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One hole in a three-band model of a high-temperature superconductor

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Abstract

We study a three-band effective model of a CuO_2 plane for the simple case of a single hole, with the aim of investigating the nature of the ground state stabilized by the motion of a single hole on the CuO_2 lattice. Our model is derived from, and retains the essential physics of the Anderson lattice model, but is more amenable to further investigation by virtue of the lifting of the spin degeneracy on the copper sites provided by perturbation theory.

We use the Lanczos algorithm to numerically solve a series of finite systems which tend to the CuO_2 plane in the thermodynamic limit. Although we only study finite systems, the largest systems are sufficiently large to demonstrate behaviour that is independent of the boundary conditions, and is hence representative of the behaviour in the thermodynamic limit. In order to gain a good understanding of the competing energy scales, we consider only a single hole at $T = 0$.

Our calculations predict that the ground state of the three-band model for a single hole is strongly quantum, dominated by short-range dimer correlations, reminiscent of a resonating valence bond state. There is no evidence for a discontinuity in the occupation number, indicating that the system is not a Fermi liquid.

These predictions are in contrast to those of the t–J model, where the hole motion alone predicts Nagaoka ferromagnetism for the planar system, and one must include magnetic exchange terms in order to obtain the experimentally observed low-spin ground state.

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1. Introduction

In this paper, we present a numerical investigation of a single hole in a high-temperature superconductor. We elect to study a limit of the natural Anderson lattice model (ALM) for the cuprates which retains the three-band character of the ALM. This model allows us to study the competition between energy scales that underpins the unusual behaviour of the cuprates. Several other authors have studied similar models [1, 2], but none appears to have numerically studied the single-hole limit. Although this may seem to be a very simple system, it allows us to investigate the nature of the ground state induced by the motion of a single hole on the CuO_2 lattice. It is this apparently simple question which we will attempt to answer in this paper.

The natural Hamiltonian for the CuO_2 planes is the ALM, given by

$$\mathcal{H}_{\mathrm{ALM}} = -\Delta \sum_{i\sigma} d^{\dagger}_{i\sigma} d_{i\sigma} + U \sum_{i} d^{\dagger}_{i\uparrow} d_{i\uparrow} d^{\dagger}_{i\downarrow} d_{i\downarrow} + V \sum_{\langle i,j\rangle\sigma} \left(d^{\dagger}_{i\sigma} p_{j\sigma} + p^{\dagger}_{j\sigma} d_{i\sigma} \right) \tag{1}$$

where we have chosen a uniform phase for the hybridization for convenience, equivalent to translating reciprocal space by (π, π). The operators $d^{\dagger}_{i\sigma}$ and $p^{\dagger}_{j\sigma}$ create copper holes on site i and oxygen holes on site j, respectively, with σ labelling the spin of the hole. Δ is the energy gained by putting holes on copper sites, U is the Coulomb penalty for double occupation of copper sites, and V is the copper–oxygen hybridization energy. The on-site oxygen energy has been set to zero for simplicity as we are only considering constant doping in this work. In the undoped limit, each copper site in the CuO_2 planes is Cu^{2+} and so is occupied by a *hole* of spin one-half. The oxygen states are O^{2-} and are filled shell. In the hole-doped cuprates, doping occurs on the oxygen sites with O^{2-} ions becoming O^{-}.

The most popular approach to this model was first proposed by Zhang and Rice [3]. Their idea was to examine all possible *local* states involving a single oxygen hole delocalized around a copper ion. They found that the lowest energy of this local state was obtained when the oxygen hole was tightly bound in a local singlet with the copper local moment. This idea allows the ALM to be reduced to a single band, $U = \infty$ Hubbard model on the copper sites only: the t–j model, which has been extensively studied (see, for example, the review article by Dagotto [4]).

