Cluster approach to the three-band Hubbard model of the Cu-O plane: Superconducting pairs

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(Received 18 July 1997; revised manuscript received 29 August 1997)

The fully symmetric Cu-O clusters with a central Cu have two-hole singlet eigenstates which are not affected by the on-site repulsion. These pairs become bound states if the number of holes in the cluster is appropriate; moreover, the quantization at half-integer values of a magnetic test flux is consistent with a superconducting pattern. To investigate the mechanism of pairing, we study the symmetric clusters with $\leqslant 21$ atoms and four holes by exact diagonalization. Further, by the diagrammatic expansion of the ground-state energy and of the scattering amplitude, we obtain the spin-flip diagrams for the effective interaction, which is attractive for singlets and repulsive for triplets. A criterion for pairing in cluster calculations is thereby obtained. [S0163-1829(97)04446-9]

I. INTRODUCTION

The three-band Hubbard model has been widely used for electron spectroscopy studies[1] of the Cu-O plane of high-T_c superconductors and for investigating their electronic properties, and the relevant region of the parameter space has been fairly well estabilished by *ab initio* calculations[2] as well. The infinite-U limit of this model gives *d-wave* superconductivity mediated by an attractive effective interaction.[3,4] The possibility that bound pairs arise from purely electronic processes, like the exchange of spin fluctuations, has been suggested long ago;[5–7] in the related *t-J* model, finite cluster calculations have also provided evidence of pairing[8] and diagrammatic studies[9,10] have supported a mechanism based on the exchange of spin waves with large momentum transfer. Since the three-band Hubbard model makes direct reference to the individual atomic orbitals and to the on-site interaction, we believe that it can provide a useful viewpoint on the correlation effects that allow the holes to overcome repulsion and convert it into an effective attraction; exact calculations on finite models may bring to light interesting local aspects of the microscopic dynamics, and can be used as tests for the analytical approximations.

Recently,[11–14] we have pointed out that a class of highly symmetric clusters exhibit hole pairing in real space; as a criterion for pairing we used[15] $\Delta(n)<0$, where $\Delta(n)=E(n)+E(n-2)-2E(n-1)$, and $E(n)$ is the ground-state energy of the system with n holes. In this paper, we first extend the previous findings by performing numerical calculations on larger clusters. Next, we investigate the nature of the effective interaction which explains the results of the exact diagonalizations; it is clear that a thorough understanding of the microscopic processes is needed before attempting to address the thermodynamic limit. Similar to other approaches, the mechanism which operates in our clusters involves spin flip; in our description, the effective interaction is a correlation effect which depends in an essential way on the point symmetry of the plane. The pairing interaction and the symmetry of the wave function remove the ambiguity in the $\Delta(n)<0$ criterion; in fact, negative Δ by itself does not necessarily mean pairing, and in unsymmetric geometries and exotic parts of the parameter space it can arise from different mechanisms. The above criterion has also been questioned[16] because when one introduces the nuclear degrees of freedom, the sign of Δ can be reversed. This can happen when the ground state with $n-1$ holes is degenerate, and Jahn-Teller distorts, gaining energy by the distortion; the pairing then looks like an artifact due to the neglect of vibrations. Ultimately, the argument runs into trouble because it depends on a comparison of ground-state energies with different hole numbers n.

In this paper, we begin by using $\Delta(n)<0$ as a preliminary criterion, but the analysis will lead us eventually to propose a new one, which is free from the above criticisms. When the actual point symmetry of the system is accounted for, as in our model, the effective interaction provides the dynamical explanation for $\Delta(n)<0$ and for the fact that it does imply pairing in this case. Finally, still by exact diagonalization, we study the quantization of a test flux in these clusters, and find a behavior which is consistent with superconducting pairing.

