}_y$. *Nature*, 356(6372):775–776, 1992.

[14] J. Zaanen, G. A. Sawatzky, and J. W. Allen. Band gaps and electronic structure of transition-metal compounds. *Phys. Rev. Lett.*, 55:418–421, Jul 1985.

[15] C. Weber, C. Yee, K. Haule, and G. Kotliar. Scaling of the transition temperature of hole-doped cuprate superconductors with the charge-transfer energy. *EPL (Europhysics Letters)*, 100(3):37001, nov 2012.

[16] Hirofumi Sakakibara, Hidetomo Usui, Kazuhiko Kuroki, Ryoataro Arita, and Hideo Aoki. Two-Orbital Model Explains the Higher Transition Temperature of the Single-Layer $\text{Hg}$-Cuprate Superconductor Compared to That of the La-Cuprate Superconductor. *Phys. Rev. Lett.*, 105:057003, Jul 2010.

[17] D. C. Peets, D. G. Hawthorn, K. M. Shen, Young-June Kim, D. S. Ellis, H. Zhang, Seiki Komiya, Yoichi Ando, G. A. Sawatzky, Ruixing Liang, D. A. Bonn, and W. N. Hardy. X-Ray Absorption Spectra Reveal the Inapplicability of the Single-Band Hubbard Model to Overdoped Cuprate Superconductors. *Phys. Rev. Lett.*, 103:087402, Aug 2009.

[18] D. C. Peets, D. G. Hawthorn, K. M. Shen, Young-June Kim, D. S. Ellis, H. Zhang, Seiki Komiya, Yoichi Ando, G. A. Sawatzky, Ruixing Liang, D. A. Bonn, and W. N. Hardy. Erratum: X-Ray Absorption Spectra Reveal the Inapplicability of the Single-Band Hubbard Model to Overdoped Cuprate Superconductors [Phys. Rev. Lett. 103, 087402 (2009)]. *Phys. Rev. Lett.*, 104:169903, Apr 2010.

[19] Steven R. White and D. J. Scalapino. Phase separation and stripe formation in the two-dimensional $t-J$ model: A comparison of numerical results. *Phys. Rev. B*, 61:6320–6326, Mar 2000.

[20] Yokoyama, Hisatoshi and Tanaka, Yukio and Ogata, Masao and Tsuchiura, Hiroki. Crossover of Superconducting Properties and Kinetic-Energy Gain in Two-Dimensional Hubbard Model. *Journal of the Physical Society of Japan*, 73(5):1119–1122, 2004.

[21] M. Capone and G. Kotliar. Competition between $d$-wave superconductivity and antiferromagnetism in the two-dimensional Hubbard model. *Phys. Rev. B*, 74:054513, Aug 2006.

[22] Yokoyama, Hisatoshi and Ogata, Masao and Tanaka, Yukio and Kobayashi, Kenji and Tsuchiura, Hiroki. Crossover between BCS Superconductor and Doped Mott Insulator of $d$-Wave Pairing State in Two-Dimensional Hubbard Model. *Journal of the Physical Society of Japan*, 82(1):014707, 2013.

[23] Misawa, Takahiro and Imada, Masatoshi. Origin of high-$T_c$ superconductivity in doped Hubbard models and their extensions: Roles of uniform charge fluctuations. *Phys. Rev. B*, 90:115137, Sep 2014.

[24] Philippe Corboz, T. M. Rice, and Matthias Troyer. Competing States in the $t$-$J$ Model: Uniform $d$-Wave State versus Stripe State. *Phys. Rev. Lett.*, 113:046402, Jul 2014.

[25] Junya Otsuki, Hartmut Hafermann, and Alexander I. Lichtenstein. Superconductivity, antiferromagnetism, and phase separation in the two-dimensional Hubbard model: A dual-fermion approach. *Phys. Rev. B*, 90:235132, Dec 2014.

[26] Zhao, Hui-Hai and Ido, Kota and Morita, Satoshi and Imada, Masatoshi. Variational Monte Carlo method for fermionic models combined with tensor networks and applications to the hole-doped two-dimensional Hubbard model. *Phys. Rev. B*, 96:085103, Aug 2017.

[27] Bo-Xiao Zheng, Chia-Min Chung, Philippe Corboz, Georg Ehlers, Ming-Pu Qin, Reinhard M. Noack, Hao Shi, Steven R. White, Shiwei Zhang, and Garnet Kin-Lic Chan. Stripe order in the underdoped region of the two-dimensional Hubbard model. *Science*, 358(6367):1155–1160, 2017.

[28] Kota Ido, Takahiro Ohgao, and Masatoshi Imada. Correlation-induced superconductivity dynamically stabilized and enhanced by laser irradiation. *Science Advances*, 3(8):e1700718, 2017.

