

## Hole dynamics in the two-dimensional strong-coupling Hubbard Hamiltonian

Efthimios Kaxiras

*IBM Research Division, Thomas J. Watson Research Center, Yorktown Heights, New York 10598*

Efstratios Manousakis

*Supercomputer Computations Research Institute, Florida State University, Tallahassee, Florida 32306*

(Received 10 March 1988)

The effective Hamiltonian, obtained from the Hubbard model in the strong-coupling limit, is diagonalized exactly for a periodic two-dimensional square lattice of ten sites. We obtain the ground state of the system for any filling of the lattice and the  $k$ -space excitation spectrum for a single hole. Within our finite-size system, it is found that for very small coupling ratio  $t/U$ , each hole creates a local ferromagnetic environment at least of the size of our system, whereas for increasing  $t/U$ , the holes tend to form clusters, induced by antiferromagnetic spin correlations. A range of  $t/U$  exists for which pairing of the holes may be possible.

The recently discovered superconducting copper oxides,<sup>1</sup> which exhibit prominent two-dimensional (2D) features, provided new impetus<sup>2</sup> for the study of various approximations or limits of the Hubbard model. This model describes electrons in a single-band crystal and consists of a hopping matrix element ( $-t$ ) between neighboring lattice sites, and an on-site repulsion ( $U$ ) between electrons of opposite spin. In the simplest case, the ubiquitous Cu-O planes of the superconducting oxides are modeled by a 2D square-lattice Hubbard Hamiltonian, with each lattice site representing a CuO<sub>2</sub> unit cell. Theoretical investigations of this single-band model have not produced a thorough understanding of its ground-state properties, and whether or not it exhibits a coherent state that gives rise to superconductivity remains equivocal.

For  $t/U \ll 1$ , a convenient reference state is the half-filled lattice with one electron in each lattice site. There are two kinds of excitations from this state, holes introduced by doping and spin excitations. Both kinds of excitations and their interplay may be essential to the possible existence of a condensate. In the present work, we obtain the ground state of the single-band, strong-coupling Hubbard Hamiltonian by exact diagonalization of the Hamiltonian matrix for a ten-site periodic lattice. We shall study the ground-state energy of the system at all possible fillings of the lattice, the  $k$ -space excitation spectrum of single holes, and the dynamic interaction of pairs of holes. Our results indicate that for a range of values of the coupling ratio  $t/U$ , and for light doping the holes tend to form pairs. This pairing tendency is observed while the system exhibits antiferromagnetic correlations. As the doping increases or  $t/U$  increases, the holes tend to form clusters larger than the pairs. For very small  $t/U$  on the other hand, each hole creates a local ferromagnetic environment at least of the size of our finite system. It is difficult to predict how these conclusions might be affected in the limit of an infinite-size lattice.

We begin with a brief discussion of the effective Hamiltonian, which is obtained from the Hubbard model in the strong-coupling (large- $U$ ) limit by second-order perturbation theory,<sup>3,4</sup> and acts on the Hilbert space of states with

each lattice site occupied by at most one electron. The effective Hamiltonian, defined on a 2D square lattice with  $M$  sites and  $N$  electrons, can be conveniently separated in three terms  $H_{\text{eff}} = H_1 + H_2 + H_3$ , with distinct physical content. The first term  $H_1$  allows the holes to hop by one site; the second term  $H_2$  counts the number of nearest-neighbor (NN) pairs with opposite spins or exchanges their spins; and the third term  $H_3$  allows hopping of the holes by two sites, via NN pairs of opposite spin, with or without spin exchange. For a detailed discussion of the Hamiltonian see Ref. 5. At half-filling ( $N=M$ ),  $H_{\text{eff}}$  is equivalent to a Heisenberg antiferromagnet with coupling  $J=4t^2/U$ . Here we diagonalize  $H_{\text{eff}}$  for a square lattice with ten sites and periodic boundary conditions and obtain the ground-state energy and wave function for all possible fillings of the lattice. Details of the calculations have been discussed elsewhere.<sup>5</sup> In the following all energies will be given in units of  $t$ . This leaves the ratio  $t/U$  as the only parameter, which is proportional to the antiferromagnetic coupling.