Although the t–J model is attractive for its inherent simplicity, it is not clear whether this model retains the essential physics of the ALM. Certainly, it does not if one ignores superexchange, with the resulting t-model giving Nagaoka ferromagnetism on the square lattice [5], and hence superexchange is *required* to give the experimentally observed low-spin ground state. The addition of t' and t'' has been found to reproduce the experimental band structure more faithfully [6], and it is now more common to study the t–t'–t''–J model. We criticize this modification on the basis that it is based on phenomenological considerations rather than a clear understanding of the underlying physical processes. On these grounds, we have chosen to study the ALM directly, retaining the three-band character of the model, with the aim of gaining a clear physical picture of the nature of the ground state induced by hole motion alone.

In the undoped parent compounds, there is massive spin degeneracy due to the presence of a single hole on each copper site. Upon doping, there is additional degeneracy from the position of the oxygen hole, and its spin. This degeneracy is lifted by second-order perturbation theory in V, and we obtain a new effective Hamiltonian, the t–X–J model:

$$\mathcal{H}_{\mathrm{eff}} = X \sum_{\langle ij\rangle\sigma} \sum_{\langle ij'\rangle\sigma'} d^{\dagger}_{i\sigma} p^{\dagger}_{j'\sigma'} p_{j\sigma} d_{i\sigma'} - t \sum_{\langle ij\rangle\sigma} \sum_{\langle ij'\rangle} p^{\dagger}_{j\sigma} p_{j'\sigma} + \frac{J}{2} \sum_{\langle ii'\rangle} \mathbf{S}_i \cdot \mathbf{S}_{i'} \tag{2}$$

where $X = \frac{UV^2}{\Delta(U-\Delta)}$, $t = \frac{V^2}{U-\Delta}$ and $J = \frac{4V^4}{\Delta^3} + \frac{4V^4}{\Delta^2 U}$. This reduction to an effective model requires that $U - \Delta, \Delta \gg V$, and physically corresponds to the elimination of the copper charge degree of freedom. This is experimentally relevant as the system is a robust Mott insulator. Note that we retain the basic three-band picture inherent in the physical material, with a flat, non-bonding oxygen band sandwiched between bonding and anti-bonding hybrid bands.

In this paper, we restrict our attention to the case $t = 0$ for simplicity. This corresponds to the physical limit $U - \Delta \gg \Delta$, which is appropriate to the real physical system. We have studied non-zero values of t, and have found that the nature of the ground state at $t = 0$ is preserved up to $t/X = \Delta/U \approx 0.25$. Above this, the ground state does not have the experimentally observed low total spin, and the model is equivalent to the Nagaoka problem. At $t = X$, this equivalence can be rigorously demonstrated. In the real physical system, we expect that $t/X \approx 0.1$. We also set $J = 0$ since it is much smaller than both t and X, and correlations induced by the hole motion will dominate over superexchange correlations. In fact, we will see that the hole motion alone induces a low-spin ground state which is *significantly* different to that induced by the Heisenberg interactions, and we wish to highlight the basic nature of this state.

We have deliberately chosen not to include other physical terms such as direct oxygen–oxygen hopping t_p, Coulomb repulsion on the oxygen sites U_p, and inter-atom Coulomb repulsion U_{pd} in order to keep the model as simple as possible. In fact, both first- and second-neighbour oxygen hopping is present in the second-order effective Hamiltonian , and the direct inclusion of t_p amounts to an anisotropic reparametrization of t. We ignore U_p and U_{pd} on the assumption that U is much larger than each of these and will dominate the correlation effects.

We are also limited to studying $T = 0$ since our chosen technique, the Lanczos algorithm, fails at finite temperature. Recent work by Long *et al* [7] indicates that there may be a solution to this difficulty.

2. Method of study

The principal method we use to study the t–X model is *exact diagonalization* using the Lanczos algorithm. Like most other models of strongly correlated systems, the model is not amenable to an exact solution and we must rely on approximate methods. Exact diagonalization is the first step towards formulating a controlled approximation scheme which encompasses the major features of the model. We choose to exactly solve finite systems using the Lanczos algorithm, and then attempt to scale the results to infinite system size. Of crucial importance to good scaling is the correct choice of geometry: we require a geometry which scales to the square lattice as we take the infinite limit. Whilst one may choose to study square clusters (3×3, 4×4, etc), one can only study a very few of these systems before the problem becomes too large for the computer. To overcome this difficulty we choose to study the linear chain with extra bonds of length p, which corresponds to the square lattice in the limit that both the system size N and the length of the extra bond p are taken to infinity. The case of additional fifth-neighbour bonds ($p = 5$) is depicted in figure 1, which clearly shows that such systems are best considered as a 'spiral'.

