Pairing Interaction in Two-Dimensional CuO_2

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We show that planar CuO_2 clusters with strong repulsive Coulomb interactions can exhibit an effective attractive pairing interaction. This interaction arises from a disruption of the local lattice charge distribution produced when a hole is added and occurs for a wide range of parameter values, in the region where both the on-site Cu repulsion and the nearest-neighbor Cu-O repulsion are appreciable. A signature of this mechanism is that the occupation of Cu sites *decreases* as the system is doped. We suggest that this effective attractive interaction is responsible for high-T_c superconductivity in oxides.

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To make progress in the theoretical understanding of high-T_c superconductivity in oxides it is imperative to identify unambiguously the dominant mechanism giving rise to pairing. At present, calculations[1] of the electron-phonon interaction in these materials suggest that phonons alone cannot explain high-T_c superconductivity. This is also supported by isotope-effect measurements[2,3]: As T_c becomes higher, the isotope effect becomes smaller, suggesting that another pairing mechanism, not involving the ions, becomes dominant as T_c increases. This leads one naturally to consider alternative pairing mechanisms involving interactions between electrons only.

Experimental observations of antiferromagnetism and antiferromagnetic spin fluctuations in these systems[4] suggest the existence of a large on-site repulsion energy on the Cu d orbitals, U. Various mechanisms for superconductivity involving U in an essential way have been proposed: singlet pair condensation,[5,6] spin-fluctuation–induced superconductivity,[7,8] resonating valence bonds,[9] strings,[10] and spin bags,[11] among others. However, recent numerical calculations[12,13] suggest that a Hubbard U by itself can give rise to antiferromagnetism but not to superconductivity, at least in the simple two-dimensional models and for the interesting temperature regime.

Varma, Schmitt-Rink, and Abrahams[14] have first suggested that a nearest-neighbor repulsion V between Cu and O plays an important role in high-T_c superconductivity. Their model stresses the importance of Cu-O charge fluctuations, but does not involve U in an essential way. A strong-coupling argument suggesting pairing in the presence of large U and V, and calculations on one-dimensional clusters, were discussed in Ref. 13. Here we study the role of U and V in an extended Hubbard model for two-dimensional CuO_2 clusters. We find that in the presence of a strong on-site Cu U, a near-neighbor Cu-O repulsion V can give rise to an effective attractive interaction between holes on the O sites of the cluster. The addition of a hole produces a local reorganization of the ion lattice charge distribution which requires a significant energy. The tendency for pairing results from the fact that it costs less energy to add a second hole in the vicinity of the first, rather than to create the separate local charge reorganization required for two isolated holes.

Figure 1 shows two CuO_2 clusters that we will study. If we take the vacuum state to be Cu^{+} $3d^{10}$ and O^{-2} $2p^{6}$, the nominally undoped state of a cluster can be considered as having one hole per Cu. With use of Cu

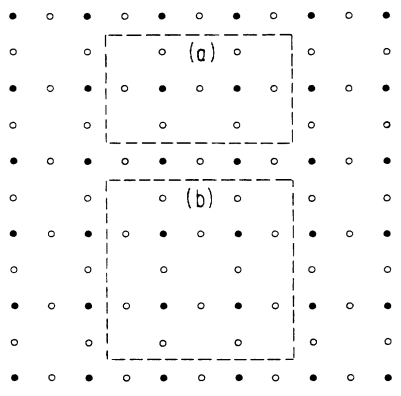

FIG. 1. Two-dimensional CuO_2 lattice. The O atoms are denoted by open circles, the Cu atoms by full circles. The regions enclosed by dashed lines show the (a) nine-site and (b) sixteen-site clusters studied by exact diagonalization, with the occupation of the Cu atoms outside the clusters next to the boundary taken into account in a mean-field way. The twelve-site unit referred to in the text is (b) with the top (right) O atoms being the same as the bottom (left) O atoms.

$d_{x^2-y^2}$- and O p_x, p_y-like orbitals, an extended Hubbard model describing the dynamics of holes for such a single orbital per site lattice has the form

$$H=\sum \epsilon_{ij} C_{i\sigma}^{\dagger} C_{j\sigma} + \tfrac{1}{2} \sum U_{ij} C_{i\sigma}^{\dagger} C_{i\sigma} C_{j\sigma'}^{\dagger} C_{j\sigma'} \quad (1)$$

