Pairing correlations with s^* and d symmetry: exact results for the Cu_4O_8 cluster

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The pairing correlation functions in the Cu_4O_8 cluster, which is an elementary fragment of a CuO_2 plane, a common element of the crystal lattice of high-T_c superconductors, have been numerically calculated by exact diagonalization. Cases corresponding to s, s^*, and d symmetry of the Cooper pair have been considered. It has been shown that at realistic values of the parameters of a model Hamiltonian and a nearly optimal doping level ($x=0.25$ excess carriers per copper atom) there are no pairing correlations with s symmetry and that the correlations with s^* symmetry are considerably weaker than the correlations with d symmetry. This is true for both hole and electron doping. The predominance of pairing correlations in the s^* channel is consistent with the experimental data on the influence of nonmagnetic defects on high-T_c superconductors. When $x=0.5$, which corresponds to the nonsuperconducting metallic state of a CuO_2 plane, the pairing correlations in all the channels either vanish or are strongly suppressed. © *1994 American Institute of Physics.*

1. INTRODUCTION

Despite the intensive work of numerous experimental and theoretical teams, the mechanism of the superconductivity of high-T_c superconductors has not yet been established. This is attributable both to the ambiguous nature of the experimental data,[1] which hampers their interpretation, and to the complexity of the physicochemical structure of high-T_c superconductors: the nature of the interactions that play the main role in forming the superconducting state is unclear.

The small magnitude of the isotope effect and the fact that high-T_c superconductors have several anomalous properties, even at temperatures above the critical temperature,[1] provided a basis for postulating a nonphonon mechanism for high-T_c superconductivity. There have been various concrete realizations of this hypothesis: the spin-bag model,[2] the resonating-valence-bond model,[3] the model of superconductivity induced by charge instability,[4] the model of a nonlinear exchange interaction,[5] the kinetic model,[6] etc. In the recent period the phenomenological model of superconductivity caused by the interaction of quasiparticles with spin fluctuations[7] has become popular.

Practically all the theoretical studies were based on some simplifying assumptions, whose applicability was not always proved. It is often difficult to understand how strongly (and in what way) such approximations influence the final result. One of the techniques used in the theoretical work, which makes it possible to approach the problem from a somewhat different standpoint, is to utilize experimental data in the calculations. For example, in Ref. 7 the spectral density of the spin fluctuations was taken from nuclear magnetic resonance experiments. One of the unquestionable advantages of such an approach is that it makes it possible not only to take into account the common features of different high-T_c superconducting systems, but also to reveal the differences between them (for example, between 123, 124, 201, and other phases). However, this approach is still phenomenological to a considerable degree. It depends on the accuracy of the specific experimental data and still does not make it possible to resolve several existing contradictions.

In this situation great importance is attached to numerical methods, which make it possible to obtain additional information on the properties of the system under investigation by means of an exact calculation of the characteristics of a particular model Hamiltonian without making any assumptions regarding the nature of the ground state, without employing expansions in small parameters, etc. Such methods include, first of all, the quantum Monte Carlo method and exact diagonalization. One shortcoming of numerical methods is the existence of the restrictions imposed on the size of the system investigated by the speed and size of the internal memory of the computer. The Monte Carlo method makes it possible to study comparatively large clusters consisting of $N_a=50$–200 atoms, but the calculation error increases dramatically with decreasing temperature, so that the range at ~ 100 K, which is most interesting for high-T_c superconductivity, cannot be examined. Under exact diagonalization the restrictions on N_a are significantly more severe ($N_a=10$–20), but exact diagonalization makes it possible to investigate the properties of the ground state of the system ($T=0$).

Exact diagonalization has been used in several studies[8–11] to calculate the binding energies of the excess carriers in Cu–O clusters. It was shown that the binding energy is negative, i.e., there is a tendency for the formation of a bound state of excess carriers, over a broad range of the model parameters. This is true for both hole[8–10] and electron[11] doping, providing an argument in favor of identical nonphonon mechanisms (or at least mechanisms having similar features) for the superconductivity of p-type and n-type high-T_c superconductors.

If the calculations are performed with the Emery model[12] (which, in our opinion, is the most realistic model of the electronic structure of CuO_2 planes, the largest cluster having the symmetry of a CuO_2 plane and allowing the use of exact diagonalization is the 12-site Cu_4O_8 cluster. Due to the small dimensions of the cluster, the subject of quantita-

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tive agreement between the numerical calculations and experiment should not, of course, even be mentioned. However, since the coherence length in a high-T_c superconductor is small, amounting to several lattice periods, it may be hoped that the qualitative picture will be correct. In fact, the results of the numerical calculations that we performed for the Cu_4O_8 cluster using exact diagonalization[13–17] are in fairly good agreement with experiments on the effects of radiation on high-T_c superconductors, the influence of magnetic and nonmagnetic impurities on T_c, and the dependence of T_c and the optical gap on the pressure. There is also good correspondence[18] with the analytical data obtained in several limiting cases for an infinite CuO_2 plane.[19,20] All this suggests that numerical investigations of even tiny clusters can provide valuable information on the characteristics of an infinite system.

The question of the symmetry of the superconductive gap in high-T_c superconductors is presently a very crucial problem. Its solution might significantly reduce the size of the list of discussible theoretical models, which may be hypothetically divided into two groups: models with s-wave and d-wave symmetry of the gap. For example, the spin-fluctuation model of high-T_c superconductors predicts that the gap has $d_{x^2-y^2}$ symmetry,[7] and, according to one of latest papers of P. W. Anderson *et al.*,[21] the gap does not change sign, is highly anisotropic, and has the symmetry of the crystal. The experimental studies have not provided an unequivocal answer to the question of the gap symmetry, although many investigators have arrived at the conclusion that the gap has $d_{x^2-y^2}$ symmetry.[22–24] It has not, however, been ruled out that the superconducting state in high-T_c superconductors is a mixture of states with different symmetries.[25]

In numerical cluster calculations information on the gap symmetry is provided by the pairing susceptibilities or pairing correlation functions calculated for various pairing channels.[26] Their sign makes it possible to assess the presence or absence of pairing interactions in the respective channel,[26,27] and their absolute value makes it possible to determine in which channel the tendency to form a superconducting state is strongest. In general, drawing an unequivocal conclusion regarding superconductivity in a particular pairing channel requires proving that the pairing correlators in configuration space do not vanish as the dimensions of the system increase.[28,29] Only under this condition will a pairing correlation function in momentum space diverge in the thermodynamic limit.

The divergence of a pairing correlation function in an s pairing channel was, in fact, observed for the single-band two-dimensional (2D) Hubbard model with on-site attraction[30] (this is consistent with the well established fact that superconductivity exists in this model). Conversely, in the single-band 2D Hubbard model with repulsion, long-range superconducting order was not discovered in the s channel.[28] Frick *et al.*[31] reached the same conclusion regarding the 2D Emery model. The negative results in Refs. 28 and 31 should be approached with some caution, since they were obtained by the Monte Carlo method, i.e., at a finite temperature which might not be low enough for superconducting correlations to appear in the system. In general, the presence of three atoms in the unit cell of a CuO_2 plane renders the problem of calculating pairing correlators in clusters with different dimensions very difficult (the Monte Carlo method is usually applied to clusters consisting of 16 and 36 cells); therefore, the question of the character of the variation of the pairing correlators as the dimensions of the system increase cannot yet be resolved unequivocally.