II. MODEL HAMILTONIAN

The key ingredient of our approach is cluster geometry, while the semiempirical Hubbard Hamiltonian that we use is quite standard.[11] It reads $H=h+H_c$, where h is the one-body part and H_c is the interaction. Here,

$$h=\sum \epsilon_{ij} c_{i\sigma}^{\dagger} c_{i\sigma}, \tag{1}$$

where $c_{i\sigma}$ are hole annihilation operators, $\varepsilon_{ii}=\varepsilon_p$ for an O site, $\varepsilon_{ii}=\varepsilon_d=0$ for a Cu site, $\varepsilon_{ij}=t$ for a Cu-O bond, and $\varepsilon_{ij}=t_{ox}$ for an O-O bond. Here, we are concerned primarily with the dynamical meaning of $\Delta<0$, and since the O-O hopping integral t_{ox} is marginal (maximum binding occurs for vanishing t_{ox} in all cases), we limit the present analysis to $t_{ox}=0$. We split the interaction part H_c as follows:

$$H_c=W+H_{\text{off-site}}, \tag{2}$$

where the on-site part is

$$W=\sum_i U_i n_{i+} n_{i-}, \tag{3}$$

n is a number operator, $U_i=U_p$ or U_d for O and Cu sites, respectively. We have included the off-site repulsion $H_{\text{off-site}}$ in our model;[11] it was seen that O-Cu interactions favor pairing and O-O interactions oppose it; with reasonable values of U_p and U_d the effect of $H_{\text{off-site}}$ does not change matters in any important way and we are going to neglect it altogether in the following for the sake of simplicity. Below, we use the following parameter values (in eV): Cu-O hopping $t=1.3$, on-site interactions $U_d=5.3$, $U_p=6$ for Cu and O, respectively; atomic energy-level difference ε_p-$\varepsilon_d=3.5$. Such values compare well with theoretical predictions[2] and with evidence from electron spectroscopy, as we have discussed elsewhere.[11]

III. $W=0$ PAIRS, ''ALLOWED'' CLUSTERS, AND $\Delta<0$

Two holes in the same degenerate level of the one-body Hamiltonian h may form several singlet pairs; diagonalization of the on-site repulsion W in this space yields pair eigenstates of the total Hamiltonian H. The $W=0$ pairs are *two-hole* singlet eigenstates of H which are also exact eigenstates of W with eigenvalue 0. While triplet pairs are trivially not affected by the on-site repulsion in this model, the same property for singlet pairs results from a configuration interaction which yields $W=0$; a null result, however, can only arise because of symmetry. In the infinite Cu-O plane, such pairs exist,[12] along with ''normal'' singlet pair eigenstates for which $\langle W\rangle$ does not vanish. This suggests that finite clusters can faithfully represent the hole-hole correlations that occur in the plane only if they possess the full C_{4v} symmetry. We found[11] that this condition is necessary but not sufficient; only if the cluster is centered around a Cu ion, some of the degenerate levels (but not all of them) always yield $W=0$ pairs.

Thus, for short, we term ''allowed'' clusters those centered around a Cu atom and having the full C_{4v} symmetry. The $W=0$ pairs have far reaching consequences on the many-body properties, and single out the allowed clusters as good models of the plane. The present discussion does not apply to the forbidden geometries, like those examined by Hirsch *et al.*[15] and Balseiro *et al.*[17] The Cu_4O_4 geometry considered by Ogata and Shiba[18] has the C_{4v} symmetry of the lattice, but lacks the central Cu, and therefore it is forbidden (the pairs on degenerate levels feel the on-site repulsion, as one can see by performing the two-hole configuration interaction calculation[11]). The dynamics of holes in Cu_4O_4 (Ref. 19) does not lead to pairing if the on-site repulsion on oxygen is included.

