Antiferromagnetism, Localization, and Pairing in a Two-Dimensional Model for CuO_2

J. E. Hirsch
Department of Physics, University of California, San Diego, La Jolla, California 92093
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The properties of a model to describe the CuO_2 planes in the high-T_c superconductors are discussed in the strong-coupling limit. On the assumption that doping creates holes on the O rather than the Cu sites, following the recent suggestion of Emery, it is shown that antiferromagnetic ordering of the Cu moments will lead to either localization or pairing of the O holes. The effect of magnetic anisotropy on the pairing interaction is discussed, and it is suggested that it will strongly enhance superconductivity in these systems.

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There has been growing suspicion among theorists recently that a possible alternative mechanism for superconductivity may involve in some way antiferromagnetism rather than phonons.[1-16] Monte Carlo simulations and a strong-coupling expansion for the Hubbard model pointed at that possibility,[1] and summation of a selected class of diagrams within weak-coupling perturbation theory was shown to yield pairing in anisotropic singlet states near an antiferromagnetic (AF) instability.[2,3] More recently, Anderson's "resonating valence bond" model also proposed a relation between antiferromagnetic pairs and superconductivity.[4-8] Elucidation of this question may be the key to an understanding of the oxide superconductors (with or without phonons also playing a role[9]) as well as possibly the heavy-fermion superconductors and the Bechgaard salts.[10]

In this paper we discuss some properties of a model recently proposed by Emery[15] to describe a CuO_2 plane as occurs in the oxide superconductors. Following Emery, we assume the parameters are such that there is one hole per site in the d band of the Cu atoms (Cu^{++}), and that doping creates holes on the O^{--} sites rather than the Cu sites. The Hamiltonian is given by

$$H=\sum_{(i,l)} t_1(d_{i\sigma}^{\dagger}c_{l\sigma}+\text{H.c.})+(\epsilon-\mu)\sum_{l} c_{l\sigma}^{\dagger}c_{l\sigma} +U\sum_{i} n_{i\uparrow}n_{i\downarrow}-\mu\sum_{i\sigma} d_{i\sigma}^{\dagger}d_{i\sigma}, \quad (1)$$

where $d_{i\sigma}^{\dagger}$ creates holes on Cu sites and $c_{l\sigma}^{\dagger}$ creates holes on O sites; (i,l) run over nearest neighbors in the structure appropriate to Cu-O planes shown in Fig. 1. We neglect any Coulomb repulsion between holes on the O sites. If the Cu on-site repulsion U is much larger than the O level position ϵ, doping will create holes (or electrons) on the O^{--} atoms.

The effective hopping between holes on Cu atoms is $t=t_1^2/\epsilon$; the usual superexchange mechanism yields a Heisenberg antiferromagnetic exchange interaction $J=t^2/U$ between spins on Cu atoms. We will assume that because of spin-orbit coupling the magnetic interaction between Cu spins can be anisotropic, and consider an in-plane coupling J_{xy} and a perpendicular coupling J_z. The situation becomes particularly simple in the limit $J_z/J_{xy}\gg 1$, and we will discuss that limit first.

Consider a perfect antiferromagnetic arrangement of the Cu moments, as in Fig. 1, and a few O holes. Without flipping Cu spins, a given O hole can only move within a "cage" of four O atoms surrounding the nearest Cu atom of opposite spin. The effective hopping for the O hole is $t_{OO}=-t_1^2/(U-\epsilon)$, and it can hop to its nearest or next-nearest neighbor on this four-site lattice. Its ground-state wave function is

$$|\psi\rangle=\tfrac{1}{2}(|1\rangle+|2\rangle+|3\rangle+|4\rangle)$$

with energy $E=-4t_{OO}$ and it is separated by a large gap ($4t_{OO}$) from the excited states in this cell.

In order for the O hole to propagate beyond its cage, it has to exchange its spin with its corresponding Cu spin. If we assume for the moment that this is the only way in which a Cu spin flip occurs, a propagating O hole will leave behind a "string" of broken Cu-Cu antiferromagnetic bonds (Fig. 2). The cost in energy in so doing is $4J_z\times[1+(\text{distance traveled by O hole})$ (in units of Cu-Cu distance)]. The lowering in energy obtained by propagation of the O hole is only weakly dependent on the

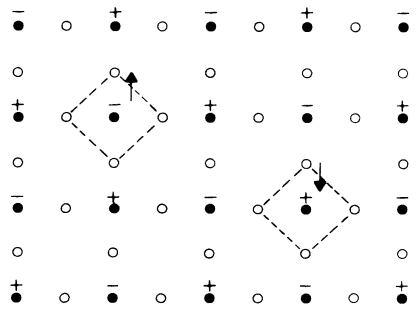

FIG. 1. Two-dimensional Cu (filled circles)-O (open circles) lattice. +,− denote orientation of Cu spins, and the O holes are represented ↑,↓. The dashed lines indicate the cages where the O holes move without flipping Cu spins. Hopping in these cages occurs between nearest neighbors and next-nearest neighbors with equal amplitude.