We note that when this geometry is considered as a modified linear chain, the associated reciprocal space can be considered to be a line which crosses the Brillouin zone of the square lattice p times, yielding a multi-part Fermi volume. This choice of geometry allows us to effectively scale the results of finite system calculations to infinite systems. The methodology is to consider each value of p individually. For a given p, we calculate for many values of the system size N, and then scale the results to $N \mapsto \infty$. This is repeated for many different

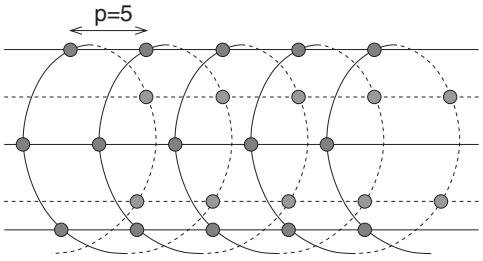


Figure 1. An example of the spiral geometries ($p = 5$) used in exact diagonalization calculations. The circles represent the location of copper ions, the oxygen ions have been omitted for clarity, and sit midway between each connected pair of copper ions.

values of p, and we then attempt to converge to the limit $p \mapsto \infty$. These calculations are complicated by the presence of two types of boundary conditions. Under the assumption that the boundary conditions are closed, the geometry is a spiral coiled around a torus, and this yields (global) boundary conditions around the entire system, and (local) boundary conditions around each 'segment' of the spiral. Each of these can be either periodic or aperiodic, giving a total of four possible combinations of boundary conditions. In the thermodynamic (infinite) limit, we expect the results to be independent of the boundary conditions.

The question in which we are most interested in this work is whether the system is a Fermi liquid, and if it is not, then to identify what it actually is. We are also interested in examining the nature of the spin correlations that are induced by the motion of a single hole. The quantities which allow us to investigate these issues are the oxygen hole occupation number, $\langle n(\mathbf{k})\rangle$ and the static copper spin correlations $\langle|\mathbf{S}_\mathbf{k}|^2\rangle$, respectively.

In order to ask whether the system is a Fermi liquid or not, we must ask what happens when a hole is added to the system. What is the distribution in reciprocal space? Is there a well-defined Fermi surface? The ability to calculate $\langle n(\mathbf{k})\rangle$ allows us to answer these questions. The calculation is straightforward, and we have to calculate

$$\langle n(\mathbf{k})\rangle = \frac{1}{N}\sum_{\sigma jn}\left\langle p^\dagger_{j,\sigma}\, p_{j+n,\sigma}\right\rangle \cos kn. \tag{3}$$

Note well that we are working with *three* atoms per unit cell, comprising one copper and two oxygen atoms, and we calculate the occupation of the oxygen sites only. This means that the quantity $\left\langle p^\dagger_{j,\sigma}\, p_{j+n,\sigma}\right\rangle$ has four components, associated with transitions between the different sublattices, and the physically interesting quantities are the eigenvalues of the associated matrix. The states in the unit cell actually split into bonding and non-bonding configurations, of which only the bonding state is ever occupied, and hence the eigenvalue associated with the non-bonding state is zero, and we are left with only one non-zero eigenvalue. Noting that we are working with only a single hole, the occupation number $\langle n(\mathbf{k})\rangle = \langle \delta n(\mathbf{k})\rangle$, the *change* in occupation number. Although the copper band is half-filled, we can consider the system to be effectively empty since we are only interested in the oxygen occupation. Consequently, rather than expecting $\langle n(\mathbf{k})\rangle$ to be a step function in a free electron picture, we would expect to see only a peak of unit height at the Fermi surface. The presence of interactions complicates this picture somewhat since this leads to the hole being distributed non-uniformly throughout the Fermi volume. In order to compare systems of different sizes we must ensure that each calculation has the same normalization. This is achieved by multiplying by the system size N

to compensate for the different numbers of points in reciprocal space across which the single particle is distributed. The quantity we actually calculate is

$$N\langle\delta n(\mathbf{k})\rangle = \sum_{\sigma jn} \left\langle p^{\dagger}_{j,\sigma} p_{j+n,\sigma} \right\rangle \cos kn. \tag{4}$$