Consider an allowed cluster in the noninteracting limit, and suppose filling the levels with n holes according to the *Aufbau* principle; when interactions are introduced, pairing is favored if n is such that the uppermost pair is a $W=0$ singlet. We reported previously exact diagonalization calculations with $n=4$ yielding $\Delta<0$ in CuO_4 (Ref. 11) and Cu_5O_4 (Ref. 13) in a physically relevant part of the parameter space. Here, we study Cu_5O_8, Cu_5O_{12}, and Cu_5O_{16} (44 100 configurations), using an improved Lanczos technique. The present paper is primarily devoted to the dynamics of the paired state in this series of clusters, namely, all the allowed ones up to 21 atoms; $n=4$ holes are enough to show the effect, because the lowest one-hole level belongs[20] to a_1 symmetry, and the next $e(x,y)$ level yields the $W=0$ pair. The latter has a spatial wave function transforming like $x(1)y(2)+x(2)y(1)$, where x and y are orbitals, and belongs to the ${}^1B_2(xy)$ representation; note, however, that the charge density of the pair has lobes along the axes, and is in line with an order parameter having the ${}^1B_1(x^2-y^2)$ symmetry[12] which is favored by experimental evidence.[21] Group theory shows that in the four-body problem, the interactions resolve the ground $e\otimes e$ configuration into ${}^3A_2$, ${}^1A_1$, ${}^1B_1$, and ${}^1B_2$; the numerical ground state of the system belongs to ${}^1B_2$ like the $W=0$ pair and $\Delta<0$. The calculated trend of Δ with the number of atoms N is shown in Fig. 1. The size of these clusters is comparable with that of pairs in high-T_c superconductors, while the magnitude of $|\Delta|$ at fixed n decreases as N is increased.

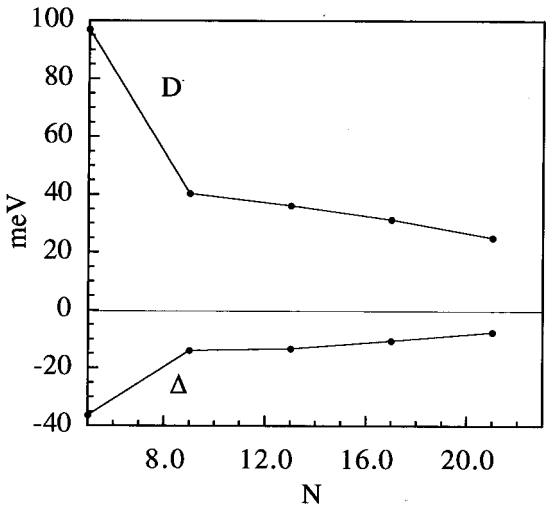

FIG. 1. Δ and singlet-triplet separation D for allowed clusters containing N atoms and four holes. The dots are the values calculated with the parameters listed in the text; $t_{ox}=0$. The curves are a guide to the eye. The clusters are CuO_4, Cu_5O_4, Cu_5O_8, Cu_5O_{12}, and Cu_5O_{16}.

Larger allowed clusters are outside the scope of the present paper. We note in passing, however, that beyond 21 atoms more than four holes become necessary; for instance, in $Cu_{13}O_{36}$ the second one-hole level, although twice degenerate, gives rise to ''normal'' pairs. Ascending the series of bonding levels, one finds that the fourth is a set of degeneracy 5 containing $e(x,y)$, a_1 (twice) and b_2 components. By two-hole configuration interaction, one obtains 6 $W=0$ pairs, and we predict pairing for even n, with $10\leqslant n\leqslant 20$. Such systems might shed light on interactions between pairs and eventually on superconductivity in the three-band Hubbard model. Unfortunately, the numbers of configurations ($>10^{12}$ already for $n=10$) puts them outside the reach of direct diagonalization methods.

IV. EFFECTIVE INTERACTION

In all the allowed clusters up to 21 atoms, the lowest one-hole level belongs[20] to a_1 symmetry, and the next

$e(x,y)$ level yields the $W=0$ pair; there are b_1 and a_1 empty orbitals, that below we shall write with the short-hand notations b and a', respectively; empty orbitals of $e(x,y)$ symmetries also exist in all cases except CuO_4. We noted above that the interactions produce a nondegenerate $^1B_2$ four-hole ground state with $\Delta<0$. Negative Δ values are usually taken as *indication* of pairing, but in principle they could result from other microscopic mechanisms;[16] however, the very fact that the present symmetry-driven mechanism involves two holes in degenerate states strongly suggests that an effective attraction between those holes is leading to bound pairs (rather than to a condensation or bag of particles). Further support for an effective hole-hole attraction comes from the observation[13] that the hole charge distribution of the pair shrinks and tends to concentrate more on oxygen when pairing prevails. Indeed, we show that in the case of dressed $W=0$ singlet pairs, $\Delta<0$ arises from an effective pairing interaction which is attractive; the same interaction is repulsive for triplet pairs. The argument rests on a comparison of the perturbation series for Δ and for the two-hole amplitude; for the dressed $W=0$ pair the two-hole amplitude actually involves Δ and yields its dynamical interpretation.

