

# Superconducting properties of the hole-doped three-band $d - p$ model studied with minimal-size real-space $d$ -wave pairing operators

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The three-band  $d$ - $p$  model is investigated by means of a variational Monte Carlo (VMC) method with the BCS-like wave-function supplemented by the Gutzwiller and Jastrow correlators. The VMC optimization leads to a  $d$ -wave superconducting state with a characteristic domelike shape of the order parameter for hole doping  $\delta \lesssim 0.4$ , in good agreement with the experimental observations. Also, the off-diagonal pair-pair correlation functions, calculated within VMC, vindicates the results obtained very recently within the diagrammatic expansion of the Gutzwiller wave function method [cf. Zegrodnik, Biborski, Fidrysiak, and Spałek, *Phys. Rev. B* **99**, 104511 (2019)]. Subsequently, the nature of the  $d$ -wave pairing is investigated by means of recently proposed *minimal-size real-space  $d$ -wave pairing operators* [Moreo and Dagotto, *Phys. Rev. B* **100**, 214502 (2019)]. An emergence of the long-range superconducting ordering for both  $d$  and  $p$  orbitals is reported by analyzing the corresponding off-diagonal pair-pair correlation functions. The dominant character of  $d$ -wave pairing on  $d$  orbitals is confirmed. Additionally, the trial wave function is used to investigate the magnetic properties of the system. The analysis of spin-spin correlation functions is carried out and shows antiferromagnetic  $\mathbf{q} = (\pi, \pi)$ , short-range order, as expected. For the sake of completeness, the charge gap has been estimated, which for the parent compound takes the value  $\Delta_{CG} \approx 1.78 \pm 0.51$  eV, and agrees with values reported experimentally for the cuprates.

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## I. INTRODUCTION

The unconventional superconductivity discovered in copper-based compounds by Bednorz and Müller in 1986 is still under intensive debate [1]. This class of systems is difficult to handle realistically by means of the most popular quantum chemistry method, i.e., density functional theory (DFT), due to the fact that the electron-electron interactions play a crucial role in the resulting physical properties. As electronic correlations cannot be described consistently within any known mean-field formalism (e.g., *double counting* problem in DFT methods), simplified models capturing the essentials of electronic structure are required. The application of the canonical single-band models used for recaption of the correlated systems (Hubbard and  $t$ - $J$  models [2,3]) allowed for the reproduction of both the Mott insulating phase at half-filling and the superconducting state for the electron- and hole-doped cases. In such approaches, the initial multi-band problem ( $d$ - $p$  model) is mapped onto a single-band picture in which the Zhang-Rice singlets [4] play the role of *quasiparticles*. It is believed that many of the unusual properties of the cuprates arise from the electronic degrees of freedom of the copper-oxygen planes, which are common for the whole cuprate family. Although the mentioned models

allow us to reproduce the selected fundamental features of the cuprates, other subtle phenomena such as charge(spín)-density-waves or nematicity appearance may directly emerge from the interplay between  $d$  and  $p$  orbitals [5]. However, the question of the minimal model which captures the cuprate physics to a satisfactory extent still remains an open issue and ongoing research of both single and multiband approaches is in place. In this respect, it is worth noting that the description of the mentioned ordered phases within the single band picture has recently led to some interesting results [6–9]. Nevertheless, microscopic insight into the pairing between  $d$ - $d$ ,  $p$ - $d$ , and  $p$ - $p$  channels can lead to better understanding of the superconducting state [10], as suggested by some of the experimental observations [11,12]. Therefore, it is natural to consider a more realistic model in which the unit cell consists of one  $d$  orbital and two  $p$  orbitals. Regardless of the number of bands considered, the exact ground state for Hubbard-type Hamiltonians (excluding selected  $1d$  cases) is not known. Therefore, approximate methods are to be used in its diagonalization procedure. Whereas exact diagonalization (ED) techniques provide an accurate numerical solution, they are limited to small systems, which essentially cannot give answers related to the presence of the long-range electronic correlations. The state-of-art density matrix renormalization group method, though computationally demanding, has been profitably exploited for studying both charge order as well as pairing in strongly correlated model systems [6,7]. The determinant Monte Carlo (DMC) calculations, despite the infamous *sign problem*, are promising for the description of

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the cuprates [13]. However, the paired state has not been explicitly included in such analysis. Another choice is the application of *variational* methods which may be considered as well balanced in view of their complexity and reliability of the obtained results. Therefore, a properly constructed trial wave function allows us to gain insight into the nature of the ground state of the particular correlated electronic system [14].

Encouraged by the results for the superconducting and nematic states obtained by means of the diagrammatic expansion of the Gutzwiler wave function (DE-GWF method) approach for the three-band  $d$ - $p$  model [15,16], we have decided to characterize the superconducting properties, particularly in view of the spatial dependence of the correlation functions (CFs) obtained by means of the variational Monte Carlo (VMC) calculation scheme. Numerous studies regarding this topic have been carried out [13,16–25], related to both *normal* and *superconducting* states. Here, we extend the analysis of the superconducting (SC) state with explicit calculations of the *minimal-size real-space  $d$ -wave pairing operators* (MSPOs) proposed very recently by Moreo and Dagotto [10]. To the best of our knowledge, their equal-time CFs have not been analyzed so far. We also supplement our analysis of the variational *ansatz* for the paired state within the  $d$ - $p$  model with intersite Jastrow-type correlators.

In the next section, we describe the model and sketch the method. Subsequently, in Sec. III we present the characteristics of the  $d$ -wave superconducting phase for the hole-doped case by means of the *standard* investigation, i.e., by analyzing the CFs for the  $d$ -wave pairing between holes residing on the NN  $d$  orbitals. Subsequently, we continue our analysis of the CFs defined for the MSPOs, consisting of a proper combination of the  $d$ - $p$  and  $p$ - $p$  pairing amplitudes. In Sec. IV, we also provide the spin-spin CFs and show the development of the short-range antiferromagnetic (AF) order, as well as determine the value of the charge transfer gap. We conclude our results in the last section.

## II. THREE-BAND $d$ - $p$ MODEL AND METHOD

We consider the three-band  $d$ - $p$  model described by the Hamiltonian

$$\hat{H} = \sum_{(il,jl')} t_{ij}'' \hat{c}_{il\sigma}^\dagger \hat{c}_{jl'\sigma} + \sum_{il} \epsilon_l \hat{n}_{il} + \sum_{il} U_l \hat{n}_{il\uparrow} \hat{n}_{il\downarrow}, \quad (1)$$

where  $\hat{c}_{il\sigma}^\dagger$  ( $\hat{c}_{il\sigma}$ ) are creation (annihilation) fermionic operators acting on orbital  $l \in \{d_{x^2-y^2}, p_x, p_y\}$  related to  $i$ th unit cell. As in our previous works [15,16], hoppings are limited to the NN orbitals (cf. Fig. 1). The values of hopping amplitudes  $t_{ij}''$  as well as atomic energy levels are set to  $t_{pp} = 0.49$  eV,  $t_{pd} = 1.13$  eV,  $\epsilon_p = -3.57$  eV, and  $\epsilon_d = 0$ , which are typical values for the cuprates [3]. The repulsive intraorbital Hubbard interactions are  $U_{p_x} = U_{p_y} = 4.1$  eV and  $U_{d_{x^2-y^2}} = 10.3$  eV for oxygen and copper orbitals, respectively. This set of microscopic parameters was utilized recently in our DE-GWF solution [16] and their values are similar to those applied by Kung *et al.* [13].