Note that for a pure free electron model, this results in a peak at the Fermi level which diverges linearly as a function of system size, $N\langle\delta n(\mathbf{k}_{\mathrm{F}})\rangle \sim N$. In an interacting Fermi liquid, where a finite fraction of the particle enters at the Fermi level, there is still a linearly divergent peak, but some of the weight is redistributed across the remainder of the Fermi volume. For a Luttinger liquid, the particle is distributed across the whole Fermi volume. We still expect a divergence at the Fermi level, but in this case, it should diverge as $N\langle\delta n(\mathbf{k}_{\mathrm{F}})\rangle \sim N^{\alpha}$, where $0 \leqslant \alpha \leqslant 1$. A divergent peak is indicative of long-range order, whereas short-range correlations converge to a limit which is independent of system size.

The evaluation and interpretation of the spin correlations is rather more straightforward. Experimentally, one measures the copper–copper spin correlations via neutron scattering, and the relevant operator is

$$\langle|\mathbf{S}_{\mathbf{k}}|^2\rangle = \frac{1}{N} \sum_{jn} \langle \mathbf{S}_j \cdot \mathbf{S}_{j+n} \rangle \cos kn \tag{5}$$

where the labels j and n are for the copper sites only. Although this quantity is experimentally relevant, we do not attempt to make experimental predictions from these simplified calculations.

Before analysing the results of the numerical calculations for the t–X model, let us consider what a naïve free electron picture would provide. The crucial quantity one must consider is the band structure, or equivalently, the structure factor. For the spiral systems we study here, the structure factor is given by

$$\gamma(k) = \cos k + \cos pk. \tag{6}$$

We must be careful when comparing the correlated tight-binding model that we study to the simple free electron picture. In the correlated model, there are localized holes on each copper site, and a single delocalized oxygen hole, and these are distinct. This means we only need consider the number density of the single oxygen hole, and $\langle n(\mathbf{k})\rangle = \langle\delta n(\mathbf{k})\rangle$ in the correlated picture. In a free electron picture, there is no localization, and we must consider all the holes. The system is therefore *half-filled*, and the structure factor allows us to deduce which regions we would expect to be filled. In all of the results presented here, we provide the free electron structure factor for comparison.

3. Results

We studied a variety of different systems with system sizes ranging from $10 \leqslant N \leqslant 22$ and spiral pitches ranging from $2 \leqslant p \leqslant 5$. For each system we calculated the occupation number, $N\langle\delta n(\mathbf{k})\rangle$, and the spin density $\langle|\mathbf{S}_{\mathbf{k}}|^2\rangle$. For each value of p, we group systems of different N in order to determine how the quantities of interest scale with system size for a given spiral pitch.

As we explained previously, we study each value of p independently, and then search for convergence at $p \mapsto \infty$. The signature of convergence to the thermodynamic limit is the equivalence of periodic and aperiodic boundary conditions. At $p = 2$ and 3, we see good convergence of the calculations with respect to N, but changing the local boundary conditions alters the results drastically, and these systems are certainly not representative of the limit

$p \mapsto \infty$ in which we are interested. For the systems with higher values of p which we study, the results indicate that these systems may be representative of the $p \mapsto \infty$ limit. In figure 2, we show the results for the $p = 4$ spiral, and in figure 3 we show the results of the $p = 5$ spiral.

