Superconductivity in the multiband Hubbard model with long-range Coulomb repulsion

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A multiband CuO Hubbard model is studied which incorporates long-range (LR) repulsive Coulomb interactions. In the atomic limit, it is shown that a charge transfer from copper to oxygen ions occurs as the strength of the LR interaction is increased. The regime of phase separation is replaced by a uniform state with doubly occupied oxygens. As the holes become mobile a superfluid condensate is formed, as suggested by a numerical analysis of pairing correlations and flux quantization on one-dimensional chains. The competition between charge-density wave and superconductivity is also analyzed. It is argued that the results are also applicable to two dimensions.

I. INTRODUCTION

The study of high-T_c superconductors[1] continues attracting much attention. Recently, progress has been made in the theoretical search for ground-state superconductivity in one-band electronic models. In particular, the two-dimensional (2D) t-J model has shown indications of d-wave superconductivity[2] when analyzed near the phase separation (PS) regime[3] at intermediate densities. It would be desirable to extend these observations to more realistic three-band versions of the Hubbard model,[4,5] especially in the region of parameters where the mapping to the t-J model[6] is not valid. Although many experimental results point to the presence of d-wave superconductivity in the cuprates, and it is generally accepted that spin-fluctuation-mediated superconductivity is d wave, it is also important to explore mechanisms of superconductivity in purely electronic models other than those based on spin fluctuations. Not much is known about the superconducting properties of these models. Weak-coupling and large-N mean-field calculations have indeed shown that a region of superconductivity exists near phase separation in the multiband Hubbard model as in the one-band case.[7] However, these techniques are only approximated and should be supplemented by unbiased computational studies, like exact diagonalization and quantum Monte Carlo methods.[8,9]

Varma, Schmitt-Rink, and Abrahams[5] have suggested that the inclusion of a short-range Coulomb repulsion induces a charge-transfer process, leading to the formation of tightly bound hole pairs on the oxygen ions. Unfortunately, the charge-transfer mechanism seems systematicallly correlated with a PS process, as was recently discussed in the atomic limit.[10] Thus, these pairs are not mobile and the system cannot become superfluid. In this sense, PS is an unwelcomed effect in this model. In spite of this problem, Sudbø *et al.*[11] argued that the one-dimensional (1D) model exhibits superconducting correlations immediately before phase separation. Similar results were reported by Sano and Ono[12] in the $U_d=\infty$ limit. These papers analyze the 1D problem by studying the parameter K_ρ used in conformal field theory rather than pairing correlations. In fact, in a previous study,[13] it was found that the tail in the pairing correlations is negligible in the regime of parameters considered in Refs. 11 and 12 (see Fig. 7 below). In any case, it would be difficult to predict the validity of the results obtained by the analysis of Ref. 11 in the more realistic two-dimensional problem, since no intuitive picture supporting the presence of superconductivity was provided. Thus, it would be desirable to have a model which exhibits robust superconducting pairing correlations in the ground state, and whose main features can be understood intuitively, allowing its extension to two dimensions where explicit numerical simulations are very difficult.

In this paper, a simple modification of the standard multiband model is presented that addresses these issues. The multiband Hubbard model for the Cu-O planes is defined by[4,5]

$$H=-t_{\rm pd}\sum_{\langle ij\rangle\sigma}(c^\dagger_{i\sigma}c_{j\sigma}+\text{H.c.})+U_d\sum_i n_{i\uparrow}n_{i\downarrow}$$
$$+U_p\sum_j n_{j\uparrow}n_{j\downarrow}+\epsilon_d\sum_i n_i+\epsilon_p\sum_j n_j\ , \qquad (1)$$

where $c^\dagger_{i\sigma}, c^\dagger_{j\sigma}$ are creation operators of holes ($\sigma=\uparrow,\downarrow$), $n_{l\sigma}=c^\dagger_{l\sigma}c_{l\sigma}$, and $n_l=n_{l\uparrow}+n_{l\downarrow}$. The label i corresponds to Cu sites and j to O sites. U_d and U_p are the Coulomb on-site repulsion energies for the copper oxygen sites, respectively, and ϵ_d and ϵ_p are the ion energies. The charge-transfer energy is defined by $\Delta=\epsilon_p-\epsilon_d$. It is generally accepted that U_d should be larger than Δ and U_p in the cuprates, so we will work in this regime. The doping fraction is $x=n_h/N$, where $n_h=(N_h-N)$ is the number of doped holes, N_h is the total number of holes, and N is the number of Cu-O cells. At half-filling ($x=0$) and in

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this region of parameters, the model presents antiferromagnetic order as in the undoped parent compounds. Most of the results presented here correspond to intermediate doping ($0.25 \leq x \leq 0.66$).