The magnetic properties of this system were investigated in Ref. 5. Suffice to say that the system exhibits four phases as a function of the doping fraction  $x=X/M$  ( $X=M-N$ ) and the coupling ratio  $t/U$ . In two phases, which we shall call FC and AFC, the system exhibits short-range ferromagnetic and antiferromagnetic correlations, respectively. In a third phase, called AF, the spin-spin correlations are consistent with antiferromagnetic order extending to the largest possible distance of our finite lattice. Finally, we have verified that for small enough  $t/U$  and the smallest possible doping ( $x=0.1$ , one hole in our finite lattice), the system orders ferromagnetically ( $F$  phase), as predicted by Nagaoka's theorem.<sup>6</sup> In an infinite system, a single hole is expected to induce long-range ferromagnetic order only for vanishing small  $t/U$ . The occurrence of this phase at nonzero values of  $t/U$  is connected to finite-size effects (one hole is equivalent to 10% doping).

We consider first the ground-state energy of the system as a function of lattice filling at various values of  $t/U$ . It is very easy to extend the discussion to fillings of the lat-

tice higher than one electron per site by noticing that the system with  $X$  doubly occupied sites maps to that with  $X$  holes.<sup>6</sup> Thus, the total energy of the system with  $X$  doubly occupied sites and that with  $X$  holes differ exactly by  $XU$ . The ground-state energy  $E(n)$  as a function of the band-filling factor, defined as  $n=N/M$ , is shown in Fig. 1.  $E(n)$  is independent of  $t/U$  for  $t/U \leq 0.01$ , and has a global minimum at  $n=0.5$ , which is consistent with exact results for finite lattices in the  $U \rightarrow \infty$  limit of the Hubbard Hamiltonian.<sup>7</sup> At  $n=0.5$ , half of the sites are unoccupied and the other half are occupied by a single electron. Thus, on the average, each electron has a NN hole and full advantage can be taken of the hopping energy. Adding electrons to this system restricts their motion without any significant gain from coupling energy ( $t/U$  is very small); on the other hand, reducing the number of electrons leaves too few carriers in the system. In either case, the total energy is increased, making the quarter-filled band the global ground state for small  $t/U$ . For  $t/U \geq 0.25$ , a different shallow minimum appears at  $n=1$  which eventually becomes the global minimum. For  $t/U=0.40$   $E(n)$  looks remarkably similar to the "insulating case" described by Anderson,<sup>8</sup> though at these values of  $t/U$  it is not clear how well  $H_{\text{eff}}$  approximates the Hubbard model.

We discuss next the properties of the system at small dopings, namely  $x=0.1-0.2$ .<sup>9</sup> Let us define

$$h_i(X) = \langle \psi_0(X) | H_i | \psi_0(X) \rangle - \langle \psi_0(0) | H_i | \psi_0(0) \rangle, \quad i=1,2,3, \quad (1)$$

where  $|\psi_0(X)\rangle$  is the ground-state wave function of the system with  $X$  holes. The total energy, relative to the ground-state energy of the half-filled state, is given by

$$\epsilon(X) = h_1(X) + h_2(X) + h_3(X). \quad (2)$$

Notice that  $\langle \psi_0(0) | H_i | \psi_0(0) \rangle = E_0 \delta_{i2}$ , where  $E_0$  is the ground-state energy of the half-filled state. For the case  $x=0.1$ , our ten-site system contains one hole and the eigenfunctions can be characterized by a  $\mathbf{k}$  vector:

$$|\psi_{\mathbf{k}}(1)\rangle = \frac{1}{\sqrt{M}} \sum_i e^{i\mathbf{k}\cdot\mathbf{r}_i} \sum_{\alpha} c_{\alpha} |\alpha, \mathbf{r}_i\rangle, \quad (3)$$

where  $i$  runs over the lattice sites,  $\mathbf{r}_i$  is the position of the hole, and  $\alpha$  runs over all possible spin configurations.