As mentioned, we employ VMC approach in this study. This method exhibits both advantages as well as drawbacks which are common for the whole family of variational

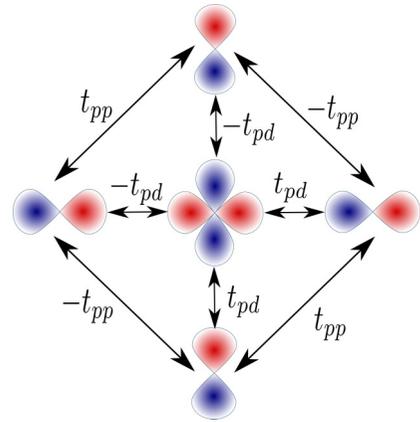


FIG. 1. The hopping parameters included in three-band  $d$ - $p$  model described by Hamiltonian defined in Eq. (1). Central orbital is  $d_{x^2-y^2}$ , states for to the copper atom whereas, remaining one are the oxygen  $p_x/p_y$  orbitals.

methods. Particularly, the proper choice of wave-function *ansatz* is crucial to obtain reasonable output. Thus, one cannot expect to obtain properties of the system which are not encoded in the variational wave function since the solution is narrowed to the subspace in the Hilbert space. Therefore, the specific form of the wave function should be carefully chosen. This issue does not appear in the determinant quantum Monte Carlo (DQMC), which in principle (disregarding controllable approximations) provides an *exact* solution for the nonzero temperature. However, DQMC suffers from the infamous *sign problem* and is more complex from the algebraic perspective. It must be stressed that the variational wave function in the VMC approach can be formulated in the manner which ensures reasonable computational costs and flexibility which is necessary to describe correlated system in many cases. Also, one of the main advantages of VMC in the context of strongly correlated systems is its *universality*, e.g., long-range interactions, as well as three- and four-center two-body terms which can be encompassed almost effortlessly when the dose of generality is applied during the process of code development.

Our many-body trial wave-function is taken in the following manner [14,26]:

$$|\Psi_T\rangle \equiv \hat{P}_G \hat{P}_J \hat{\mathcal{L}}_{S_{\text{tot}}}^c \hat{\mathcal{L}}_{N_e} |\Psi_0\rangle, \quad (2)$$

where  $\hat{P}_G$  is the Gutzwiller-type correlator given in the form

$$\hat{P}_G \equiv \exp \left[ - \sum_l g_l \sum_i \hat{n}_{il\uparrow} \hat{n}_{il\downarrow} \right], \quad (3)$$

with  $g_{p_x} = g_{p_y}$  due to the equivalency of the oxygen orbitals. The interorbital correlations are captured by the symmetric Jastrow density-density correlator:

$$\hat{P}_J \equiv \exp \left[ - \sum_{il,jl'} \lambda_{il,jl'} \hat{n}_{il} \hat{n}_{jl'} \right]. \quad (4)$$

Both  $\{g_l\}$  and  $\{\lambda_{il,jl'}\}$  are the subsets of variational parameters. When performing calculations for the  $z$  component of the total spin, we set  $S_{\text{tot}}^z = 0$  and constant number of electrons  $N_e$ , as well as the projectors  $\hat{\mathcal{L}}_{S_{\text{tot}}}^c$  and  $\hat{\mathcal{L}}_{N_e}$  are applied during sam-

pling procedure. The noninteracting part  $|\Psi_0\rangle$  is constructed from eigenstates of the BCS variational Hamiltonian  $\hat{\mathcal{H}}_{\text{eff}}$  defined as

$$\hat{H}_{\text{eff}} = \sum_{(il,jl')} \tilde{t}_{ij}^{ll'} \hat{c}_{il\sigma}^\dagger \hat{c}_{jl'\sigma} + \sum_{il} (\tilde{\epsilon}_l - \tilde{\mu}) \hat{n}_{il} + \sum_{il,jl'} \tilde{\Delta}_{ij}^{ll'} \hat{c}_{il\uparrow}^\dagger \hat{c}_{jl'\downarrow} + \text{H.c.} \quad (5)$$

Note that parameters with tildes are *different* than those in the noninteracting part of the  $d$ - $p$  Hamiltonian [Eq. (1)] as they are considered as variational parameters to be optimized. More precisely, the above effective Hamiltonian defines the uncorrelated wave function  $|\Psi_0\rangle$ . The choice of both the hopping terms and the pairing amplitudes is thus identical to our DE-GWF study [16], i.e., it allows for the emergence of  $d$ -wave pairing. It is worth mentioning that during preliminary studies we have analyzed both  $d$ - and  $s$ -wave pairing scenarios within the DE-GWF approach, and, the  $d$ -wave turned out to be the stable one. Therefore, we have  $\Delta_{i+a_x,j}^{dd} = -\Delta_{i,j+a_y}^{dd}$ , where  $a_x/a_y = a$  refers to the nearest neighbor (NN)  $d$  orbital in  $x$  and  $y$  directions, respectively. The diagonalization of Hamiltonian Eq. (5), which in turn allows us to compose the many-electron part  $|\Psi_0\rangle$ , is divided into two stages. First, the following transformation of creation (annihilation) operators is applied. The spin-down sector is converted to the hole picture, i.e.,  $\hat{c}_{il,\downarrow}^\dagger \rightarrow \hat{f}_{il,\downarrow}$  and  $\hat{c}_{il,\downarrow} \rightarrow \hat{f}_{il,\downarrow}^\dagger$ . Whereas the spin-up-sector operators are subject to the identity transformation, i.e.,  $\hat{c}_{il,\uparrow}^\dagger (\hat{c}_{il,\uparrow}) \rightarrow \hat{f}_{il,\uparrow}^\dagger (\hat{f}_{il,\uparrow})$ . The spin-down-sector transformation leads to the form of variational Hamiltonian, which can be directly diagonalized [14], by finding (numerically) the unitary transformation for the  $N$ -orbital system.