As explained before, Varma and co-workers[5,10,11] studied the Hamiltonian Eq. (1) supplemented by a nearest-neighbor Coulomb repulsion (regulated by a parameter V) which induces phase separation [note that this new parameter can be estimated from an analysis of the experimentally measured dielectric constant and it is of order 2–3 eV s (Ref. 14)]. To avoid the phase-separation problem, we have extended the range of the density-density interactions. The relevance of the long-range part of the Coulomb interaction between the holes, which prohibits PS, was emphasized in a recent work by Emery and Kivelson.[15] In particular, if the interactions are of infinite range, then phase separation is certainly eliminated.[16] This leads us to add to Eq. (1) a more general interaction

$$H_C = \frac{V}{2} \sum_{l \neq m} \frac{n_l n_m}{d_{lm}} e^{-d_{lm}/\lambda} , \tag{2}$$

where $d_{lm} = |\mathbf{r}_l - \mathbf{r}_m|$ and λ is the range of the screened Coulomb repulsion. Since periodic boundary conditions are used, d_{lm} is defined as the shortest distance between points l and m. A finite λ represents the screening effects produced by degrees of freedom not included explicitly in the Hamiltonian (like other planes, impurities, etc.)

II. ATOMIC LIMIT

To motivate the introduction of a long-range term in the Hamiltonian it is convenient to first consider the atomic limit $t_{pd} = 0$.[17] In the 1D case, it is not difficult to guess which are the possible states with lowest energy for different values of V. In Fig. 1(a) we show the ground state in the regime of small V. This state (denoted by I), has a hole at every copper ion, and the doped holes are on singly occupied oxygens. On increasing V, a phase-separated state [denoted by II and shown in Fig. 1(b)] becomes energetically competitive with state I.[10,13] It contains a region of doubly occupied oxygens and another separated region with one hole per copper ion. However, there is a third possibility which becomes energetically favorable due to the long-range Coulomb interaction. In Fig. 1(c) we show a state (III) where all the holes are on doubly occupied oxygens forming a periodic pattern. The reason for considering this state is that charge tends to spread uniformly over the lattice in the presence of long-range forces, rather than separating into different densities (Wigner crystal). State III is a configuration with "preformed" pairs on the oxygens and a finite hopping amplitude may render it superfluid as in the attractive Hubbard model at large $|U|/t$. Unfortunately, this analogy shows that charge-density-wave (CDW) states will compete against superconductivity, since in the negative-U Hubbard model at half-filling, CDW and superconductivity are degenerate. A study of the 1D t-J model with $1/r$ interactions have shown already the strong competition CDW-superconductivity.[18] Then, it is not enough to add a long-range interaction to make a superfluid a phase-separated state. Whether CDW or superconductivity dominates is a highly nontrivial issue which requires a controlled calculation (as described below).

To verify that the states of Fig. 1 are indeed the correct ground states of the pure long-range ($\lambda = \infty$) model in the atomic limit, we have compared the energies of states I, II, and III numerically for 12, 24, and 48 CuO cells. We have considered $U_d = 7$, $U_p = 1$, $\Delta = 1.5$, in arbitrary units, and $x = 0.33$. For the largest cluster considered there is a direct crossing from state I to state III at $V_c \approx 3.38$. In the 2D case, we have carried out explicit calculations for 4×4 and 6×6 CuO_2 periodic clusters at a hole concentration $x = 0.25$. In this case the ground state is not obvious, and it is difficult to determine it by calculating the energy of each possible configuration because their number grows exponentially with the number of atoms. For this reason we developed a simulated an-