Nevertheless, indications of the existence of superconducting pairing correlations in singlet s^* and $d_{x^2-y^2}$ channels (s^* denotes nonlocal s pairing) in the 2D Emery model at a finite temperature have been obtained in several studies (see, for example, Refs. 26 and 27). In this connection it would be interesting to perform calculations of the corresponding pairing correlators using exact diagonalization. The size of the clusters accessible to investigation is, of course, smaller, but it now becomes possible to study the case of $T=0$. In addition, exact diagonalization makes it possible to perform calculations with a fixed number of particles in the system, i.e., to find the dependence of pairing correlators on the degree of doping in an explicit form (when the determinantal algorithm of the quantum Monte Carlo method[26,27] is used, the concentration of particles is assigned indirectly by selecting an appropriate value of the chemical potential).

In the present work exact diagonalization was used to calculate the pairing correlators in the s, s^*, and $d_{x^2-y^2}$ pairing channels for the Cu_4O_8 cluster, which can be described by the Emery model. It was shown that at a nearly optimal doping level for high-T_c superconductors ($x=0.25$ charge carriers per copper atom) the pairing correlations in the s^* channel predominate. This is true for both hole and electron doping of the cluster. In the case of hole doping, there are also pairing correlations in the $d_{x^2-y^2}$ channel, but they are considerably weaker than those in the s^* channel. When $x=0.5$ (which corresponds to the nonsuperconducting metallic state of a CuO_2 plane) there are no pairing correlations at all or they are strongly suppressed in all the pairing channels. We also investigated the undoped insulating state. The results of our calculations are in good agreement with the published Monte Carlo data obtained at a finite temperature for Cu–O clusters consisting of 48 and 108 atoms (although there are still some discrepancies). As a whole, these results attest to s^* pairing of the excess carriers (both holes and electrons) in a CuO_2 plane. It has not, however, been ruled out that under hole doping the $d_{x^2-y^2}$ channel also makes a certain contribution to the superconducting correlations.

2. BASIC DEFINITIONS

To numerically calculate the pairing correlators in the Cu_4O_8 cluster with periodic boundary conditions we used the Emery model,[12] whose Hamiltonian has the form

$$H=-t\sum_{\langle ik\rangle,\sigma}(d_{i\sigma}^{+}p_{k\sigma}+\text{h.c.})+\varepsilon\sum_{k,\sigma}n_{k\sigma}+U_d\sum_{i}n_{i\uparrow}n_{i\downarrow}$$

$$+U_p\sum_{k}n_{k\uparrow}n_{k\downarrow}+V\sum_{\langle ik\rangle,\sigma,\sigma'}n_{i\sigma}n_{k\sigma'}, \qquad (1)$$

where $d_{i\sigma}^{+}$ and $p_{k\sigma}^{+}$ are the creation operators of a hole in the $d_{x^2-y^2}$ and $p_{x(y)}$ states, respectively ($\sigma=\downarrow$ or $\uparrow$), $\langle ik\rangle$ denotes

summation over the nearest neighbors, the index i refers to the copper sites, the index k refers the oxygen sites, $n_{i\sigma}=d_{i\sigma}^{+}d_{i\sigma}$ and $n_{k\sigma}=p_{k\sigma}^{+}p_{k\sigma}$ are the occupancies of the copper and oxygen orbitals, t is the matrix element for the hopping of holes between copper and oxygen sites, $\varepsilon=\varepsilon_p-\varepsilon_d$ is the difference between the energies of a hole at the oxygen and copper sites, and U_d, U_p, and V are the Coulomb repulsion energies of holes at the copper and oxygen sites and between them, respectively. The term describing direct oxygen-oxygen hopping was omitted in (1) (we assume that this simplification of the model does not significantly influence the results, since the corresponding matrix element for hopping in high-T_c superconductors is several times smaller than t).

Since we use the hole representation of Hamiltonian (1), the number of particles (holes) N in the undoped insulating state of the Cu_4O_8 cluster is equal to the number of copper sites, i.e., $N=4$ (the $Cu^{2+}O^{2-}$ or "holes on copper" valence state). An increase (or decrease) in N corresponds to hole (or electron) doping of the CuO_2 plane in the high-T_c superconductor. For the relative (per copper atom) concentrations x of excess carriers we have $x=0$ when $N=4$ (original undoped state), $x=0.25$ when $N=5$ (hole doping) and $N=3$ (electron doping), and $x=0.5$ when $N=6$ (hole doping) and $N=2$ (electron doping). We note that the value $x=0.25$ is close to the optimal doping level x_0, at which T_c of the specific high-T_c superconducting compound reaches its maximum (although the maximum value of T_c differs for different high-T_c superconducting systems and phases): $x_0=0.15$–0.25 in p-type high-T_c superconductors[32] and $x_0=0.14$–0.17 in n-type high-T_c superconductors.[33] The value $x=0.5$ corresponds to the metallic nonsuperconducting state of the CuO_2 plane.

The pairing correlators in the α pairing channel have the following form (see, for example, Ref. 28):

$$P_\alpha=\sum_{\mathbf{r},\mathbf{r}'}\langle\Delta_\alpha^{+}(\mathbf{r})\Delta_\alpha(\mathbf{r}+\mathbf{r}')\rangle, \tag{2}$$

where the averaging $\langle\ldots\rangle$ is carried out either with respect to the ground state (at $T=0$) or with respect to a Gibbs ensemble (at $T\neq 0$) and

$$\Delta_\alpha^{+}(\mathbf{r})=\frac{1}{\sqrt{N_0}}\sum_{\boldsymbol{\rho}} g_\alpha(\boldsymbol{\rho})C_{\mathbf{r},\uparrow}^{+},C_{\mathbf{r}+\boldsymbol{\rho},\downarrow}^{+}. \tag{3}$$

The summation over $\mathbf{r}$ and $\mathbf{r}'$ in (2) is carried out over all the unit cells, whose number is equal to N_0 (in a simple lattice N_0 is the number of sites, and in a CuO_2 plane it is the number of copper atoms). The operator $C_{\mathbf{r},\sigma}^{+}$ in (3) describes the creation of a particle with a projection of the spin σ with a coordinate $\mathbf{r}$ in the unit cell. If there are several atoms in the unit cell, $\mathbf{r}$ denotes the coordinates of equivalent sites. In the case of a CuO_2 plane, either the copper atoms with the coordinates $\mathbf{R}_{Cu}$ or the oxygen atoms with the coordinates $\mathbf{R}_{Cu}+(a/2)\mathbf{e}_x$ or $\mathbf{R}_{Cu}+(a/2)\mathbf{e}_y$, where a is the lattice period (the distance between two neighboring copper atoms), can be chosen as such equivalent sites. In principle, the operators $C_{\mathbf{r},\sigma}^{+}$ may be linear combinations of the creation operators of particles at different sites in the unit cell.

The form of the function $g_\alpha(\boldsymbol{\rho})$ in (3) is determined by the symmetry of the pairing state (below we shall consider only the cases of $\alpha=s$, $\alpha=s^*$, and $\alpha=d_{x^2-y^2}$). For s pairing $g_\alpha(\boldsymbol{\rho})=1$ when $\boldsymbol{\rho}=0$, and $g_\alpha(\boldsymbol{\rho})=0$ for other values of $\boldsymbol{\rho}$. For s^* pairing $g_\alpha(\boldsymbol{\rho})=1$ when $\boldsymbol{\rho}=\pm a\mathbf{e}_x$ or $\boldsymbol{\rho}=\pm a\mathbf{e}_y$, and $g_\alpha(\boldsymbol{\rho})=0$ for other values of $\boldsymbol{\rho}$. For $d_{x^2-y^2}$ pairing $g_\alpha(\boldsymbol{\rho})=1$ when $\boldsymbol{\rho}=\pm a\mathbf{e}_x$, $g_\alpha(\boldsymbol{\rho})=-1$ when $\boldsymbol{\rho}=\pm a\mathbf{e}_y$, and $g_\alpha(\boldsymbol{\rho})=0$ for other values of $\boldsymbol{\rho}$.

