Photoemission spectra of Sr$_2$CuO$_2$Cl$_2$: a theoretical analysis

A. Nazarenko,$^1$ K.J.E. Vos,$^2$ S. Haas,$^1$ E. Dagotto,$^1$ and R.J. Gooding$^2$

1. Department of Physics and National High Magnetic Field Lab, Florida State University, Tallahassee, FL 32306, USA

2. Department of Physics, Queen’s University, Kingston, Ontario, Canada K7L 3N6

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Abstract

Recent angle resolved photoemission (ARPES) results for the insulating cuprate Sr$_2$CuO$_2$Cl$_2$ have provided the first experimental data which can be directly compared to the (theoretically) well–studied problem of a single hole propagating in an antiferromagnet. The ARPES results reported a small bandwidth, providing evidence for the existence of strong correlations in the cuprates. However, in the same experiment some discrepancies with the familiar 2D $t-J$ model were also observed. Here we discuss a comparison between the ARPES results and the quasiparticle dispersion of both (i) the $t-t'-J$ Hamiltonian and (ii) the three–band Hubbard model in the strong–coupling limit. Both model Hamiltonians show that the experimentally observed one–hole band structure can be approximately reproduced using reasonable values for $t'$, or the direct oxygen hopping amplitude $t_{pp}$.

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The dispersion of the hole quasiparticles in the normal state of the two–dimensional (2D) CuO$_2$ planes is of crucial importance in the search for a microscopic theory of the high temperature superconductors. To be specific, comparison between experimentally observed and theoretically predicted quasiparticle dispersions allows for the scrutiny of Hamiltonians proposed to describe the carriers in these planes. Recently, angle resolved photoemission (ARPES) data \cite{1} have shown that for Bi2212 and YBCO, both near optimal doping, the quasiparticle bandwidth is only $\sim 0.3$eV. In addition, the dispersion contains an interesting flat region around X = ($\pi$, 0) and Y = (0, $\pi$) (in the notation of the 2D square lattice). The apparent universality of these results among different hole-doped compounds implies that their origin must lie in the CuO$_2$ planes. The experimentally observed small bandwidth suggests that strong correlations are crucial for a proper description of carriers in the cuprates. Further, calculations based upon the t $-$ J model \cite{2} have shown that the flat regions near X and Y are naturally obtained for holes moving in an antiferromagnetic (AFM) background, the latter being a direct consequence of the effect of strong correlations at half filling. Then, for doping levels such that the AFM correlation length, $\xi_{AF}$, is robust, the dispersion obtained from the 2D t $-$ J model should approximate the observed quasiparticle dispersion.

In a recent letter, Wells et al. \cite{3} reported ARPES measurements on Sr$_2$CuO$_2$Cl$_2$. This material is an insulating layered copper oxide that is difficult to dope. These results are extremely important because this experiment precisely corresponds to the well–studied theoretical problem of a single hole moving in a quantum antiferromagnet, and thus is a problem that serves as a test case for any proposed microscopic Hamiltonian, as we now discuss. From Ref. \cite{3} the main message for theorists can be summarized in two items: (i) on the one hand, the ARPES spectra shows that the 2D t $-$ J model accurately describes the results along the diagonal (0, 0) to ($\pi$, $\pi$) in momentum space. The state with the highest energy in the valence band is obtained at $\bar{M} = (\pi/2, \pi/2)$ as expected from many calculations \cite{4}. Further, the bandwidth of the quasiparticle is very close to the bandwidth obtained for one hole in the 2D t $-$ J model using various techniques \cite{4,5}. This result is of considerable
importance for theories of high–Tc as it supports the use of one–band models to describe
the electronic properties of the cuprates; (ii) on the other hand, Wells et al. [3] noticed that
the 2D t − J model prediction fails near X = (0, π) since the experiments do not confirm
that the energies of the M and X points are nearly degenerate, as is predicted by t − J model
calculations [4,5].