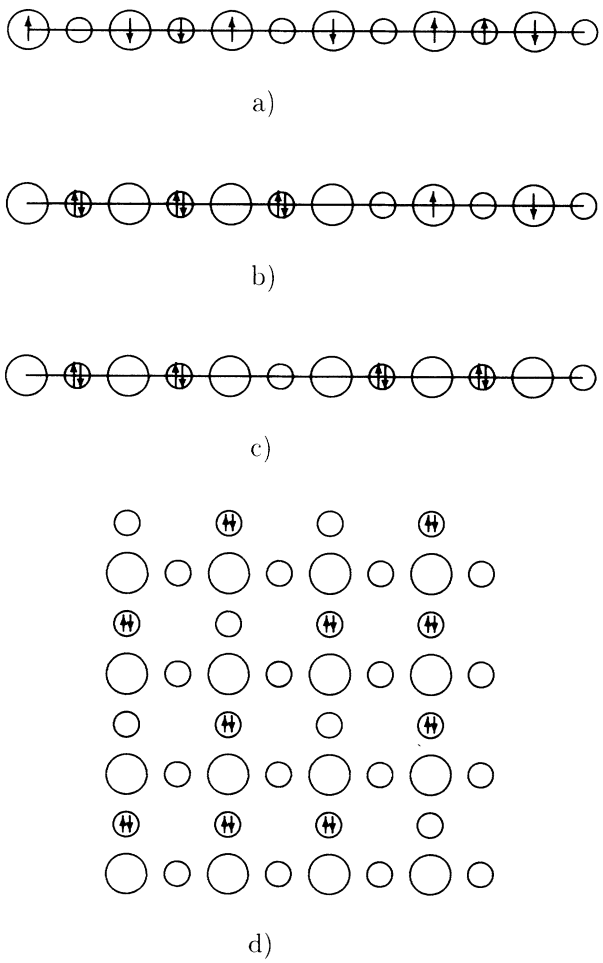

FIG. 1. (a) Ground state of the Hamiltonian Eqs. (1) and (2) for small V, in the atomic limit. The copper ions are represented by large circles and the oxygens by small circles. In this and the following figures the alignment of spins is arbitrary. (b) Ground state in the region of small λ (short-range interactions) and large V. This state has phase separation and was previously discussed in Ref. 10. (c) New state discussed in this paper. It becomes the ground state in the case of large λ and sufficiently large V. (d) Generalization of the state shown in (c) to a 2D lattice.

nealing[19] program to find the ground state. This algorithm can be described as a sequence of Monte Carlo runs, each one generated at a fixed value of the temperature. The temperature is decreased between runs until the ground state is reached. At each step of a Monte Carlo run, a possible move consisting of the hopping of a hole to a nearest-neighbor site is offered. The local variation of energy ΔE is computed and the move is accepted by comparing its corresponding Boltzmann factor $\exp(-\beta\Delta E)$ with a random number. Typically, the ground state was reached with Monte Carlo runs of 100 000–200 000 sweeps over the lattice and for the lowest temperature $T_{\min}\approx 2$ in units of U_p. Using this technique, we have determined the ground state of this cluster for the same set of parameters U_d, U_p, and Δ as before. For small V, the ground state has one hole per Cu atom and the doped holes are in O sites located as far as possible from each other. As V is increased, a charge transfer from Cu to O sites starts to develop. For $V\approx 3$, a state with doubly occupied O sites separated from a region of single-occupied Cu sites becomes stable. For larger values of V, the ground state has the form illustrated in Fig. 1(d), which is the analog of Fig. 1(c).

In order to find the λdependence of our results, numerical calculations were carried out using the simulated annealing technique on chains of 12 and 24 CuO cells and for $x=0.33$. Periodic boundary conditions are used throughout in this study. We have considered the same parameters as above. As an order parameter we used the number of doubly occupied O sites. The results are shown in Fig. 2(a). We observed that the states I, II, and III are dominant in a large region of parameter space. Only in narrow regions near the phase boundaries, especially for large values of λ, other types of states become competitive. However, these states are simple variations of states I, II, and III (for example, including small local fluctuations). Note that, for the 12-cell chain, there is a particular value of $\lambda\sim 8$ where the PS regime becomes very narrow, and it may disappear when the hopping is switched on. This value of λ reduces to ~ 4 for the 24-cell chain. We conjecture therefore that the PS regime does not exist for $\lambda > 4$ or perhaps for even smaller values of λ, so it is not necessary to have a fully unscreened $1/r$ interaction to obtain the effects described here. This value of λ corresponds to two Cu-Cu lattice spacings, and thus it is not physically unrealistic. The main features in Fig. 2(a) are also present for the case of $x=0.66$. Similar calculations for the 4×4 and 6×6 CuO_2 clusters, for $x\approx 0.25$, give essentially the same phase diagram, as can be seen in Fig. 2(b).