As was noted in Ref. 28, P_α is the Fourier component taken when $\mathbf{k}=0$

$$P_\alpha(\mathbf{k})=\sum_{\mathbf{r}'} P_\alpha(\mathbf{r}')\exp(i\mathbf{k}\mathbf{r}'),$$

where

$$P_\alpha(\mathbf{r}')=\sum_{\mathbf{r}}\langle\Delta_\alpha^{+}(\mathbf{r})\Delta_\alpha(\mathbf{r}+\mathbf{r}')\rangle.$$

The divergence of $P_\alpha\equiv P_\alpha(\mathbf{k}=0)$ in the thermodynamic limit attests to the presence of long-range off-diagonal order, i.e., superconductivity, in the system. In a restricted system (a cluster) P_α is always finite. To determine the presence or absence of superconducting correlations in a cluster, it is convenient to introduce the quantity[26]

$$\tilde{P}_\alpha=\frac{1}{\sqrt{N_0}}\sum_{\mathbf{r},\mathbf{r}',\boldsymbol{\rho},\boldsymbol{\rho}'} g_\alpha(\boldsymbol{\rho})g_\alpha(\boldsymbol{\rho}')\langle C_{\mathbf{r},\uparrow}^{+}C_{\mathbf{r}+\mathbf{r}',\uparrow}\rangle \times\langle C_{\mathbf{r}+\boldsymbol{\rho},\downarrow}^{+}C_{\mathbf{r}+\mathbf{r}'+\boldsymbol{\rho}',\downarrow}\rangle, \tag{4}$$

which is obtained from (2) by not taking into account the anomalous averages $\langle C^{+}C^{+}\rangle$ and $\langle CC\rangle$. If $P_\alpha>\tilde{P}_\alpha$, there are pairing correlations in the α channel; if $P_\alpha<\tilde{P}_\alpha$, there are no pairing correlations in that channel. It may turn out that $P_\alpha>\tilde{P}_\alpha$ in several channels. In that case the pairing correlations are stronger in the channel for which the difference $P_\alpha-\tilde{P}_\alpha$ is larger.

Some investigators[26,34] calculated the correlator

$$P'_\alpha=\sum_{\mathbf{r},\mathbf{r}'}\langle\Delta_\alpha(\mathbf{r})\Delta_\alpha^{+}(\mathbf{r}+\mathbf{r}')\rangle,$$

rather than the correlator P_α defined by Eq. (2). Its value does not coincide with P_α in the general case. It can, however, be shown that $P'_\alpha-\tilde{P}'_\alpha=P_\alpha-\tilde{P}_\alpha$, where the partially uncorrelated correlator $\tilde{P}'_\alpha$ is obtained from P'_α precisely as $\tilde{P}_\alpha$ is obtained from P_α. We shall henceforth consider only P_α and $\tilde{P}_\alpha$, which are defined by expressions (2) and (4).

In the calculation of P_α and $\tilde{P}_\alpha$ we assumed that the operators $C_{\mathbf{r},\sigma}^{+}$ are the creation operators $d_{\mathbf{R}_{Cu},\sigma}^{+}$ of holes at the copper sites, i.e., $\mathbf{r}=\mathbf{R}_{Cu}$. The physical basis of this choice is the fact that in high-T_c superconductors the pairing wave function has an amplitude at the copper sites that is comparable to the amplitude at the oxygen sites.[35] Therefore, P_α and $\tilde{P}_\alpha$ should not be significantly dependent on the specific location of the sites with the coordinate $\mathbf{r}$ in the unit cell.

The results of calculations of the difference $P_\alpha-\tilde{P}_\alpha$ in the $\alpha=s,s^*$, and $d_{x^2-y^2}$ channels for Cu_4O_8 clusters with $N=2$, 3, 4, 5, and 6 holes are presented below. Since Hamil-

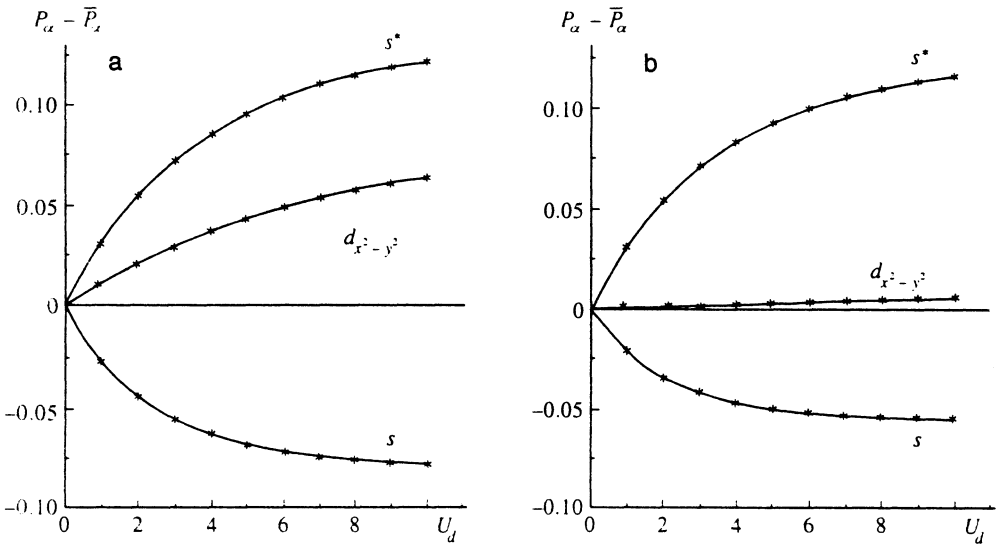

FIG. 1. Dependence of $P_\alpha - \bar{P}_\alpha$ on U_d in the s,s^*, and $d_{x^2-y^2}$ channels for the optimally doped states of the Cu_4O_8 cluster when $\varepsilon=1$, $U_p=V=0$, and $t=1$: a) $N=5$ (hole doping); b) $N=3$ (electron doping).

tonian (1) is a model, there is hardly any sense in referring to some fixed set of values of its parameters (t, ε, U_d, U_p, and V) corresponding to real compounds (furthermore, these values may be different for different high-T_c superconducting systems, since the electronic state of CuO_2 planes is greatly dependent on their specific environment in a three-dimensional unit cell). For this reason we investigated a fairly broad range of parameters of Hamiltonian (1).

3. "OPTIMALLY" DOPED STATES ($N=3$ AND 5; $x=0.25$)

As was noted in the introduction, the addition of one hole ($N=5$) or one electron ($N=3$) to the insulating state of the Cu_4O_8 cluster corresponds to a concentration of excess carriers $x=0.25$ (per copper atom), which is close to the optimal (corresponding to the highest T_c) doping level of a CuO_2 plane. Therefore, the calculation of pairing correlators in the Cu_4O_8 cluster with $N=3$ and $N=5$ is of fundamental interest from the standpoint of the question of the nonphonon mechanism of high-T_c superconductivity and the gap symmetry corresponding to this mechanism.

In numerical cluster calculations of various characteristics in systems with strong Coulomb correlations it is useful to ascertain whether the values of the characteristics for a cluster coincide with the corresponding values for an infinite system in the limiting case of the absence of interparticle interactions. For example, the binding energies of two excess holes or two excess electrons introduced into the Cu_4O_8 cluster upon doping vanish when $U_d=U_p=V=0$ (Ref. 11). This is certainly also true for an infinite CuO_2 plane, since bound two-particle states cannot be described by a single-particle Hamiltonian. The physical reasonableness of the numerical results obtained in this special case (and some other cases) creates some hope that cluster calculations will also reflect the properties of the infinite system to some extent in the presence of interparticle interactions (see also Refs. 18–20).

