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PAIRING IN CuO_2 DRIVEN BY EXCHANGE INTERACTIONS BETWEEN CARRIERS AND LOCALIZED Cu SPINS

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Calculations using a realistic multi-band extended Hubbard hamiltonian, which correctly describes the Heisenberg excitation spectrum and superexchange energy of the insulating phase of CuO_2, show that the two largest energies relating to the carrier quasiparticle are the carrier bandwidth (> 5 eV) and an exceptionally large (~3 eV times a density of states factor as in an RKKY interaction) effective exchange energy between a delocalized carrier hole and a localized Cu hole. The Cu holes are found to remain localized in the superconducting materials. Calculations using a model pairing hamiltonian show that this exchange energy induces (through partial spin-polarization of the Cu holes) an attraction between carriers. This attraction is strong enough to overcome a realistic Coulomb repulsion, hence leading to net attractive pairing. Triplet p-wave (or possibly singlet d-wave) pairing is implied.

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1. INTRODUCTION

We previously derived[1], from semi-empirical and ab initio sources, a multi-band extended Hubbard hamiltonian. It includes all important energies needed to describe $1\pm x$ holes in otherwise closed shell $Cu(d^{10})O(p^6)_2$ sheets and hence is "realistic". Important conclusions include: (1) Locally these materials screen not as metals but as dielectrics. This fact, evident from Auger data on these and related compounds, implies large intra- and inter-atomic Coulomb energies, hence strong correlation between holes. This occurs in all materials with nearly filled valence shells. Evidence for this is also seen in bandstructure calculations in, for example, La_2CuO_4[2], in which, over most of the Brillioun zone, there is a relatively large gap (~3eV) from the highest occupied to the lowest unoccupied bands. This necessarily implies poor <u>local</u> screening and (2) Oxygen-oxygen direct transfer integrals, t_{pp}, are about half as large and twice as many as the Cu-O transfer. This is immediately apparent from the fact that O^{2-} ions are big. Thus at the small lattice distances in these materials the overlap of neighboring O-pσ orbitals is larger than half the overlap of an O-pσ with a neighboring Cu-d(x^2-y^2) orbital. Therefore this energy is crucial to any realistic model. Scaling up by the ratio of overlaps from the known (from photoemission data) t_{pp} (~0.25 eV) in MgO, an absolute value of $t_{pp} \approx 0.65$ eV is obtained in CuO_2. (The lattice distance in CuO_2 is 10% smaller than in MgO.) The consequence is a wide carrier bandwidth (> 5 eV) even in the <u>absence</u> of Cu-O interactions; thus, the carriers are delocalized.

Not only are the major conclusions of this work

supported by several independent ab initio studies of the energy parameters[3,4], but so are the semi-quantitative values for parameters which were obtained using scaling arguments to compare related compounds.

Our model correctly predicts properties of the insulating material which corresponds to one hole per CuO_2 unit. Calculations (using parameters from Ref. 3 plus direct exchange energies[1]) on clusters of up to 5 Cu and 16 O atoms with one hole per Cu demonstrate that the Heisenberg excitation spectrum and the measured value of the superexchange energy are well reproduced.

Computations including carriers[5] show that they occupy the O-pπ orbitals in only the smallest cluster (Cu-O-Cu with three holes). In larger clusters, the O-pσ orbitals are preferred. This energy preference increases monotonically with cluster size, which justifies omitting the pπ orbitals from the hamiltonian describing the carrier quasiparticle. Examination of the low-lying excitation spectrum and eigenfunctions of clusters with carriers shows that the first holes per CuO_2 unit, localized by correlation effects largely on the Cu sublattice remain localized in the presence of carriers. This implies that a spin-1/2 antiferromagnetic Heisenberg system exists in the superconducting materials and a description of the carrier quasiparticle must include coupling between the carrier and spin subsystems. The nature of this interaction has been studied using exact solutions of the multi-band extended Hubbard model for finite systems[6].

The interaction between delocalized carrier holes and localized Cu holes can be described as an effective exchange energy. The excitation spectrum of a CuO_4

cluster with two holes shows that the magnitude of this energy is as large as ~3 eV (derived from the singlet-triplet energy splitting), despite the fact that the carrier and localized holes are to a large degree on different sublattices. The sign of this exchange is antiferromagnetic. The exchange energy of a CuO cluster is, however, considerably smaller, showing that the delocalized property of the carriers is essential. Because the carrier is delocalized this energy is reduced by a density of states factor. The interaction is similar to an RKKY interaction.

2. PAIRING MODEL

Using the above facts, we have deduced[6] an approximate pairing model which contains the essential physics: (1) The carriers have wide bands and do not require deviations from antiferromagnetic order in the Cu-sublattice to have high mobility. (2) The effective exchange between carriers and localized Cu holes acts to spin-polarize the Cu sublattice while the Cu-Cu superexchange energy counteracts such polarization. Thus, the net result is locally a partial alignment of the Cu spins as opposed to the anti-aligned ground state which occurs in the absence of carriers.

Pairing models based on electronic excitations sufficiently high in energy that a carrier cannot move a distance of the screening length during the virtual excitation lifetime, must also include the Coulomb energies in order to determine whether any resulting attraction can overcome the Coulomb repulsion; net attraction is presumed to be a precondition for superconductivity. Because of local dielectric screening in CuO_2 sheets, the screening length is approximately the average separation between charge carriers. Therefore the Coulomb repulsion is important as the pairing interaction is not fully retarded.

Our pairing model consists of two sub-hamiltonians[6], a carrier hamiltonian in which the localized Cu holes are treated in mean field with averaged spin orientation, and a spin hamiltonian which contains the carrier-Cu effective exchange and the Cu-Cu superexchange energies. The carrier hamiltonian determines the Coulomb repulsion while the spin hamiltonian determines the attractive component of the total carrier-carrier interaction.

Solving these hamiltonians with zero, one and two carriers in boxes of N CuO_2 units and periodic boundary conditions determines the pairing energy. Varying N varies the carrier concentration.

We find net attractive p-wave triplet and repulsive s-wave pairing. Pairing is in the Cooper sense in that the attraction is between delocalized carriers. Singlet d-wave pairing is not tested nor ruled out. Our major conclusion is net attractive pairing interactions occur due to the large ratio of the effective Cu-delocalized O exchange energy to the superexchange energy. In the simplest sense delocalized carriers tend to partially align localized Cu spins, thereby producing regions of attractive exchange potentials which two carriers of similar alignment can share.

We conclude by noting that definitive experimental evidence indicating the symmetry of the gap does not exist. There is some evidence (although not definitive) that the gap is nodeless. However, this does not preclude triplet p-wave in which a nodeless gap is allowed. Finally, there is some evidence for a p-wave or possibly a d-wave[7] gap.

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