A. Diagrams for Δ

The perturbation series for Δ in powers of W is obtained by the well-known diagrammatic expansion[22] of the ground-state energy E of a many-body system. This is based on the theorem

$$E=E^{(0)}+i\lim_{t\to\infty(1-i\eta)}\frac{d}{dt}\ln\widetilde{R}(t), \tag{4}$$

where $E^{(0)}$ is the noninteracting ground-state energy, $\eta=+0$;

$$\widetilde{R}(t)=\langle\Phi_0|U(t)|\Phi_0\rangle, \tag{5}$$

$|\Phi_0\rangle$ is the noninteracting ground state, and $U(t)$ is the time evolution operator in the interaction representation. One then expands the correlation function $\widetilde{R}(t)$ using the linked cluster theorem, that simplifies the expansion, but the diagrams that violate the Pauli principle and/or the number of particles must be retained. The diagrammatic rules are readily applied to the $n=2$ case, when $|\Phi_0>$ is a nondegenerate single-determinant state. For $n=3$ and $n=4$, the non interacting ground state is degenerate, while the derivation of Eq. (4) assumes that the interactions do not modify the symmetry of the ground state; therefore we take $|\Phi_0\rangle$ as the x component of the ^2E irreducible representation for $n=3$ and $^1B_2$ for $n=4$; we know from direct diagonalization that these symmetries are correct for the whole series of clusters. The $^1B_2$ component of the noninteracting ground state reads

$$|\Phi_0\rangle=\frac{|xy\rangle+|yx\rangle,}{\sqrt{2}} \tag{6}$$

where in terms of orbitals $|xy\rangle$ is the single determinant $|x_+y_-a_+a_-|$. Since this cannot be written as a single determinant, some care is necessary in the diagrammatic expansion; the correlation function is

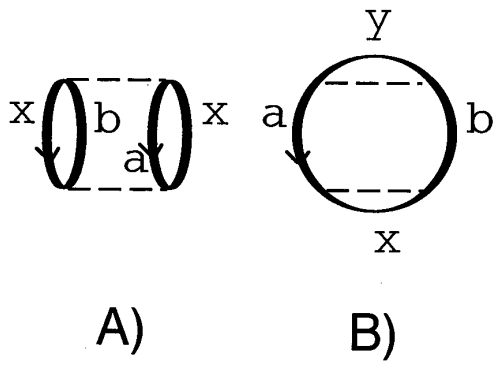

FIG. 2. Linked diagrams contributing to the second-order expansion of $E(4)$. Diagram (A) is normal, while (B) is anomalous. The a, b, x, and y lines are labeled according to the representations of the C_{4v} group.

$$\widetilde{R}(t)=\langle xy|U(t)|xy\rangle+\langle yx|U(t)|xy\rangle. \tag{7}$$

Applying the linked cluster theorem,[22] the first term leads to the standard diagrams, while the second is anomalous, in the sense that it vanishes in the noninteracting limit. Thus, we find all the diagrams contributing to the expansion for $U(t)$ averaged over $|x_+y_-a_+a_-|$, plus the anomalous or spin-flip diagrams that have entering x_+ and y_- and outgoing y_+ and x_- lines (Fig. 2). Let $W(k,l,m,n)=\langle k_+l_-|W|m_+n_-\rangle$; the anomalous diagram [Fig. 2(b)] can be obtained from the normal one [Fig. 2(a)], which is proportional to $W(a,b,x,x)^2$, simply by letting $W(a,b,x,x)\to W(a,b,y,y)$ in the upper interaction line. A sign change follows, in agreement with the standard diagrammatic rules. In summary, one starts the expansion as if the ground state were the single determinant $|x_+y_-a_+a_-|$; the diagrams that involve propagating x and y lines get corrections, which are obtained by exchanging x and y in the upper interaction line where the x or y lines end; the loose lines should then be joined, and the diagram so obtained carries a minus sign because of the change in the number of loops. After some algebra, we obtain the second-order approximation

$$\Delta^{(2)}=-2\left[\sum_b\frac{W(a,b,x,x)^2}{(\varepsilon_b-\varepsilon_a)}-\sum_{a'}\frac{W(a,a',x,x)^2}{(\varepsilon_{a'}-\varepsilon_a)}\right], \tag{8}$$

where ε_a is the one-hole energy of the a orbital and so on; the sums run over all empty states of the appropriate symmetries (a_1 and b_1), while no contributions arise from the empty e orbitals since the relevant W matrix elements vanish. The sign of Δ is seen to depend on the relative weight of the virtual transitions to states of different point symmetry, and ultimately on the parameters in the Hamiltonian.