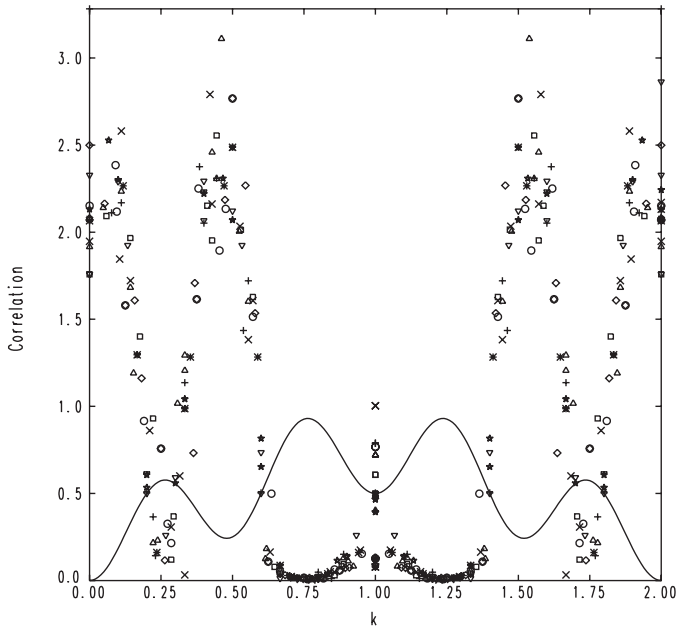
In both figures 2 and 3 we only show the results for periodic boundary conditions around the spiral segments, with both periodic and aperiodic boundary conditions around the whole system. In fact, the results for aperiodic boundary conditions around the spiral show essentially the same behaviour for both $p = 4$ and 5. We can identify several interesting features from these data.

Let us first examine the occupation numbers shown in figures 2(a) and 3(a). As we discussed in the previous section, we can quite easily compare the results of these calculations to what we would expect from the free electron gas. In the free electron picture we expect the occupied regions to occur in the minima of the structure factor. Comparing the numerical results (symbols) with the structure factors (solid lines), we see that the occupied regions in the correlated model are consistent with those expected in the free electron model, and the expected Fermi surfaces have many parts. However, there is no indication of any divergence at the Fermi points, indicating that the additional hole does not enter principally at the Fermi level, but is spread across the Fermi volume. The largest correlations are not provided by the largest systems, indicating that the high peaks are truly non-divergent. This is evidence for non-Fermi liquid behaviour in the three-band model at low doping.

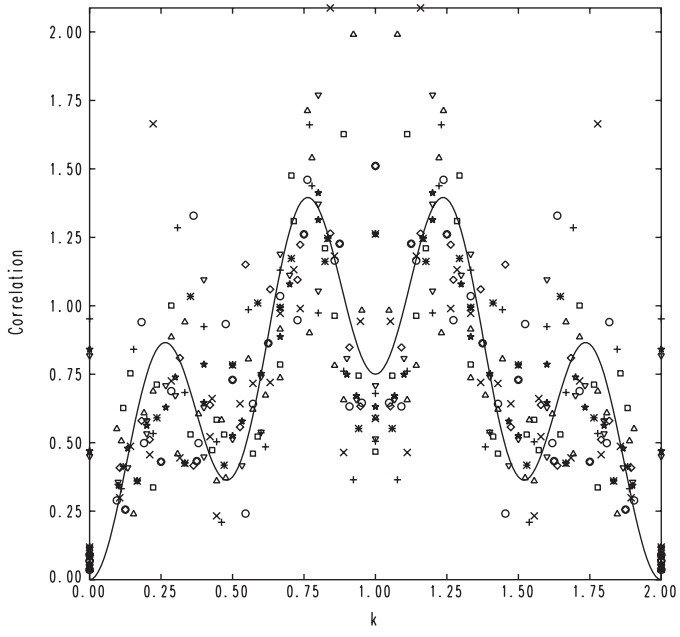
We may also obtain much useful information from the spin correlations shown in figures 2(b) and 3(b). The numerical data are indicated by the symbols, whilst the solid lines represent the free electron structure factor scaled to have area $s(s + 1) = \frac{3}{4}$. The similarity between the numerical data and the structure factor is striking. There is no sign of any divergent spin correlations, and hence there is no long-range order. The resemblance of the numerical data to the structure factor indicates that the correlations are primarily nearest neighbour, and this is indicative of a *spin-dimerized* ground state. Such a state is reminiscent of the RVB state proposed by Anderson [8], and is highly quantum with no classical analogue.

