Generalized one-band model based on Zhang-Rice singlets for Tetragonal CuO

I. J. Hamad and L. O. Manuel
Instituto de Física Rosario (CONICET) and Universidad Nacional de Rosario, Bv. 27 de Febrero 210 bis, 2000 Rosario, Argentina

A. A. Aligia
Centro Atómico Bariloche and Instituto Balseiro, CNEA, CONICET, 8400 Bariloche, Argentina

Tetragonal CuO (T-CuO) has attracted attention because of its structure similar to that of the cuprates. It has been recently proposed as a compound whose study can give an end to the long debate about the proper microscopic modeling for cuprates. In this work, we rigorously derive an effective one-band generalized $t-J$ model for T-CuO, based on orthogonalized Zhang-Rice singlets, and make an estimative calculation of its parameters, based on previous ab initio calculations. By means of the self-consistent Born approximation, we then evaluate the spectral function and the quasiparticle dispersion for a single hole doped in antiferromagnetically ordered half-filled T-CuO. Our predictions show very good agreement with angle-resolved photoemission spectra and with theoretical multiband results. We conclude that a generalized $t-J$ model remains the minimal Hamiltonian for a correct description of single-hole dynamics in cuprates.

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More than three decades after their discovery, high temperature superconductors still give rise to many debates. On the theoretical side, one of the most long-standing and important discussions is about the proper microscopic model for describing superconductivity. In this respect and from the outset, attention was focused on the spectral function of a single-hole doped on the parent half-filled compounds, whose quasiparticle (QP) dispersion relation is directly measured in angle-resolved photoemission (ARPES) experiments. Experimental evidence shows that this doped hole resides on the O 2p orbitals \([1] \text{ and } [2]\). For the CuO planes that build up the cuprates, Zhang and Rice \([4]\) proposed that a singlet, called Zhang-Rice (ZR) singlet, is formed between the spin of a cooper atom and the spin of the hole residing in a linear combination of four ligand oxygen orbitals around the cooper atom. Integrating out the oxygen orbitals, a one-band effective model was proposed in which the effective holes (representing ZR singlets) reside on the cooper atoms and propagate emitting spin excitations, magnons. In this model, adding two holes as nearest-neighbors in an antiferromagnetic background costs less energy than if they are added far apart. This is a simplified view of the pairing glue of magnetic origin \([22]\).

Since the proposal of Zhang and Rice, an unclosed debate about the validity of one-band effective models has taken place \([3] \text{ and } [4]\). Several authors sustain that only the three-band model \([1] \text{ and } [2]\) is valid for describing the physics of the cuprates correctly, where the three bands come from two O 2p orbitals and one Cu 3d\(_{x^2−y^2}\) orbital, not only for the insulating parent compound at half-filling, but also for many other phases of the rich phase diagram of the cuprates and related compounds. This issue is of central importance since many investigations have been done in one-band models and hence their validity is, at least partially, questioned.

Recently, tetragonal CuO (T-CuO) has been synthesized, by growing epitaxially CuO planes on a substrate \([001] \text{ SrTiO}_3\) \([13]\). T-CuO can be considered as two interpenetrating CuO\(_2\) sublattices sharing one oxygen atom and hence has two degenerate antiferromagnetic ground states, as shown in Fig. 3. ARPES experiments were performed on this compound \([20]\), showing substantial intralayer coupling between these two sublattices and a similar dispersion (with some differences) to that of the cuprate \({\text{Sr}_2\text{Cu}_2\text{O}_2\text{Cl}_2}\). This material was addressed in a recent work \([15]\) as a good candidate to discern whether one-band models, based on ZR singlets, are valid for describing the physics of CuO planes or if, instead, three-band models should be used.

In this Letter, we rigorously derive an effective one-band model for T-CuO and compare its QP dispersion with experimental ARPES results and theoretical predictions for the three-band model. Using a procedure based on previous derivations of generalized one-band effective Hamiltonians \([10]\), we start from a spin-fermion model for T-CuO and we obtain then its effective one-band model for the ZR singlets. The parameters of the model were calculated starting from parameters determined by constrained-density-functional computations for \({\text{La}_2\text{CuO}_4}\) \([17]\) and estimating their variations for the T-CuO case. We find an effective hopping to first nearest neighbors (NN) between CuO\(_2\) sublattices, and effective hoppings to first, second, and third NN in the same sublattice, together with superexchange parameters \(J\) (the usual NN antiferromagnetic one for CuO\(_2\) planes) and a ferromagnetic \(J'\) (NN in T-CuO, belonging to different CuO\(_2\) sublattices).

Using this model, we calculate the QP dispersion by means of the self-consistent Born approximation.
The coordinate versors point in the directions of $c$ of phases (Fig. S1 of Ref. 23) the Hamiltonian reads

$$H_{1J}^s = -\sum_{\kappa=0}^3 t_{\kappa} \sum_{iv_{\kappa}\sigma} \left( c_{iv_{\kappa}\sigma}^\dagger c_{iv_{\kappa+1}\sigma} + \text{H.c.} \right) +$$
$$+ \frac{J}{2} \sum_{iv_{\kappa}} S_i \cdot S_{i+\kappa},$$  \hspace{1cm} (2)

where the subscript $\kappa = 0$ refers to intersublattice hopping of NN Cu atoms in the T-CuO structure, while $\kappa = 1, 2, 3$, refer to first, second, and third NN within each CuO$_2$ sublattice, respectively. Instead of using arbitrary values for the parameters, we have calculated them, keeping the states corresponding to orthogonalized ZR singlets and using results from constrained-density-functional calculations [15]. These values are very similar to those corresponding to the model used by Adolphs et al. [13], as shown in Table 3 of Ref. 23. We have checked that the results for both sets are quite similar. To simplify the discussion we present here only the results for the latter. The parameters in meV are $t_0 = -184$, $t_1 = 369$, $t_2 = -11$, $t_3 = 65$, $J = 150$, and $J' = 0$. This effective model was proposed previously by Moser et al. [20]. Here we provide its justification and determine its parameters.

The spectral functions were calculated by means of the SCBA [32–35], a semianalytic method that has been proven to compare very well with exact diagonalization (ED) results on finite clusters in different systems [32, 36, 37, 39, 41]. It is one of the more reliable and checked methods up to date to calculate the hole Green’s function, and in particular itsQP dispersion relation. However, some care is needed to map the QP weight between different models [37]. In order to do such calculation, we follow standard procedures [39]. On one hand the magnetic dispersion relation is obtained treating the magnetic part of the Hamiltonian at the linear spin-wave level, since the system we study has long-range order, and hence its magnetic excitations are semiclassical magnons. On the other hand, the electron creation and annihilation operators in the hopping terms are mapped into holons of a slave-fermion representation (details in Ref. 23).

Within SCBA, we arrive to an effective Hamiltonian:

$$H_{\text{eff}} = \sum_k c_k h_k^\dagger h_k + \sum_k \omega_k \theta_k^\dagger \theta_k +$$
$$+ \frac{1}{\sqrt{N}} \sum_{kq} \left( M_{kq} h_k^\dagger h_{k-q} + \text{H.c.} \right),$$  \hspace{1cm} (3)

$$c_k = 2t_0 \cos(k \cdot c) + 4t_2 \cos(ak_x) \cos(ak_y) + 2t_3 \cos(2ak_x) + \cos(2ak_y),$$
$$\omega_k = \sqrt{A_k^2 - 4B_k^2},$$
$$M_{kq} = 2t_0 \{ \cos[ (k - q) \cdot c] u_q - \cos( k \cdot c) u_q \} + 2t_1 \{ u_q \xi(k - q) - v_q \xi(k) \},$$  \hspace{1cm} (4)
where $\epsilon_k$ is the bare hole dispersion (with no coupling to magnons), $\omega_k$ is the magnon dispersion relation, with $A_k = 2J - J' \cos(c \cdot k)$, $B_k = \frac{4}{\pi} \sum v_i \cos(v_1 \cdot k) - \frac{J}{2} \cos(d \cdot k)$, and $M_{k\ell}$ is the vertex that couples the hole with magnons. Here $\zeta(k) = \cos(ak_x) + \cos(ak_y)$, and $c = b(x + y)$, $d = b(-x + y)$, being $a = 2b$ the distance between Cu atoms in the CuO$_2$ planes. The vectors $c$ and $d$ are indicated in Fig. 3. We now compare our results with ARPES experiments performed on T-CuO, specifically with the those in Figs. 2 and 3 of Ref. [20]. A broadening equivalent to 20 meV was applied to the spectral functions (see text).