B. Diagrams for the two-hole amplitude

Let G denote the two-hole amplitude for holes of opposite spin in the degenerate (x,y) orbitals. Singlet and triplet arise from the space spanned by the noninteracting states $|xy\rangle$ and $|yx\rangle$, which are connected by C_{4v} operators. In the Nambu formalism, G is a matrix

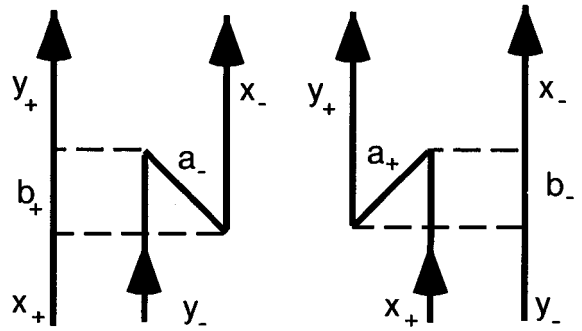

FIG. 3. Typical spin-flip diagrams for the anomalous propagator $A_{\rm sf}$.

$$G(\omega)=\begin{pmatrix} A & A_{\rm sf} \\ A_{\rm sf} & A \end{pmatrix}; \tag{9}$$

here A is a normal propagator, while $A_{\rm sf}$ involves a spin flip and vanishes in the noninteracting limit. When interactions are included, the first-order contribution to the scattering amplitude vanishes because the product of x and y orbitals vanishes on all sites. Therefore, the spin-flip process, $A_{\rm sf}$, shown in Fig. 3, is the lowest-order $x-y$ scattering, and produces the effective interaction to second order.

Evaluating $A_{\rm sf}$ according to the standard rules, we find that near $\omega=2\varepsilon_x=2\varepsilon_y$, $A_{\rm sf}\approx i\Delta^{(2}/(\omega-2\epsilon_x)^2$ +less singular terms; moreover, to second order, $A=i/(\omega-2\epsilon_x)$. We know from symmetry that $G(\omega)$ has singlet and triplet eigenvectors $1/\sqrt{2}(1,\pm 1)$; for the singlet and triplet we get

$$A+A_{\rm sf}=\frac{i}{\omega-\eta_s}, \quad A-A_{\rm sf}=\frac{i}{\omega-\eta_t}, \tag{10}$$

where $\eta_{s,t}$ are the new eigenvalues. Therefore, for the singlet the expansion is

$$\frac{1}{\omega-\eta_s}\approx\frac{1}{\omega-2\epsilon_x}+\frac{\Delta^{(2}}{(\omega-2\epsilon_x)^2}\approx\frac{1}{\omega-2\epsilon_x-\Delta^{(2)}}, \tag{11}$$

that is $\eta_s=2\varepsilon_x+\Delta^{(2)}$; the triplet receives the opposite correction, and the ${}^1B_2-{}^3A_2$ separation is $D=2|\Delta|$. Negative Δ means that the spin-flip interaction is attractive for ${}^1B_2$ which is pushed down by $|\Delta|$ and becomes the ground state, while the triplet is pushed up by the same amount.

Since U_p and U_d are not small compared to the Cu-O hopping term t, the second-order is generally a poor approximation; interestingly, $W=0$ pairs are an exception, because the large interactions are *dynamically* small. The first-order term vanishes, and the second order is of the order of tens of meV. Comparison with exact numerical diagonalization results shows that the second-order approximation for D and Δ is already rewarding, and In Fig. 1 we show how D and Δ scale with the cluster size; they are indeed very closely proportional, although their ratio is somewhat larger than 2.