It is the purpose of this paper to provide evidence that item (ii) above can be naturally
accounted for in two different ways, and we shall discuss each of these ideas separately.
Firstly, if the t − J model is enlarged to contain hole hopping amplitudes at distances larger
than one lattice spacing, one can indeed fit the ARPES data. There is no symmetry con-
straining the hole hopping to just nearest-neighbors, and in several studies it has been shown
that to reproduce the spectra of three–band Hubbard models using one–band t − J models
on small clusters, it is necessary to include a “t′-hopping” along the plaquette diagonals [3].
As long as the ratio |t′/t| is small, this term does not affect the AFM properties of the model
near half-filling, since this hopping only connects sites that are on the same sublattice of a
Néel ordered state. However, such terms will clearly affect the quasiparticle dispersion.

It has, in fact, become standard to use Hamiltonians including t′ hoppings to describe
several compounds. Some authors [7] have argued that to fit the ARPES Fermi surface
of different materials it is necessary to select t′ = −0.2t for La2−xCuxCuO4; t′ = −0.45t for
YBCO; and t′ = +0.2t for Nd2−xCexCuO4, and there seems to be some justification for this
approach. For example, it was recently shown [8] that the differing magnetic properties of
the LaSrCuO and NdCeCuO systems could be accounted for only if such a next–nearest–
neighbour hopping term was included. Since all these compounds have the same CuO2
planes, the strength of these effective “t′-terms” changes among the different cuprates likely
due to three dimensional effects (bi-layer hopping, influence of apical oxygens, influence of
orbitals usually neglected in the 3 and 1 band descriptions of the cuprates, etc.). Then, it is
reasonable to expect that the t − t′ − J model can mimic the dispersion of Sr2CuO2Cl2 with
a properly selected amplitude t′. Although it is clear that fitting data with an arbitrary
parameter is less satisfying than microscopically deducing the value of t′, in the absence of
a microscopic theory to compute $t'$ the best we can do is show that a reasonable value of this amplitude leads to agreement with the experiments. Note that we only have one free parameter to fit an entire function of momentum (dispersion), and thus not much extra freedom is introduced in the model with a $t'$ amplitude.

The $t - t' - J$ model Hamiltonian is defined as

$$
H = -t \sum_{\langle ij \rangle} (\bar{c}_{i\sigma} \bar{c}_{j\sigma} + \text{h.c.}) - t' \sum_{\langle\langle ii'\rangle\rangle} (\bar{c}_{i\sigma}^\dagger \bar{c}_{j\sigma}^\dagger + \text{h.c.}) + J \sum_{\langle ij \rangle} \left( S_i \cdot S_j - \frac{1}{4} n_i n_j \right),
$$

where $\bar{c}$ are hole operators, $\langle \rangle$ refers to n.n. sites, $\langle\langle \rangle\rangle$ refers to n.n.n. sites (along the diagonal of the plaquettes), and the rest of the notation is standard. Using the Born approximation (extensively discussed in the literature [9,5]) the dispersion of one hole in the $t - t' - J$ model was here calculated for several values of $t'$ [10]. To select $t'$ the experimentally observed isotropy of the dispersion of Sr$_2$CuO$_2$Cl$_2$ near the top of the band at ($\pi/2, \pi/2$) [3] was used. We followed this criterion since it is the most clearly defined feature in the data of Ref. [3] (concentrating our effort on the measured behavior near X would be dangerous since for these momenta the ARPES data only provide a bound on the quasiparticle energy). We observed that to reproduce the ($\pi/2, \pi/2$) isotropy, an amplitude $t' \sim -0.35t$ is necessary (supplemented by $J = 0.125eV$ as energy scale and a ratio $J/t$ around 0.3–0.4, as is usually assumed). This value of $t'$ is similar to that found in earlier comparisons based on small cluster exact diagonalization results [8] to the ARPES data [11]. The resulting dispersion is shown in Fig. 1, and is compared with the experimental data. Also shown is the dispersion produced by the $t - J$ model. The agreement near the top of the band is much improved by the inclusion of $t'$, especially in the $(0, \pi) \rightarrow (\pi, 0)$ direction. It is clear that along this direction the $t - J$ and $t - t'(= -0.35t) - J$ models have substantially different dispersion relations. On the other hand, along the $(\pi, 0) \rightarrow (0, 0)$ line our results have more structure than the reported experiments. However, experimentally along this line it is not possible to extract a reliable quasiparticle dispersion (see Fig. 2a of Ref. [3]) and thus we do not consider
this disagreement with states having an energy much less than the chemical potential a serious problem. Actually, it is possible that only the quasiparticles near \( \bar{M} \) are relevant for normal state and superconducting properties [12]. Nevertheless, it is important to fix this quantitative discrepancy, and for this purpose we turn to our second technique for producing a quasiparticle dispersion that agrees with the data of Ref. [8].