In effect,  $|\Psi_0\rangle$  is defined in the standard manner, namely,

$$|\Psi_0\rangle_{\{\tilde{t}_{ij}^{ll'}, \tilde{\epsilon}_l, \tilde{\mu}, \tilde{\Delta}_{ij}^{ll'}\}} = \prod_{p=1}^{p=\tilde{n}} \hat{\gamma}_p^\dagger |\tilde{0}\rangle, \quad (6)$$

where  $|\tilde{0}\rangle$  is the vacuum state for operators  $\hat{\gamma}_p^\dagger$  ( $\hat{\gamma}_p$ ) representing quasiparticles for which the variational Hamiltonian  $\hat{\mathcal{H}}_{\text{eff}}$  can be written in the diagonal form. Note that index  $p = 1, 2, \dots, \tilde{n}$  runs over first  $\tilde{n}$  single-particle eigenstates of the variational Hamiltonian with  $\tilde{n} = N + \sum_{il} (\hat{n}_{il\uparrow} - \hat{n}_{il\downarrow})$ , resulting directly from the particle-hole transformation for the down spins. The sampling procedure is executed in the standard manner. Configurations representing the distribution of  $\tilde{n}$  particles among  $N$  orbitals,  $\{|x\rangle\}$ , are sampled by means of the Metropolis-Hastings [14] algorithm according to the probability density  $\rho(x) \propto |\langle x | \Psi_T \rangle|^2$ . Physical quantities related to operators  $\{\hat{O}\}$  are estimated as an average of their so-called *local values* [14]  $O_{\text{loc}}(x)$ ,

$$\langle \hat{O} \rangle \approx \frac{1}{M} \sum_{m=1}^M \frac{\langle x_m | \hat{O} | \Psi_T \rangle}{\langle x_m | \Psi_T \rangle} \equiv \sum_{m=1}^M O_{\text{loc}}(x_m), \quad (7)$$

with  $|x_m\rangle$  generated with respect to the probability density  $\rho(x)$ . In particular, the expectation value of the system energy, i.e.,  $\langle \hat{\mathcal{H}} \rangle$  can be computed for a given set of variational parameters. At least two commonly exploited strategies for the wave-function optimization exist: *variance optimization* and *energy optimization*. Sorella *et al.* elaborated the ef-

ficient procedure—*stochastic reconfiguration* (SR) method [14], which benefits in simultaneous optimization steps for the whole set of variational parameters. We have implemented the SR-based approach in our self-developed code (also recently used in a different context [27]) as it is regarded as the state-of-art method in the field of interest [26].

### III. RESULTS

In our computations, the system is represented by the square cluster containing  $L \times L = 64$  unit cells, each consisting of one  $d$  orbital and two  $p$  orbitals ( $p_x$  and  $p_y$ ). This results in 64 copper and 128 oxygen atoms represented by appropriate orbitals. For the sake of clarity, we define doping parameter  $\delta$ ,

$$\delta \equiv 5 - \frac{N_e}{L^2}, \quad (8)$$

i.e., the parent compound refers to  $\delta = 0$  with five electrons per  $\text{CuO}_2$  complex, and  $\delta > 0$  corresponds to the hole-doped complex with  $N_e < 5$ . We assume  $S_{\text{tot}}^z = 0$ ; therefore minimal doping resolution is  $\Delta\delta = 2/64 = 0.03125$ . The trial wave function is minimized with respect to the set of variational parameters by means of the SR method, and probed averages  $\langle \hat{O} \rangle$  are sampled within  $M \propto 10^7$  Monte-Carlo (MC) steps. Also, since VMC operates in the real-space representation and the considered cluster is *finite*, we apply periodic boundary conditions.

#### A. Superconducting correlations

Within the VMC approach, the superconducting properties of the system are typically determined by analyzing the appropriate anomalous CFs (equal-time two-body Green's functions). However, the choice of CF is not unique and one may find a particular form more suitable than another in the given methodological context [10]. First, we analyze the pairing between two  $d$ - $d$  holes by means of *standard* equal-time CFs [10,18,19,28] commonly used in the analysis of superconducting states in real space. This part is regarded as validation of the applied method in view of our earlier DE-GWF solution [16]. Next, we have applied the recently proposed [10] MSPOs by means of their spatial dependency of CFs, to determine the  $d$ -wave pairing properties within the three-band  $d$ - $p$  model. Specifically, these pairing operators refer to the possibility of intra- $p$  Cooper pair formation [10,23,29].

#### 1. Standard correlation functions

To inspect fundamental superconducting properties, as well as to compare the results obtained by means of VMC with those of DE-GWF solutions, we first analyzed the spatial dependence of *standard* off-diagonal pair-pair CFs for  $d$ - $d$  pairs, which is defined as

$$D_{\alpha\beta}^{dd}(\mathbf{R}) \equiv \frac{1}{L^2} \sum_{\mathbf{r}} \langle \hat{\Delta}_{\alpha}^{\dagger}(\mathbf{r} + \mathbf{R}) \hat{\Delta}_{\beta}(\mathbf{r}) \rangle, \quad (9)$$

with  $\alpha, \beta \in \{x, y\}$  and

$$\hat{\Delta}_{\alpha}^{\dagger}(\mathbf{r}) \equiv \frac{1}{\sqrt{2}} (\hat{c}_{i(\mathbf{r})d\uparrow}^{\dagger} \hat{c}_{j(\mathbf{r}+\mathbf{a}_{\alpha})d\downarrow}^{\dagger} - \hat{c}_{i(\mathbf{r})d\downarrow}^{\dagger} \hat{c}_{j(\mathbf{r}+\mathbf{a}_{\alpha})d\uparrow}^{\dagger}). \quad (10)$$

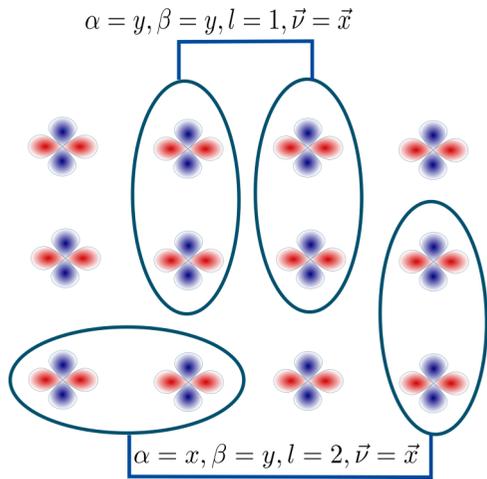


FIG. 2. Schematic representation of exemplary  $d$ - $d$  pair-pair terms present in the correlation functions defined in Eq. (9).

Function  $i(\mathbf{r})$  maps the position of the center of the given orbital  $d$  onto the index  $i$ . Vectors  $\mathbf{a}_\alpha$  are given as

$$\mathbf{a}_x = \begin{pmatrix} a \\ 0 \end{pmatrix}, \mathbf{a}_y = \begin{pmatrix} 0 \\ a \end{pmatrix}, \quad (11)$$

where  $a$  is the lattice parameter. The functions defined in Eq. (9) describe the spatial distribution of anomalous pair-pair correlations, where each pair consists of two  $d$  orbitals separated by the lattice constant  $a$  in  $x$  or  $y$  directions (cf. Fig. 2). Note that as we analyze pure  $d$ -wave pairing, the relation  $D_{\alpha\beta}^{dd}(\mathbf{R}) = D_{\beta\alpha}^{dd}(\mathbf{R})$  holds. It also should be mentioned that maximal distance refers to  $\mathbf{R}_{\max} = (\frac{L}{2}, \frac{L}{2})$  as we apply the periodic boundary conditions to the system. In Fig. 3, we present the spatial dependence of  $D_{\alpha\beta}^{dd}$  for the selected direction  $\mathbf{R} \parallel \mathbf{x}$ . It comes out that  $D_{\alpha\alpha}^{dd} \approx -D_{\alpha\beta}^{dd}$  within the limit of attainable distance. Moreover, the values for  $|\mathbf{R}| \geq 2a$  approach saturation, though for the high doping regime, correlations do not decay to zero (within the statistical error  $\propto 10^{-3}$ ). However, in the accessible maximal distance, we obtain very good agreement when compared to our recent