with $C_{i\sigma}^{\dagger}$ the hole creation operator for Cu or O depending on the site index i. Here ϵ_{ij} includes on-site energies ϵ_d for Cu and ϵ_p for O, and a Cu-O hopping t, while U_{ij} describes the on-site Coulomb energies U and U_p of Cu and O, respectively, and the intersite Cu-O Coulomb interaction V. The point of view we take can be illustrated by consideration of the cluster shown in Fig. 1(b). Suppose the parameters are such that the first four holes go largely onto the Cu sites where they are antiferromagnetically coupled through a superexchange interaction. Call the ground-state energy of this state $E_0(4)$. Adding one and then two additional holes, we compute the ground-state energies $E_0(5)$ and $E_0(6)$, respectively. Then a "binding energy" Δ is defined by

$$\Delta = E_0(6) + E_0(4) - 2E_0(5). \quad (2)$$

We will use Δ as a measure of the pairing interaction.[15] Clearly, the value of Δ depends on the size of the cluster. Figure 2(a) shows the binding energy versus $|U|$ for different sized 1D lattices for a quarter-filled negative-U Hubbard model. For a single site ($N=1$), $\Delta = |U|$, while for $N \to \infty$, Δ is the binding energy of a pair. The results shown in Fig. 2(a), obtained by exact diagonalization, show a smooth transition as N increases. For a 2D lattice, with two particles interacting through an attractive on-site interaction $-|U| n_{i\uparrow} n_{i\downarrow}$, the binding energy is given by

$$|U|^{-1} = N^{-1} \sum_n [2(\epsilon_k + 4t) + |\Delta|]^{-1} \quad (3)$$

with $\epsilon_k = 2t(\cos k_x + \cos k_y)$. Taking a bandwidth $8t$ of order an electronvolt and a binding energy for the $N \to \infty$ lattice appropriate to the oxide superconductors gives $|U|/t = 5$. Figure 2(b) shows the binding energy $|\Delta|$ versus the linear lattice size N. Just as for the 1D case, the binding energy evolves smoothly towards the infinite-lattice limit. As Δ is the zero-frequency limit of the effective interaction, $\Delta > 0$ in a cluster could still give rise to superconductivity in the infinite system through retardation effects. Indeed, as our cluster size increases we find that the conditions on the parameters to obtain $\Delta < 0$ become less stringent.

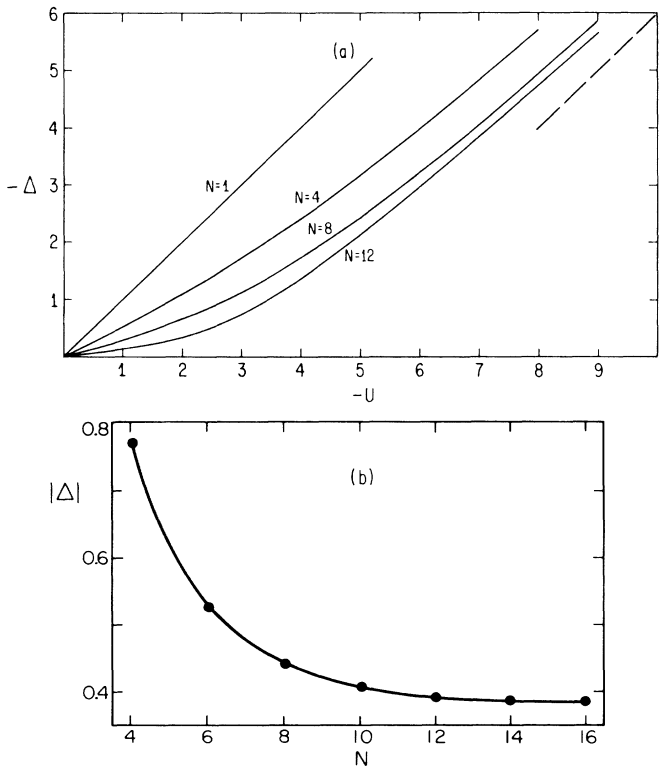

FIG. 2. (a) Binding energy for a pair of electrons added onto a quarter-filled N-site one-dimensional attractive Hubbard model. The dashed line denotes the large-U limit for $N > 1$. (b) Binding energy for a pair of electrons on a two-dimensional lattice interacting through an attractive one-site interaction $U/t = -5$.