The experimental dispersion, and hence our results can distinguish between the possible degenerate magnetic orders by the ARPES matrix elements. In the previous paragraph, it is worth to mention that, on one hand, at some points in the experimental curve the effect of the ARPES matrix elements is very strong, especially around the $X/M'$ point, where there is no intensity at all in the ARPES data, and on the other hand a $\beta$ band seems to merge with the QP band, specially at the $X/M'$ point but also possibly around the $\Gamma$ point. For this calculation a broadening equivalent to 170 meV was applied, in order to make the intensity plot softer. The results are shown in Fig. 3. The similarities with the experimental curve follow the trends explained in the previous paragraph. It is worth to mention that, on one hand, at some points in the experimental curve the effect of the ARPES matrix elements is very strong, especially around the $X/M'$ point, where there is no intensity at all in the ARPES data, and on the other hand a $\beta$ band seems to merge with the QP band, specially at the $X/M'$ point but also possibly around the $M$ point. So at these two points, in particular around the $X/M'$ points, the comparison of our calculation with the experiment is obscured by these experimental facts. Finally, it is worth to mention that in the case that the illuminated area in the ARPES experiments contains domains with both magnetic $Q = (\pi, 0)$ and $Q = (0, \pi)$ vectors (as mentioned above, they are degenerate), the QP dispersion should be a superposition of both curves shown in Fig. 2 which does not seem to be what is observed in the experiment.

![Figure 2](image_url)

**FIG. 2:** (Color online) Quasiparticle dispersion relation (relative to $\Gamma$) along the path marked in the inset, the same as the one measured in the ARPE experiment in Ref. [20]. A broadening equivalent to 20 meV was applied to the spectral functions (see text).

![Figure 3](image_url)

**FIG. 3:** (Color online) SCBA intensity map along the same path as in Fig. 2. The assumed magnetic order is $(\pi, 0)$. The agreement is very good, except in the last case. This discrepancy is quite likely due to missing quasiparticle peaks with small weight in the experiment (see also Fig. S4 of Ref. [22]). In that sense, we note that the $\Gamma$ point (and points located in its vicinity) shows a very broad spectrum (see Figs. 2 and 3 in Ref. [20]), and hence there may be some uncertainty in the determination of the QP energy which could explain this discrepancy. The bandwidth of the QP dispersion, along this path, taken from our SCBA calculation is 0.3 eV, very similar to the bandwidth of the experimental dispersion, approximately 0.4 eV. We have also calculated an intensity curve along...
FIG. 4: (Color online) SCBA hole’s dispersion relation in units of $J$ (0.15 eV) along the path marked in the inset. Black dashed line: result corresponding to $t_0 = 0$ (decoupled sublattices). Blue full line: full result with $t_0 = −184$ meV.

the experiment [42]. The intensity curve Fig. 3 should also change accordingly, but in our case we have checked that the only noticeable changes occur around the $X/M$ point, at which nevertheless there is no intensity in the ARPES data corresponding to the band ascribed to ZR singlets [20].

In general, the spectral function corresponding to a definite momentum contains, in the hole picture, a low energy pole, whose energy defines the QP energy, and a high energy part which is related to the incoherent movement of the hole, having its origin in multimagnon processes [32]. When the quasiparticle weight is significant, the brighter areas in Fig. 3 will coincide with the energy of the QP in Fig. 2. On the contrary when the incoherent part of the spectral function takes most of the spectral weight, this will not happen. Points like $\Gamma$ and $\Delta$ have low QP weight, while on the contrary for the lines $B−B′$, $A′−A$ the QP weight is relatively high (some spectral functions can be seen in Fig S4 of ref. 23).

It was claimed previously that the one-hole dispersion in T-CuO requires a three-band model to be described correctly [13]. The evidence presented came from a variational calculation on the spin-fermion model Eq. (2), whose results a one-band model supposedly cannot capture. In particular, it was shown that the minimum that the QP dispersion has at $(\pi/2, \pi/2)$ for CuO$_2$ (or, equivalently in T-CuO, if the two CuO$_2$ sublattices are disconnected), shifts along the diagonal $\Gamma \equiv (0,0) − (\pi, \pi)$, towards the $\Gamma$ point, when the two CuO$_2$ sublattices are connected to form T-CuO. This happens for $Q = (0,0)$. Alternatively, the shift is along the antidiagonal towards $X/M$ for $Q = (\pi,0)$. This is what we have shown in figure 2. These results are in line with previous investigations for CuO$_2$ planes [15], where it was claimed that a one-band $t−t′−t''−J$ model has a minimum at $(\pi/2, \pi/2)$ that along the diagonal of the Brillouin zone is controlled by spin fluctuations, while in the three-band model the variational method used in Ref. 10 does not need to include spin fluctuations in order to have an absolute minimum at $(\pi/2, \pi/2)$.

Using the generalized $t−J$ model [Eq. (37)] derived from $H_{sf}$ [Eq. (2)] we now calculate the QP dispersion along the same path as in Ref. 15 and with the corresponding parameters (set B of Table III of Ref. 23), and $Q = (0, \pi)$. Results are shown in Fig. 4 plotted adopting the hole’s picture. As before, a broadening equivalent to 20 meV was applied to the spectral functions, but the results do not depend significantly on this (unless broadenings an order of magnitude larger are applied). It is clear that when both sublattices are connected through the $t_0$ term, the QP dispersion relation derived from $H_{sf}$ is recovered. In particular, we obtain a shift of the QP minimum along the diagonal towards the $\Gamma$ point, although this shift is lower (about half) in magnitude than the one obtained with the three-band model. This difference might be due to the different theoretical treatments used by Adolphs et al. to solve $H_{sf}$ [Eq. (2)] and by us to solve $H_{sf}$ [Eq. (37)]. In this respect, we remark it is very difficult to decide which theoretical treatment gives more accurate results from quantitative differences of this kind, since on one hand both compare very well with ED results in finite clusters, while on the other hand no experiment so far could even measure this shift in the QP dispersion relation. We also remark that varying $t_2$, the QP dispersion relation is not changed apart from a constant shift (in agreement with previous results [16]). This is important since $t_2$ is the parameter obtained with less accuracy.

The shift in our model is not caused by the coupling of the hole with spin fluctuations, which in fact conspires against it. This can be seen from the effective Hamiltonian Eq. (4), since the bare-hole dispersion (i.e. with no coupling to magnons) $\epsilon_k = 2t_0 \cos(k \cdot c) + 4t_2 \cos(ak_x) \cos(ak_y) + 2t_3 [\cos(2ak_x) + \cos(2ak_y)]$ has a minimum, along the diagonal $k_x = k_y$, that shifts from $(\pi/2, π/2)$ towards the $\Gamma$ point when the intersublattice hopping $t_0$ is turned on. For example, the bare hole minimum is at $(0.4\pi, 0.4\pi)$ for the parameter set we used. However, when the interaction of the bare hole with spin fluctuations (magnons) is taken into account through the vertex $M_{kq}$, the minimum shifts back towards $(\pi/2, \pi/2)$. The shift obtained is about 10% of the distance between...
the $A$ and $\Gamma$ points. Note that the SCBA contains an infinite number of spin fluctuations while only a few are included in the treatment of Ref. [12]. In any case, we have shown that a ZR one-band model can explain a shift in the QP minimum at ($\pi/2, \pi/2$), and that the interaction of the bare hole with spin fluctuations is not responsible for this shift. Finally, the QP bandwidth along this path is, in our one-band model, of the order of $3J$, slightly less than the result from the variational method in the three-band model Eq. [2] [15].

