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EFFECTIVE TRANSFER FOR SINGLETS FORMED BY HOLE DOPING IN THE HIGH-T_c SUPERCONDUCTORS

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By comparing the exact results of an extended multiband Hubbard Hamiltonian for Cu_2O_7 and Cu_2O_8 clusters to those obtained from an effective single band Hamiltonian, we show that the low energy scale physics is very well described by a t–t'–J model which describes the motion of singlets in an antiferromagnetic background of spins. We obtain values for t, t' and J for both hole and electron doping and show that these are different. We also study the dispersion relations and density of states within the quasi particle approximation both of which show rather interesting characteristics which could be relevant for high T_c superconductors.

1. Introduction

Since the discovery of the new high T_c superconductors, a number of model Hamiltonians have been proposed to explain their physical properties [1]. The most simple one including explicitly electron correlation effects is the single band Hubbard model, as proposed by Anderson [2]. This involves only Cu sites, as it is supposed that the oxygen atoms effectively renormalize the parameter values and can be projected out. Subsequent experimental evidence, in particular from spectroscopies like XPS, EELS and XAS [3,4], indicated that holes introduced in the CuO_2 planes by doping the antiferromagnetic compounds go largely on the oxygen sites. Therefore it seems more appropriate to classify these materials, in the Zaanen–Sawatzky–Allen (ZSA) scheme [5], as charge transfer instead of Mott–Hubbard insulators. This suggests that a more involved Hamiltonian, like the extended Hubbard model [6,7], is needed, which includes the oxygen sites explicitly.

It was suggested by Zhang and Rice (Z&R) [8] and by ourselves [4] that the problem can be simplified for certain parameter ranges. Although a hole goes on oxygen, it can stabilize its energy by several eV by forming a symmetric singlet with one of the Cu spins already present. This hole increases its effective Cu–O transfer matrix element by a factor of two by delocalizing over the four oxygens surrounding a Cu atom, and will at the same time gain half the maximum available delocalization energy in the oxygen lattice. The appearance of such local singlet instead of triplet states is closely related to the low symmetry, or two dimensionality of the problem. By elimination of all excited local hole states Z&R then arrived at the so-called t–J model, a one band Hamiltonian containing again only Cu sites. It contains a Heisenberg term, describing the Cu spin–spin interaction, and a kinetic term for the motion of holes (singlets) in the spin lattice. If the oxygen bandwidth is neglected, as Z&R did, the singlet hole can only hop between nearest neighbor Cu sites. Inclusion of direct oxygen-oxygen transfer matrix elements introduces the possibility of next nearest neighbor hopping and as already pointed out by McMahan et al. [9] further lowers the (kinetic) energy of the symmetric singlet state. As recently pointed out by Lee [10], this so-called t–t'–J model may lead to different physics. After division of the Cu lattice in antiferromagnetic spin sublattices t in-

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volves intersublattice hopping whilst t' involves intrasublattice hopping. In case of a Néel-like state the first process is strongly renormalized because the singlet leaves behind a wake of misoriented spins. The second (t') process is not hindered by the antiferromagnetic order, leading to a free motion of the charge carriers.

In this paper our aim is twofold. First we obtain estimates of t, t' and J and investigate whether the split off singlet band retains its identity. Secondly we investigate the dispersion relation and density of states of this singlet band, for both holes and electrons, in the quasi particle approximation.

2. The calculations

We consider the two clusters Cu_2O_7 and Cu_2O_8 as shown in fig. 1. In the calculations we include the O–2p and Cu–3d orbitals. The many body Hamiltonian is solved exactly by numerical diagonalization, giving all energy eigenvalues and eigenstates. With respect to the "vacuum state" $3d^{10}$–$2p^6$, we are interested in the two hole solutions describing the undoped system, the three hole solutions describing the hole doped system and the one hole solutions describing the electron doped system.