[29] Ido, Kota and Ohgao, Takahiro and Imada, Masatoshi. Competition among various charge-inhomogeneous states and $d$-wave superconducting state in Hubbard models on square lattices. *Phys. Rev. B*, 97:045138, Jan 2018.

[30] Andrew S. Darmawan, Yusuke Nomura, Youhei Yamaji, and Masatoshi Imada. Stripe and superconducting order competing in the Hubbard model on a square lattice studied by a combined variational Monte Carlo and tensor network method. *Phys. Rev. B*, 98:205132, Nov 2018.

[31] Takahiro Ohgao, Motoaki Hirayama, Takahiro Misawa, Kota Ido, Youhei Yamaji, and Masatoshi Imada. *Ab initio* study of superconductivity and inhomogeneity in...
a Hg-based cuprate superconductor. *Phys. Rev. B*, 101:045124, Jan 2020.

[32] Motoaki Hirayama, Takahiro Misawa, Takahiro Ohgoe, Youhei Yamaji, and Masatoshi Imada. Effective Hamiltonian for cuprate superconductors derived from multiscalar *ab initio* scheme with level renormalization. *Phys. Rev. B*, 99:245155, Jun 2019.

[33] S. N. Putilin, E. V. Antipov, Ol. Chnaissim, and M. Marezio. Superconductivity at 94 K in HgBaCuO$_{4+\delta}$. *Nature*, 362(6417):226–228, 1993.

[34] Masatoshi Imada and Takashi Miyake. Electronic Structure Calculation by First Principles for Strongly Correlated Electron Systems. *Journal of the Physical Society of Japan*, 79(11):112001, 2010.

[35] F. Aryasetiawan, M. Imada, A. Georges, G. Kotliar, S. Biermann, and A. I. Lichtenstein. Frequency-dependent local interactions and low-energy effective models from electronic structure calculations. *Phys. Rev. B*, 70:195104, Nov 2004.

[36] F. Aryasetiawan, K. Karlsson, O. Jepsen, and U. Schönberger. Calculations of Hubbard $U$ from first-principles. *Phys. Rev. B*, 74:125106, Sep 2006.

[37] Philipp Werner, Rei Sakuma, Fredrik Nilsson, and Ferdi Aryasetiawan. Dynamical screening in La$_2$CuO$_4$. *Phys. Rev. B*, 91:125142, Mar 2015.

[38] Seung Woo Jang, Hirofumi Sakakibara, Hiori Kino, Takao Kotani, Kazuhiyo Kuroki, and Myung Joon Han. Direct theoretical evidence for weaker correlations in electron-doped and Hg-based hole-doped cuprates. *Scientific Reports*, 33997-6-1, 2014-2322, 2016.

[39] Shingo Teranishi, Kazutaka Nishiguchi, and Koichi Kusakabe. Material-dependent screening of Coulomb interaction in single-layer cuprates. *Journal of the Physical Society of Japan*, 87(11):114701, 2018.

[40] F. Nilsson, K. Karlsson, and F. Aryasetiawan. Dynamically screened Coulomb interaction in the parent compounds of hole-doped cuprates: Trends and exceptions. *Phys. Rev. B*, 99:075135, Feb 2019.

[41] Shingo Teranishi, Kazutaka Nishiguchi, and Koichi Kusakabe. Material Optimization of Potential High-$T_c$ Superconducting Single-Layer Cuprates. *Journal of the Physical Society of Japan*, 90(5):054705, 2021.

[42] Shingo Teranishi, Kazutaka Nishiguchi, Seiji Yunoki, and Koichi Kusakabe. Effect of On-site Coulomb Repulsion on Ferromagnetic Fluctuations in Heavily Overdoped Cuprates. *Journal of the Physical Society of Japan*, 90(9):094707, 2021.

[43] Motoaki Hirayama, Takashi Miyake, and Masatoshi Imada. Derivation of static low-energy effective models by an *ab initio* downfolding method without double counting of Coulomb correlations: Application to SrVO$_4$, FeSe, and FeTe. *Phys. Rev. B*, 87:195144, May 2013.

[44] Motoaki Hirayama, Takahiro Misawa, Takashi Miyake, and Masatoshi Imada. *Ab initio* studies of magnetism in the iron chalcogenides FeTe and FeSe. *Journal of the Physical Society of Japan*, 84(9):093703, 2015.

[45] Motoaki Hirayama, Takashi Miyake, Masatoshi Imada, and Silke Biermann. Low-energy effective Hamiltonians for correlated electron systems beyond density functional theory. *Phys. Rev. B*, 96:075102, Aug 2017.