Overall, we conclude that our effective generalized one-band model, rigorously derived from orthogonalized Zhang-Rice singlets, and without free parameters, not only does recover characteristics of the three-band model, but also its predictions agree qualitatively and quantitatively with ARPES experiments in tetragonal CuO.

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* Electronic address: aligia@cab.cnea.gov.ar

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[42] Note that in the exact diagonalization of the QP dispersion, unless some small anisotropy is put by hand, the two magnetic degenerate ground states coexist in the calculation and, hence, the results should reflect this fact.
Supplemental Material: Generalized one-band model based on Zhang-Rice singlets for Tetragonal CuO

I. J. Hamad, L. O. Manuel, A. A. Aligia

THE STARTING MODEL

It is known that for energies below 1 eV, the physics of the superconducting cuprates is described by the three-band Hubbard model $H_{3b}$, which contains the $3d_{x^2-y^2}$ orbitals of Cu and the $2p_\sigma$ orbitals of O \cite{1-3}. We denote by $\hat{\mathbf{x}}$ and $\hat{\mathbf{y}}$ the unit vectors along the directions of nearest-neighbor (NN) Cu atoms in the CuO$_2$ planes (which are second NN in the T-CuO structure) and $a$ their distance. Experimental evidence about the symmetry of holes in cuprate superconductors \cite{5-7} shows that the undoped system has one hole in each $3d_{x^2-y^2}$ orbital, so that all Cu atoms are in the oxidation state 2+, while added holes enter the O $2p_\sigma$ orbitals. Therefore, it is natural to eliminate the Cu-O hopping $t_{pd}$ and the states with Cu$^{+}$ and Cu$^{+3}$ (keeping them as virtual states) by means of a canonical transformation \cite{8,9}. The resulting effective Hamiltonian, which consists of Cu 1/2 spins and O holes is usually called spin-fermion model. As usual, we change the phases of half the Cu and O orbitals so that the Cu-O hopping has the same sign independent of direction (see Fig. 1)

$$d_{i\sigma} \rightarrow e^{iQ \cdot (\mathbf{R}_i - \mathbf{R}_0)} d_{i\sigma}, \quad p_{j\sigma} \rightarrow e^{iQ \cdot (\mathbf{R}_j - \mathbf{R}_0)} p_{j\sigma}, \quad Q = \frac{\pi}{a} (\hat{x} - \hat{y}),$$ (1)

where $\mathbf{R}_i^0$ ($\mathbf{R}_j^0$) is a fixed Cu (O) position. After this transformation, for one hole added to the undoped system, the model can be written as \cite{9,10}

$$H_{sf}^P = \sum_{i\delta\delta'\sigma} p_{i+\delta'\sigma}^\dagger p_{i+\delta\sigma} \left[ (t_1^{sf} + t_2^{sf})(\frac{1}{2} + 2\mathbf{S}_i \cdot \mathbf{s}_{i+\delta}) - t_2^{sf} \right] - J_d \sum_{i\delta} \mathbf{S}_i \cdot \mathbf{s}_{i+\delta} - t_{pp} \sum_{j\gamma\sigma} p_{j+\gamma\sigma}^\dagger p_{j\sigma} + \frac{J}{2} \sum_{i\delta} \mathbf{S}_i \cdot \mathbf{S}_{i+2\delta}. \quad (2)$$

Here $i$ ($j$) labels the Cu (O) sites and $i + \delta$ ($j + \gamma$) label the four O atoms nearest to Cu atom $i$ (O atom $j$). The spin at the Cu site $i$ (O orbital 2p$_\sigma$ at site $i + \delta$) is denoted as $\mathbf{s}_i$ ($\mathbf{s}_{i+\delta}$). The first term corresponds to an effective O-O hopping with possible spin flip with a Cu spin, $t_1^{sf}$ ($t_2^{sf}$) correspond to virtual processes through Cu$^+$ (Cu$^{+3}$). When both NN vectors coincide ($\delta = \delta'$), the second term contains a term of the form of the second one. The total Cu-O NN exchange is $J_K = 2(t_1^{sf} + t_2^{sf}) - J_d$. In second-order perturbation theory, $J_d$ vanishes if the on-site O repulsion is neglected \cite{8,9}, but in general $J_d > 0$. The third term is the direct O-O hopping and the last one is the exchange between nearest Cu atoms.

FIG. 1: Scheme of the $3d_{x^2-y^2}$ (squares) and $2p_\sigma$ (number 8) orbitals of the T-CuO planes. Blue and red orbitals belong to different CuO$_2$ sublattices. Empty and filled parts of the orbitals have opposite signs.
It has been shown that $H_{sf}^p$ with parameters slightly renormalized to fit the energy levels of a CuO$_4$ cluster in some symmetry sectors (solving small matrices) reproduces Cu and O photoemission and inverse photoemission spectra and spin-spin correlations functions of the three band model $H_{3b}$ in a Cu$_2$O$_8$ cluster [10]. This was later extended to angle-resolved Cu and O photoemission intensities [11]. Therefore we assume that $H_{sf}^p$ is an accurate representation of the low-energy physics of $H_{3b}$.

The system of tetragonal CuO (T-CuO) consists of two interpenetrating CuO$_2$ sublattices, one displaced with respect to the other in a vector $\gamma$ connecting two NN O ions (see Fig. 1). One of the sublattices can be described by Eq. (2). The O orbitals of the other sublattice lie on the same site as the previous ones but are orthogonal to them. We label $i'$ the Cu orbitals of the second sublattice and $q_{i'\sigma\delta}$ the annihilation operators of the four O 2p$_{\sigma}$ orbitals nearest to Cu site $i$. The Hamiltonian that describes the second sublattice $H_{sf}^q$, has the same form as $H_{sf}^p$ with $i$ replaced by $i'$ and the O $p$ operators by the $q$ ones. Including the NN O-O hopping and the NN Cu-Cu exchange between both sublattices, the Hamiltonian reads

$$H_{sf} = H_{sf}^p + H_{sf}^q + t'_{pp} \sum_{\gamma \sigma} s_{\gamma} \left( p_{i\gamma+\sigma}^\dagger q_{j\sigma} + \text{H.c.} \right) - \frac{J'}{2} \sum_{i\gamma} S_{i\gamma} \cdot S_{i+\gamma},$$

(3)

where $s_{\gamma} = -1$ for $\gamma \parallel \hat{x} + \hat{y}$ and $s_{\gamma} = 1$ in the perpendicular direction (see Fig. 1) and $t'_{pp} \approx 0.6t_{pp}$ [12]. This is essentially the same Hamiltonian as that considered by Adolphs et al [12]. The last term is originated by perturbation theory in fourth order in the Cu-O hopping $t_{pd}$ involving two O atoms, each one forming a Cu-O-Cu angle of 90 degrees, and virtual states with an O occupied by two holes in perpendicular orbitals (one $p_{j\sigma}$ and one $q_{j\sigma'}$). It is ferromagnetic due to the Hund rules at the O atoms. Estimating the difference between singlet and triplet two-hole states from that between $^1$D and $^3$P states in atomic O (1.97 eV [14]) and taking the rest of the parameters from constrained-density-functional calculations for La$_2$CuO$_4$ (Ref. 13) we obtain $J' = 2.7$ meV. This value is very sensitive to the Cu-O charge transfer energy $\Delta$ and to the Coulomb repulsion $U_{pd}$ between Cu and O. For example changing $U_{pd}$ from 1.2 eV to 0, $J'$ increases to 34 meV.

In Table I we show an estimation of the parameters of $H_{sf}$ based on previous results [11] of the low-energy reduction procedure from $H_{3b}$ with parameters derived from constrained-density-functional calculations (set A) and the parameters used by Adolphs et al [12] (set B).