The model parameters used in the calculations are

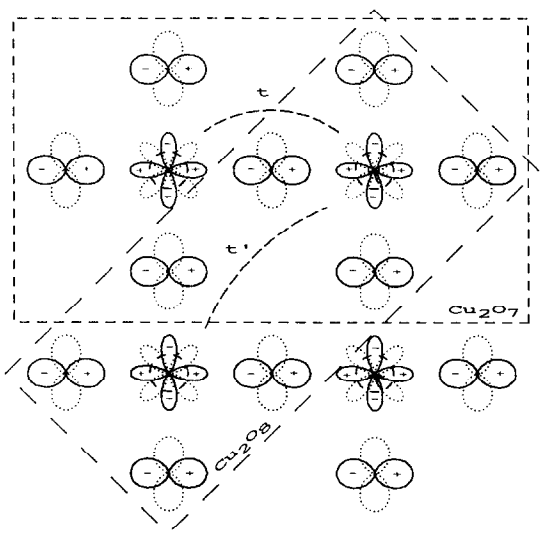

Fig. 1. The clusters considered in the calculations.

given in table I. Here we also give a translation from our parametrization to the parameters that appear in Hubbard-like model Hamiltonians. Apart from the values listed in table I we have also investigated the effect of selected deviations from this "standard set". The one hole tight binding parameters are defined according to Slater and Koster [11]. In general, for transition metal oxides, the p–d interactions satisfy the relation $pd\sigma \approx -0.5\ pd\pi$ [12]. Note that, in going from electrons to holes, these parameters have changed sign. The value for $pd\sigma$ is taken from fits to photoemission [13]. The value thus obtained is in close agreement with LDA calculations [14,9]. The oxygen p–p interactions satisfy the relation

$$pp\pi - pp\sigma = W/4$$

where W is the oxygen bandwidth, known to be about 5 eV. The energy difference between the O–2p and Cu–3d orbitals ($\epsilon_p - \epsilon_d$) is directly related to the conduction gap. In order to compensate for the banding of levels in going from a cluster to a periodic lattice [15], we take this charge transfer energy to give a gap value which is slightly larger than the measured value of 1.5–2 eV. The Coulomb interaction on Cu can be obtained fairly accurately by a combination of resonant photoemission and inverse photoemission data. The peak corresponding to two holes on Cu (d^8) is found at about 11 eV below E_F in both the high T_c materials and CuO, indicating a similar value for both compounds. From a fit to data on CuO we obtained the value given in table I. Here it is important to include all d^8 multiplets, as fully specified in terms of the Racah A, B and C parameters. For the B and C parameters the unscreened atomic values are taken, as explained elsewhere [13]. U_d is then the Coulomb expectation value for two holes in the $d_{x^2-y^2}$ orbital, in terms of A, B and C. Values for the O–O (Slater F_0 and F_2 integrals) and Cu–O (U_{pd}) Coulomb interaction are estimated from

Table I
The standard parameter set. All energies in eV.

ϵ_d	0	A	6.5	$U_d = A+4B+3C = 8.8$
ϵ_p	3.5	B	0.15	$U_p = F_0+0.16F_2 = 6.0$
$pd\sigma$	1.5	C	0.58	
$pd\pi$	−0.7	F_0	5	$t_{pd} = \frac{1}{2}\sqrt{3}\ pd\sigma = 1.3$
$pp\sigma$	−1.0	F_2	6	$t_{pp} = \frac{1}{2}(pp\pi - pp\sigma) = 0.65$
$pp\pi$	0.3	U_{pd}	0	

Auger measurements on Cu_2O [16]. U_{pd} is found to be smaller than 1 eV. For completeness we report the result for both $U_{pd}=0$ and $U_{pd}=1$ eV.

The size of the matrix is reduced by noting that the Hamiltonian commutes with both the total spin S and its z component S_z. A further reduction of about a factor of 4 is achieved by using the spatial symmetry of the problem. The appropriate point group for both clusters is D_{2h}.