Therefore, it would be interesting to examine whether the equality $P_\alpha=\bar{P}_\alpha$, which holds for an infinite undoped CuO_2 plane in all the α channels, is satisfied for the Cu_4O_8 cluster when $U_d=U_p=V=0$. Figure 1 presents plots of the dependence of $P_\alpha-\bar{P}_\alpha$ on U_d/t when $\varepsilon/t=1$ and $U_p=V=0$. It is seen that $P_\alpha-\bar{P}_\alpha\to 0$ as $U_d\to 0$ in all the pairing channels in the cases of both hole ($N=5$) and electron ($N=3$) doping of the Cu_4O_8 cluster. We obtained the same results for other values of ε/t when $U_d=U_p=V=0$ (as we shall see below, the condition $P_\alpha-\bar{P}_\alpha\to 0$ when $U_p=V=0$ and $U_d\to 0$ does not hold for the insulating undoped state of the cluster with $N=4$ holes). Thus, it may be expected that the finite dimensions of the Cu_4O_8 cluster will not have a decisive influence on the results in doped states and that the dependences of the pairing correlators on the parameters of Hamiltonian (1) will, therefore, reflect (at least qualitatively) their dependences in a CuO_2 plane. We note that, as follows from the calculations, when $U_d=U_p=V=0$, the ground states of the Cu_4O_8 clusters with $N=3$ and $N=5$ holes are quadruply degenerate (doubly with respect to the overall projection of the spin and doubly with respect to the total momentum) and that the interparticle interaction does not remove this degeneracy [our calculated data make it possible to determine the degree of degeneracy of the ground state, which we found for each set of values of the parameters of Hamiltonian (1)].

We now proceed to a discussion of the magnitude and sign of $P_\alpha-\bar{P}_\alpha$ in different pairing channels. In the s channel P_α is always smaller than $\bar{P}_\alpha$ (we investigated fairly broad ranges of values of the model parameters: $\varepsilon/t=0$–5, $U_d/t=0$–10, $U_p/t=0$–4, and $V/t=0$–4), i.e., there are no superconducting correlations in the s channel. This is due to the fact that the charge carriers experience strong on-site Coulomb repulsion, which naturally prevents local pairing. We shall henceforth consider only the s^* and $d_{x^2-y^2}$ channels, in which $P_\alpha>\bar{P}_\alpha$.

As follows from Fig. 1, $P_\alpha-\bar{P}_\alpha$ is two to three times greater in the s^* channel than in the $d_{x^2-y^2}$ channel under hole doping and one to two orders of magnitude greater in the former channel than in the latter channel under electron doping. Thus, even if superconducting correlations make some contribution to $P_\alpha-\bar{P}_\alpha$ in the $d_{x^2-y^2}$ channel, they are considerably stronger in the s^* channel. In order to verify

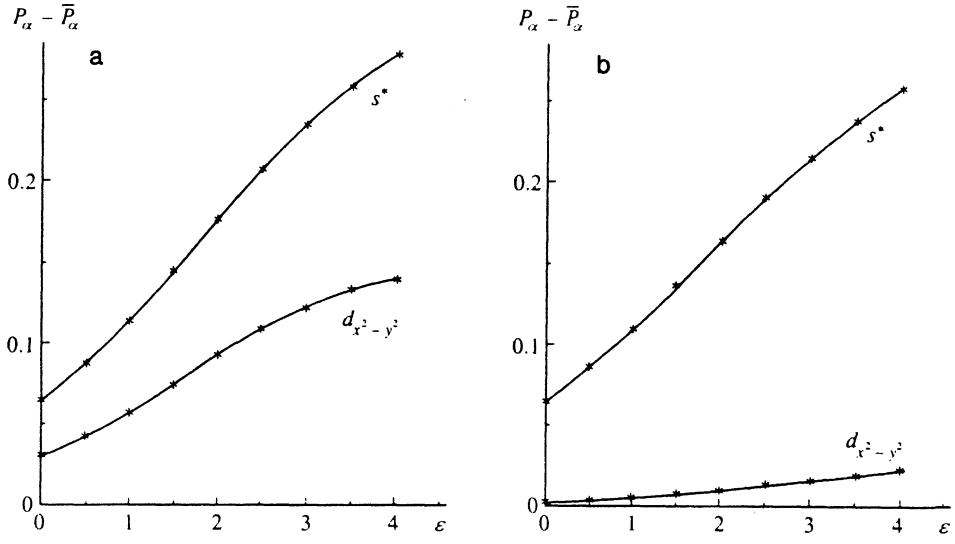

FIG. 2. Dependence of $P_\alpha - \bar{P}_\alpha$ on ε in the s^* and $d_{x^2-y^2}$ channels for the optimally doped states of the Cu_4O_8 cluster when $U_d=8$, $U_p=V=0$, and $t=1$: a) $N=5$ (hole doping); b) $N=3$ (electron doping).

whether this is also true in other regions of the space of the model parameters, we calculated $P_\alpha - \bar{P}_\alpha$ for various values of ε, U_d, U_p, and V.

Figure 2 presents plots of the dependence of $P_\alpha - \bar{P}_\alpha$ in the s^* and $d_{x^2-y^2}$ channels on ε/t when $U_d/t=8$ and $U_p=V=0$ for $N=5$ and $N=3$. In both channels $P_\alpha - \bar{P}_\alpha$ increases with increasing ε/t, although $P_\alpha - \bar{P}_\alpha$ is always greater in the s^* channel than in the $d_{x^2-y^2}$ channel (as ε/t increases, this difference becomes increasingly more significant). We note that at other values of U_d, U_p, and V the dependence of $P_\alpha - \bar{P}_\alpha$ on ε/t is also monotonic. Unlike Scalettar *et al.*,[27] we did not discover a maximum for $P_\alpha - \bar{P}_\alpha$ at $\varepsilon/t \approx 2$ in the insulating or doped states. This may be due to the fact that the calculations in Ref. 27 were performed by the Monte Carlo method at a finite temperature.

Figure 3 presents plots of the dependence of $P_\alpha - \bar{P}_\alpha$ on U_p/t when $\varepsilon/t=1$, $U_d/t=8$, $V=0$. It is seen that U_p/t has a very weak influence on $P_\alpha - \bar{P}_\alpha$ in both the s^* channel and the $d_{x^2-y^2}$ channel (under both hole doping and electron doping). An increase in ε/t results in even greater weakening of the dependence of $P_\alpha - \bar{P}_\alpha$ on U_p/t. It should be stressed that the binding energies E_b of two excess holes or two excess electrons introduced into the Cu_4O_8 cluster upon doping[11] are, conversely, very sensitive to the value of U_p/t: the binding energies for the same values of the parameters ($\varepsilon/t=1$, $U_d/t=8$, $V=0$) change sign from plus to minus when $U_p/t \approx 0.5$ (for $N=3$) and $U_p/t \approx 1$ (for $N=5$). Thus, a correspondence between E_b and $P_\alpha - \bar{P}_\alpha$ cannot be established. This is apparently due to the fact that the correlator $P_\alpha - \bar{P}_\alpha$ characterizes the state of a cluster with a fixed doping level ($x=0.25$ when $N=3$ or $N=5$), while E_b contains information on the characteristics of the three states with $x=0$, 0.25, and 0.5 at once [we recall that for the cluster under consideration $E_b=E(4)+E(6)-2E(5)$ under hole doping and $E_b=E(4)+E(2)-2E(3)$ under electron doping, where $E(N)$ is the ground-state energy of the cluster with N holes[11]]. Thus, the ground-state energies of the excess holes