4. Discussion and conclusions

In this work, we have provided evidence for the possible non-Fermi liquid behaviour of a single hole in a high-temperature superconductor. Starting from the natural three-band model of the CuO_2 planes, we derived a low-energy effective Hamiltonian which retains the essential character of the ALM. We then proceeded to numerically solve this model on a sequence of geometries which are equivalent to the square lattice in the thermodynamic limit. Although computational constraints prevent this limit from ever being truly reached, we have been able to perform calculations on sufficiently large systems to remove any dependence on boundary conditions. This suggests that our largest systems are representative of the infinite limit. These calculations have demonstrated that a single oxygen hole on the CuO_2 lattice generates highly quantum short-range spin correlations, yielding a strongly dimerized ground state. The hole motion strongly lifts the spin degeneracy to provide a low-spin ground state where all spins are paired in local singlets. This is in direct contrast to the popular one-band t–J model, where one expects the hole motion alone ($J = 0$) to provide Nagaoka ferromagnetism, and one must rely on the high-order superexchange in order to provide the observed low-spin ground state. Moreover, we note that the Heisenberg interactions stabilize an ordered antiferromagnet, physically quite distinct from the state we have studied, which contains only short-range correlations and no long-range order.



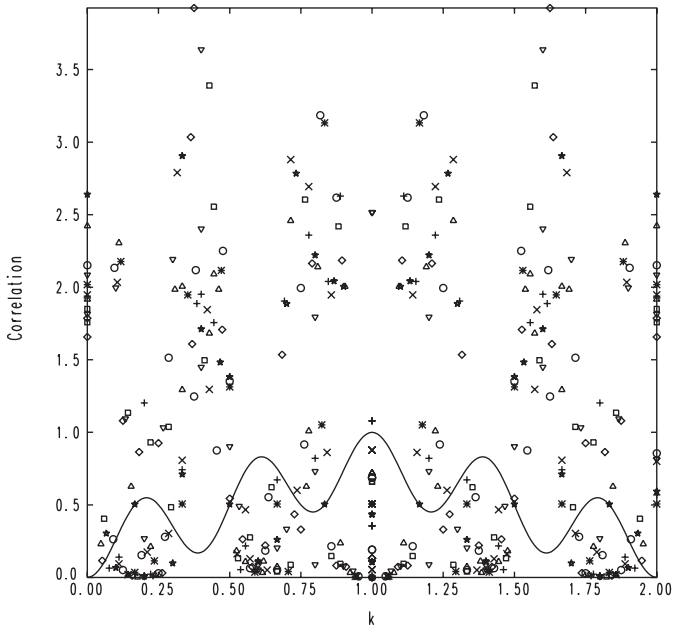
(*a*)



(*b*)

Figure 2. Finite size scaling of (*a*) $N\langle\delta n(\mathbf{k})\rangle$, (*b*) $\langle|\mathbf{S}_\mathbf{k}|^2\rangle$ on the $p = 4$ spiral. The solid lines represent the free electron structure factor scaled to (*a*) unit area, (*b*) area $s(s+1) = \frac{3}{4}$.

This suggests that the single-band picture of the CuO_2 planes does not correctly reproduce the ground state of the ALM.



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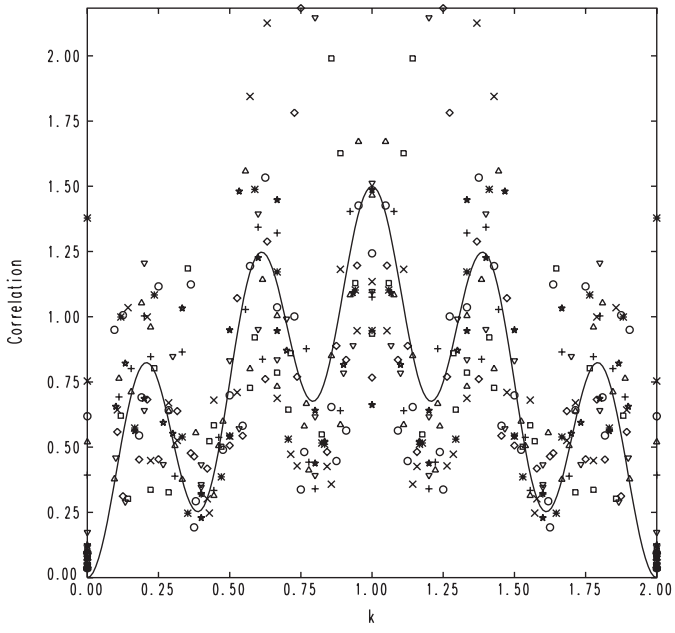