3. Undoped case: two holes

We first consider the case of the (undoped) antiferromagnetic insulating materials, where there is one hole per Cu unit. So for our clusters this corresponds to a two hole calculation. The quantity of most interest is then the superexchange interaction for nearest neighbors (J_{nn}) and next nearest neighbors (J_{nnn}), corresponding to the Cu_2O_7 and Cu_2O_8 cluster, respectively. The superexchange is here defined as the energy difference between the singlet and triplet or, for nearest neighbors,

$$H=J_{nn}\sum_{<i,j>,j>i} \boldsymbol{S}_i\cdot\boldsymbol{S}_j . \tag{1}$$

An order of magnitude estimate may be obtained from the fourth order perturbation expression:

$$J_{nn}=4\frac{t_{pd}^4}{\Delta^2}(1/U_d+2/(2\Delta+U_p)) . \tag{2}$$

For the standard parameter set this gives $J_{nn}=0.25$ eV. Results of the calculations are shown in tables II and IV. As seen from table IV J_{nn} is considerably reduced by U_{pd} and as pointed out by Stechel and Jennison [17] is further reduced by a direct Cu–O exchange obtaining finally the experimental value of 0.12 eV. An important observation is that orbitals other than $d_{x^2-y^2}$ and p_σ do not play any significant role for the low energy states. In particular the occupation of the Cu $d_{3z^2-r^2}$ orbital turns out to be much less than 1% in both the lowest singlet and lowest triplet states. Note that similar calculations have been done by Annet et al. [18] who embedded small clusters in a large "host" of uncorrelated oxygen atoms. We are able to reproduce their results to within less than 5% for J_{nn} and 25% for J_{nnn}. This good agreement can be understood by noting that the two holes are almost completely located at the Cu sites and their nearest neighbor oxygens. This holds of course only for the low lying oxygen states.

4. Hole doped case: three holes

We now proceed to the case of the (p-doped) (super)conducting materials. So we put one additional hole into our cluster, which leads to a three hole calculation. Results are shown in table III. The ground state is a spin doublet, spatially symmetric (antisymmetric) in the Cu_2O_7 (Cu_2O_8) cluster, and is separated by more than 1.5 eV from the lowest quartet. Again, for the low energy states the $d_{x^2-y^2}$ and p_σ orbitals turn out to be dominant, but less exclusively so than in the undoped case. For example, in the Cu_2O_7 ground state the occupation of $d_{3z^2-r^2}$ (per Cu) is about 1% and restricting the basis set to $d_{x^2-y^2}$ and p_σ orbitals raises the ground state energy by only 0.06 eV, while for the first excited state the corresponding numbers are 10% and 0.14 eV. We will see below that in a band description the excited state would correspond to the point $\boldsymbol{k}=(\pi/a, 0)$ in the Brillouin zone, and further that for full translational symmetry the doped holes are expected to go into states near the zone boundary. These states would thus contain up to 10% $d_{3z^2-r^2}$ character, as has actually been found by $\boldsymbol{q}$ dependent EELS by Nücker et al. [3]. The second excited state, which lies about 0.3 eV above the first excited state, contains a considerable amount of $d_{3z^2-r^2}$ character and no equivalent state is found when using only the restricted basis.

Next, let us address the question if the obtained states can be interpreted in terms of a local singlet formed by the added hole with a copper spin. For that purpose we explicitly construct three-hole wave functions from the one-hole (doublet) and two-hole (singlet) ground state wave functions of a CuO_4 cluster, by taking direct products

$$|\mathrm{R},\uparrow\rangle=|\Phi(1),\uparrow\rangle_{\mathrm{L}}\otimes|\Phi(2),\mathrm{S}\rangle_{\mathrm{R}} \tag{3}$$

Here R(L) indicates the CuO_4 unit on the right (left) hand side in the cluster, and the three-hole state is labeled by the position of the singlet. The set of orbitals in such a CuO_4 unit is restricted to $d_{x^2-y^2}$ and the four p_σ orbitals. Note that the hybridization

Table II
Results of the two hole calculations. Parameter values not mentioned in column 2 are taken from table I. "O" indicates the central oxygen (Cu_2O_7, Cu_2O) or one of the four equivalent oxygens between the two Cu atoms (Cu_2O_8). Column 3 contains the ground state (singlet) energy in eV.