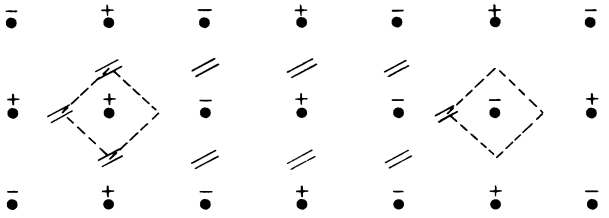

FIG. 2. Cu spin configuration after a single hole has propagated from its initial cage (to the left) to its final cage (to the right). The broken Cu-Cu antiferromagnetic bonds are indicated by the double parallel lines.

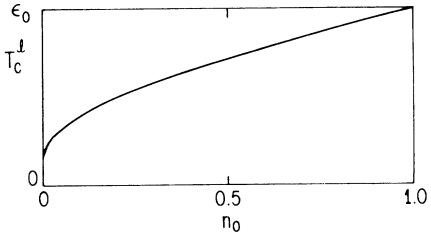

FIG. 3. Dependence of localization temperature on density of O holes.

distance traveled (the maximum kinetic energy for an O hole in this lattice is $E_{\rm kin}=-7t_{OO}$), so that it is clear that a single O hole cannot propagate very far in this manner. However, a second O hole can follow the first and flip all Cu spins back to their original orientation. This then provides a strong attractive interaction between two O holes. In the limit $J_{xy}\rightarrow 0$, it yields a confining potential between two O holes that increases linearly with their separation. Note that this would not occur in a one-dimensional Cu-O chain, as the number of broken bonds would be independent of the distance between the holes.

At high temperatures (but lower than the Cu-Cu exchange so that there is AF order in the Cu lattice) the O holes will be localized in their cages, since the entropy gain in putting an up and a down hole in arbitrary cages in the lattice outweighs the energy gain in having them close by where they can propagate as described above. As the temperature is lowered, one will have a delocalization transition below which pairs of up and down holes will propagate closely coupled. At still lower temperatures, the pairs will condense into a superconducting state. One may also have parameter regimes where the superconducting and delocalization transition occur together. The behavior of the resistivity versus temperature in the oxide superconductors, where in some cases ρ increases as T is lowered and drops suddenly to zero at the superconducting transition, while in other cases ρ decreases smoothly as one approaches T_c, suggests that both situations described above occur in these systems.

We can obtain a rough estimate of the dependence of the delocalization transition temperature on the concentration of O holes, n_O. The difference in entropy between having the holes unpaired and paired is roughly $\Delta S\sim k\ln V_O$, where $V_O=1/n_O$ is the available volume per hole. The difference is energy between the localized holes and the propagating paired holes is roughly $\Delta E\sim\epsilon_O(1-n_O)$, where ϵ_O is the lowering in energy obtained by propagation of the hole pair, of order t_{OO}^2/J as discussed below. We obtain then, as an estimate of the delocalization temperature,

$$T_c^l\cong\epsilon_O(1-n_O)/\ln(1/n_O), \tag{2}$$

which is schematically plotted in Fig. 3. As the density of holes increases the delocalized region increases; the dominant effect is that the gain in entropy in localizing the holes decreases as the volume per hole decreases. We have neglected here the fact that an increasing concentration of O holes will weaken the AF order, making delocalization even more likely as n_O increases.

Let us now consider the propagation of a pair of O holes. In the large-J_z limit, as the first hole flips the Cu spin and hops to the next plaquette, the second hole immediately follows, flipping the Cu spin back, as depicted in Fig. 4. The intermediate state has sixteen sites available for both holes (now of parallel spin), and its energy is $6J_z-1.09t_{OO}$ (if we take only the lowest two single-particle states of the sixteen-site system into account). Eliminating the intermediate state $|\alpha\rangle$ we obtain an effective hopping for closely coupled O holes:

$$t_p=-0.16t_{OO}^2/(6J_z-1.09t_{OO}). \tag{3}$$

We can think of the pair of holes as occupying the O site that is common to both O-hole nearest-neighbor cages. An effective Hamiltonian for the propagation of the pair of holes in the strong-coupling limit is then

$$H_{\rm eff}=\sum_{(l,l')} t_p(b_l^\dagger b_{l'}+\text{H.c.}), \tag{4}$$

where l,l' run over the square lattice of O sites. This square lattice should be imagined as a checkerboard, where on the black squares there is a diagonal hopping in addition to the nearest-neighbor hopping. The operators $b_l^\dagger, b_l$ create (destroy) hard-core bosons. Strictly speaking, two pairs of holes can occupy the same neighboring plaquettes, but the increase in energy in occupying the higher states in the plaquette is so high that this situation can be neglected. There is similarly a repulsion between pairs on neighboring sites, which effectively creates an excluded volume around each pair in this strong-coupling limit.