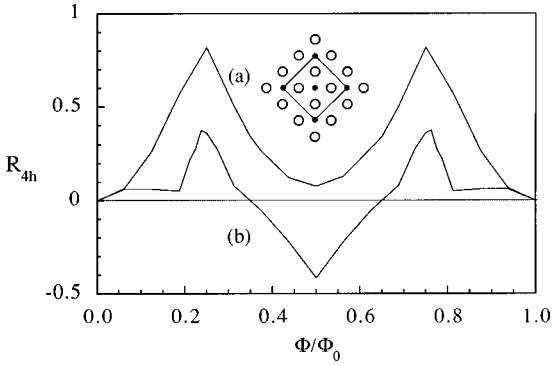

FIG. 4. Four-hole response function R versus the normalized flux, for (a) Cu_5O_4 and (b) Cu_5O_{16}. The parameter values are the same as in Fig. 1. The inset shows the geometry of Cu_5O_{16} with the closed path.

V. FLUX QUANTIZATION

We test the paired state by exposing the system to a vector potential **A(r)** according to the Peierls prescription,

$$\epsilon_{ij}\rightarrow\epsilon_{ij}\exp\left[\frac{2\pi i}{\phi_0}\int_{r_i}^{r_j}\mathbf{A}\cdot d\mathbf{r}\right], \tag{12}$$

where $\phi_0=hc/e$ is the flux quantum, and looking for superconducting correlations. In a macroscopic experiment, one makes a sample with a large hole, and inserts a magnetic field; the ground-state energy $E(\phi)$ is trivially a periodic function of ϕ/ϕ_0, where ϕ is the flux in the tube, and $\phi=0$ is an extremum. Superconductors quantize the flux by allowing integer and half-integer multiples of ϕ_0. The reason is that a flux $\phi_0/2$ corresponds to a superconducting ground state with pairs having different symmetry than those prevailing at $\phi=0$, and the system is stable; arbitrary ϕ values are not allowed because they cost large amounts of (free) energy.

Canright and Girvin have shown[23] that cluster calculations can be used to obtain qualitative insight on the occurrence of superconductivity, by looking for a tendency to flux quantization. The signature[24,25] is present when $\phi=0$ is a minimum of $E(\phi)$ and the only other minimum of comparable depth occurs at $\phi/\phi_0=\frac{1}{2}$; the barriers separating the minima are small in a small system but one expects them to increase with size, leaving the flux quantized in units of $\phi_0/2$. Canright and Girvin[23] used a square lattice of rectangular geometry and periodic boundary conditions along one of the axes; to demonstrate the effect, they assumed an *attractive* on-site interaction and observed superconducting flux quantization for even hole numbers and strong enough attraction. Since W is purely *repulsive*, we cannot modify the geometry in analogy with Ref. 23, because the cluster would become very unsymmetric and forbidden. We must insert the flux tube inside allowed clusters, but since no closed path encircling the central Cu is available for the holes, there is no response to a central flux tube. Therefore, we use an *external* device providing a closed path around the flux tube. The device, however, cannot be chosen arbitrarily, because the cluster must remain allowed and the singlet pair must remain one with $W=0$. To fulfill this condition, we introduce an infinitesimal hopping t_d between the external Cu's (see the inset of Fig. 4) and study the linear response to this perturbation. The relevant response function R is