(*b*)

Figure 3. Finite size scaling of (*a*) $N\langle\delta n(\mathbf{k})\rangle$, (*b*) $\langle|\mathbf{S}_\mathbf{k}|^2\rangle$ on the $p = 5$ spiral. The solid lines represent the free electron structure factor scaled to (*a*) unit area, (*b*) area $s(s+1) = \frac{3}{4}$.

We also observed non-Fermi liquid behaviour in these calculations, with the results providing no evidence for a discontinuity at the Fermi points expected from Fermi liquid theory. Although the occupied regions are broadly in line with the Fermi picture, the lack of a

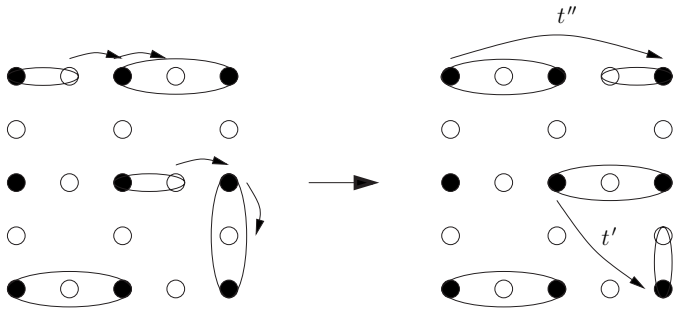

Figure 4. An illustration of how effective t'–t'' hopping can result from the X-term of the t–X–J model, in a specific spin background.

discontinuity means that the hole does not necessarily enter the system at the Fermi point, but is non-uniformly distributed across the entire Fermi volume. The strong correlations present in the model appear to be sufficient to destroy the Fermi liquid behaviour that would otherwise be seen.

It is now useful to re-examine the assumptions and approximations we made at the beginning of this investigation. We have chosen to neglect a variety of physical interactions (for example, t_p) for simplicity, and it is not clear, given our omission of any hopping other than V, whether the model implicitly contains t' or t'' (in a square-lattice Hubbard picture) which are known [6] to correspond more closely to experiment than t alone. In fact, we find that these terms *are* present, but are controlled by the nature of the spin state induced by the hole motion. Our model clearly allows Zhang–Rice singlets to hop to nearest neighbours, and one must consider the nature of the ground state to see that t' and t'' are also present. In figure 4, we show how singlets between copper local moments allow Zhang–Rice singlets to hop to second neighbours.

Note well that these hops only occur when there is a singlet in the relevant place in the copper spin background. The calculations in this paper indicate that we expect the ground state of the model to contain such singlets, and hence we predict the existence of an effective t' and t''. The sizes of these parameters are determined by the spin correlations induced by the hole motion in the vicinity of the hole, and can be deduced from a calculation, rather than being imposed.

In conclusion, these results provide preliminary evidence for (a) the breakdown of Fermi liquid theory, (b) the existence of an RVB-like ground state in the three-band model of the high-temperature superconductors, due to correlations induced solely by the motion of a single oxygen hole. This is in direct contrast to the predictions of the t–J model, where the hole motion leads to Nagaoka ferromagnetism, suggesting that the one-band description is inadequate for describing the rich behaviour of the cuprates. Furthermore, we demonstrate that the three-band model we study implicitly contains Hubbard t' and t'' terms due to the nature of the spin background induced by the motion of the hole. This provides some explanation for the need to include t' and t'' that has been noted in the literature.

Since this study has only been of a single hole, we can make no statement about possible superconductivity in the three-band model, and much further investigation is required.

Acknowledgments

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