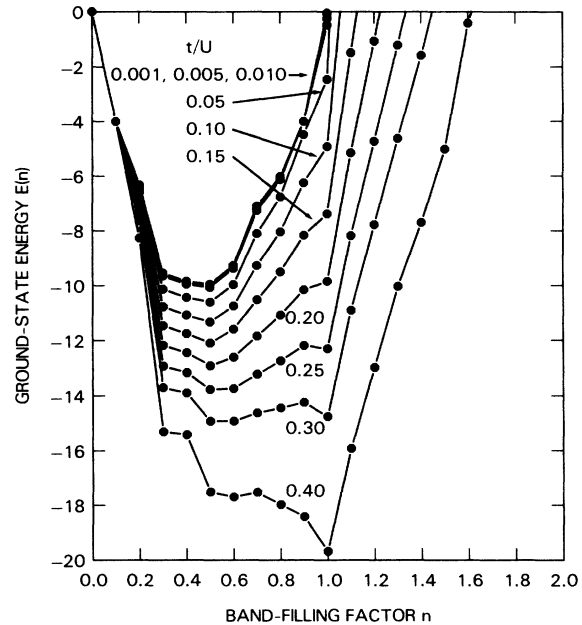


FIG. 1. The ground-state energy  $E(n)$  of the system vs filling of the band  $n=N/M$  for various values of  $t/U$ .

Since we are dealing with a finite system, the  $k$  space consists of ten independent vectors: four of them are generated from  $(3\pi/5, \pi/5)$  by  $\pi/2$  rotations, another four are generated from  $(2\pi/5, 4\pi/5)$  by  $\pi/2$  rotations, and the set is completed by the  $(0,0)$  and  $(\pi, \pi)$  vectors. The symmetry of the real-space lattice results in equal eigenvalues for vectors connected by  $\pi/2$  rotations. The energy of the one-hole system relative to the energy of the half-filled state, at various  $k$  vectors can be obtained from Eqs. (1) and (2) by replacing  $|\psi_0(X)\rangle$  for  $X \neq 0$  in Eq. (1) with the wave function of Eq. (3). The total energy given in Table I for representative values of  $t/U$  in each magnetic phase, shows very small dispersion in  $k$ . This corresponds to a large effective mass for the hole. In Table I we also show the contribution of the different  $h_i$  terms to the total energy for  $t/U=0.1$  where the dispersion is somewhat more pronounced. A comparison of the  $h_i$  values at the

TABLE I. The total energy at different  $k$  vectors in the various magnetic phases for the one-hole system. For  $t/U=0.1$  the contributions of the different terms  $h_1, h_2, h_3$  to the total energy are also given.

$t/U$	Phase	$(k_x, k_y)$				
		(0,0)	$(3\pi/5, \pi/5)$	$(4\pi/5, 2\pi/5)$	$(\pi, \pi)$	
0.032	F	-2.426	-2.417	-2.371	-2.400	
0.034	FC	-2.354	-2.331	-2.300	-2.348	
0.036	AFC	-2.289	-2.285	-2.250	-2.295	
0.040	AF	-2.160	-2.206	-2.181	-2.190	
0.1	AF	-0.715	-1.323	-1.272	-0.727	
	$h_1$	-2.927	-2.505	-2.623	-2.838	
	$h_2$	2.494	1.514	1.683	2.317	
	$h_3$	-0.282	-0.332	-0.331	-0.205	
0.2	AF	1.252	-0.312	-0.098	1.206	

different  $k$  vectors indicates that the dispersion comes predominantly from the  $H_2$  term which gives rise to the antiferromagnetic coupling of spins. The ground state is characterized by  $\mathbf{k}=(0,0)$  in the F and FC phases, by  $\mathbf{k}=(\pi,\pi)$  in the AFC phase and by  $\mathbf{k}=(3\pi/5,\pi/5)$  (or its  $\pi/2$  rotated partners) in the AF phase. This last vector of our finite  $k$ -space set is the one closest to the  $|k_x| + |k_y| = \pi$  boundary in the 2D Brillouin zone, where the Fermi level of the noninteracting ( $U=0$ ) Hubbard model is located at half-filling.