Since the structure of T-CuO is different from that of the cuprates, the estimation of the parameters is very approximate. It would be desirable to have estimations for the parameters of $H_{3b}$ for T-CuO, in particular the charge-transfer energy $\Delta$. In absence of them one can estimate the hopping terms taking into account that the CuO distance is increased from the value $b = a/2 = 1.895$ Å used in Ref. [13] to $b = 1.9525$ Å in T-CuO [10], using the scaling $t_{pd} \propto d^{-7/2}$ and $t_{pp} \propto d^{-2}$ for the dependence on the distance $d$ of the hopping parameters [18]. This leads to a reduction of $t_{pp}$ by a factor 0.94 and using that for small $t_{pd}$, $t_{sf} \propto t_{pd}^2$ a reduction of these effective hoppings by a factor 0.81 might be expected, neglecting the influence of the change in on-site energies and repulsions.

**TABLE I: Parameters of the spin-fermion model for T-CuO in eV.**

| set | $t^f_1$ | $t^f_2$ | $J_d$ | $t_{pp}$ | $t'_{pp}$ | $J$ | $J'$ |
|-----|--------|--------|------|--------|---------|-----|-----|
| A   | 0.37   | 0.08   | 0.28 | 0.56   | 0.336   | 0.13| 0.0027|
| B   | 0.45   | 0        | 0.48 | 0.615  | 0.369   | 0.15| 0     |

**THE GENERALIZED $t - J$ MODEL FOR CUO$_2$ PLANES.**

Zhang and Rice proposed that the low-energy physics of the cuprates is dominated by the now called Zhang-Rice singlets (ZRS) [19]. In the language of $H_{sf}^p$, for which fluctuations via Cu$^+$ and Cu$^{+3}$ are included virtually, for each Cu site $i$ these singlets have the form

$$|i\bar{s}\rangle = \frac{1}{\sqrt{2}} \left( \tilde{\pi}_{i\uparrow} d_{i\downarrow}^\dagger - \tilde{\pi}_{i\downarrow} d_{i\uparrow}^\dagger \right) |0\rangle,$$

(4)

$$\tilde{\pi}_{i\sigma} = \frac{1}{2} \sum_{\delta} p_{i+\delta\sigma},$$

(5)
where $d^\dagger_{i\sigma}$ creates a hole at the 3d$_{x^2-y^2}$ orbital of site $i$. Retaining only ZRS and neglecting the rest of the states (or including them perturbatively) and mapping these states $|i\tilde{s}\rangle \leftrightarrow |i0\rangle$ to the vacuum at site $i$ (which corresponds to a full 3d shell) leads to a one-band generalized $t-J$ model. Several systematic studies of this mapping were made starting for either $H_{3d}$ or $H_{sf}^p$, which include more terms than just the NN hopping $t$ and the exchange $J$. See for example Refs. [20–22]. A difficulty with the states $|i\tilde{s}\rangle$ is that they have a finite overlap for NN Cu sites $i$ and $i+\delta$. Using these non-orthogonal singlets Zhang proved that the mapping from $H_{sf}^p$ to the $t-J$ model is exact for $t_1^{sf} = t_{pp} = 0$. [23] This procedure was generalized to include the other terms of $H_{sf}^p$, leading to additional terms in the generalized $t-J$ model [20].

However, orthogonalizing the states leads to a simpler mapping procedure which is in general preferred and is more accurate when $t_1^{sf} > t_2^{sf}$ (fluctuations via Cu$^+$ dominate) [20], which is in general the case. The trick to obtain orthonormal states is to transform Fourier the $\tilde{\pi}_{i\sigma}$ operators, normalize in wave-vector space, and transform back [19], leading to

$$\pi_{i\sigma} = \frac{1}{N} \sum_k e^{-i\mathbf{k} \cdot \mathbf{R}_i} \beta_k \sum_m e^{i\mathbf{k} \cdot \mathbf{R}_m} \tilde{\pi}_{m\sigma}, \quad \beta_k = [\cos^2(k_x b) + \cos^2(k_y b)]^{-1/2},$$

(6)

where $\mathbf{R}_i$ is the two-dimensional position of the Cu site $i$ and $b = a/2$, where $a$ is the lattice parameter of the CuO$_2$ planes. The new operators $\pi_{i\sigma}$ satisfy canonical anticommutation rules. The mapping is now different:

$$|i0\rangle \leftrightarrow |i\tilde{s}\rangle = \frac{1}{\sqrt{2}} \left( \pi_{i\uparrow}^\dagger d_{i\downarrow}^\dagger - \pi_{i\downarrow}^\dagger d_{i\uparrow}^\dagger \right) |0\rangle.$$  

(7)

Inverting Eq. (6), one has for the two $2p_{\sigma}$ O orbitals per unit cell

$$p_{i+b\delta\sigma} = \frac{1}{N} \sum_k \beta_k e^{-i\mathbf{k} \cdot \mathbf{R}_i} e^{-i\mathbf{k} \cdot \mathbf{b}} \sum_m e^{i\mathbf{k} \cdot \mathbf{R}_m} \left[ \cos(k_x b) \pi_{m\sigma} + \cos(k_y b) \gamma_{m\sigma} \right],$$

$$p_{i+b\delta'\sigma} = \frac{1}{N} \sum_k \beta_k e^{-i\mathbf{k} \cdot \mathbf{R}_i} e^{-i\mathbf{k} \cdot \mathbf{b}} \sum_m e^{i\mathbf{k} \cdot \mathbf{R}_m} \left[ \cos(k_y b) \pi_{m\sigma} - \cos(k_x b) \gamma_{m\sigma} \right],$$

(8)

where the $\gamma_{m\sigma}$ correspond the so called non-bonding O orbitals which do not mix with the Cu 3d$_{x^2-y^2}$ orbitals by symmetry. They are defined asking that the Fourier transforms $\tilde{\pi}_{k\sigma}$ and $\gamma_{k\sigma}$ anticommute. In any case we neglect these non-bonding orbitals in what follows.

Using Eqs. (5) and (8) one can write

$$\tilde{\pi}_{i\sigma} = \sum_m \lambda(\mathbf{R}_m) \pi_{i+m\sigma},$$

(9)

$$\lambda(\mathbf{R}_m) = \frac{1}{N} \sum_k \left[ \cos^2(k_x b) + \cos^2(k_y b) \right]^{1/2} \cos(\mathbf{k} \cdot \mathbf{R}_m) =$$

$$= \frac{1}{N} \sum_k \left[ 1 + (\cos(k_x a) + \cos(k_y a)) / 2 \right]^{1/2} \cos(k_x x_m) \cos(k_y y_m).$$

(10)

As expected, the sum in Eq. (9) is dominated by $\lambda(\mathbf{R}_m) \simeq 0.96$ and the other terms decrease rapidly with distance (see Table I).

The part independent of spin of the first term in Eq. (2) is

$$\frac{1}{2} (t_1^{sf} - t_2^{sf}) \sum_{i\delta\delta'\delta} p_{i+b\delta\sigma} p_{i+b\delta'\sigma} = 2(t_1^{sf} - t_2^{sf}) \sum_{i\sigma} \tilde{\pi}_{i\sigma}^\dagger \tilde{\pi}_{i\sigma} = 2(t_1^{sf} - t_2^{sf}) \sum_{i\sigma} \nu(\mathbf{R}_i) \pi_{i+\delta\sigma}^\dagger \pi_{i\sigma},$$

(11)

where using Eqs. (9), (10) and symmetry

$$\nu(\mathbf{R}_i) = \sum_m \lambda(\mathbf{R}_i + \mathbf{R}_m) \lambda(-\mathbf{R}_m) = \frac{1}{N^2} \sum_{kq} (\beta_k \beta_q) e^{-i\mathbf{k} \cdot \mathbf{R}_i} e^{i\mathbf{q} \cdot \mathbf{R}_m} =$$

$$= \frac{1}{N} \sum_k (\beta_k)^2 e^{-i\mathbf{k} \cdot \mathbf{R}_i} = \frac{1}{N} \sum_k \left[ 1 + (\cos(k_x a) + \cos(k_y a)) / 2 \right] \cos(k_x x_i) \cos(k_y y_i).$$

(12)
It is easy to see that \( \nu(0) = 1 \) (contributing to a constant energy of the \( \pi \) orbitals which we drop), \( \nu(a\hat{x}) = \nu(a\hat{y}) = 1/4 \), and other \( \nu(R_i) = 0 \). Calculating the matrix element \( \langle B|\pi_{j}^{\pi}\pi_{i}\rangle = -1/2 \), where \( |A\rangle = d_{ij}^\dagger |is\rangle \) and \( |B\rangle = d_{il}^\dagger |js\rangle \), one realizes that the mapping Eq. 7 leads to

\[
P\pi_{j}^{\pi}\pi_{i}\pi_{i}P \leftrightarrow -d_{il}^\dagger d_{ij}/2, \tag{13}
\]

for the corresponding operators, and the same interchanging spin up and down, where \( P \) is the projector on the low-energy subspace of Zhang-Rice singlets (LESZRS). Thus, the spin independent part of the first term in Eq. 2 provides a contribution

\[
-\frac{1}{4}(t_1^{sf} - t_2^{sf}) \sum_{i,s} d_{i+s\sigma}^\dagger d_{i\sigma}, \tag{14}
\]

to the NN hopping of the one-band model.