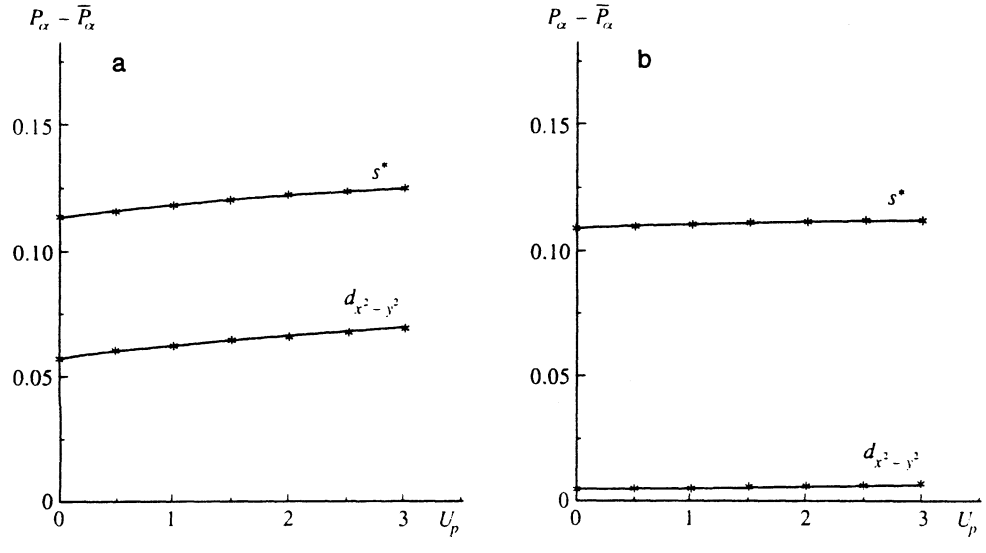

FIG. 3. Dependence of $P_\alpha - \bar{P}_\alpha$ on U_p in the s^* and $d_{x^2-y^2}$ channels for the optimally doped states of the Cu_4O_8 cluster when $\varepsilon=1$, $U_d=8$, $V=0$, and $t=1$: a) $N=5$ (hole doping); b) $N=3$ (electron doping).

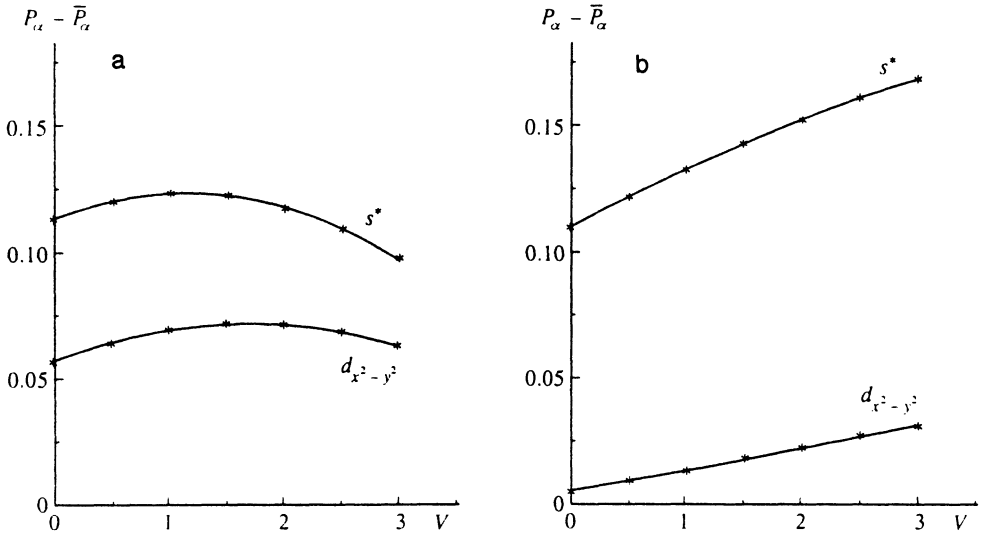

FIG. 4. Dependence of $P_\alpha - \bar{P}_\alpha$ on V in the s^* and $d_{x^2-y^2}$ channels for the optimally doped states of the Cu_4O_8 cluster when $\varepsilon=1$, $U_d=8$, $U_p=0$, and $t=1$: a) $N=5$ (hole doping); b) $N=3$ (electron doping).

or electrons correspond to the doping level $x=0.25$ only "on the average."

The exact-diagonalization calculation of $P_\alpha - \bar{P}_\alpha$ at various values of V/t is of special interest, since the Monte Carlo method does not make it possible to investigate the case of $V>0.5t$ due to a sharp increase in the calculation error.[27] As can seen from Fig. 4, for hole doping the plots of $P_\alpha - \bar{P}_\alpha$ versus V/t in both channels exhibit small maxima in the vicinity of $V/t=1$–2 when $\varepsilon/t=1$, $U_d/t=8$, and $U_p=0$. We note that these maxima vanish when ε/t increases from 1 to 3 (they may simply shift toward larger values of V/t). For electron doping the value of $P_\alpha - \bar{P}_\alpha$ for the same values of ε/t, U_d/t, and U_p/t increases monotonically as V/t increases in both pairing channels. A similar effect was discovered in Ref. 27 when the insulating state of the $Cu_{16}O_{32}$ cluster was investigated by the Monte Carlo method in the restricted range $V/t<0.5$.

4. STRONGLY DOPED STATES ($N=2$ AND 6; $x=0.5$)

The Cu_4O_8 clusters with two excess holes ($N=6$) and with two excess electrons ($N=2$) correspond to a strongly doped CuO_2 plane ($x=0.5$ carriers per copper atom). At this doping level neither p-type nor n-type high-T_c superconductors have superconducting properties, and they are normal.[32,33] Therefore, if the pairing correlators $P_\alpha - \bar{P}_\alpha$ in the Cu_4O_8 cluster do, in fact, contain information regarding the presence or absence of superconducting correlations in a CuO_2 plane, when $N=2$ and $N=6$ we should observe either a change in the sign of $P_\alpha - \bar{P}_\alpha$ from plus to minus or at least a decrease in $P_\alpha - \bar{P}_\alpha$ in the s^* and $d_{x^2-y^2}$ channels. When the pairing correlators are calculated by the Monte Carlo method, the region of large values of x is generally not investigated,[27] the calculations being restricted to $x<0.25$.

Figure 5 presents plots of the dependence of $P_\alpha - \bar{P}_\alpha$ on

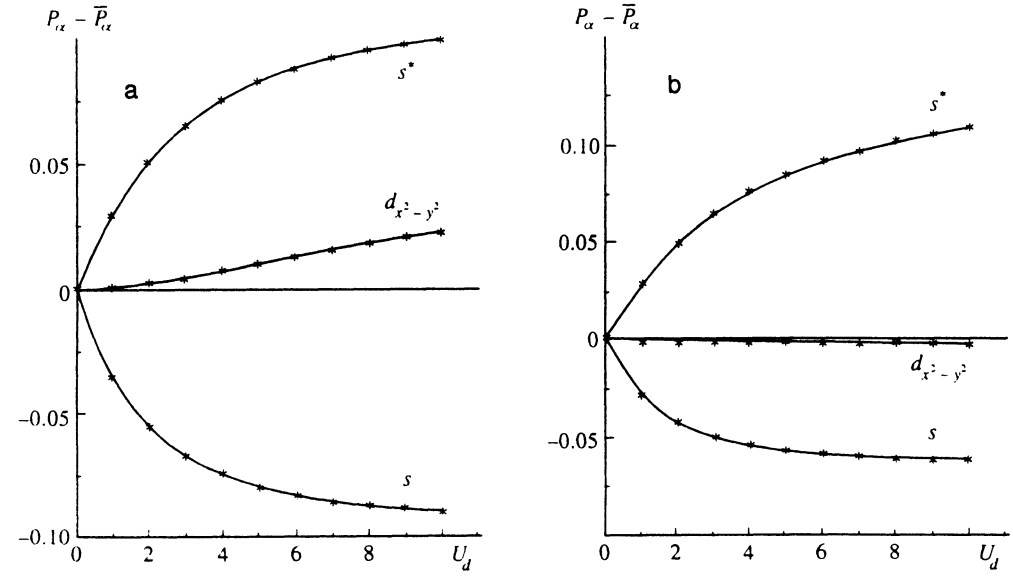

FIG. 5. Dependence of $P_\alpha - \bar{P}_\alpha$ on U_d in the s,s^*, and $d_{x^2-y^2}$ channels for strongly doped states of the Cu_4O_8 cluster when $\varepsilon=1$, $U_p=V=0$, and $t=1$: a) $N=6$ (hole doping); b) $N=2$ (electron doping).