We now consider the nine-site cluster shown in Fig. 1(a). We diagonalize the Hamiltonian Eq. (1) exactly for this unit for $n = 2$, 3, and 4 holes and obtain the ground-state energy $E(n)$ in each sector. The binding energy Δ for this unit is then given by $\Delta = E(2) + E(4) - 2E(3)$. We take free-end boundary conditions, but in order to simulate the outside environment we take for ϵ_p on the O atoms next to the boundary $\epsilon_{p\text{bound}} = \epsilon_p + V\langle n_{cu}\rangle$. Here $\langle n_{cu}\rangle$ is found from a separate calculation for a twelve-site lattice with four holes and periodic boundary conditions [Fig. 1(b)]. We believe that this value of $\langle n_{cu}\rangle$ should be very close to that of the infinite system near the half-filled-band case. This is based on the fact that numerical simulations on twelve- and 48-site lattices with U only show very little change of $\langle n_{cu}\rangle$ with lattice size.[13]

Results for Δ obtained from the nine-site cluster of Fig. 1(a) are shown for a range of parameters in Figs. 3(a)–3(d), versus the on-site Cu interaction U for several values of the on-site O interaction U_p and four different Cu-O interactions $V = 0$, 2, 4, and 6. For $V \gtrsim 4$, Δ becomes negative for large U. Note that in this region, Δ is relatively insensitive to U_p. Examination of ground-state expectation values shows that for small U_p pairing occurs on the center O site in Fig. 1(a): The expectation value $\langle n_\uparrow n_\downarrow\rangle - \langle n_\uparrow\rangle\langle n_\downarrow\rangle$ is positive for four holes in the cluster when $\Delta < 0$. This suggests that the resulting superconducting state will be s-wave like. As U_p increases, double occupation of O sites decreases and pairing occurs increasingly between holes on neighboring O sites. We have studied the dependence of Δ on the relative difference between O and Cu single-particle energies $\epsilon = \epsilon_p - \epsilon_d$ and found that the largest binding occurs for $\epsilon \cong 0$ for a wide range of parameters. This zero value

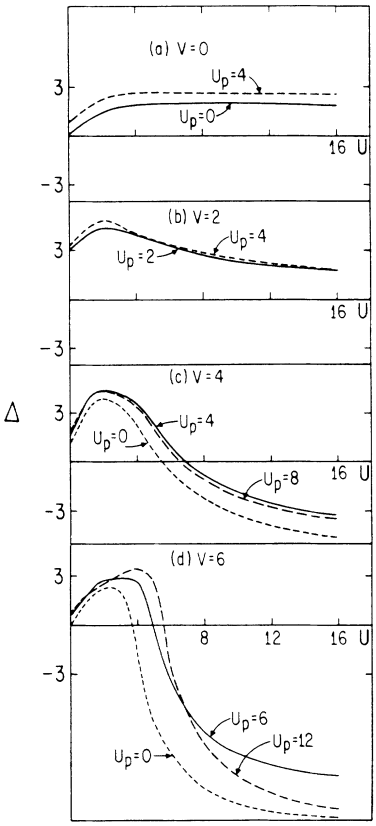

FIG. 3. Binding energy for a pair of holes added on the nine-site CuO_2 cluster [Fig. 1(a)] with two holes. $t=1$, $\epsilon=0$. (a) $V=0$, (b) $V=2$, (c) $V=4$, (d) $V=6$.

was used for the results shown in Fig. 3. We have also examined the effect of a direct O-O hopping and found that it enhances pairing up to a value around $t/3$ and then suppresses it.

Similar calculations of Δ were carried out for the sixteen-site cluster shown in Fig. 1(b) with use of a Lanczos method. Here too the single-particle energies of the outer oxygens are given by $\epsilon_{p\text{bound}}$. Figure 4 shows results for Δ vs U with $\epsilon_p-\epsilon_d=0$ and $U_p=8$, $V=4$. For this larger cluster, Δ is reduced in magnitude as expected, but shows a similar behavior to that found on the nine-site cluster. Once again, we see the important role of U. Variations with the other parameters were similar to those shown in Fig. 3 for the nine-site cluster, except that in all cases the condition for negative Δ was found to be less stringent for the larger cluster.

A common feature found in all the cases where Δ was negative was that the occupation of Cu sites was reduced rather than increased as the cluster was doped. For example, for $U=10$, $U_p=8$, $\epsilon=0$, $V=4$, the average Cu occupation in the nine-site cluster was 0.69, 0.50, and 0.31 hole for 2, 3, and 4 holes in the cluster. This feature, which only occurs in the presence of large U and V, appears to be the key necessary element to obtain a

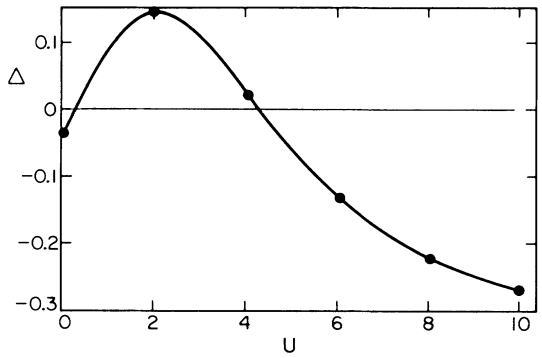

FIG. 4. Binding energy for a pair of holes added on the sixteen-site CuO_2 cluster [Fig. 1(b)] with four holes. $t=1$, $\epsilon=0$, $V=4$, $U_p=8$.

negative Δ.