The spin dependent part of first term in Eq. 2 is

\[
(t_1^{sf} + t_2^{sf}) \sum_{i,s,s'} p_{i+s\sigma}^\dagger p_{i\sigma} \sigma_{s's} \cdot S_i = 4 \sum_{i,s} \pi_{ls}^{\pi} \pi_{ls}^{\pi} \sigma_{s's} \cdot S_i, \tag{15}
\]

where \( \sigma_{s's} \) are the matrix elements between spins \( s' \) and \( s \) of a vector constructed from the three Pauli matrices. Replacing Eq. 9 in Eq. 15 one obtains several terms. Note that for at most one added hole in the system \( \sum_{s,s'} \pi_{ls}^{\pi} \pi_{ms} \sigma_{s's} \cdot S_i = 2 \sum_{s} \pi_{ls}^{\pi} \pi_{ms} S_m \cdot S_i = 2s \cdot S_i \sum_{s} \pi_{ls}^{\pi} \pi_{ms} \), where \( s_l = \sum_{s,s'} \pi_{ls}^{\pi} \pi_{ls}^{\pi} \sigma_{s's}/2 \) is the spin of the Wannier function \( \pi \) at site \( l \). Then if either \( i = l \) or \( i = m \), projection of this term in the LESZRS Eq. 7 leads to \( s_i \cdot S_i = -3/4 \), and this term reduces to a hopping. Using \( \lambda(-R_m) = \lambda(R_m) \) and neglecting as before the on-site energy correction one obtains for the sum of all terms of this form

\[
6(t_1^{sf} + t_2^{sf}) \sum_{i,m} \lambda(R_m)\lambda(0)d_{l+m\sigma}^\dagger d_{i\sigma}. \tag{16}
\]

For the rest of the terms one can use \( s^z_m = -S^z_m \) in the LESZRS, Eq. 13 and the mappings

\[
P\pi_{j}^{\pi}\pi_{i}P \leftrightarrow d_{il}^\dagger d_{ij}^\dagger S^+_{j}/2, \quad P\pi_{j}^{\pi}\pi_{i}P \leftrightarrow d_{il}^\dagger d_{ij}/2 = d_{il}^\dagger d_{j\bar{i}} S^-_{j}/2, \tag{17}
\]

leading to the following three-site terms

\[
4(t_1^{sf} + t_2^{sf}) \sum_{l \neq i \neq m} \lambda(R_l - R_i)\lambda(R_m - R_i)d_{l\sigma}^\dagger d_{m\sigma} S_i \cdot S_m. \tag{18}
\]

Using Eqs. 5 and neglecting non-bonding states absent in the LESZRS, the second term of Eq. 2 becomes

\[
-J_d \sum_{l \notin m,s'} \eta(R_l - R_i, R_m - R_i)\pi_{ls}^{\pi} \pi_{ms} \sigma_{s's} \cdot S_i, \tag{19}
\]

where

\[
\eta(R_l, R_m) = \sum_{\alpha=x,y} [A_{\alpha}(R_l)A_{\alpha}(R_m) + B_{\alpha}(R_l)B_{\alpha}(R_m)],
\]

\[
A_{\alpha}(R_l) = \frac{1}{N} \sum_{k} \beta_k \cos(k_x x_l) \cos(k_y y_l) \cos^2(k_{\alpha} b),
\]

\[
B_{\alpha}(R_l) = -\frac{1}{2N} \sum_{k} \beta_k \sin(k_x x_l) \cos(k_y y_l) \sin(k_{\alpha} a),
\]

\[
B_{\alpha}(R_l) = -\frac{1}{2N} \sum_{k} \beta_k \cos(k_x x_l) \sin(k_y y_l) \sin(k_{\alpha} a). \tag{20}
\]
As before we can separate from the sum the terms with either \( i = l \) or \( i = m \), for which we can use \( s_i \cdot S_i = -3/4 \) in the LESZRS, and the rest. Using also Eqs. (13), (17) and \( A_{\alpha}(-R_l) = A_{\alpha}(R_l), B_{\alpha}(-R_l) = -B_{\alpha}(R_l) \), one obtains

\[
- \frac{3}{2} J_d \sum_{\tilde{i}\sigma} \eta(R_{\tilde{i}}, 0) d_{\tilde{i}+1\sigma} d_{\sigma} - J_d \sum_{\tilde{l} \neq i \neq m\sigma} \eta(R_l - R_i, R_m - R_i) d_{l\sigma}^d d_{m\sigma} S_i \cdot S_m.
\]

Finally, using Eqs. (5) and neglecting non-bonding states the term in \( t_{pp} \) of Eq. (2) becomes

\[
\frac{t_{pp}}{2} \sum_{\tilde{i}\sigma} \mu(R_l) d_{\tilde{i}+1\sigma} d_{\sigma},
\]

where

\[
\mu(R_l) = \frac{8}{N} \sum_{\mathbf{k}} \delta_{\mathbf{k}}^2 \cos^2(k_x b) \cos^2(k_y b).
\]

Including the Cu-Cu exchange term and adding Eqs. (14), (16), (18), (21), and (22), one realizes that \( PH_{sf}^p \) can be mapped into the following generalized \( t - J \) model:

\[
H^p_{t,J} = \frac{t^{sf}_{12} - t^{sf}_{11}}{4} \sum_{\tilde{i}\sigma} d_{\tilde{i}+2\sigma} d_{\sigma} + \sum_{\tilde{i}\sigma} \left[ 6(t^{sf}_{11} + t^{sf}_{12}) \lambda(R_{\tilde{i}}) \lambda(0) - \frac{3}{2} J_d \eta(0, 0) + \frac{t_{pp}}{2} \mu(R_{\tilde{i}}) \right] d_{\tilde{i}+1\sigma} d_{\sigma} + \sum_{\tilde{l} \neq i \neq m\sigma} \left[ 4(t^{sf}_{11} + t^{sf}_{12}) \lambda(R_l - R_i) \lambda(R_m - R_i) - J_d \eta(R_l - R_i, R_m - R_i) \right] d_{l\sigma}^d d_{m\sigma} S_i \cdot S_m + \frac{J}{2} \sum_{\tilde{i}\delta} S_i \cdot S_{\tilde{i}+1\delta}.
\]

The main two-dimensional integrals that enter this expression are displayed in Table II. Note that \( \lambda(R_l) \) and \( \mu(R_l) \) are symmetric under the operations of the point group \( C_{4v} \), while \( A_{\alpha}(-R_l) = A_{\alpha}(R_l) \) and \( B_{\alpha}(-R_l) = -B_{\alpha}(R_l) \). Some of these integrals were given previously [24]. There are small differences in some \( \mu(R_l) \). We believe that our results are more accurate.

| \( R/a \) | \( \lambda \) | \( A_x \) | \( A_y \) | \( B_x \) | \( B_y \) | \( \mu \) |
|---------|---------|---------|---------|---------|---------|---------|
| (0,0)   | 0.9581  | 0.4791  | 0.4791  | 0       | 0       | 1.4535  |
| (1,0)   | 1.4011  | 0.1989  | -0.05877| 0.2802  | 0       | 0.5465  |
| (1,1)   | -0.02351| -0.01753| -0.01753|         | 0.2441  |         |
| (2,0)   | -0.01373| -0.02643| 0.01270 |         | -0.1277 |         |

**THE GENERALIZED \( t - J \) MODEL FOR T-CUO**

Naturally, the one-band model for the other CuO\(_2\) sublattice \( H^p_{t,J} \) (the mapping of \( PH_{sf}^p \) to a generalized \( t - J \) model) has the same form as \( H^p_{t,J} \) above, with the only difference that \( i \) refers to Cu sites of the other sublattice. In addition, the exchange term proportional to \( J' \) in Eq. (3) retains the same form in the one-band model. Therefore, the remaining task is to map the term proportional to \( t'_{pp} \).