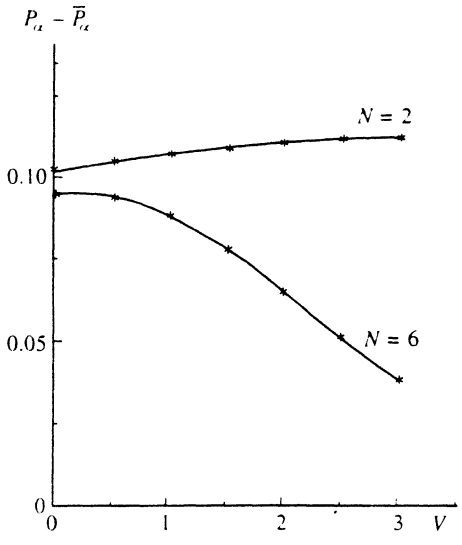

FIG. 6. Dependence of $P_\alpha - \bar{P}_\alpha$ on V in the s^* channel for strongly doped states of the Cu_4O_8 cluster with $N=6$ and $N=2$ when $\varepsilon=1$, $U_d=8$, $U_p=0$, and $t=1$.

U_d/t when $\varepsilon/t=1$ and $U_p=V=0$ for the cases of hole ($N=6$) and electron ($N=2$) doping of the Cu_4O_8 cluster. As in the case of optimal doping, $P_\alpha \to \bar{P}_\alpha$ when $U_d/t \to 0$ and $U_p=V=0$ in all pairing channels, providing some hope that the effects of the finite size are small in the states with $N=6$ and $N=2$.

In the s channel $P_\alpha < \bar{P}_\alpha$, i.e., an increase in the concentration of carriers of either sign does not result, as would be expected, in the appearance of superconducting correlations.

Under electron doping ($N=2$), the value of $P_\alpha - \bar{P}_\alpha$ in the $d_{x^2-y^2}$ channel is negative and very small in absolute value ($\sim 10^{-3}$ when $U_d/t=8$). Under hole doping ($N=6$), $P_\alpha > \bar{P}_\alpha$ in the same channel, i.e., the superconducting correlations are maintained, but they are strongly suppressed in comparison with the optimally doped state, since the difference $P_\alpha - \bar{P}_\alpha$ is three to four times smaller when $N=6$ than when $N=5$.

A different situation is observed for the pairing correlators in the s^* channel. Although the values of $P_\alpha - \bar{P}_\alpha$ for $N=6$ and $N=2$ are smaller than the values for $N=5$ and $N=3$, respectively, the differences amount to only 6–8% under electron doping and 10–20% under hole doping (see Figs. 1 and 5). This, however, is true only when $U_p=V=0$. While an increase in U_p/t does not radically alter the situation, when $N=6$, an increase in V/t results in a decrease in $P_\alpha - \bar{P}_\alpha$, but when $N=2$, the dependence of $P_\alpha - \bar{P}_\alpha$ on V/t is very weak. The corresponding plots are presented in Fig. 6. If we recall (see Sec. 3 and Fig. 4) that the value of $P_\alpha - \bar{P}_\alpha$ in the s^* channel scarcely depends on V/t when $N=5$ and increases with increasing V/t when $N=3$, at nonzero values of V/t we see that $P_\alpha - \bar{P}_\alpha$ is significantly greater in the Cu_4O_8 cluster with one excess carrier ($x=0.25$) than in the cluster with two excess carriers ($x=0.5$) under both hole and electron doping. At sufficiently large values ($V/t=2$–3) this difference reaches 50–100%.

Moreover, since the pairing correlators $P_\alpha - \bar{P}_\alpha$ defined by Eqs. (2)–(4) contain four fermionic operators, when their dependence on the concentration of carriers in the cluster is investigated, it is useful to redefine $P_\alpha - \bar{P}_\alpha$ somewhat by normalizing it to the square of the occupancy of the copper sites with excess carriers (holes when $N<4$ and electrons when $N>4$). This, of course, applies only to doped states. Since the occupancies increase with increasing x,[11] the value of $(P_\alpha - \bar{P}_\alpha)_{x=0.25}/(P_\alpha - \bar{P}_\alpha)_{x=0.5}$ is greater than the ratio calculated from Eqs. (2)–(4).

We shall not present the corresponding plots, since the establishment of quantitative relationships between the correlators for different doping levels of a CuO_2 plane is beyond the scope of the present work. The qualitative result is that the normalization of $P_\alpha - \bar{P}_\alpha$ in doped states to the occupancy results in a 10–60% increase in $(P_\alpha - \bar{P}_\alpha)_{x=0.25}/(P_\alpha - \bar{P}_\alpha)_{x=0.5}$, thereby attesting to significant weakening of the superconducting correlations in the Cu_4O_8 cluster upon the transition from the optimally doped state ($x=0.25$) to the strongly doped state ($x=0.5$).

Thus, the main result of this section is that upon the transition from strong (corresponding to the normal state of a CuO_2 plane) doping of the Cu_4O_8 cluster with holes or electrons to optimal doping, the pairing correlators $P_\alpha - \bar{P}_\alpha$ in the s^* and $d_{x^2-y^2}$ channels either change sign from plus to minus or decrease strongly. In our opinion, this supports the idea that when $x=0.25$, the superconducting correlations do, in fact, make a significant contribution to $P_\alpha - \bar{P}_\alpha$. With respect to the s^* channel, a necessary condition for an increase in $(P_\alpha - \bar{P}_\alpha)_{x=0.25}/(P_\alpha - \bar{P}_\alpha)_{x=0.5}$, i.e., the condition for suppression of the superconducting correlations under strong doping, is that $V/t>1$.

We note that the use of the variational Monte Carlo method in Ref. 36 revealed a decrease in the energy of the superconducting ground state with s^* symmetry to a level below the energy of the normal state of the $Cu_{36}O_{72}$ cluster when $V/t>1$ and $\varepsilon/t=1$–2, from which it was concluded that there may be a nonphonon mechanism of superconductivity in the s^* pairing channel in high-T_c superconductors. The reason for this may be, for example, the closeness of the system to charge instability when $\varepsilon/t \sim 1$ and V/t is large.[4] It is interesting that the exact-diagonalization calculations of the binding energy E_b of excess holes in the Cu_4O_8 cluster[8,11] revealed both a sharp increase in $|E_b|$ with increasing $V/t>1$ and the appearance of instability with respect to phase separation, i.e., the formation of stable bound states of three of more holes when $V/t>2$ (see also Refs. 18–20). These effects are not observed in the case of electron doping.[11]

5. UNDOPED STATE ($N=4$, $x=0$)

Let us dwell briefly on the undoped state of the Cu_4O_8 cluster ($N=4$, $x=0$), which corresponds to the original undoped compounds La_2CuO_4, $YBa_2Cu_3O_6$, Nd_2CuO_4, $SrCuO_2$, etc. As we know, these compounds are insulators. This state is interesting, since the Monte Carlo investigations of finite Cu–O clusters (see, for example, Ref. 27) revealed very strong pairing correlations with $d_{x^2-y^2}$ symmetry in them (although in an infinite undoped CuO_2 plane there should apparently be no superconducting correlations in any channel).