The underlying physics of carrier transport in weakly doped CuO\(_2\) planes is governed by oxygen hole motion in an AFM background, and thus one might expect that the three–band Hubbard model in the strong–coupling limit [13] should be employed. However, as is now well known, a renormalization procedure approximately maps the low–energy physics of the three–band Hubbard model into that of the one–band \( t - J \) model [14]. Subsequently, a perturbative calculation [15] prescribed conditions under which further–than–nearest–neighbour hopping terms must be included. As demonstrated above, if one wishes to fit the ARPES data of Ref. [3] at least the next–nearest–neighbour hopping term \( t' \) is required.

One possible caveat to the reduction to one–band models has been discussed in previous papers [14–16]. To be specific, the reduction from the three–band Hubbard model to the \( t - J \) model eliminates the spin degree of freedom of the oxygen carriers, effectively producing spinless vacancies. However, if the direct oxygen–oxygen hopping is large enough, the oxygen hole can acquire a non–zero spin. As a demonstration of the potential inadequacies of any renormalization to a one–band model based on carriers possessing no spin, consider firstly that unlike the \( t - t' - J \) model without the \( t' \) term, holes described by the three–band model moving in a rigid Ising-like AFM are mobile [17], and thus these carriers possess a spin degree of freedom. However, in order to enhance the delocalization of these holes in a quantum AFM, the spins become distorted, and the spin of the carrier is quenched [14,16], approximating the spinless vacancies of the one–band \( t - t' - J \) model. If one then includes a hopping process that is independent of the magnetic background, such as a direct oxygen-oxygen hopping term, and the energy scale of this additional hopping term is larger than any of the other terms, the spin of the carrier is no longer quenched [16], and one should then not expect that such carrier motion (in a strongly correlated magnetic background)
should be describable by the spinless vacancies of the $t - t' - J$ model.

Motivated by this observation, we have examined the usefulness of fitting the experimental results with the microscopic Hamiltonian of Ref. [16]. We have studied the analogue of the three-band $t - J$ model found from the strong coupling limit of the three-band Hubbard model. This is a complicated microscopic Hamiltonian, and we refer the reader to Ref. [16] for a detailed discussion of Eq. (2) of that paper. In addition to this, here we append the Hamiltonian of Ref. [16] with a direct oxygen-oxygen hopping term, something that we found to be crucial to our success in fitting the ARPES data [3]. The parameter values for this Hamiltonian [18] can be estimated from the parameters of the three-band Hubbard model [6], with the Cu–O exchange interaction reduced by a direct exchange term [19]. Then, following the notation of Ref. [16], one finds

$$J_1 = 0.032, \quad J_2 \approx 0.16, \quad t_a = 0.38, \quad t_b = 0.43,$$

and $t_{pp} = -0.65$  \hfill (2)

Extrapolating the analysis of Shastry [15] to include the direct oxygen hopping, and according to the above argument concerning the potential inadequacies of the renormalization to a one-band $t - t' - J$ model, the important ratio of hopping parameters is $t_{pp}/|t_a + t_b| \approx 0.8$. This implies that the renormalization to spinless vacancies may not be entirely justified.

We have used these parameters in an exact diagonalization study of the Hamiltonian of Ref. [16] for a $4 \times 4$ CuO$_2$ cluster with periodic boundary conditions. Our results are shown in Fig. 2. Note that the shape of the quasiparticle dispersion (at least for the small number of wave vectors that we can access) is identical to that of Wells’ data, which is encouraging. However, the bandwidth is roughly half of that found experimentally. This latter result is not that surprising given that for a large $t_{pp}$, important finite-size effects are to be expected [20].