**Mapping using non-orthogonal singlets**

We define the nonorthogonal ZRS for the second CuO\(_2\) sublattice in analogy to Eqs. (4) and (5):
FIG. 2: Orbitals involved in the hopping between non-orthogonal ZRS belonging to different CuO$_2$ sublattices.

$$|i\tilde{s}\rangle = \frac{1}{\sqrt{2}} \left( \hat{\rho}_{i\uparrow}^d d_{i\uparrow}^\dagger - \hat{\pi}_{i\sigma}^d d_{i\sigma}^\dagger \right) |0\rangle,$$

(25)

$$\hat{\rho}_{i\sigma} = \frac{1}{2} \sum_{q_{i+\delta\sigma}} q_{i+\delta\sigma},$$

(26)

Adolphs et al. argue that the orbitals $\hat{\pi}_{i\sigma}$ and $\hat{\rho}_{i\sigma}$ at NN sites do not mix [12]. However, in spite of a partial cancellation, the result is nonzero. An example is shown in Fig. 2 for $R_n = R_i + b(\hat{x} - \hat{y})$. In terms of the numbers of the figure

$$\hat{\pi}_{i\sigma} = \frac{1}{2} \sum_{i=1}^{4} p_{i\sigma}, \quad \hat{\rho}_{n\sigma} = \sum_{i=3}^{6} q_{i\sigma},$$

(27)

Then

$$t'_{pp} \sum_{ij\gamma\sigma} s_{\gamma} \left( p^\dagger_{j+\gamma\sigma} q_{j\sigma} + \text{H.c.} \right) \hat{\pi}_{i\sigma}^\dagger |0\rangle = \frac{t'_{pp}}{2} (q_{i\sigma}^\dagger + q_{i\sigma}^\dagger) |0\rangle + ... = \frac{t'_{pp}}{2} \hat{\rho}_{n\sigma}^\dagger |0\rangle + ...$$

(28)

It is easy to see that the same value $t'_{pp}/2$ is obtained for $R_n - R_i = -b(\hat{x} - \hat{y})$, while the result is $-t'_{pp}/2$ for $R_n - R_i = \pm b(\hat{x} + \hat{y})$. There are also contributions $\pm t'_{pp}/4$ at fourth NN. The mapping $|i\tilde{s}\rangle \leftrightarrow |0\rangle$ leads to a factor -1/2 [similar to Eq. (13)] plus some corrections due to non-orthogonality of the ZRS [20]. The details are beyond the scope of this work. In the following subsection, we derive the rigorous result using orthogonal ZRS. In any case, the simpler results presented here show that the effective hopping is not zero.

Mapping using orthogonal singlets

The term in $t'_{pp}$ of Eq. (3) can be written in the form

$$H' = t'_{pp} \sum_{ij\gamma\sigma} s_{\gamma} \left( p^\dagger_{i+\gamma\sigma} q_{j\sigma} + \text{H.c.} \right) =$$

$$= t'_{pp} \sum_{i\sigma} p^\dagger_{i+b\hat{x}\sigma} (q_{i+b\hat{y}\sigma} + q_{i+a\hat{y}-b\hat{y}\sigma} - q_{i-b\hat{y}\sigma} - q_{i-a\hat{y}-b\hat{y}\sigma}) +$$

$$+ p^\dagger_{i+b\hat{y}\sigma} (q_{i+b\hat{x}\sigma} + q_{i-b\hat{x}+a\hat{x}\sigma} - q_{i-b\hat{x}\sigma} + q_{i+b\hat{x}+a\hat{x}\sigma}) + \text{H.c.},$$

(29)
where the sum runs over all sites of the first CuO$_2$ sublattice.

Using Eqs. (8) and the corresponding ones for the second CuO$_2$ sublattice:

\[ q^{n+b\xi\sigma} = \frac{1}{N} \sum_k \beta_k e^{-ik R_n} e^{-iky b} \sum_m e^{ik R_m} [\cos(k_x b) \rho_{m\sigma} + ...], \]

\[ q^{n+b\zeta\sigma} = \frac{1}{N} \sum_k \beta_k e^{-ik R_n} e^{-ik_y b} \sum_m e^{ik R_m} [\cos(k_y b) \rho_{m\sigma} - ...], \]  

(30)

one obtains after some algebra

\[ H' = t''_{pp} \xi(\mathbf{R}_\tau) \sum_{\mathbf{R}_i \sigma} \left( \pi^\dagger_{i\sigma} \rho_{i+\tau\sigma} + \text{H.c.} \right), \]  

(31)

where \( \tau \) denotes the vectors connecting both CuO$_2$ sublattices (\( x_\tau \) and \( y_\tau \) below are both odd multiples of \( b \)) and

\[ \xi(\mathbf{R}_\tau) = -\frac{4}{N} \sum_k \sin(k_x b) \sin(k_y b) \sin(k_x x_\tau) \sin(k_y y_\tau). \]  

(32)

It is easy to see that \( \xi(\mathbf{R}_\tau) = -1 \) if \( \mathbf{R}_\tau = \pm b(\hat{x} + \hat{y}) \), \( \xi(\mathbf{R}_\tau) = 1 \) if \( \mathbf{R}_\tau = \pm b(\hat{x} - \hat{y}) \), and \( \xi(\mathbf{R}_\tau) = 0 \) for other \( \mathbf{R}_\tau \).

Therefore

\[ H' = t''_{pp} \sum_{\mathbf{R}_i \gamma \sigma} s_{\gamma} \left( \pi^\dagger_{i\sigma} \rho_{i+\gamma\sigma} + \text{H.c.} \right). \]  

(33)

using the mapping Eq. (13) and adding the other terms, the complete generalized \( t-J \) model for T-CuO takes the form

\[ H_{tJ} = H_{tJ}^p + H_{tJ}^q - t''_{pp} \sum_{\mathbf{R}_i \gamma} s_{\gamma} \left( \pi^\dagger_{i\sigma} \rho_{i+\gamma\sigma} + \text{H.c.} \right) - J' \sum_{\mathbf{R}_i \gamma} \mathbf{S}_i \cdot \mathbf{S}_{i+\gamma}. \]  

(34)

To compare with experiment it is convenient to write the Hamiltonian in terms of the following operators

\[ c_{i\sigma} = e^{iQ \cdot (\mathbf{R}_i - \mathbf{R}_0)} d_{i\sigma}, \]  

(35)

which restores the original phases of the Cu orbitals [changed before in Eq. (1)]. If the phases are not restored, the problem is of course equivalent, but the wave vectors are displaced by \( Q \) complicating the comparison with experiment. This transformation within each CuO$_2$ sublattice changes the sign of the NN hopping (at distances \( \pm a \hat{x}, \pm a \hat{y} \)) leaving second and third NN hopping unchanged. In addition also the sign of the intersublattice hopping at distances \( \pm b(\hat{x} - \hat{y}) \) is changed, keeping the sign in the perpendicular direction, so that the corresponding term in Eq. (34) becomes

\[ H_{NN} = \frac{t''_{pp}}{2} \sum_{\mathbf{R}_i \gamma \sigma} \left( c^\dagger_{i\sigma} c_{i+\gamma\sigma} + \text{H.c.} \right). \]  

(36)

**SIMPLIFIED GENERALIZED \( t-J \) MODEL**

The state of the art technique for studying the dynamics of one hole in an antiferromagnet is the self-consistent Born approximation (SCBA) \[25, 28\]. It compares very well with exact diagonalization of small clusters \[25, 27, 28, 32\], while permitting an extensions to larger clusters. From previous studies for the antiferromagnetic order of CuO$_2$ planes, one knows that the propagation of the hole is easier through each sublattice with spins pointing in the same
direction, in particular for hopping involving second and third NN, while it is inhibited for first NN in spite of the fact the corresponding hopping is larger, because the hopping distorts the antiferromagnetic alignment.