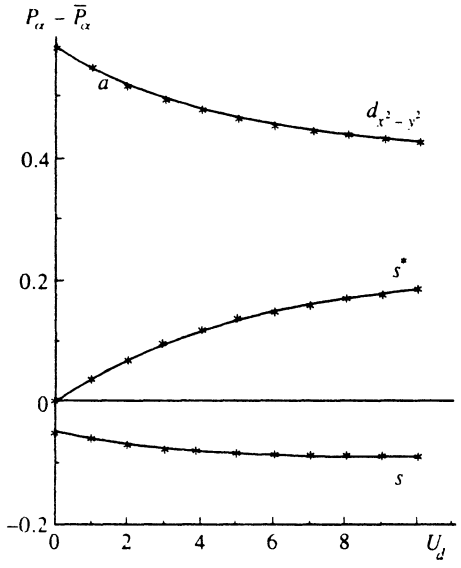

FIG. 7. Dependence of $P_\alpha - \bar{P}_\alpha$ on U_d in the s,s^*, and $d_{x^2-y^2}$ channels for the insulating undoped state of the Cu_4O_8 cluster with $N=4$ holes when $\varepsilon=1$, $U_p=V=0$, and $t=1$.

Figure 7 presents plots of the dependence of $P_\alpha - \bar{P}_\alpha$ on U_d/t when $\varepsilon/t=1$ and $U_p=V=0$. It can be seen that $P_\alpha - \bar{P}_\alpha<0$ in the s channel, but that the values of $P_\alpha - \bar{P}_\alpha$ in the s^* channel and especially in the $d_{x^2-y^2}$ channel are positive and considerably larger than the values in the optimally doped cluster (see Sec. 3 and Fig. 1). These results are consistent with the numerical investigations of the single-band 2D Hubbard model[26] and the $Cu_{16}O_{32}$ cluster.[27] As Scalettar *et al.*[27] suggest, the fact that P_α is greater than $\bar{P}_\alpha$ in the $d_{x^2-y^2}$ channel in the undoped state at low temperatures is a consequence of the appearance of an energy gap in the quasiparticle spectrum due to the formation of a spin density wave. We performed analytical calculations of $P_\alpha - \bar{P}_\alpha$, using a modified mean-field approximation and taking into account the presence of only antiferromagnetic (but not superconducting) correlations in the cluster. The very good quantitative agreement between the results of our calculations and the numerical data for large values of U_d indicates that in the case of $U_d>t$ magnetic interactions do, in fact, cause a sharp increase in $P_\alpha - \bar{P}_\alpha$ in the $d_{x^2-y^2}$ channel when $N=4$. This is probably true for the s^* channel, in which $P_\alpha - \bar{P}_\alpha$ increases rapidly with increasing U_d.

However, unlike the case of doped states, in the case of $N=4$ the results of the cluster calculations are very strongly influenced by effects caused by the finite size of the cluster. This follows from the fact that $P_\alpha - \bar{P}_\alpha \to 0$ when $U_p=V=0$ and $U_d \to 0$ only in the $\alpha=s^*$ channel, while in the $\alpha=s$ and $\alpha = d_{x^2-y^2}$ channels $P_\alpha - \bar{P}_\alpha$ tends to nonzero values as $U_d \to 0$, and that in the $d_{x^2-y^2}$ channel the value of $P_\alpha - \bar{P}_\alpha$ is positive and increases with decreasing U_d (although, if we start out from the assumption that large values of $P_\alpha - \bar{P}_\alpha$ are caused only by antiferromagnetic interactions,[27] a decrease in $P_\alpha - \bar{P}_\alpha$ with decreasing U_d would be expected due to the weakening of the antiferromagnetic correlations). We stress that $P_\alpha - \bar{P}_\alpha$ is finite at any, no matter how small, nonzero value of U_d/t (the extreme left-hand points in Fig. 7 correspond to $U_d/t=0.01$). When $U_d=U_p=V=0$, $P_\alpha - \bar{P}_\alpha$ is strictly equal to zero. The nonanalytic dependence of $P_\alpha - \bar{P}_\alpha$ on U_d/t in the s and $d_{x^2-y^2}$ channels is attributed to the fact that in the absence of an interparticle Coulomb interaction the ground state of the Cu_4O_8 cluster with $N=4$ holes is quadruply degenerate, and a nonzero value for at least one of the parameters U_d, U_p, and V results in complete removal of this degeneracy. The undoped state is probably more sensitive to the size of the cluster than are the doped states.

6. DISCUSSION AND CONCLUSIONS

In the present work we made an attempt to approach the question of the symmetry of the superconducting state in a high-T_c superconductor by avoiding uncontrollable analytical approximations and solving the many-particle Schrödinger equation exactly, but for a very small Cu–O cluster consisting of only 12 sites. Unlike the determinantal quantum Monte Carlo method, whose specific problem ("the minus-sign problem") precludes studying the characteristics of CuO_2 planes in the most interesting temperature range ($T<100$ K), the exact diagonalization of the Hamiltonian employed in the present work allowed us to calculate the pairing correlators $P_\alpha - \bar{P}_\alpha$ in the s, s^*, and $d_{x^2-y^2}$ pairing channels for the ground ($T=0$) state of the Cu_4O_8 cluster with various numbers of excess holes and excess electrons.

As follows from the results which we obtained, at the optimal doping level ($x=0.25$) the correlators $P_\alpha - \bar{P}_\alpha$ are negative in the s channel due to on-site Coulomb repulsion of the carriers. In the s^* and $d_{x^2-y^2}$ channels $P_\alpha - \bar{P}_\alpha>0$ under both hole and electron doping. The large value of $P_\alpha - \bar{P}_\alpha$ in the s^* channel indicates the presence of strong superconducting correlations with s^* symmetry in the optimally doped cluster. Under hole doping the $d_{x^2-y^2}$ channel makes a definite contribution to these correlations, but the correlator $P_\alpha - \bar{P}_\alpha$ in the $d_{x^2-y^2}$ channel is considerably smaller than that in the s^* channel. These results point out a fundamental difference between the multiband 2D Emery model and the single-band 2D Hubbard model, for which there are pairing correlations only in the $d_{x^2-y^2}$ channel and for which they are strongly suppressed in the s and s^* channels.[26]

In the case of strong doping ($x=0.5$), there are no pairing correlations in the s channel, and in the $d_{x^2-y^2}$ channel they are absent under electron doping and are very strongly suppressed under hole doping. In the s^* channel the value of $P_\alpha - \bar{P}_\alpha$ for $x=0.5$ is slightly smaller than the value for $x=0.25$, although a sharp decrease in $P_\alpha - \bar{P}_\alpha$ could have been expected in this pairing channel, since the state of a CuO_2 plane is not superconducting when $x=0.5$. The following facts should, however, be borne in mind. First, when the Cu_4O_8 cluster is investigated by exact diagonalization, the permissible carrier concentration values are highly discrete: $x=0$, 0.25, 0.5. It has not been ruled out that the value $x=0.25$ may differ quite strongly from the "optimal" value of x_0, at which the correlator $P_\alpha - \bar{P}_\alpha$ is maximal. This remark applies mainly to the case of electron doping,[33] since the maximum of T_c is achieved in n-type high-T_c supercon-