Cluster	Parameters	Energy	occupation number			
			singlet		triplet	
			x^2-y^2	O	x^2-y^2	O
Cu_2O_7	-	−3.7	0.66	0.21	0.70	0.13
	$U_{pd}=1$ eV	−3.5	0.68	0.17	0.70	0.11
	$\Delta=2.5$ eV	−4.4	0.57	0.26	0.62	0.16
	$U_d, U_p=\infty$	−3.4	0.69	0.14	0.70	0.13
Cu_2O_8	-	−3.5	0.68	0.08	0.69	0.08
	$U_{pd}=1$ eV	−3.5	0.68	0.08	0.69	0.08
	$\Delta=2.5$ eV	−4.2	0.60	0.11	0.61	0.10
	$U_d, U_p=\infty$	−3.5	0.69	0.08	0.69	0.08

Table III
Results of the three hole calculations. Shown are the three lowest lying energy states. Column 3 gives the spatial symmetry of the three hole states for the D_{2h} point group. For notations see table II.

Cluster	State	Symmetry	Energy	occupation number		
				x^2-y^2	$3z^2-r^2$	O
Cu_2O_7	1	A_g	−3.3	0.78	0.01	0.37
	2	B_{1u}	−2.5	0.73	0.10	0.26
	3	B_{1u}	−2.2	0.52	0.36	0.32
Cu_2O_8	1	B_{2u}	−3.2	0.77	0.00	0.20
	2	B_{3g}	−2.8	0.79	0.00	0.17
	3	B_{1u}	−2.0	0.46	0.43	0.18

$|\Phi(1), \uparrow\rangle$ and the relative amplitudes of $d^9\underline{L}$, d^8 and $d^{10}\underline{L}^2$ in $|\Phi(2), S\rangle$ are those pertaining to a CuO_4 cluster, and are not treated as variable parameters in these three-hole wave functions. For the Cu_2O_8 cluster the definition (3) is adequate, but for the Cu_2O_7 cluster, where the central oxygen atom is common to both CuO_4 units, wave function components corresponding to double occupation of an orbital on that atom must be dropped, and the wave function renormalized. With these states we write down the Hamilton eigenvalue problem as a 2×2 matrix equation. In the case of Cu_2O_7 we must take into account that $|R, \uparrow\rangle$ and $|L, \uparrow\rangle$ have a nonzero overlap of about 2% for the standard set.

The eigenstates are

$$|E, \uparrow\rangle = \frac{1}{\sqrt{N}}(|R, \uparrow\rangle + |L, \uparrow\rangle)$$

and

$$|O, \uparrow\rangle = \frac{1}{\sqrt{N}}(|R, \uparrow\rangle - |L, \uparrow\rangle) \quad (4)$$

where N, the normalization factor, is not necessarily equal to 2. These two eigenstates are now compared with the ground state and first excited state obtained in the full three-hole calculations. We should point out that the comparison is made using the restricted basis set in the full cluster calculation, because we are interested here in the adequacy of the local singlet picture, and not so much in the issue of the basis set. We find that the overlap of $|E, \uparrow\rangle$ or $|O, \uparrow\rangle$ (whichever is appropriate) with the ground state is 98.9% (98.2%) for Cu_2O_7 (Cu_2O_8), and with the first excited state is 98.8% (98.0%), while the obtained energy values deviate by 0.08 eV (0.07 eV). From this we conclude that the lowest energy states are indeed well described by a singlet state at one, and a

spin on the other Cu site. This implies that the effect of holes introduced in the antiferromagnetic compounds is not to induce a direct ferromagnetic exchange between nearest neighbor Cu spins [19], but is more accurately described as the removal of a spin from the lattice, it being replaced by a spinless charged particle.

The hopping matrix elements of such a singlet quasiparticle can also be obtained unambiguously because of the nearly perfect representation of the lowest two three-hole cluster states by the singlet-spin product states. For the latter states the energy difference between $|E, \uparrow\rangle$ and $|O, \uparrow\rangle$ corresponds to twice the effective transfer matrix element (effective, if $|R, \uparrow\rangle$ and $|L, \uparrow\rangle$ overlap). The nearest neighbor (t) and next nearest neighbor (t') hopping matrix elements are therefore taken as half the energy difference between the first excited state and the ground state, in Cu_2O_7 and Cu_2O_8 respectively, which yields the values shown in table IV. Here also the results for the conduction gap are given, which is defined by

$$E_{gap} = E_0(1\text{ hole}) + E_0(3\text{ holes}) - 2E_0(2\text{ holes}) . \quad (5)$$

As can be seen from the table, the results for t and t' are not sensitive to changes in the inter-site Coulomb interaction U_{pd} and the change transfer energy Δ. The last two lines in the table are given to illustrate the effect of the oxygen–oxygen hopping t_{pp} and the on site Coulomb repulsion. Recently Maekawa et al. [20] have also estimated the value of t from calculations on a Cu_2O_{11} cluster.