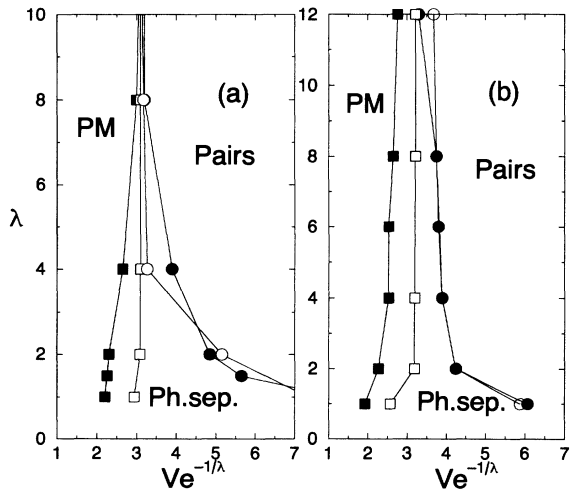

FIG. 2. (a) Phase diagram of the Hamiltonian Eqs. (1) and (2) in the atomic limit, obtained from 12- (filled symbols) and 24- (open symbols) cell chains, $U_d=7$, $U_p=1$, and $\Delta=1.5$, as a function of λ and V. The doping fraction is $x=0.33$. "PM" denotes a region dominated by the states like the one shown in Fig. 1(a); "Ph.Sep." denotes the region where state Fig. 1(b) dominates; and "Pairs" is the region with preformed pairs, as exemplified by Figs. 1(c) and 1(d). (b) Same as (a) for the 4×4 (filled symbols) and 6×6 (open symbols) cell clusters and $x=0.25$.

III. COMPETITION CDW-SUPERCONDUCTIVITY

The results summarized in Figs. 1 and 2 were obtained in the atomic limit. To study the effect of a finite kinetic energy ($t_{pd}\neq 0$) and to establish the assumed existence of a superconducting regime, Lanczos diagonalization techniques were used to obtain the ground state of the Hamiltonian Eqs. (1) and (2) explicitly on a finite chain. Due to the large number of states per unit cell the lattices that can be studied numerically are limited to a small number of cells. In this paper we studied six cells (12 atoms) with various hole numbers. Fortunately, our chains are large enough for the study of correlations using local pairing operators as has been repeatedly shown in the case of the attractive Hubbard model. For the results shown below we chose $U_d=7$, $U_p=1$, and $\Delta=1.5$ in units of t_{pd}. These are reasonably realistic values for the cuprates[20] and they have been used before by Sudbø and coworkers[10,11] in their analysis of the charge-transfer scenario. Then it is natural to work in the same region of parameter space. We have also observed that our results are qualitatively similar over a broad range of parameters.

First, let us numerically consider the issue of charge transfer and phase separation. A sharp indication of charge transfer from Cu to O sites is given by the susceptibility associated with the expected occupation of O sites,[9]

$$\chi_{CT}=\langle n_O^2\rangle-\langle n_O\rangle^2 , \tag{3}$$

where

$$n_O=\sum_{j=1}^{N} n_j$$

is the number of holes on the oxygens. The results for this susceptibility are shown in Fig. 3(a) for $n_h=2$ and $n_h=4$ on the six-cell chain and a long-range (LR) interaction. Although these chains are small, we expect that the information they provide is qualitatively correct, as has been shown in several examples.[8] In both cases a peak is observed at a particular value of $V\approx 4$. This peak signals the onset of a charge-transfer process in this model.