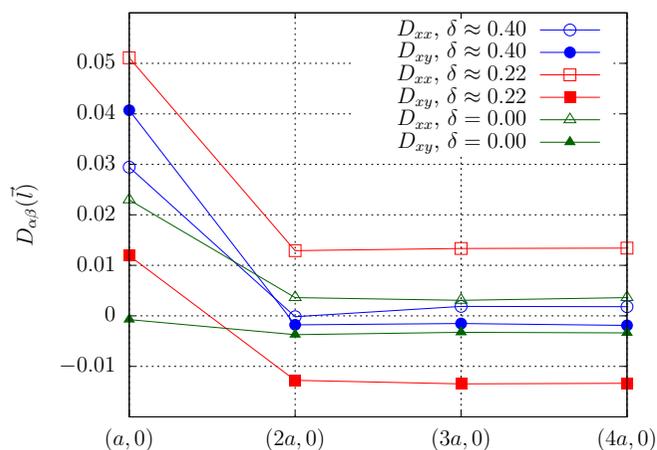


FIG. 3. Correlation functions defined in Eq. (9), for the three representative hole dopings and with  $\mathbf{R}$  parallel to  $x$  direction.

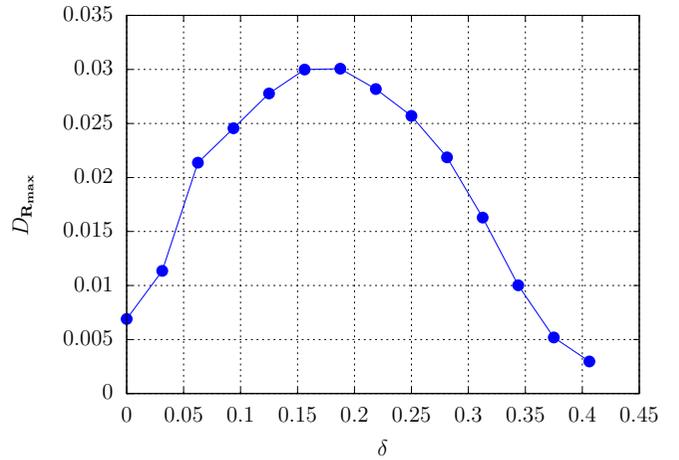


FIG. 4. Superconducting order parameter  $D_{\mathbf{R}_{\max}}$ , the line is guide for the eye.

analysis [16]. In Fig. 4, we present a superconducting order parameter for the  $d$ -wave-pairing defined as

$$D_{\mathbf{R}_{\max}} \equiv \sum_{\alpha\beta} (-1)^{1-\delta_{\alpha\beta}} D_{\alpha\beta}^{dd}(\mathbf{R} = \mathbf{R}_{\max}). \quad (12)$$

We obtain a qualitative agreement when compared to the DE-GWF solution [16], namely, the maximal amplitude of the order parameter appears at  $\delta \approx 0.15 - 0.2$ . As already mentioned, the nonzero amplitude is present for each considered doping. This fact is due to a slow convergence of variational parameters in the high hole-doping regime and/or related to the limited cluster dimension. Contrary to the DE-GWF solution, we find it less problematic to optimize wave function for  $\delta$  in the vicinity of the parent compound. In spite of  $D(\delta = 0) > 0$ , an abrupt decrease of the order parameter for  $\delta \lesssim 0.1$  occurs and the obtained values of the order parameter form the dome-like shape as a function of  $\delta$ , characteristic of the cuprate family. We compare both methods quantitatively by computing the expectation values of NN ( $i, j$ )  $d$ -orbital pairs, namely,

$$\Delta_{dd} \equiv \langle \hat{c}_{id\uparrow}^\dagger \hat{c}_{jd\downarrow}^\dagger \rangle, \quad (13)$$

which is the measure of the superconducting order in the infinite system size, i.e., when

$$|\langle \hat{c}_{i(\mathbf{r})d\uparrow}^\dagger \hat{c}_{j(\mathbf{r}+\mathbf{a})d\downarrow}^\dagger \rangle|^2 \approx \lim_{|\mathbf{R}| \rightarrow \infty} \langle \hat{\Delta}^\dagger(\mathbf{r} + \mathbf{R}) \hat{\Delta}(\mathbf{r}) \rangle. \quad (14)$$

The comparison of  $\Delta_{dd}$  for both methods shows that DE-GWF and VMC provide quantitatively similar results (cf. Fig. 5), as expected, since both approaches have been supplied with the similar form of variational ansatz. It also suggests that the presence of Jastrow terms in our wave function does not affect the solution, at least in view of  $d$ -wave pairing on  $d$  orbitals. The discrepancies appearing for higher hole doping are possibly caused by the presence of the finite-size effects [30] which are absent in the DE-GWF solution (excluding diagrams summations radius in the real space) or optimization issues, as mentioned above. We also observe nonzero pairing amplitudes for  $\delta = 0$ . We estimated previously [16] that  $U_d \gtrsim 13$  eV possibly leads to the full reduction of

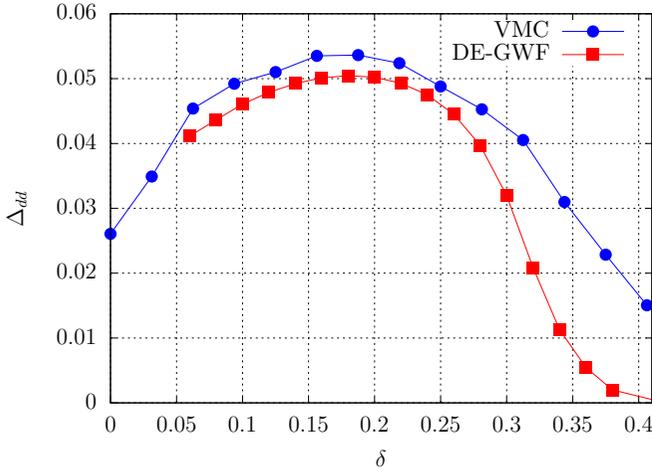


FIG. 5. The comparison of  $\Delta_{dd}(\delta)$  between DE-GWF [16] and VMC approaches. The microscopic parameters are listed in Sec. II.

*d*-wave pairing for the parent compound. The comparison of the results obtained from both methods validates the solution procured for the *assumed form of the wave-function ansatz*. However, characteristics related to the magnetic properties, as well as estimation of charge gap (CG) value (presented in the following subsections) may suggest that the adopted form of the variational state allows us to reproduce the main features of the three-band *d* – *p* model.