The expectation values  $h_1(X)/X$ ,  $h_2(X)/X$ ,  $h_3(X)/X$ , and the total energy  $\epsilon(X)/X$ , per hole, are shown in Fig. 2 as a function of  $t/U$  for the systems with one ( $x=0.1$ ) and two ( $x=0.2$ ) holes. There are pronounced breaks in the three expectation values when the system changes magnetic phase. The different magnetic phases are indicated on the  $h_1$  curves of the one- and two-hole systems. The total energy per hole is lower in the two-hole system<sup>10,11</sup> in all phases (FC, AFC, AF), except when the one-hole system develops perfect ferromagnetic order for  $t/U < 0.033$  (F phase). This fact can be interpreted as indication that in an infinite system, at sufficiently low  $t/U$ , each hole will induce a local ferromagnetic environment, at least equal to the size of our finite system, which facilitates its hopping. This conjecture does not exclude the possibility that two such ferromagnetic "polarons" are bound in a larger antiferromagnetic environment.<sup>12</sup> Going from ferromagnetic to antiferromagnetic correlations, the hopping energy  $h_1$  is reduced both in the one- and in the two-hole systems, reflecting the relative difficulty of holes to hop by a single site. For  $t/U > 0.065$  the two-hole system gains more energy per hole from the hopping term than the one-hole system [ $h_1(2)/2 < h_1(1)$ , see Fig. 2]. Thus, in the antiferromagnetic environment

the presence of two holes enhances their individual hopping energy, which may be attributed to a correlated motion of the holes.

The term  $h_2$  gives the potential energy of the hole relative to the half-filled state, which increases with the coupling ratio  $t/U$ , since the existence of holes breaks antiferromagnetic bonds. In the two-hole system, the potential energy is smaller than in the one-hole system (see Fig. 2), because the two holes may be found with a finite probability at NN sites thereby reducing the number of broken antiferromagnetic bonds relative to being at further neighbor distances. Finally, the term  $h_3$  is insignificant with respect to the other two terms for small  $t/U$ . This term begins to make a non-negligible contribution to the total energy only at the highest values of  $t/U$  considered, and its magnitude is almost identical in the one- and two-hole systems.

The total energy per hole provides insight to the behavior of holes at higher doping fractions ( $x > 0.2$ ) as well. Let us define the binding energy per hole as

$$b(X) = [\epsilon(X) - X\epsilon(1)]/X. \quad (4)$$

The quantity  $b(X)$  is shown in Fig. 3 for  $X=2, \dots, 5$  as a function of  $t/U$ . An interesting feature is the existence of a range of  $t/U$  for which  $b(2)$  is lower than any other  $b(X)$  (see Fig. 3). The two-hole distribution function for the various distances (multiplied by the number of neighbors at each distance in the unit cell, i.e., four neighbors at  $r=1$ , four at  $r=\sqrt{2}$  or 2, and one at  $r=\sqrt{5}$ , see Ref. 5) is shown as an inset at the lower left corner of Fig. 3. When the two-hole system develops antiferromagnetic correlations ( $t/U > 0.04$ , see Fig. 2), the probability of finding the holes at NN sites ( $r=1$ ) starts to increase rapidly at the expense of further neighbor ( $r=\sqrt{2}, 2, \sqrt{5}$ ) occupation