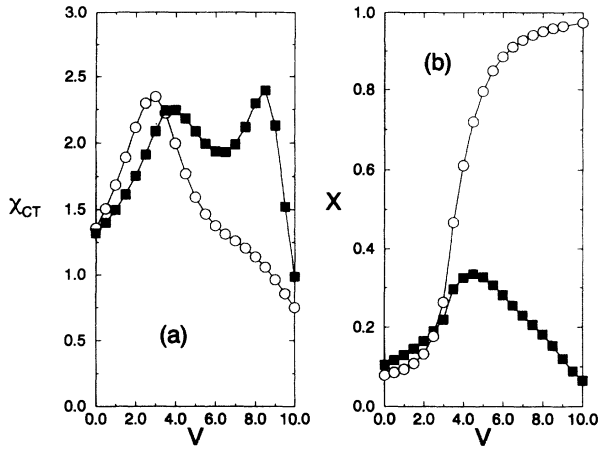

FIG. 3. (a) Susceptibility χ_{CT} [Eq. (3)] as a function of V for a six-cell chain, $t_{pd}=1$ and other parameters as in Fig. 2(a). The full squares denote $n_h=2$ and the circles $n_h=4$. (b) Order parameter X (see text) used to study phase separation versus V. The full squares denote $\lambda=\infty$, and the open circles are for the nearest-neighbor interaction. The number of holes is 8 ($n_h=2$).

However, χ_{CT} does not distinguish between the states II and III in Fig. 1. (Note that for $V>U_d$, a second peak develops corresponding to the transition to a state with doubly occupied Cu and O sites, but this is clearly an unphysical region of parameters.) Thus, to confirm that phase separation does not occur in the presence of long-range interactions, we have measured the long-wavelength component of the susceptibility associated with the correlations of pairs of holes on O sites:[21]

$$X=\frac{1}{N^2}\sum_{j=1}^{N}\sum_{i=1}^{N}\langle n_{j\uparrow}n_{j\downarrow}n_{i\uparrow}n_{i\downarrow}\rangle e^{(2\pi/N)(j-i)} . \quad (4)$$

It can be easily verified that this long-wavelength component is maximum for a fully phase-separated state, i.e., a state where all the doubly occupied oxygens form a single cluster [as that shown in Fig. 1(b) for $n_h=2$]. This quantity is normalized such that it is equal to zero for a uniform state and it is equal to one for a fully phase-separated state. In Fig. 3(b), X is shown for the case $n_h=2$ on the six-cell chain. With a short-range interaction, $X\rightarrow 1$ as V is increased. On the other hand, in the presence of long-range interactions, X goes through a maximum, and then decreases at large V, showing the absence of phase separation in this limit. The peak at intermediate V is due to the proximity of the PS state in the spectrum (as discussed previously in the atomic limit). At this point it is important to remark that the critical value of $V(V_c)$ at which the charge-transfer process occurs (namely $\sim 3t_{pd}$), since it is larger than U_p, implies that the effective U in O sites is attractive. Whether such a regime is realistic for the cuprates needs more experimental work.

Now let us investigate whether state III of Fig. 1(c) becomes superfluid when the holes acquire mobility in the presence of LR interactions. For this purpose, we consider the pairing correlation,

$$C(m)=\frac{1}{N}\sum_{j}\langle\Delta_{j+m}^{\dagger}\Delta_j\rangle , \quad (5)$$

where the pairing operator is defined as $\Delta_j=c_{j\uparrow}c_{j\downarrow}$, and $j, j+m$ indicates O sites. In Figs. 4(a) and 4(b), the pairing correlations are plotted as a function of distance for two different densities, for $t_{pd}=1$, $U_d=7$, $U_p=1$, and $\Delta=1.5$, and several values of V. For $n_k=2$ the correlations decay rather rapidly at large distances, indicating the absence of superconductivity. This example shows that it is not obvious that once phase separation is destroyed it will be replaced by superconductivity. As discussed earlier, there is a competition between superconductivity and CDW, and the absence of superconductivity at this density could be explained by a CDW order dominating over superconductivity. On the other hand, for $n_h=4$ ($x=0.67$), the pairing correlation is very *strong* even at the largest distances available on our finite clusters. The signal monotonically increases with V, for $V<U_d$. At this filling, we should be able to detect a suppression of CDW order.