## 2. Minimal-size real-space *d*-wave pairing operator correlation functions

Moreo and Dagotto [10] emphasize that the local *d*-wave operators can provide a more suitable description of the paired holes in cuprates. Their arguments are based on the recent experimental observation of a surprisingly small real-space extension [12] of the Cooper pairs. They also analyze this issue in view of the *p* – *p* Cooper pair formation within the single *plaquette*. In this paper, we compute the MSPO CFs in the framework of the VMC method.

All four MSPOs preserve *d*-wave symmetry. The intrasite *p*-orbital pair correlation operator is defined as

$$\hat{\Delta}_{D_0}^\dagger(\mathbf{r}) \equiv \sum_{\mu} \gamma_{\mu} \hat{c}_{i(\mathbf{r}+\mathbf{a}_{\mu}/2)\uparrow}^\dagger \hat{c}_{i(\mathbf{r}+\mathbf{a}_{\mu}/2)\downarrow}^\dagger, \quad (15)$$

where  $\gamma_{\mu} = \text{sgn}(\mu)$ . The analysis of time evolution of  $\hat{\Delta}_{D_0}^\dagger$  (i.e., Heisenberg equation  $-i\frac{d\hat{\Delta}_{D_0}^\dagger}{dt} = [\hat{H}, \hat{\Delta}_{D_0}^\dagger]$ ) also provides other pairing operators in the systematic and elegant manner. Namely, the *d*-*p* pairing operator is given as

$$\hat{\Delta}_{D_{pd}}^\dagger(\mathbf{r}) \equiv \sum_{\mu,\sigma} f_{\sigma} \gamma_{\mu} \alpha_{i(\mathbf{r}),\mu} \hat{c}_{i(\mathbf{r})\sigma}^\dagger \hat{c}_{i(\mathbf{r}+\mathbf{a}_{\mu}/2)\bar{\sigma}}^\dagger, \quad (16)$$

where  $\mu \in \{\pm x, \pm y\}$ , and  $f_{\sigma} = \text{sgn}(\sigma)$  with  $\text{sgn}(\sigma) = -\text{sgn}(\bar{\sigma})$ , and  $\alpha_{i(\mathbf{r}),\mu} = \pm 1$ , consistent with the *d*-*p* hopping sign convention (cf. Fig. 1). The other two MSPOs of intra-*p* type,  $\hat{\Delta}_{D_{pp}}^\dagger$  and  $\hat{\Delta}_{D_{plaq}}^\dagger$ , are also obtainable in such a procedure and are defined as

$$\hat{\Delta}_{D_{pp}}^\dagger(\mathbf{r}) \equiv \sum_{\mu,\sigma} f_{\sigma} \gamma_{\mu} \hat{c}_{i(\mathbf{r}+\mathbf{a}_{\mu}/2)\sigma}^\dagger \hat{c}_{i(\mathbf{r}-\mathbf{a}_{\mu}/2)\bar{\sigma}}^\dagger \quad (17)$$

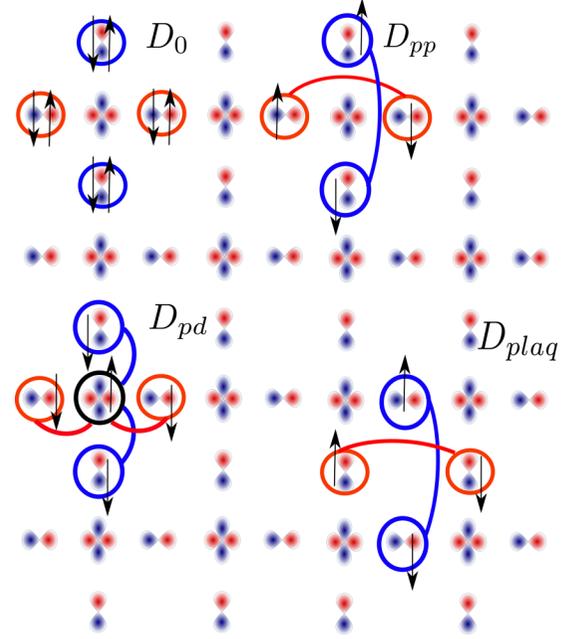


FIG. 6. Pairing operators provided in Ref. [10] for which the correlation functions have been computed. Cooper pairs are assigned by connecting lines (excluding  $D_0$  for which pair occupies single *p* orbital). Relative phase signs are marked by colors.

and

$$\hat{\Delta}_{D_{plaq}}^\dagger(\mathbf{r}) \equiv \sum_{\mu,\sigma} f_{\sigma} \gamma_{\mu} \hat{c}_{i(\mathbf{r}+\mathbf{a}_{\mu}/2)\sigma}^\dagger \hat{c}_{i(\mathbf{r}+\mathbf{a}_{\bar{\mu}}+\mathbf{a}_{\mu}/2)\bar{\sigma}}^\dagger, \quad (18)$$

where  $\mu \perp \bar{\mu}$ . A schematic representation of the above operators is shown in Fig. 6. It should be noted that these operators are not independent *by construction*. Also, if the ground state reflects the *d*-wave superconductivity, simultaneous emergence of all the long-range orderings encoded in Eqs.(15)–(18) is expected. This important feature can be utilized for the characterization of the system ground state. We define the CFs of these pairing operators in the *standard* manner case [cf. Eq. (9)], i.e.,

$$D_{D\tau}(\mathbf{R}) \equiv \frac{1}{L^2} \sum_{\mathbf{r}} \langle \hat{\Delta}_{D\tau}^\dagger(\mathbf{r} + \mathbf{r}) \hat{\Delta}_{D\tau}(\mathbf{r}) \rangle, \quad (19)$$

where  $\tau \in \{0, pd, pp, plaq\}$ .

In Figs. 7(a)–7(d), we present spatial dependency of MSPO CFs for the representative set of dopings. Disregarding fluctuations originating both from sampling and optimization effects, we observe the saturation of their values within the relatively short distance, i.e.,  $\mathbf{R} \approx (3a, 0)$ . Furthermore, all amplitudes fit the picture resulting from *standard* analysis. Namely, the highest amplitude for the most distant pair corresponds to  $\delta \approx 0.2$ , i.e., the optimal doping. Importantly, for the parent compound, we obtain a nearly vanishing value of  $D_{D_0}$  [cf. Fig. 7(a)]. The residual nonzero values for  $D_{D_0}$  and  $D_{D_{pd}}$  at high hole dopings are present, nevertheless they are significantly smaller than for  $\delta \approx 0.2$ .