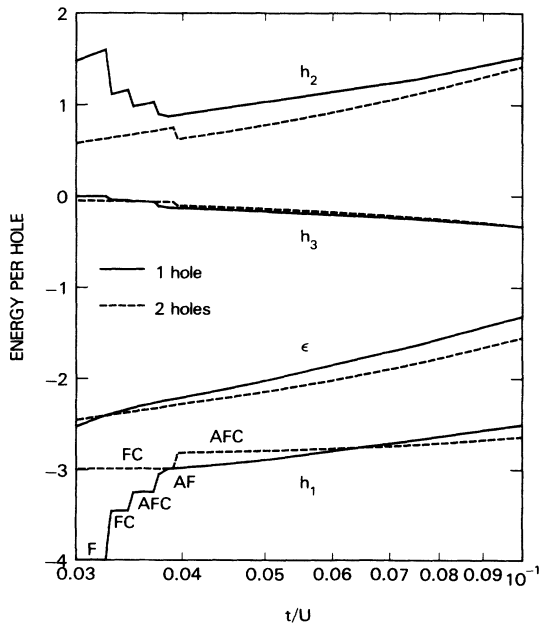


FIG. 2. The expectation values  $h_1$ ,  $h_2$ ,  $h_3$  and the total energy  $\epsilon$ , per hole, vs  $t/U$  for the one- and two-hole systems. The magnetic phases are indicated on the  $h_1$  curves.

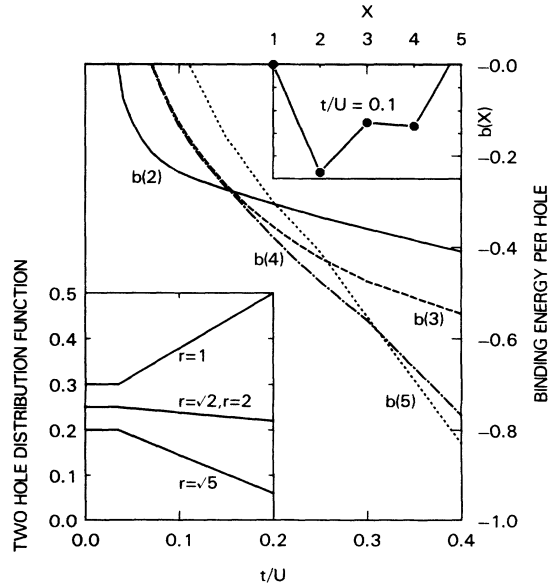


FIG. 3. Binding energy  $b(X)$  per hole vs  $t/U$  for various dopings  $X$ . The inset at the lower left corner is the two-hole distribution function for  $X=2$ . The inset at the upper right corner is the binding energy  $b(X)$  vs  $X$  for a particular  $t/U$ .

probabilities (Fig. 3, inset). Furthermore, it is striking that  $b(4) < b(3)$  for any value of  $t/U$  despite the higher hole concentration at  $X=4$ , indicating that in a system with even number of holes the binding energy per hole is lowered due to pairing. This feature of  $b(X)$  is shown more clearly in the inset at the upper right corner of Fig. 3 for a particular value of  $t/U$ . The shape of  $b(X)$  vs  $X$  remains the same throughout the region  $0.08 \leq t/U \leq 0.16$ . In this region, the system may be thought of as a collection of pairs with repulsive interpair interactions manifested by  $|b(4)| < |b(2)|$ . These results suggest that when pairing of all the holes is possible the energy of the system is minimized, and that pairing at short distances tends to be favored in the antiferromagnetic regions.<sup>13</sup>

For values of  $t/U$  larger than 0.16, Fig. 3 shows that  $b(2)$  is no longer the lowest binding-energy curve. As  $t/U$  increases, so does the value of  $X$  for which  $b(X)$  is lowest, which suggests that clusters of more than two holes are preferred. Gathering the holes into clusters creates the least possible disturbance in the antiferromagnetic background, by breaking the smallest number of bonds. Thus, for large  $t/U$  the strong antiferromagnetic correlations induce the formation of clusters. For somewhat lower values of  $t/U$ , due to competition from hopping energy, the antiferromagnetic background can only support the

formation of pairs. The pairs are the smallest mobile units that possibly condense to the zero-momentum state.