The generalized $t - J$ model for CuO$_2$ planes, as described above, contains three-site terms which combine second and third NN with spin-flip processes. These so-called correlated hopping processes are argued to play an important role for superconductivity [29, 30]. However, the above argument indicates that for the propagation of the hole, only the spin-conserving part is important. Therefore, to simplify the model and bring it amenable to the SCBA treatment we retain only hopping up to third NN in the CuO$_2$ planes and approximate $\mathbf{S}_i \cdot \mathbf{S}_m \simeq (\mathbf{S}_i^z \mathbf{S}_m^z)$ in Eq. (35). This leads to a simplified effective model for T-CuO similar to that considered by Moser et al [17].

$$H_{ij}^{\kappa} = -\frac{\sum_{\kappa=0}^3 t_{i\kappa} \sum_{i\kappa} e^i_{\kappa} c_{i\kappa}^\dagger c_{i\kappa} + H.c.}}{\kappa} + \frac{J}{2} \sum_{iv} \mathbf{S}_i \cdot \mathbf{S}_{i+v} - \frac{J'}{2} \sum_{iv_0} \mathbf{S}_i \cdot \mathbf{S}_{i+v_0},$$

where the subscript $\kappa = 0$ refers to intersublattice hopping of NN Cu atoms in the T-CuO structure (connected by the vectors $v_0 = \pm b(\hat{x} \pm \hat{y})$), while $\kappa = 1, 2, 3$, refer to first, second, and third NN within each CuO$_2$ sublattice. Comparison with Eqs. (27), (30) and using Eq. (34) leads to

$$t_0 = -\frac{t_{pp}}{2},$$
$$t_1 = \frac{t_{1f}^s - t_{1f}^c}{4} + 6(t_1^f + t_2^f)\lambda(0)\lambda(a\hat{x}) - \frac{3}{2} J_d \eta(a\hat{x}, 0) + \frac{t_{pp}}{2} \mu(a\hat{x}),$$
$$t_2 \simeq 6(t_1^f + t_2^f)\lambda(0)\lambda(a(\hat{x} + \hat{y})) - \frac{3}{2} J_d \eta(a(\hat{x} + \hat{y}), 0) + \frac{t_{pp}}{2} \mu(a(\hat{x} + \hat{y})) + 2\langle \mathbf{S}_i^z \mathbf{S}_{i+v_1}^z \rangle \left[ 4(t_1^f + t_2^f)\lambda^2(a\hat{x}) - J_d \eta(a\hat{x}, a\hat{y}) \right],$$
$$t_3 \simeq 6(t_1^f + t_2^f)\lambda(0)\lambda(2a\hat{x}) - \frac{3}{2} J_d \eta(2a\hat{x}, 0) + \frac{t_{pp}}{2} \mu(2a\hat{x}) + \langle \mathbf{S}_i^z \mathbf{S}_{i+v_1}^z \rangle \left[ 4(t_1^f + t_2^f)\lambda^2(a\hat{x}) - J_d \eta(a\hat{x}, -a\hat{x}) \right].$$

Using Eqs. (20), Table II and $\langle \mathbf{S}_i^z \mathbf{S}_{i+v_1}^z \rangle = -0.186$ for the NN expectation value for the Heisenberg model in the square lattice (see next section), one obtains

$$t_1 \simeq 0.555 t_{1f}^s + 1.055 t_{2f}^s + 0.273 t_{pp} - 0.101 J_d,$$
$$t_2 \simeq 0.161(t_{1f}^s + t_{2f}^s) - 0.122 t_{pp} - 0.0173 J_d,$$
$$t_3 \simeq 0.0935(t_{1f}^s + t_{2f}^s) + 0.0638 t_{pp} - 0.0033 J_d.$$

The fact that $t_{1f}^s + t_{2f}^s$ and $t_{pp}$ enter with different sign in $t_2$ leads to a large relative error in this parameter. Fortunately, the results seem to be rather insensitive to $t_2$.

Using the estimated parameters for the spin-fermion model based on previous constrained-density-functional calculations (set A) or given by Adolphs et al. [12] (set B), tabulated in Table II we obtain the results presented in Table III.

| set | $t_0$ | $t_1$ | $t_2$ | $t_3$ | $J$ | $J'$ |
|-----|------|------|------|------|-----|-----|
| A   | -168 | 417  | -2   | 69   | 130 | 3   |
| B   | -184 | 369  | -11  | 65   | 150 | 0   |

**THE SELF-CONSISTENT BORN APPROXIMATION**

As Adolphs et al. [12], we assume the antiferromagnetic order of T-CuO given in the left of Fig. 3. The NN Cu atoms connected by the vectors $\pm \mathbf{c}$ ($\pm \mathbf{d}$), have parallel (antiparallel) spins, where $\mathbf{c} = b(\hat{x} + \hat{y})$ and $\mathbf{d} = b(-\hat{x} + \hat{y})$. 
FIG. 3: (Color online) The two possible magnetic ground states for T-CuO $Q = (0, \pi)$ (left) and $Q = (\pi, 0)$ (right). The vectors $c$ and $d$ are indicated in the left figure.

The primitive translation vectors in the plane, which also define the unit cell, are $c$ and $2d$. The unit cell has the same size as that of the CuO$_2$ planes but it is different.

Following Martínez and Horsch [25], we perform the transformation $c_{i\sigma} \rightarrow c_{i-\sigma}$ in the sublattice in which the spins are pointing down [31], in such a way that all spins are pointing up after the transformation. Then we define the spin excitations $a_i^\dagger = c_{i\uparrow}c_{i\downarrow}$, and the holon operators $h_i$ such that

$$c_{i\uparrow} = h_i^\dagger, \quad c_{i\downarrow} = h_i^\dagger a_i,$$

in the Hilbert subspace we are considering.

Spin waves

We first diagonalize the exchange part of the Hamiltonian Eq. (37) for the undoped system. In terms of the spin excitations, it takes the form

$$H_e = \frac{J}{2} \sum_{i \neq v_1} S_i \cdot S_{i+v_1} - \frac{J'}{2} \sum_{i \neq v_0} S_i \cdot S_{i+v_0} = -2JN +$$

$$+ \frac{J}{4} \sum_{i \neq v_1} \left( a_i^\dagger a_i + a_{i+v_1}^\dagger a_{i+v_1} + a_i a_{i+v_1} + a_{i+v_1} a_i^\dagger \right) -$$

$$- \frac{J'}{2} \sum_i \left( a_i^\dagger a_i + a_{i+d}^\dagger a_{i+d} + a_i a_{i+d} + a_{i+d} a_i^\dagger \right) +$$

$$+ \frac{J'}{2} \sum_i \left( a_i^\dagger a_i + a_{i+c}^\dagger a_{i+c} - a_{i+c} a_i^\dagger \right).$$

(40)

Using the Fourier transform $a_i = N^{-1/2} \sum_k e^{-ik \cdot R_i} a_k$, one obtains

$$H_e + 2JN = \sum_k \left[ A_k a_k^\dagger a_k + B_k (a_k a_{-k} + H.c.) \right],$$

$$A_k = 2J - J' \cos(c \cdot k), \quad B_k = \frac{J'}{4} \sum_{v_1} \cos(v_1 \cdot k) - \frac{J'}{2} \cos(d \cdot k).$$

(41)