It is reasonable to compare the above amplitudes with the *dominant d*-*d* gap. The order parameter defined in Eq. (12) is normalized by  $(1/\sqrt{2})^2$  factor, what is not the case for

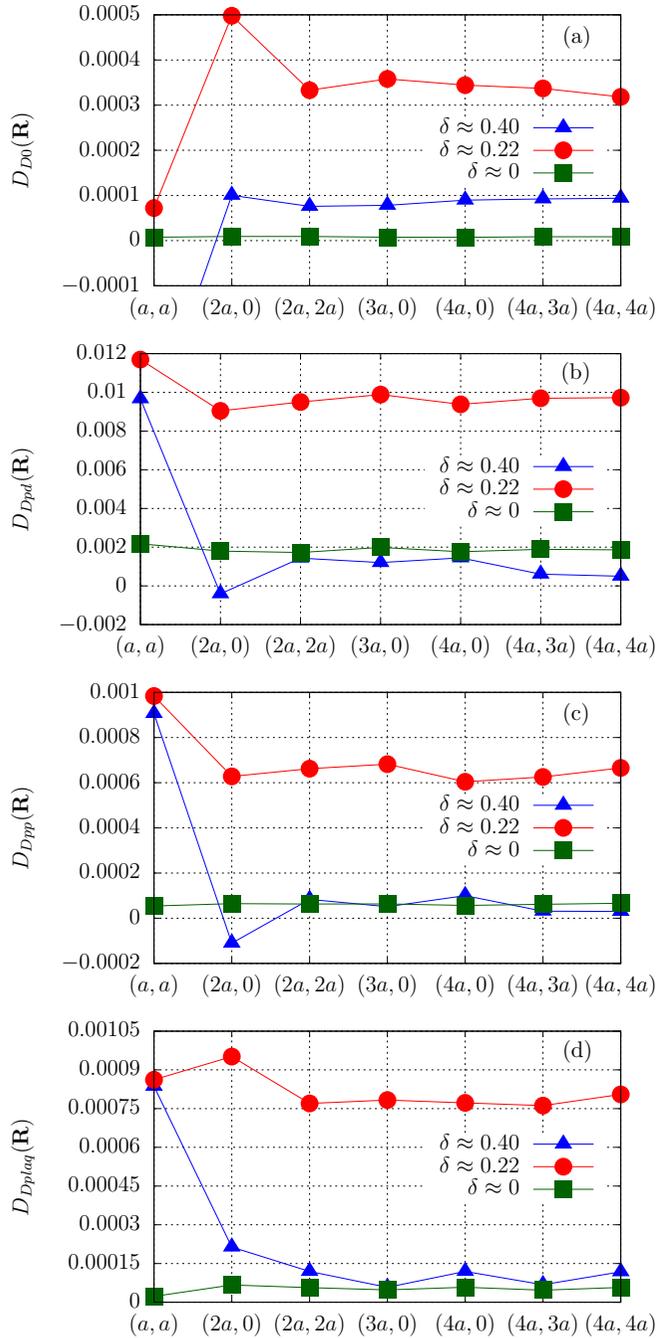


FIG. 7. Spatial dependence of the correlation functions  $D_{D0}(\mathbf{R})$ ,  $D_{Dpd}(\mathbf{R})$ ,  $D_{Dpp}(\mathbf{R})$ , and  $D_{Dplaq}(\mathbf{R})$  for three representative dopings. The amplitude for  $\delta \approx 0.22$  dominates with increasing  $|\mathbf{R}|$  when compared to that for parent compound and high doping cases, as expected.

the MSPO CFs. Therefore,  $D_{Ddd} \equiv 2 \times D_{\mathbf{R}_{\max}}$  should be compared to  $D_{D\tau}$  at the  $(4a, 4a)$  distance. This results in the ratio  $D_{Ddd}/D_{Dpd}$  being  $\approx 6$  for  $\delta \approx 0.2$ , which is in a good agreement with results obtained from DE-GWF approach [16]. Thus  $d$ - $d$  pairing can indeed be regarded as the dominant one for the considered form of the variational state.

In Fig. 8, we present the values of all MSPO CFs for the maximal attainable  $R = (4a, 4a)$  as a function of hole doping.

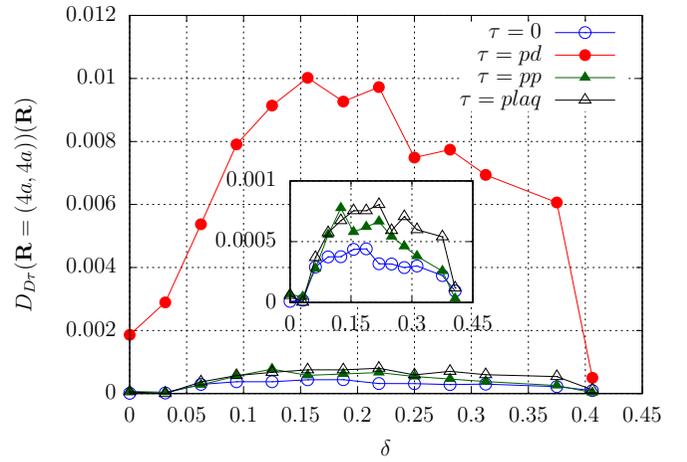


FIG. 8. The  $D_{D\tau}$  values as function of hole doping obtained for the maximally distanced pairing operators, i.e.,  $R = (L/2, L/2)$ . As amplitude of  $D_{Dpd}$  dominates by order of magnitude over other CFs, we present domelike shape for  $\tau \in \{0, pp, plaq\}$  in the inset.

Note that we excluded the values for  $N_e = 298$  ( $\delta \approx 0.34$ ) as for all four CFs considered here the obtained values were unexpectedly high due to optimization issues. As one can see, the domelike shape for all  $D_{D\tau}$  is reproduced. Nonetheless, the amplitude of  $D_{Dpd}$  is one order of magnitude higher. Detailed analysis of  $D_{D0}$ ,  $D_{Dpp}$ , and  $D_{Dplaq}$  (see inset in Fig. 8) provides evidence of domelike shape existence in the same range of  $\delta$  as observed for  $D_{Dpd}$  and  $D_{Ddd}$  and thus confirms the  $d$ -wave superconducting nature of the ground state. Note that the whole numerical analysis is performed for  $S_{\text{tot}}^z = 0$ , so the local diagonal correlations contain Zhang-Rice spin-singlet correlations at local scale.

## B. Spin-spin correlations

The VMC method allows us to determine the characteristics of the spin and charge ordering. Though the variational Hamiltonian does not include explicitly AF terms, the short-range correlations of this type can be expected. The existence of the AF order for both hole- and electron-doped cuprates is one of the main features of their phase diagram. Therefore, we investigate if the considered *ansatz* is able to reproduce such a tendency.