At zero temperature in the strictly two-dimensional system, the hole pairs will condense into a superconducting state (Bose condensation); the Hamiltonian of Eq. (4) is equivalent to a two-dimensional quantum XY model, which is known to have long-range order on the plane

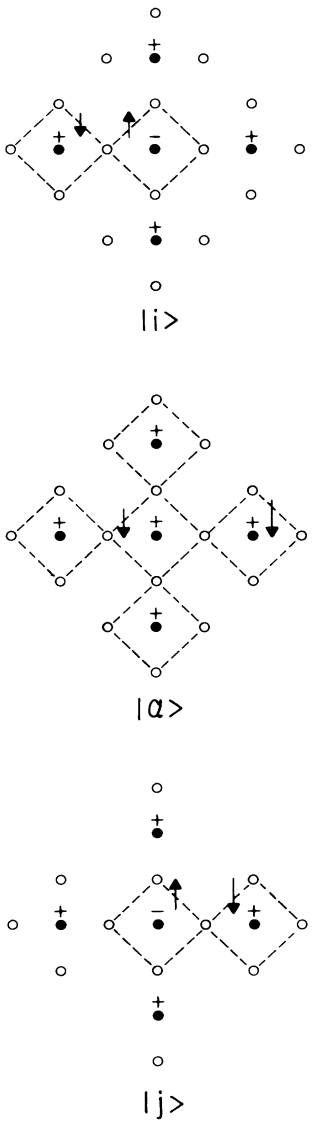

FIG. 4. Propagation of a strongly coupled hole pair. $|i\rangle$=initial state, $|\alpha\rangle$=intermediate state, $|j\rangle$=final state. The dashed lines indicate the regions available to the holes.

at $T=0$.[17] At finite temperatures, there will be a transition to a state of quasi long-range order at a temperature T_c of order $t_p n_O$, which will become true long-range order in the presence of some three-dimensional coupling.

What happens now when we relax the extreme conditions assumed in the preceding analysis? As J_z is decreased, T_c will first increase as t_p [Eq. (3)] increases but eventually it will decrease again as the binding energy of the holes becomes small.[18] When J_z is not much larger than T_{OO}, the pairs will not be tightly bound to nearest-neighbor plaquettes but will have a certain coherence length ξ_p. One can obtain an estimate for ξ_p by balancing the gain in kinetic energy in making the pair more extended with the cost in potential energy, of the form $\xi_p \sim \text{const}(t_{OO}/J_z)^{1/2}$. We do not expect the Bose condensation to be qualitatively different in that case. We can also relax the condition of having long-range antiferromagnetic order of the Cu spins. In the presence of only short-range order over a length scale ξ_m we still expect the pairing mechanism to be effective provided $\xi_p \lesssim \xi_m$. As J_z is decreased, ξ_p increases and ξ_m decreases at fixed temperature so that pairing becomes increasingly ineffective. The pairing interaction is also weakened when the number of O holes becomes large enough to reduce significantly the antiferromagnetic correlations between the Cu spins.[19] Thus, we expect a maximum in T_c at an intermediate O hole concentration.

Finally, we discuss qualitatively the effect of an in-plane Cu-Cu coupling J_{xy}. This will cause spin-exchange processes between the Cu spins, also weakening the AF order. Still, in the isotropic Heisenberg case, the long-range order is known to be 50% of the perfect Néel order at zero temperature.[17] More importantly, the spin-flip term provides another mechanism for healing the defects created by a single propagating hole. Thus, if the second hole comes by after a time longer than $1/J_{xy}$, the Cu lattice will heal by itself and the pairing interaction will be suppressed. As a function of the distance between the holes, the pairing interaction increases linearly until a separation $\xi_{\max} \sim v_h/J_{xy}$ (v_h =velocity of propagation of the hole) when it drops to zero. These considerations suggest that a uniaxial magnetic anisotropy should yield a more effective pairing mechanism than the isotropic case.

Concerning the nature of the superconducting state, in the present model there is no constraint in having two holes of opposite spins occupy the same site so that we expect the superconducting state to be isotropic s-wave-like as in the usual Bardeen-Cooper-Schrieffer theory.

In summary, we have discussed a mechanism for localization and pairing in a model for two-dimensional CuO_2. While detailed analytic as well as numerical calculations are imperative, I believe that my arguments strongly suggest that this scenario may be relevant for the oxide superconductors.

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[19]There are two ways in which this can happen. First, as noted by Emery, an O hole between two Cu spins partially blocks the exchange between them, so that antiferromagnetism will disappear as the number of O holes becomes some fraction of the number of Cu spins. Second, in the case where J_z is smaller than T_{OO}, it can become advantageous to have a region of parallel Cu spins around an O hole where it can move freely. The size of that region is proportional to $(t_{OO}/J_z)^{2/3}$ (from balance of potential and kinetic energies), so that for $n_O \sim \text{const}(J_z/t_{OO})^{2/3}$ the antiferromagnetism (and the pairing tendency) will disappear. The second mechanism is more important only if $J_z < t_{OO}$.