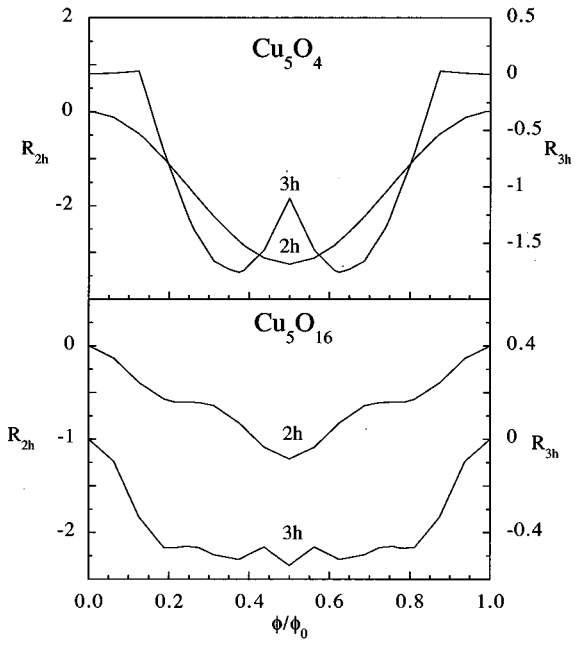

FIG. 5. Two-hole and three-hole response functions R versus the normalized flux, for Cu_5O_4 and Cu_5O_{16}. The parameter values are the same as in Fig. 1.

$$R(\phi)=\frac{E(\phi)-E(0)}{t_d} \tag{13}$$

and depends on the test flux ϕ (in units of ϕ_0). The results for Cu_5O_4 and Cu_5O_{16} with four holes are shown in Fig. 4; the same trend is obtained for all clusters. The minimum at $\phi_0/2$ is the microscopic precursor of the superconducting flux quantization; by looking at Δ and analyzing the numerical ground-state wave functions at $\phi_0/2$ we find that it corresponds to pairs of $^1A_1$ symmetry which are bound in the presence of half a flux quantum. Quite different patterns in which the flux quantization is absent are obtained for two and three holes (Fig. 5). Also, it is clear that it is W that forces the paired holes to screen the vector potential like a charge $2e$; actually, the noninteracting case is quite different (Fig. 6). If the input data are modified in such a way that Δ becomes positive by severe distortion of the symmetry, the characteristic central minimum is lost altogether.

VI. CONCLUSIONS

The three-band Hubbard cluster models having allowed geometries and appropriate hole numbers predict *d-wave* pairing; the geometry and the hole number n must be such that the two least bound holes occupy a $W=0$ singlet state. We have diagonalized exactly the Hamiltonian for a series of clusters, confirming and extending the earlier finding that this symmetry enhances correlation effects to the point that repulsion generates attraction; the paired ground state belongs to $^1B_2$. In the presence of a vector potential, a suitable response function measures the strength of superconducting correlations. We find that along with the $^1B_2$ paired state which holds for no flux, a second $^1A_1$ paired state corresponds to half a flux quantum; the change of symmetry is similar to what happens with BCS pairs.[24]

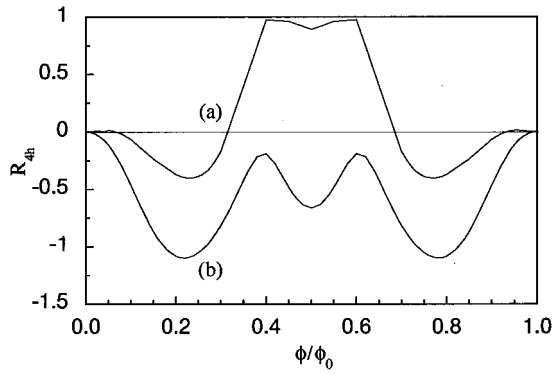

FIG. 6. Four-hole response function R versus the normalized flux, for (a) Cu_5O_4 and (b) Cu_5O_{16} in the absence of interactions. The parameter values are the same as in Fig. 1, except $U_p=U_d=0$.

The numerical results are a test ground for the diagrammatic analysis and allow us to identify the pairing mechanism, at least for the models at hand. The spin-flip second-order term in the scattering amplitude provides the effective interaction, which is attractive for singlets and leads to bound states; the same interaction is repulsive for triplets. The effective interaction can be monitored by the $^1B_2$ ground state it produces, while its strength is measured by the singlet-triplet separation; thus we can safely interpret $\Delta<0$ as pairing without having to depend on a comparison between systems with different hole numbers.

The basic correlation effect depends crucially on the point symmetry of the cluster. We believe that owing to their symmetry these allowed clusters share important many-body properties of the infinite plane, and definitely deserve further study.

ACKNOWLEDGMENTS

This work was supported by the Istituto Nazionale di Fisica della Materia. We acknowledge A.R. Bishop, Los Alamos Laboratories, New Mexico, and R. Del Sole and G. Onida, Universita' di Roma Tor Vergata, for useful discussions.

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