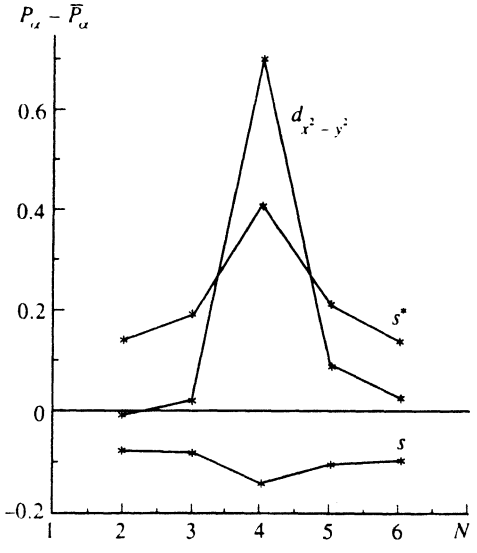

FIG. 8. Dependence of $P_\alpha - \bar{P}_\alpha$ on the number of holes N in the Cu_4O_8 cluster in the s, s^*, and $d_{x^2-y^2}$ channels when $\varepsilon=2$, $U_d=6$, $U_p=2.5$, $V=1.5$, and $t=1$: $N=4$ (undoped state); $N=5$ and 6 (hole doping); $N=2$ and 3 (electron doping).

ductors at $x_0=0.15$–0.17. Second, as was noted in Sec. 4, consideration of the variation of the occupancies of the copper orbitals in cases of doping (by means of the appropriate normalization of $P_\alpha - \bar{P}_\alpha$) results in a 10–60% increase in $(P_\alpha - \bar{P}_\alpha)_{x=0.25}/(P_\alpha - \bar{P}_\alpha)_{x=0.5}$, which specifies the degree of suppression of the pairing correlations upon the transition from the state with $x=0.25$ to the state with $x=0.5$.

In addition, the ratio $(P_\alpha - \bar{P}_\alpha)_{x=0.25}/(P_\alpha - \bar{P}_\alpha)_{x=0.5}$ is greatly dependent on the specific choice of the parameters of Hamiltonian (1). The values given for these parameters in different papers[37–42] differ quite strongly from one another, making the choice among them for numerical calculations highly speculative. Figure 8 presents plots of the dependence of $P_\alpha - \bar{P}_\alpha$ in the s, s^*, and $d_{x^2-y^2}$ pairing channels on the number of holes in the Cu_4O_8 cluster when $\varepsilon/t=2$, $U_d/t=6$, $U_p/t=2.5$, and $V/t=1.5$ (these values of the parameters were taken from Ref. 39). It has not been ruled out, however,[36] that V/t may be much greater than the value predicted by the calculations based on the local density functional approximation. An increase in V/t from 1.5 to 3 results in an increase in $(P_\alpha - \bar{P}_\alpha)_{x=0.25}/(P_\alpha - \bar{P}_\alpha)_{x=0.5}$ in the s^* channel from 1.50 to 1.84 under hole doping and from 1.36 to 1.53 under electron doping, i.e., $P_\alpha - \bar{P}_\alpha$ is considerably smaller when $x=0.5$ than when $x=0.25$, although complete suppression of the pairing correlations in the s^* channel does not occur even when $x=0.5$. We note that a conclusion regarding the predominance of the superconducting correlations specifically in the s^* pairing channel in a doped CuO_2 plane at high values of V/t was also drawn in Refs. 36 and 43.

The finite nature of the cluster which we investigated certainly has a definite influence on the results of the calculation, which is especially evident in the case of the undoped insulating state. If, for example, pairing correlations would be discovered for some particular set of model parameters or for some fixed carrier concentration, these data should, of course, be confronted with great skepticism.

However, the physical feasibility and reliability of the results of the cluster calculations are supported by the entire body of data that we obtained for the doped states of the cluster: the tendency of $P_\alpha - \bar{P}_\alpha$ to vanish in all pairing channels as the interparticle Coulomb interactions weaken, the absence of pairing correlations in the s channel at any doping level, the presence of pairing correlations in the s^* or $d_{x^2-y^2}$ channels for states "optimally" doped with either holes or electrons over a broad range of values of the parameters of the multiband 2D Emery model, the suppression of these correlations upon the transition from optimal doping to strong doping, and, finally, the good agreement of our results with the numerical and analytical calculations of other authors.

Thus, with consideration of all the foregoing remarks, it may apparently be stated that there are superconducting correlations in the s^* pairing channel in the Cu_4O_8 cluster doped by holes or electrons. Under hole doping there are also pairing correlations in the $d_{x^2-y^2}$ channel (although they are weaker). The questions of whether these correlations survive the transition to the thermodynamic limit and or whether there is long-range off-diagonal order in an infinite CuO_2 plane are still open. Calculations must be performed on systems of considerably greater size to resolve these questions.

In conclusion, at least some brief comments on the current experimental situation are in order. Evidence that the superconductive gap Δ in p-type high-T_c superconductors is anisotropic and possibly has the symmetry of a $d_{x^2-y^2}$ wave has been obtained in several studies.[22–24] Nevertheless, the experimental data are equivocal, and there is some basis to suppose that the symmetry of Δ is fairly complex (and possibly mixed[25]). Recent investigation of $Bi_2Sr_2CaCu_2O_8$ single crystals performed by scanning tunneling microscopy[23] showed that Δ is anisotropic in a CuO_2 plane: it is maximal in the directions of the crystallographic a and b axes and minimal in the direction at a 45° angle to these axes. Nevertheless, the minimal values of Δ are not equal to zero, as they should have been in the case of $d_{x^2-y^2}$ symmetry. It is still not clear whether this is attributable to inadequate experimental resolution or is due to other factors.

The $d_{x^2-y^2}$ symmetry of Δ is contested by the experimental data on the influence of nonmagnetic impurities and radiation defects on high-T_c superconductors. In superconductors with $d_{x^2-y^2}$ pairing nonmagnetic impurities are known to cause Δ and T_c to vanish at comparatively small values of the impurity concentration n_i, which correspond to $1/\tau \propto T_{c0}$, where τ is the relaxation time of the momentum of the carriers after scattering on defects and T_{c0} is the critical temperature in the absence of defects. However, according to experimental data (see, for example, Ref. 44), the values of T_c in high-T_c superconductors vanish at values of n_i corresponding to $1/\tau \propto E_F \gg T_{c0}$, where E_F is the Fermi energy. This also follows from the fact that the vanishing of T_c as n_i increases coincides with the onset of the process of carrier localization, as is evidenced by the exponential increase in the resistance R (Ref. 44 and 45), i.e., T_c vanishes at a value of n_i considerably greater than the value required for de-

struction of a superconducting state with $d_{x^2-y^2}$ symmetry. In addition, as was shown in Ref. 46, in which the direct effects of radiation defects on Δ was studied using Andreev reflection, the magnitude of Δ decreases with increasing n_i, but remains finite for $1/\tau \propto E_F$, for which T_c (as determined from the condition $R=0$) is already equal to zero. At the same time, a superconducting state with s^* symmetry is suppressed to a far lesser extent by nonmagnetic defects, but this does not contradict the experimental facts cited.

It has not, however, been ruled out that in p-type high-T_c superconductors the symmetry of the gap is mixed (for example, an s^* wave+a $d_{x^2-y^2}$ wave). This hypothesis might make it possible to reconcile the data from different experiments. We stress that our calculations indicate the existence of superconducting correlations in both the s^* channel and the $d_{x^2-y^2}$ channel under hole doping, although the correlations in the $d_{x^2-y^2}$ channel are far weaker.

With respect to n-type high-T_c superconductors, we do not know of any experimental studies whose results could be interpreted as supporting $d_{x^2-y^2}$ symmetry for the gap. Our data indicate that under electron doping the superconducting correlations in the $d_{x^2-y^2}$ channel are, in fact, very small, being one or two orders of magnitude weaker than those in the s^* channel. Just this may be the reason for the lower values of T_c in n-type high-T_c superconductors in comparison with p-type high-T_c superconductors.

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