The present study provides evidence that, within the finite-size system considered here, in the region of antiferromagnetic correlations the holes show a tendency to form pairs. It is intriguing that the range of  $t/U$  at which pairs tend to form contains the values that this quantity is believed to assume in the superconducting copper oxides (0.05–0.10). Although off-diagonal long-range order cannot be studied in finite lattices like the one considered here, the tendency for pair formation is consistent with an  $h/2e$  flux quantum observed in flux quantization experiments.<sup>14</sup> In our finite system, we find pairing of the holes to be favored in the antiferromagnetic environment, but cannot exclude the possibility of ferromagnetic polarons (whose minimal size may exceed that of our system) being bound in an enlarged antiferromagnetic region.

We wish to acknowledge useful discussions with D.-H. Lee, V. R. Pandharipande, D. Pines, and N. Read, and thank T. D. Schultz for a critical reading of the manuscript. This work was supported in part by the Florida State University Supercomputer Computations Research Institute which is partially funded by the U.S. Department of Energy, Contract No. DE-FC05-85ER250000.

<sup>1</sup>J. G. Bednorz and K. A. Müller, *Z. Phys. B* **64**, 188 (1986); C. W. Chu *et al.*, *Phys. Rev. Lett.* **58**, 405 (1987).

<sup>2</sup>P. W. Anderson, G. Baskaran, Z. Zou, and T. Hsu, *Phys. Rev. Lett.* **58**, 2790 (1987); J. E. Hirsch, *Phys. Rev. B* **35**, 8726 (1987); A. E. Ruckenstein, P. J. Hirschfeld, and J. Appel, *ibid.* **36**, 857 (1987).

<sup>3</sup>J. E. Hirsch, *Phys. Rev. Lett.* **54**, 1317 (1985).

<sup>4</sup>K. Huang and E. Manousakis, *Phys. Rev. B* **36**, 8302 (1987).

<sup>5</sup>E. Kaxiras and E. Manousakis, *Phys. Rev. B* **37**, 656 (1988).

<sup>6</sup>Y. Nagaoka, *Phys. Rev.* **147**, 392 (1966).

<sup>7</sup>M. Takahashi, *J. Phys. Soc. Jpn.* **51**, 3475 (1982).

<sup>8</sup>P. W. Anderson, *Science* **235**, 1196 (1987).

<sup>9</sup>This is the region in which the copper oxides exhibit superconductivity with the highest  $T_c$ , see e.g., J. M. Tarascon, L. H. Greene, W. R. McKinnon, G. W. Hull, and T. H. Geballe, *Science* **235**, 1373 (1987).

<sup>10</sup>This stands in contrast to the results of a recent calculation

(Ref. 11), based on a very small portion of the Hilbert space. In fact, for the same  $t/U$ , the two holes are energetically more "bound" in the ten-site system than in a smaller eight-site system. Thus, we suggest that the restricted Hilbert space of Ref. 11 may be the source of discrepancy.

<sup>11</sup>S. A. Trugman, *Phys. Rev. B* **37**, 1597 (1988).

<sup>12</sup>V. L. Pokrovskii, G. V. Uimin, and D. V. Khvashchenko, *Pis'ma Zh. Eksp. Teor. Fiz. Suppl.* (to be published).

<sup>13</sup>A pairing model in antiferromagnetic environment was recently proposed by J. R. Schrieffer, X.-G. Wen, and S.-C. Zhang, *Phys. Rev. Lett.* **60**, 944 (1988).

<sup>14</sup>R. H. Koch, C. P. Umbach, C. J. Clark, P. Chaudhari, and R. B. Laibowitz, *Appl. Phys. Lett.* **51**, 200 (1987); P. L. Gammel, D. J. Bishop, G. J. Dolan, J. R. Kwo, C. A. Murray, L. F. Schneemeyer, and J. V. Waszczak, *Phys. Rev. Lett.* **59**, 2592 (1987).