To confirm this interplay between CDW and superconductivity, we have computed the order parameter associated with the CDW order expected at this density for the long-range model which is shown in Fig. 1(c). This CDW order parameter is defined by

$$\chi_{CDW}=\frac{1}{N^2}\sum_{j=1}^{N}\sum_{i=1}^{N}\langle n_{j\uparrow}n_{j\downarrow}n_{i\uparrow}n_{i\downarrow}\rangle e^{(2\pi/P)(j-i)} , \quad (6)$$

where P is the period of the CDW (in units of lattice spacings). The results for this order parameter, normalized between 0 and 1, are shown in Fig. 5 as a function of V. The parameter set considered is the same as in Figs. 4(a) and 4(b). For $n_h=2$, it can be seen that χ_{CDW} grows

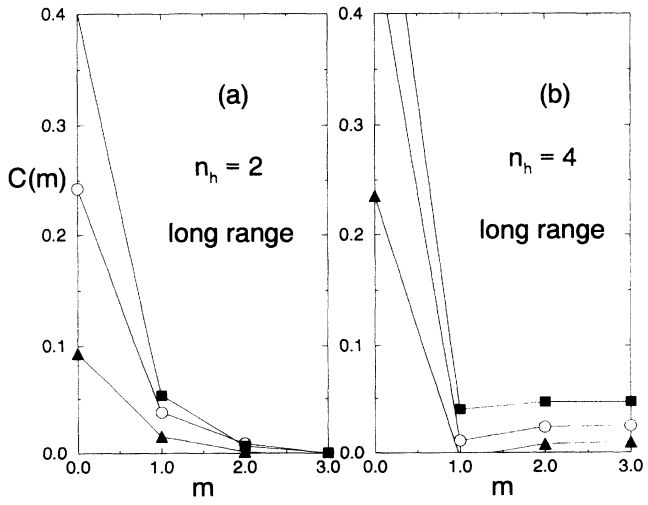

FIG. 4. (a) Superconducting correlations $C(m)$ (as defined in the text) for the long-range model at several values of V, on a six-cell chain and 8 holes ($n_h=2$). Note that m measures O-O distances. The full triangles denote results for $V=2$, the open squares $V=4$, and the full squares $V=6$. (b) As (a) but with 10 holes ($n_h=4$).

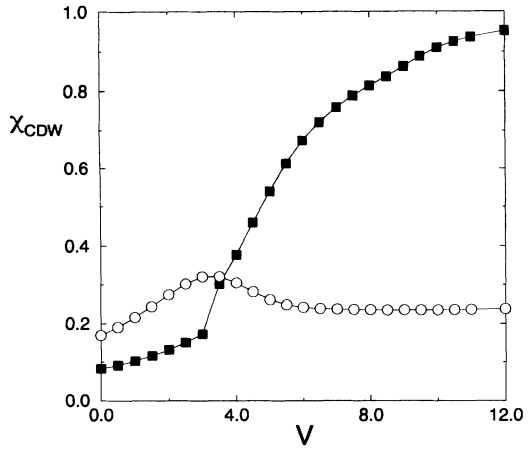

FIG. 5. CDW order parameter (see text) as a function of V for a six-cell chain for the long-range model. $t_{pd}=1$ and other parameters as in Fig. 2(a). The full squares denote $n_h=2$ and the circles $n_h=4$.

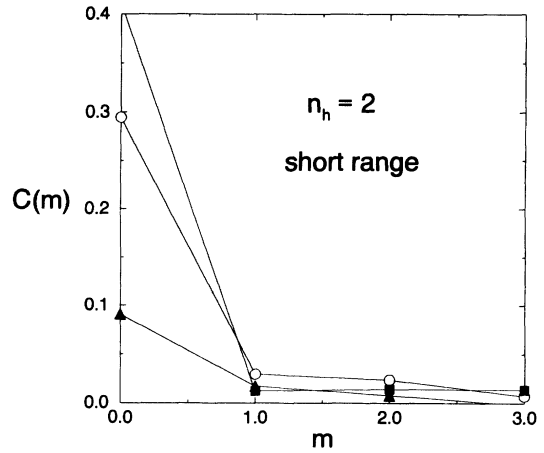

FIG. 7. Superconducting correlations $C(m)$ for the short-range model for $V=0.0$ (triangles), $V=2.667$ (squares), and $V=4.0$ (circles), on a six-cell chain and 8 holes ($n_h=2$). The parameters used are the same as in Ref. 11, i.e., $U_d=6.667$, $U_p=0$, $\Delta=1.333$ in units of t_{pd}.

monotonically with V, with a change of slope at the point where the charge transfer occurs, as shown in Fig. 3(a). On the other hand, for $n_h=4$ the CDW order parameter, after passing through a maximum at the CT instability, decreases to a constant for large V.