Dropping the constant $2JN$, $H_e$ is set into diagonal form introducing new bosonic operators:
\[ H_c = \sum_k \omega_k \theta_k^\dagger \theta_k, \quad \theta_k = u_k a_k + v_k a_k^\dagger. \]

\[ \omega_k = \sqrt{A_k^2 - 4B_k^2}, \quad u_k = \frac{1}{2} + \frac{A_k}{2\omega_k}, \quad v_k^2 = u_k^2 - 1, \]

\[ u_k > 0, \quad \text{sgn}(v_k) = \text{sgn}(B_k). \] (42)

**Spin-spin correlations**

In the spin-wave formalism, the correlation function entering Eq. (38) becomes

\[ \langle S_i^z S_{i+v_1}^z \rangle = \left( \frac{1}{2} - a_i^\dagger a_i \right) \left( \frac{1}{2} + a_{i+v_1}^\dagger a_{i+v_1} \right) = \frac{1}{4} + \langle a_i^\dagger a_i \rangle - \langle a_i^\dagger a_i a_{i+v_1}^\dagger a_{i+v_1} \rangle, \] (43)

where we have taken into account that the spins of sites \( i \) and \( i + v_1 \) point in opposite directions. Decoupling the last correlation function

\[ \langle a_i^\dagger a_{i+v_1} a_{i+v_1} \rangle = \langle a_i^\dagger a_i \rangle \langle a_{i+v_1}^\dagger a_{i+v_1} \rangle + |\langle a_i^\dagger a_{i+v_1}^\dagger \rangle|^2 + |\langle a_i^\dagger a_{i+v_1} \rangle|^2, \] (44)

we obtain

\[ \langle S_i^z S_{i+v_1}^z \rangle = -m^2 - |\langle a_i^\dagger a_{i+v_1}^\dagger \rangle|^2 - |\langle a_i^\dagger a_{i+v_1} \rangle|^2, \] (45)

where

\[ m = \frac{1}{2} - \langle a_i^\dagger a_i \rangle \] (46)

is the sublattice magnetization.

Transforming Fourier and using the inverse of the second Eq. (42)

\[ a_k = u_k \theta_k - v_k \theta_k^\dagger, \] (47)

the different correlation functions become at zero temperature

\[ \langle a_i^\dagger a_i \rangle = \frac{1}{N} \sum_{kq} \left( u_k \theta_k^\dagger - v_k \theta_k^\dagger \right) \left( u_q \theta_q^\dagger - v_q \theta_q^\dagger \right) = \frac{1}{N} \sum_k v_k^2, \]

\[ \langle a_i^\dagger a_{i+v_1} \rangle = \frac{1}{N} \sum_{kq} \left( u_k \theta_k^\dagger - v_k \theta_k^\dagger \right) e^{iq \cdot v_1} \left( u_q \theta_q^\dagger - v_q \theta_q^\dagger \right) = \frac{1}{N} \sum_k \cos(k \cdot v_1) u_q v_k, \]

\[ \langle a_i^\dagger a_{i+v_1}^\dagger \rangle = \frac{1}{N} \sum_{kq} \left( u_k \theta_k^\dagger - v_k \theta_k^\dagger \right) e^{iq \cdot v_1} \left( u_q \theta_q^\dagger - v_q \theta_q^\dagger \right) = \frac{1}{N} \sum_k \cos(k \cdot v_1) v_k^2, \] (48)

We have evaluated the two-dimensional integrals above for \( J' = 0 \). The result is \( \langle a_i^\dagger a_i \rangle = 0.19660 \), \( \langle a_i^\dagger a_{i+v_1} \rangle = 0.27558 \) and \( \langle a_i^\dagger a_{i+v_1}^\dagger \rangle = 0 \), leading to \( m = -0.30340 \) and \( \langle S_i^z S_{i+v_1}^z \rangle = -0.16799 \).

**The hopping terms**

The hopping terms of the Hamiltonian Eq. (37) can be separated in two: those involving two sites of the same sublattice (spin up or down), like the terms in \( t_2 \) and \( t_3 \), and those connecting sites of different sublattices (\( t_1 \) and
like these, the quasiparticle energy \( \text{Fig. 2 main text} \) does not coincide with the brighter areas of the intensity map weight is low, and most of the spectral weight corresponds to the incoherent part of the spectral function. In cases \( \sim \) a low broadening, equivalent to \( \sim 20 \) meV was applied (see main text). Dashed line: \( M \) spectral function with a broadening of 300 meV. With such a broadening the QP peak is washed out and only the broad peak near 0.8 eV persists, which might be misinterpreted as the QP peak.

half of the terms in \( t_0 \). The latter give rise to a holon-magnon interaction. We neglect the terms creating two spin excitations. Using the transformations introduced at the beginning of this section we obtain

\[
H_t = \sum_{\alpha=0}^{3} \sum_{\nu_{n,\sigma}} \left( c_{i\alpha}^\dagger c_{i+v_{\alpha},\sigma} + \text{H.c.} \right) = t_0 \sum_{i} \left[ h^+_i h_{i+c} + h^+_i h_{i+d} (a_i + a_{i+d}) + \text{H.c.} \right] +
\]

\[
+ t_0 \sum_{i} \left( h^+_i a_i \sum_{v_1} h_{i+v_1} + \text{H.c.} \right) + \sum_{\alpha=2}^{3} \sum_{n,\sigma} \left( h^+_i h_{i+v_{\alpha},\sigma} + \text{H.c.} \right).
\]

Using \( h_i = N^{-1/2} \sum_k e^{-ik \cdot R} h_k \), Eq. \( 47 \), and adding \( H_{s} = \sum_k \omega_k \theta^+_k \theta_k \) we obtain, after some algebra

\[
H_{s,J} = \sum_k \epsilon_k h^+_k h_k + \sum_k \omega_k \theta^+_k \theta_k + \frac{1}{\sqrt{N}} \left( \sum_{kq} M_{kq} h^+_k h_{k-q} + \text{H.c.} \right),
\]

\[
\epsilon_k = 2t_0 \cos(k \cdot c) + 4t_2 \cos(ak_x) \cos(ak_y) + 2t_3 \left[ \cos(2ak_x) + \cos(2ak_y) \right],
\]

\[
M_{kq} = 2t_0 \left\{ \cos[(k - q) \cdot c] u_q - \cos(k \cdot c) v_q \right\} + 2t_1 \left[ u_q \zeta(k - q) - v_q \zeta(k) \right],
\]

\[
\zeta(k) = \cos(ak_x) + \cos(ak_y).
\]

The holon Green function \( G_h(k, \omega) \) is obtained from the self-consistent solution of the following equations:

\[
G_h^{-1}(k, \omega) = \omega - \epsilon_k - \Sigma(k, \omega) + i\epsilon,
\]

\[
\Sigma(k, \omega) = \frac{1}{N} \sum_q M^*_{kq} G_h(k - q, \omega - \omega_q). \tag{51}
\]

In practice, the calculations are done in a large but finite system and the selfconsistency can be avoided calculating sequentially \( \Sigma(k, \omega) \) for increasing values of \( \omega \), beginning with values (near \( -4J \)) such that \( \Sigma(k, \omega - \omega_q) = 0 \) for all \( k \) and \( q \). [33]

An example of the hole spectral function calculated with the SCBA can be seen in Fig. 4 for the \( \Gamma \), \( M \) and \( A \) points. A low broadening, equivalent to \( \sim 20 \) meV was applied (see main text). For the \( \Gamma \) and \( M \) points, the quasiparticle weight is low, and most of the spectral weight corresponds to the incoherent part of the spectral function. In cases like these, the quasiparticle energy (Fig. 2 main text) does not coincide with the brighter areas of the intensity map.
usually plotted in the ARPES experiments (Fig. 3 main text). But if a very high broadening is used, the QP peak is washed out when its weight is low, and hence for these cases the dispersion might be mistakingly shifted to the energy of the incoherent resonances, coinciding with the brighter areas of Fig. 3 in the main text. This is exemplified for the $M$ point in dashed lines in fig 4. It is clear that the QP energy should be defined with a low broadening.

* Electronic address: aligia@cab.cnea.gov.ar

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