In order to better assess the quasiparticle dispersion relation that describes a single oxygen hole doped into an AFM that follows from Eq. (2) of Ref. [16], we have generated the quasiparticle dispersion using a semiclassical variational wave function that
the spin fluctuations of the Oxygen holes where the distortions of the Cu spins away from AFM order are determined variationally (see Eq. (19) of Ref. [16] for a version of this wave function that does not include Oxygen spin fluctuations). We expect that this semiclassical representation will lead to an excellent approximation to the true quasiparticle dispersion quite simply because the carrier motion has a large direct oxygen hopping. (For example, if one modifies the results of Roth [17] to include $t_{pp}$ and allows for the hopping parameters of Eq. (2) to become fitting parameters, even if one ignores the Oxygen hole spin fluctuations one can again fit the Wells’ data (shape as well as bandwidth) reasonably well — this is also displayed in Fig. 2.)

We have included these spin distortions [21,16], and numerically solved the associated variational problem to obtain the minimum energy state at each wave vector. This calculation has been carried out for an infinite lattice. Here, we have again allowed for the hopping parameters of Eq. (2) (above) to be variables that we use to fit the quasiparticle dispersion found experimentally, expecting that we obtain values close to those of the perturbation theory [16]. Our results, for $t_a \sim 0.0$, $t_b = 0.3$, and $t_{pp} = -0.35$, are shown in Fig. 3. Clearly, the agreement with experiment is quite good, and improves that found using the $t - t' - J$ model along the $(0,0) \to (0,\pi)$ branch. This may point to the importance, at least for these crystal momenta, of the spin degree of freedom of the holes.

Wells et al. [3] made the apparently reasonable assumption of grouping together the results obtained for hole-doped Bi2212 [1], where flat bands at $(0,\pi)$ near the Fermi energy were reported, with those for insulating Sr$_2$CuO$_2$Cl$_2$, concluding that as a function of doping states near $(0,\pi)$ must either be created or move up in energy in a non-rigid way. This is one possible interpretation. However, the many clear differences in the ARPES Fermi surfaces of different high-$T_c$ compounds leads us to believe that it is not correct to group Bi2212 with Sr$_2$CuO$_2$Cl$_2$ since these compounds differ in microscopic details that are relevant for quantitative comparisons — e.g., the numerical value of the $t'$ amplitude. While it is likely that the qualitative mechanism of superconductivity is the same in all hole-doped cuprates, their differences in critical temperature and ARPES data must be caused by non-universal
features. Thus, in our opinion, the data from Bi2212 and Sr$_2$CuO$_2$Cl$_2$ cannot be grouped together to obtain conclusions about the behavior of correlated electrons as a function of doping in the cuprates. To clarify this important point it will be necessary to carry out photoemission experiments on the same compound at different densities of carriers.

Summarizing, using a $t - t' - J$ model it is possible to fit the ARPES quasiparticle dispersion data for Sr$_2$CuO$_2$Cl$_2$ reasonably well at the top of the valence band. To obtain an even better quantitative agreement the use of a strong-coupling three-band Hubbard model is required [16].

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**Figure Captions**

1. Quasiparticle dispersion of the $t - t' - J$ model calculated in the Born approximation for an infinite lattice using $t' = -0.35t$ (solid line), $J/t = 0.3$ and $J = 0.125$eV, compared against the experimental ARPES data (open circles) for Sr$_2$CuO$_2$Cl$_2$ of Ref. [3]. The dashed line is the $t - J$ model result of Ref. [5].

2. Quasiparticle energies (stars) for an oxygen hole described by the effective Hamiltonian of Eq. (2) of Ref. [10] found from an exact diagonalization study on a $4 \times 4$ CuO$_2$ cluster, compared against the Sr$_2$CuO$_2$Cl$_2$ ARPES data (open circles); the zero of energy of the exact diagonalization numbers is offset for clarity. Also, the solid line denotes the band of energies for hole motion in a rigid (viz., Ising like) AFM background using hopping parameters $t_a = 0.12$, $t_b = 0.17$, and $t_{pp} = -0.18$.

3. Quasiparticle energies (solid diamonds) of an oxygen hole in the effective Hamiltonian of Eq. (2) of Ref. [10] for an infinite lattice using a semiclassical variational wave function that includes the spin fluctuations of the Oxygen carriers. The quasiparticle dispersion measured in Ref. [3] is also shown (open circles). The hopping parameters that produce this quasiparticle dispersion are $t_a = 0.0$, $t_b = 0.3$, and $t_{pp} = -0.35$. 