Calculations for a periodic twelve-site CuO_2 cluster in the undoped state (one hole per Cu) show antiferromagnetic correlations, persisting in the region where additional added O holes bind. Figure 5(a) shows the binding energy, and in Fig. 5(b) we plot the nearest-neighbor and next-nearest-neighbor equal-time spin-spin correlations versus V for $U=10$, $U_p=8$, and $\epsilon_p-\epsilon_d=0$. Note that antiferromagnetic correlations in the undoped (half-filled) case persist unchanged for the parameter range in which the doped system exhibits a negative value for Δ.

Our calculations suggest that the model discussed here, which shares many of the features of high-T_c ox-

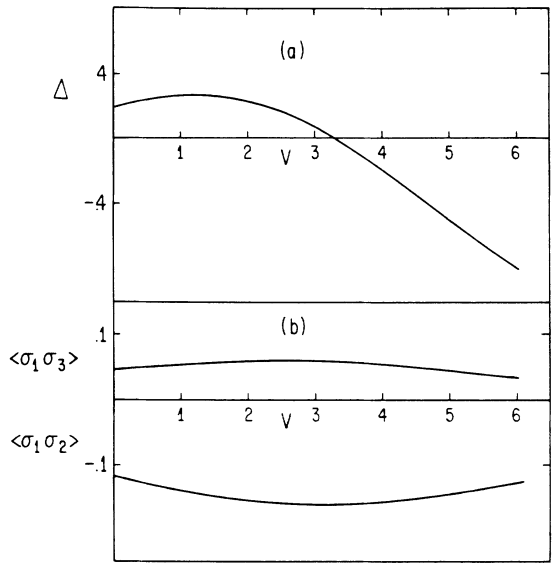

FIG. 5. (a) Binding energy for a pair of holes on the nine-site cluster vs V, for $U=10$, $U_p=8$, $\epsilon=0$. (b) Nearest-$\langle\sigma_1\sigma_2\rangle$ and next-nearest-neighbor $\langle\sigma_1\sigma_3\rangle$ spin-spin correlations for a twelve-site cluster with periodic boundary conditions; same parameters as (a).

ides, is perhaps the simplest two-dimensional model with short-range repulsive interactions that will exhibit high-temperature superconductivity.[16,17] Our cluster calculations showed that pairs of holes moving on the O sites in the presence of strong on-site and intersite Coulomb interactions bind. In the same parameter regime, the half-filled system exhibits strong antiferromagnetic correlations, suggesting that antiferromagnetism at half-filling and superconductivity away from half-filling can be manifestations of the same underlying interactions. We have also calculated the energy to add three holes to the undoped (1 hole/Cu) clusters and find that the pair and a separate hole state are energetically favored. As the system is doped away from half-filling we expect the antiferromagnetic correlations to be rapidly suppressed; in our cluster calculations, the staggered magnetization dropped rapidly as holes were added, and this was also found in Monte Carlo simulations with U only[13]; at the same time, we expect T_c to increase as the number of carriers (O holes) increases, as the pairing mechanism discussed here is local, not dependent on extended antiferromagnetic correlations. Furthermore, as discussed above, the proportion of Cu^+ relative to Cu^{++} should increase rather than decrease as the system is doped with holes if the parameters are such that Δ is negative; observation of this effect would provide strong support for the mechanism discussed here.

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Note added—After submission of this paper we received a preprint of C. Balseiro *et al.*[18] who independently discuss a very similar mechanism.

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[15]For low concentrations of holes, a pairing interaction ($\Delta < 0$) will lead to superconductivity. However, at denser fillings, repulsive interactions between pairs can lead to CDW correlations.

[16]V. J. Emery, contribution to the Workshop on Mechanisms of High Temperature Superconductors, University of Minnesota, October 1987 (unpublished), has discussed another pairing mechanism for a similar model.

[17]H. B. Schüttler (to be published) has recently discussed the formation of Cu^+O^- strings for a similar model.

[18]C. Balseiro, A. G. Rojo, E. R. Gagliano, and B. Alascio, to be published.