We perform analysis of the  $z$  component of spin-spin CFs defined in a standard manner [13,31,32], namely,

$$S_l^z(\mathbf{R}) = \frac{1}{L^2} \sum_{i, \mathbf{R}} \langle (\hat{n}_{i\uparrow} - \hat{n}_{i\downarrow})(\hat{n}_{j(\mathbf{R})\uparrow} - \hat{n}_{j(\mathbf{R})\downarrow}) \rangle, \quad (20)$$

and the static spin-spin susceptibility, which has the form

$$S_l^z(\mathbf{q}) = \sum_{\mathbf{R}} e^{i\mathbf{q}\cdot\mathbf{R}} S_l^z(\mathbf{R}), \quad (21)$$

with  $\mathbf{q}$  being the ordering vector given in  $1/a$  units. In Fig. 9, we present  $S_l^z(\mathbf{q})$  for vectors  $\mathbf{q} = \{(\pi, 0), (\pi, \pi), (\frac{\pi}{2}, \frac{\pi}{2}), (0, 0)\}$ . As one can see, the amplitude for the ordering vector  $\mathbf{q} = (\pi, \pi)$  dominates over others as the hole doping is reduced in the system. This corresponds to the tendency of establishing the magnetic state with *staggered*

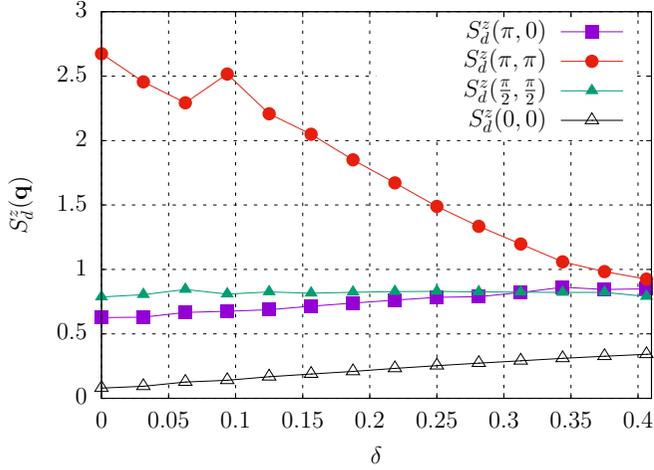


FIG. 9. Static spin-spin susceptibilities as a function of hole doping  $\delta$  for the  $d$  orbitals.  $S_d^z(\pi, \pi)$  is dominant, particularly with decreasing  $\delta$ , and attains maximum value for the parent compound.

magnetization, at least at short range. Note, that our result is in the quantitative agreement with that obtained by means of the DQMC method [13], e.g., the value of  $S_d^z(\pi, \pi)$  at  $\delta = 0$  is  $\approx 2.6$ . However, our result refers to that procured for a larger system. Thus the ratio  $\frac{S_d^z(\pi, \pi)}{L^2}$  fits the *finite-size* scaling analysis performed by Kung *et al.* [13]. In our solution the absence of the long-range AF order is expected due to no AF terms in  $\hat{H}_{\text{eff}}$ , whereas in DQMC, where  $T > 0$ , it originates from the Mermin-Wagner theorem. Despite these circumstances both approaches reproduce similar spin physics.

The evidence of the AF correlation enhancement with decreasing hole-doping manifests itself also in terms of the real-space analysis. In Fig. 10, we present spatial CF  $S_d^z(0, a)$  and  $S_d^z(a, a)$ . As one can see, with decreasing hole-doping, NN orbitals are occupied by the antiparallel spins, whereas correlations between next-NN becomes positive, indicating

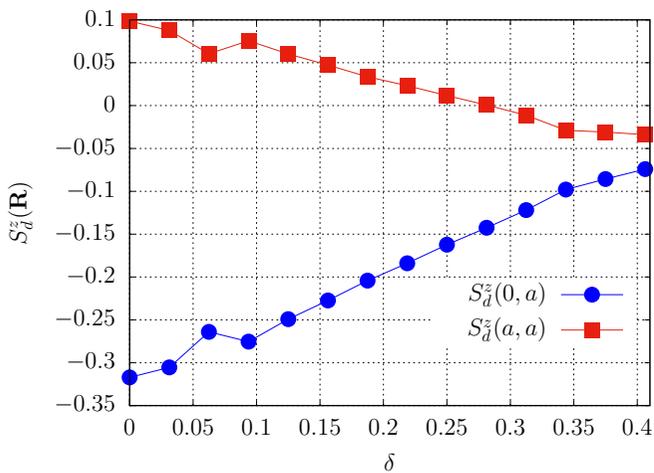


FIG. 10. The doping dependence of the correlation functions  $S_d^z(\mathbf{R})$  for the nearest- and next-nearest  $d$  orbitals. The maximal absolute values correspond to  $\delta = 0$ , and their signs correspond to the development of staggered magnetization.

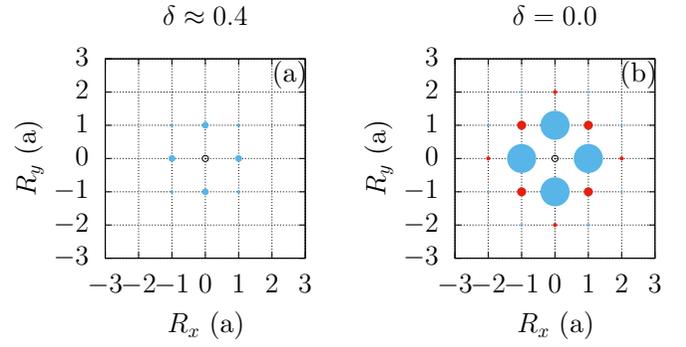


FIG. 11. The decay of spin-spin spatial correlations in real space for  $\delta \approx 0.4$  (a) and  $\delta = 0$  (b). The radius of the circles is proportional to the value of  $S_d^z(\mathbf{R})$ ; the color indicates the sign of the amplitude: positive (red) and negative (blue). For the sake of brevity, we exclude the auto-correlation function (central black dot).

parallel orientation of the  $z$  component of the further spins. In spite of the fact that the spin-spin correlations are short-ranged, in the vicinity of  $\delta = 0$  they decay slowly with the distance (cf. Fig. 11). In the analyzed doping range, we have not found any indication of crossover from AF to ferromagnetic correlations. Namely,  $S_d^z(0, a)$  remains negative and increases with increasing hole doping.

For the sake of completeness, the correlation functions related to  $p$  orbitals are presented in Fig. 12. As one can see, there is no particular spin order for each selected wave vector. Values of  $S_{p_y}^z(\mathbf{q})$  decrease monotonically with decreasing hole doping. This result agrees with that obtained by means of DQMC [13].

### C. Charge gap

As cuprates fall into the class of strongly correlated systems, the emergence of an electron-electron-induced insulating phase is characteristic of these compounds. The outcome

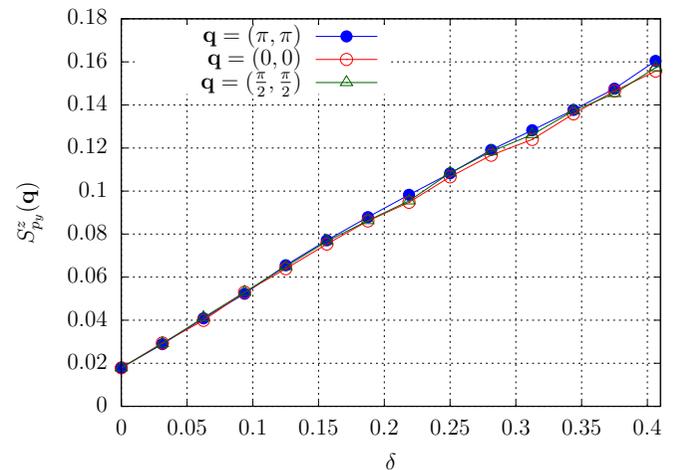


FIG. 12. Equal-time correlation functions  $S_{p_y}^z(\mathbf{q})$  for the selected wave vectors as a function of doping. In the considered range of doping, the system does not exhibit spin ordering at the  $p$  orbitals.