An important aspect is the symmetry of the states. For Cu_2O_7 the ground state has A_g symmetry (in the D_{2h} point group), and the excited state B_{1u} symmetry. Therefore the ground state corresponds to $|E, \uparrow\rangle$ and the excited state to $|O, \uparrow\rangle$, implying that the sign of t is negative. For the Cu_2O_8 cluster the ground state has B_{2u} symmetry and the excited state B_{3g} symmetry, so that $|O, \uparrow\rangle$ is the lowest energy state and consequently t' is positive. (The sign convention is the one appropriate for hole states.) For full translational symmetry this would mean that the nearest neighbor singlet transfer favours hole states at Γ and the next nearest neighbor transfer favours hole states at $k = (\pi/a, 0)$.

5. Electron doped case: one hole

Finally we turn to the n-doped materials. Here we add one electron to the cluster, which leads to a one hole calculation. In some respect the case of electron doping resembles the case of hole doping. Here again one could picture the situation as one of "spinless particles", now corresponding to full shell CuO_4 units, moving in a spin lattice. One difference lies in the symmetry of the ground state. In the one hole calculation we find B_{1u} for Cu_2O_7 and B_{3g} for Cu_2O_8. Thus the signs of t_e and t'_e, now describing the motion of full shell singlets, are reversed with respect to the hole doped case. (The sign convention is now the one appropriate for electron states.) The absolute values of these redefined hopping parameters as given in table IV resemble those obtained from the three hole calculations. Note however that both the difference in sign as well as the difference especially in t'_e from the three hole case means that the effective Hamiltonian has different parameters for the hole

Table IV
Summary of the values found for the effective singlet transfer matrix elements t and t' (hole doping), t_e and t'_e (electron doping), and the nearest and next nearest neighbor superexchange J_{nn} and J_{nnn}. E_{gap} is the calculated band gap energy. All energies in eV.

Parameters	t	t'	J_{nn}	J_{nnn}	t_e	t'_e	E_{gap}	
							Cu_2O_7	Cu_2O_8
–	−0.44	0.18	0.24	0.024	0.40	−0.10	2.0	2.0
$U_{pd}=1$ eV	−0.40	0.17	0.17	0.018	0.40	−0.10	2.6	2.6
$\Delta=2.5$ eV	−0.45	0.19	0.32	0.044	0.46	−0.13	1.6	1.5
$t_{pp}=0$ eV	−0.26	0	0.08	0	0.26	0	2.6	3.0
U_d, $U_p=\infty$	−0.27	0.17	0.05	0.005	0.40	−0.10	3.1	3.3

and electron doped case. This is a remnant of the more general multiband Hubbard Hamiltonian that we started from.

6. Quasi particle bandstructure

In fig. 2 we show the quasi particle band structure for both hole and electron doping assuming that these singlets can move freely as would be the case in a lattice without spins. Here both bands are represented in the conventional way, i.e. as electron bands. The corresponding density of states is shown in fig. 3. We note the peaks in the density of states which result from the opposite sign of t and t' and the flat dispersion between Γ and $(\pi/a, 0)$. (Shown also in figs. 2 and 3 are the "Fermi" levels corresponding to 30% hole doping which interestingly enough is at the edge of the high peak in the density of states.) Of course it is highly probable that the spin system will strongly distort this picture and perhaps even cause a breakdown of the quasi particle Fermi liquid picture. In the most simple minded picture however we might retain the quasi particle picture but now with modified parameters t and t'. We expect especially t to