[46] Motoaki Hirayama, Youhei Yamaji, Takahiro Misawa, and Masatoshi Imada. *Ab initio* effective Hamiltonians for cuprate superconductors. *Phys. Rev. B*, 98:134501, Oct 2018.

[47] Takashi Miyake, Ferdi Aryasetiawan, and Masatoshi Imada. *Ab initio* procedure for constructing effective models of correlated materials with entangled band structure. *Phys. Rev. B*, 80:155134, Oct 2009.

[48] Kazuma Nakamura, Yoshihide Yoshimoto, Yusuke Nomura, Terumasa Tadano, Mitsuaki Kawamura, Taichi Kosugi, Kazuyoshi Yoshimi, Takahiro Misawa, and Yuichi Motoyama. RESPACK: An *ab initio* tool for derivation of effective low-energy model of material. *arXiv preprint arXiv:2001.02351*, 2020.

[49] Kazuma Nakamura, Yoshihide Yoshimoto, Yoshio Nohara, and Masatoshi Imada. *Ab initio* Low-Dimensional Physics Opened Up by Dimensional Downfolding: Application to LaFeAsO. *Journal of the Physical Society of Japan*, 79(12):123708, 2010.

[50] J.B. Torrance, Y. Tokura, S.J. LaPlaca, T.C. Huang, R.J. Savoy, and A.I. Nazal. New class of high $T_c$ structures: Intergrowth of multiple copper oxide perovskite-like layers with double sheets of BiO. *Solid State Communications*, 66(7):703–706, 1988. 25th Anniversary Year.

[51] J. Karpinski, H. Schwer, I. Mangelschots, K. Conder, A. Morawska, T. Lada, and A. Paszewin. Single crystals of Hg$_{1-x}$Pb$_x$Ba$_{2-x}$Ca$_2$Cu$_3$O$_{7-2x-\delta}$ and infinite-layer Ca$_x$CuO$_{2+y}$ synthesis at gas pressure 10 kbar, properties and structure. *Physica C: Superconductivity*, 234(1):10–18, 1994.

[52] Paolo Giannozzi, Stefano Baroni, Nicola Bonini, Matteo Calandra, Roberto Car, Carlo Cavazzoni, Davide Ceresoli, Guido L Chiarotti, Matteo Cococcioni, Ismaila Dabo, Andrea Dal Corso, Stefano de Gironcoli, Stefano Fabris, Guido Fratesi, Ralph Gebauer, Uwe Gerstmann, Christos Gougoussis, Anton Kokalj, Michele Lazzeri, Layla Martin-Samos, Nicola Marzari, Francesco Mauri, Riccardo Mazzarello, Stefano Paolini, Alfredo Pasquarello, Lorenzo Paulatto, Carlo Sbraccia, Sandro Scandolo, Gabriele Scuazerio, Ari P Seitsonen, Alexander Smogunov, Paolo Umari, and Renata M Wentzcovitch. QUANTUM ESPRESSO: a modular and open-source software project for quantum simulations of materials. *Journal of Physics: Condensed Matter*, 21(39):395502 (19pp), 2009.

[53] P. Giannozzi, O Andreussi, T Brumme, O Bunau, M Buongiorno Nardelli, M Calandra, R Car, C Cavazzoni, D Ceresoli, M Cococcioni, N Colonna, I Carmimeo, A Dal Corso, S de Gironcoli, P Dehugua, R A DiStasio Jr, A Ferretti, A Floris, G Fratesi, G Fugallo, R Gebauer, U Gerstmann, F Giustino, T Gorni, J Jia, M Kawamura, H-Y Ko, A Kokalj, E Küçükbenli, M Lazzeri, M Lazzeri, M Marsili, N Marzari, F Mauri, N L Nguyen, H-V Nguyen, Oto der-la Roza, L Paulatto, S Poncè, D Rocca, R Sadowski, M Schlipf, A P Seitsonen, Alexander Smogunov, Paolo Umari, and Renata M Wentzcovitch. QUANTUM ESPRESSO: a modular and open-source software project for quantum simulations of materials. *Journal of Physics: Condensed Matter*, 21(39):395502 (19pp), 2009.

[54] Martin Schlipf and François Gygi. Optimization algorithm for the generation of oncv pseudopotentials. *Computer Physics Communications*, 196:36 – 44, 2015.

[55] John P. Perdew, Kieron Burke, and Matthias Ernzerhof. Generalized Gradient Approximation Made Simple. *Phys. Rev. Lett.*, 77:3865–3868, Oct 1996.

[56] D. R. Hamann. Optimized norm-conserving Vanderbilt pseudopotentials. *Phys. Rev. B*, 88:085117, Aug 2013.