The results presented thus far support a scenario in which s-wave superconductivity appears with increasing V if long-range interactions are included, at intermediate densities. (Although the pairing operator used in this study is local, the existence of extended s-wave pairing is not excluded. Actually both operators transform similarly under the symmetry group of the Hamiltonian.) To give further support to these claims, we have studied the dependence of the ground-state energy on an external magnetic flux Φ. To analyze this response, a phase factor $e^{i\Phi/N}$ is introduced in the hopping term in Eq. (1).[2,11] This is equivalent to allowing a magnetic flux through the Cu-O ring (chain with periodic boundary conditions). In Fig. 6, the ground-state energy measured with respect to the energy at zero flux, $\Delta E=E(\Phi)-E(\Phi=0)$, is shown

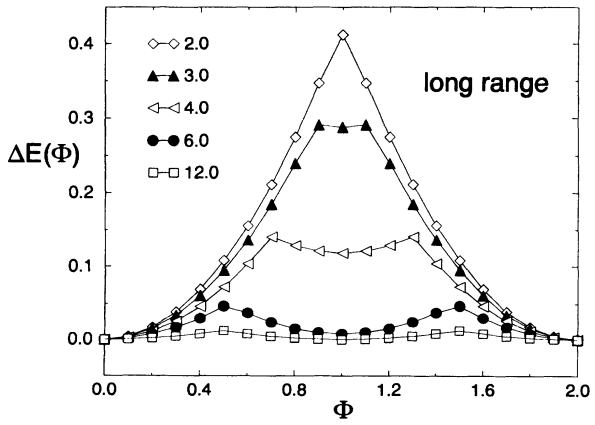

FIG. 6. The ground-state energy $\Delta E(\Phi)$ (measured with respect to the energy at zero flux) versus flux Φ for a six-cell ring for various V and a long-range interaction ($\lambda=\infty$).

as a function of Φ for $n_h=4$ and a pure LR interaction ($\lambda=\infty$). For small V the energy has a single minimum at $\Phi=0$ (mod2π). For V larger than a critical value the energy develops a second minimum at $\Phi=\pi$, indicating the existence of mobile carriers with charge $2e$ in the ground state. This anomalous flux quantization is in agreement with results obtained from the pair correlations. It is important to stress the fact that anomalous flux quantization *only* indicates the existence of pairs in the ground state, which is also compatible with CDW order. To illustrate this point, we shown in Fig. 7 the pairing correlations for the multiband Hubbard model [Eq. (1)] in the presence of only nearest-neighbor Coulomb repulsion, and for the same set of parameters considered by Sudbø *et al.*[11] In Ref. 11 was shown the presence of anomalous flux quantization by numerical calculations on the six CuO cells cluster. As it can be seen in Fig. 7, the pairing correlations for the same cluster have a very small tail compared, for example, with the ones shown in Fig. 4(b). There is an enhancement as V increases from 0.0 to 2.667 but for $V=4.0$ the pairing correlations are already suppressed. Thus, we arrive at the somewhat paradoxical conclusion that although K_ρ is larger than one, as reported in Ref. 11, the pairing correlations are suppressed. On the other hand, in our model superconducting correlations seem strong.

IV. CONCLUSIONS

In this paper, we have considered an extension of the standard Cu-O electronic Hubbard-like model for the cuprates which incorporates long-range interactions. We found that in the atomic limit a charge-transfer process exists without phase separation. The new dominant state in this regime has preformed pairs on the oxygen sites. With a hopping term that provides mobility to the holes, this state appears to become superfluid. However, there is a nontrivial competition with a CDW order. The effects observed here are intuitively evident, and are valid

both in one and two dimensions. These effects occur not only with a pure $1/r$ interaction but also for a finite-range Coulomb repulsion (λ approximately four Cu-O lattice spacings) which might correspond to the real cuprate compounds. The conclusions of this work go beyond the specific model used here, and show that it is possible to form superconducting states when $1/r$ interactions are used in a regime of phase separation. Finally, we would like to remark that the s-wave superconductivity found in this model is not incompatible with the d-wave results obtained in previous work.[2] For the parameters used in the present paper, the mapping from Eq. (1) to the t-J model is not possible. Moreover, the mechanism that led to pairing in the present model is quite different from the one in the t-J model.[2] Experiments will decide between these two possibilities.

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