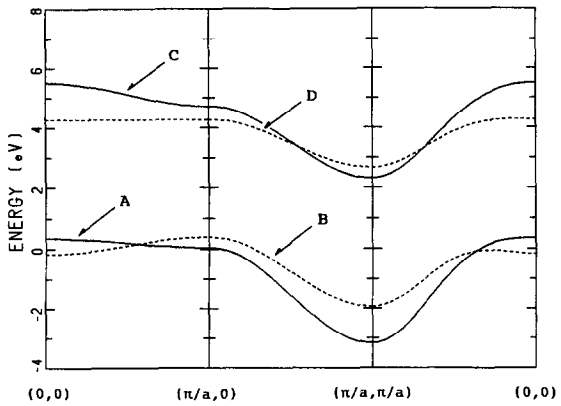

Fig. 2. One particle bandstructure for the quasiparticles as described in the text. The x and y axes correspond with the in plane Cu–O direction. The curves correspond to:
A) hole doping $t = -0.44$ eV, $t' = 0.18$ eV;
B) hole doping $t = -0.22$ eV, $t' = 0.18$ eV;
C) electron doping $t_e = -0.40$ eV, $t'_e = 0.10$ eV;
D) electron doping $t_e = -0.20$ eV, $t'_e = 0.10$ eV.

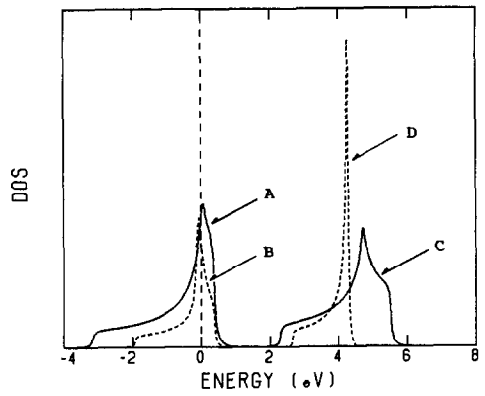

Fig. 3. One particle density of states. For notations see caption of fig. 2.

be strongly reduced because of the wake of disoriented spins left behind. We therefore also show in figs. 2 and 3 the result for t reduced to 50%. We now see that the Fermi level would be along $(0, 0) \rightarrow (\pi/a, 0)$ for 30% doping, and there could be another hole pocket between $(0, 0)$ and $(\pi/a, \pi/a)$. Also interesting is that this model predicts the direct interband transitions (charge transfer) of the insulator to be at about 3–4 eV with the indirect band gap transitions starting at 1.52–2 eV.

7. Conclusions

In conclusion, we have shown by many-body cluster calculations using an extended multiband Hubbard Hamiltonian that the low energy scale physics of the Cu_2O_7 and Cu_2O_8 clusters is well described by a simplified Hamiltonian (t–t'–J model) which involves only x^2-y^2 symmetry orbitals. We propose that this conclusion can be extended to the full CuO_2 planes for not too high doping (<0.5 hole per formula unit). In this case only the lowest energy singlet states will be of importance and they are in energy well removed from the higher lying bands of other symmetry. However the large singlet dispersion will cause this band to overlap with other symmetry bands causing a t–t'–J model description to break down at high doping. We also point out that the Cu $d_{3z^2-r^2}$ orbital could play a role well, since we found it to have appreciable ($\sim 10\%$) amplitude in

the states that would be occupied first upon doping. We expect the amount of $d_{3z^2-r^2}$ character to be even larger for points near the Brillouin zone boundary in the true crystal because each CuO_4 unit in fact has 4 nearest neighbor such units rather than only one as considered here.

The effect of the multiband character of the more general Hamiltonian as compared to a single band Hubbard Hamiltonian is fourfold:

a) It introduces the superexchange J.
b) It introduces new quasiparticles.
c) It introduces a next nearest neighbor hopping t'.
d) It causes the parameters to be different for hole and electron doping.

We have obtained values for t, t' and J and the resulting quasiparticle dispersion for both hole and electron doping. In this quasiparticle picture the change transfer band gap is indirect ≈ 1.5–2 eV and the direct gap is 3–4 eV. It is interesting that the sharp peak in the density of states below the Fermi level has an area corresponding to about 30% hole doping. The influence of the spins and J could cause a break down of such a quasi particle picture and at least cause strong renormalization of the parameters. We note that the values found here for t, J and t' seem to be in the regime leading to an attractive interaction between singlet quasiparticles [21,22]. A detailed study of the t–t'–J model and a comparison to optical, photoemission and inverse photoemission data will tell us more.

Acknowledgements

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