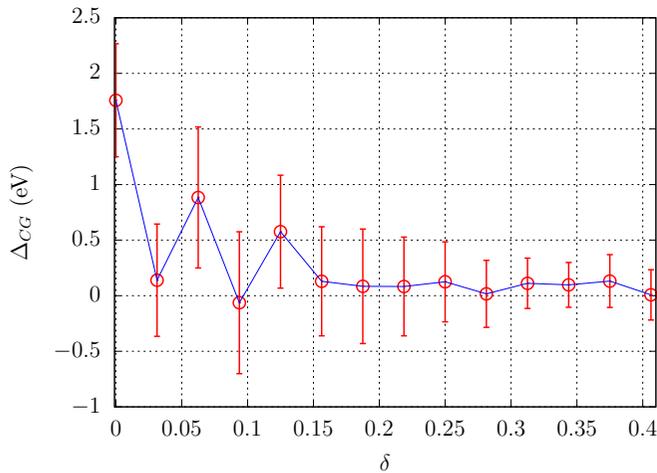


FIG. 13. Charge gap  $\Delta_{CG}$  versus  $\delta$ . The gap emerges for  $\delta \gtrsim 0.1$  attaining, its maximum for the parent compound ( $\delta = 0$ ).

of our estimation is directly comparable to other theoretical treatments as well as to the experimental results.

One of the methods for calculating the CG  $\Delta_{CG}$  (which identifies the insulating state) is based on the single-mode approximation, which has been proved to be an efficient method for the Hubbard-type systems. Within such analysis, one has to determine quantity

$$\Delta_{CG} \propto \lim_{\mathbf{q} \rightarrow 0} \frac{\chi^c(\mathbf{q})}{\mathbf{q}}, \quad (22)$$

where  $\chi^c(\mathbf{q})$  is the Fourier transform of equal-time charge-charge CF. Unfortunately, the minimal norm of the wave vector for  $L = 8$  is  $|\mathbf{q}| = \frac{\pi}{2}$ , thus we are not able to provide a firm estimate of  $\Delta_{CG}$  along these lines. Instead, we determine the value of  $\Delta_{CG}$  in a standard manner. Namely,

$$\Delta_{CG} \approx \frac{2E(N_e) - E(N_e + 2) - E(N_e - 2)}{2}, \quad (23)$$

where  $E(N_e)$  is the total energy of the system at the doping value corresponding to a particular number of electrons  $N_e$ . The above formula previously used by us in a different context [27] can be applied directly here due to the fact that in this analysis one can safely assume that  $S_{\text{tot}}^z = 0$  and  $\Delta N_e = 2$ .

In Fig. 13, we present  $\Delta_{CG}$  as a function of  $\delta$ . For  $\delta \gtrsim 0.12$ , we still obtain small but nonzero values of  $\Delta_{CG}$ , which should be considered as residual and not identified as an indicator of the insulating state. Close to zero doping, we obtain the maximal value of  $\Delta_{CG}(\delta = 0) \approx 1.78$  eV, as expected for the parent compound, which agrees well with those reported in experiments [33–37], i.e.,  $\Delta_{CG} \approx 1.32 - 2.2$  eV for the group of layered structure compounds  $X\text{-CuO}_2$ , where  $X$  refers to lanthanide (La, Sr, Nd, Ca, Sm, Tb). Kung *et al.* [13] reports the value of indirect gap  $\approx 0.77$  eV (after the extrapolation to zero temperature). The authors discuss if such a low value—when compared to the experiment—originates from *finite-size* effects or is connected with temperature extrapolation issues. The maximal size cluster taken for that study was  $6 \times 6$ , i.e., (smaller than the one examined by us) as well as  $2 \times 2$  clusters were treated at  $T = 0$  also in the framework of cluster perturbation theory and ED [13]. The latter method provided

$\Delta_{CG} \approx 1.7$ , which is very close to the value obtained by us. This may suggest that the extrapolation to  $T = 0$  for data obtained in the framework of DQMC [13] affected value of  $\Delta_{CG}$ , thus finite system-size effects seem not to be decisive in this matter. This issue needs a further analysis, since we do not have a systematic analysis of *finite-size* effects.

#### IV. CONCLUSIONS AND OUTLOOK

In this paper, we have considered a three-band  $d - p$  model of the copper-oxygen plane within the VMC approach with a wave-function *ansatz* containing both onsite Gutzwiller and intersite Jastrow correlators in real space. The analysis of superconducting pairing properties in view of the so-called *standard* analysis, i.e., the one based on  $d$ -orbital pairing CFs, provided us with results which are quantitatively consistent with our previous work, as well as qualitatively with selected experimental observations. As an extension of our previous work, we have also calculated the spatial distribution functions of pairing operators proposed by Moreo and Dagotto in their very recent report [10]. We found that the amplitude of the CF is the highest (order of magnitude higher) for the  $D_{pd}$  operator when compared to those consisting of  $p$  orbitals only. Moreover, the considered CFs show the *domelike* behavior as a function of hole doping, which is similar to the NN  $d-d$  pairing amplitude. According to our study, the CFs for MSPO parameters can be regarded as convenient observables for the characterization of the  $d$ -wave paired state in the  $d-p$  model. Recapitulating, scrutinization of pairing observables in the context of this paper, as well as analysis performed recently [16], indicate that both inter- and intraorbital pairing amplitudes are responsible for the net  $d$ -wave superconductivity in the three-band  $d-p$  model. Nonetheless, the dominant contribution to the superconducting state results from the  $d-d$  pairing [16].

For the sake of completeness, we have also determined *spin-spin* equal-time CFs. Even though the utilized *ansatz* is not supplemented with explicit AF terms, we have observed short-range AF ordering on the  $d$  orbitals. Detailed analysis brought us to conclusions similar to those obtained by Kung *et al.* [13]. Particularly, static *spin-spin* susceptibilities agree quantitatively with the DMC solution. Moreover, the estimated value of CG is  $\Delta_{CG} \approx 1.78$  eV, which fits surprisingly well experimental data [33–38].

Recapitulating, we have retrieved the main features of hole-doped cuprate compounds by means of VMC method, within compact Gutzwiller-Jastrow variational approach. According to experimental findings [5], the symmetry of charge order in the cuprates is likely to be complex and the role of  $p$  orbitals is supposed to be quite important. Nevertheless, we have not analyzed the onset of *charge* ordering [5]. This issue can be related both to the supercell size, as well as to the form of variational *ansatz*. Possibly, the application of the most general Pfaffian wave function and more distant Jastrow terms, with a minimal dose of symmetries, could provide a better understanding of this state. However, in such a scenario the number of variational parameters is large and the optimization procedure may become too complex. The recent development of dedicated VMC codes may help to overcome these difficulties [26], potentially even at the *ab initio* level

[39,40]. We should be able to see progress in this matter in the near future.

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