

Hole pairs in a spin liquid: Influence of electrostatic hole-hole repulsion

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The stability of hole bound states in the t - J model including short-range Coulomb interactions is analyzed using computational techniques on ladders with up to 2×30 sites. For a nearest-neighbor (NN) hole-hole repulsion, the two-holes bound state is surprisingly robust and breaks only when the repulsion is several times the exchange J . At $\sim 10\%$ hole doping the pairs break only for a NN repulsion as large as $V \sim 4J$. Pair-pair correlations remain robust in the regime of hole binding. The results support electronic hole-pairing mechanisms on ladders based on holes moving in spin-liquid backgrounds. Implications in two dimensions are also presented. The need for better estimations of the range and strength of the Coulomb interaction in copper oxides is remarked. [S0163-1829(99)50902-8]

The discovery of superconductivity in $\text{Sr}_{14-x}\text{Ca}_x\text{Cu}_{24}\text{O}_{41}$ (Ref. 1) under high pressure has triggered a considerable effort to understand the physics of copper-oxide ladder materials. Early theoretical studies predicted the existence of superconductivity based on a purely electronic mechanism.² The rationale for the hole-hole attraction is that each individual hole distorts the spin-liquid ground state of the undoped ladder, increasing locally the energy density. The damage to the spin arrangement is thus minimized if holes share a common distortion. Several calculations, including computational studies for the t - J model,²⁻⁵ have confirmed that an effective attraction between holes exists in environments with short-range antiferromagnetic (AF) correlations. The fact that the hole pairs transform as $d_{x^2-y^2}$ under rotations, i.e., the same channel in which the real doped two-dimensional (2D) cuprates superconduct, provides additional support to the electronic mechanisms described above.

However, studies of hole pairing in spin liquids are usually performed without the introduction of an intersite hole-hole Coulomb repulsion. The accuracy of such an approximation has been much debated, but there are few robust calculations supporting the various points of view. The neglect of electrostatic interactions is particularly important for values of J/t such as 0.3–0.4, presumed to be realistic, since in this regime the pair size is small, roughly $2a$ (where a is the lattice spacing).⁴⁻⁶ Nevertheless, note that in the real materials at least the five d orbitals of Cu, as well as the three p orbitals of O, should be taken into account. In this extended model, polarization effects caused by electron-hole excitations can effectively reduce the strength of the electrostatic repulsion. Actually, O^{2-} has a large polarizability that influences the effective Coulomb interactions of a variety of ionic insulators.⁷ This effect has been analyzed for the on-site repulsions U_d and U_p .⁸ Although the bare couplings are substantially reduced by polarization, their strength remains the dominant scale, and models with no doubly occupied sites

capture properly this effect. However, it is unclear if the additional neglect of the hole-hole repulsion at distance a remains a good approximation.

Unfortunately, the explicit calculation of the strength of screened Coulombic interactions is difficult. Using results for 2D copper oxides as guidance, a constrained-density-functional approach where the local-density approximation (LDA) bands are identified with a mean-field solution of the Hubbard model reports a repulsion $U_{pd} \sim 1.2$ eV between a pair of holes at the Cu and O ions.⁹ Since the bare value is 7.8 eV, a reduction of a factor 6.5 is effectively achieved by polarization effects. If the trend continues, then the repulsion V at distance a will be ~ 0.6 eV $\sim 4J$ [with $J \sim 0.15$ eV (Ref. 10)]. However, other estimations comparing Auger spectroscopy results with $(\text{Cu}_4\text{O}_5)^{6-}$ cluster calculations using a multiband Hubbard model report $U_{pd} < 1$ eV.⁸ In addition, for two holes in neighboring oxygens $U_{pp} \sim 0$ was observed,⁸ suggesting that the hole-hole correlations decay rapidly with distance. For this reason the above estimation 0.6 eV should be considered as an upper bound for V . Other studies based on the interaction of two neighboring Zhang-Rice singlets report $V \sim 0.2$ eV.¹¹ Finally, approaches where the dielectric constant $\epsilon \sim 30$ (Ref. 12) is used even at short distances provide $V \sim 0.13$ eV. Then, current estimations locate $V \in [J, 4J]$ as the realistic range for the hole-hole repulsion in a one-band model. Such strong repulsion is dangerous for real-space AF theories. Coulomb interactions are also a central ingredient of the “striped” scenarios.¹³

The purpose of this paper is to discuss a computationally intensive calculation of the effect of intersite hole-hole repulsion on the hole bound states of the t - J model on ladders. Since the size of the hole pairs on planes and ladders is comparable for the same J/t , our results have consequences also for studies in 2D. The calculation is performed with the density matrix renormalization group (DMRG) technique,¹⁴ as well as using a diagonalization technique in a reduced

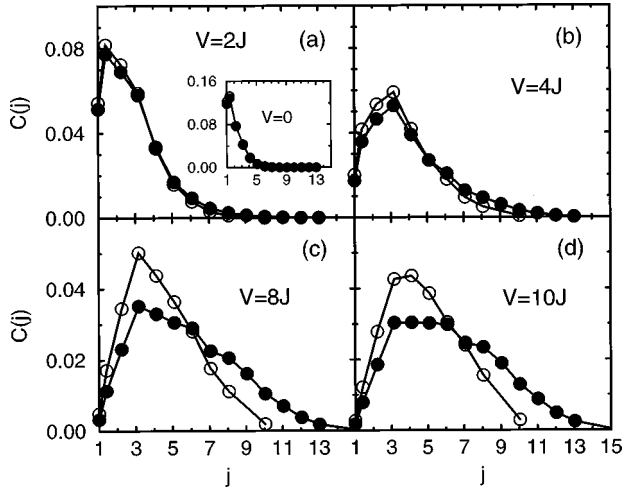


FIG. 1. Hole-hole correlation $C(j)$ (see text) for 2×20 (open circles) and 2×30 (full circles) clusters with two holes, using DMRG with OBC and $J/t=0.4$. Results for several couplings V/J are shown. The Coulomb repulsion has range $R=1$. Here \mathbf{j} is the distance between a site at the center defined as origin, and sites belonging to the opposite leg.

Hilbert space¹⁵ (the optimized reduced-basis approximation or ORBA).¹⁶ The Hamiltonian employed here is the t - J model at a realistic coupling $J/t=0.4$, supplemented by a hole-hole repulsion $V_{hh} = \sum_{ij}(V/|\mathbf{i}-\mathbf{j}|)n_i n_j$, where n_i is the hole number operator at site \mathbf{i} . The range R of the repulsion has been restricted to 1 and $\sqrt{2}$ lattice spacings, since the speed of convergence of the numerical techniques (both variational) decreases as R grows. Earlier results have been obtained in the case of static holes¹⁷ or using small clusters.¹⁸

Let us discuss first the case of two holes. Figure 1 contains the hole-hole correlation $C(\mathbf{j}) = \langle n_{\mathbf{0}} n_{\mathbf{j}} \rangle$ obtained on $2 \times N$ clusters with two holes and open-boundary conditions (OBC's) using DMRG ($m=300$ states), for the case where V_{hh} acts only at a distance of one lattice spacing (i.e., $R=1$).¹⁹ $\mathbf{0}$ is a site at the center of the cluster, and the figure shows the hole-hole correlation along the leg opposite to where $\mathbf{0}$ is located (results for the other leg are similar). $C(\mathbf{j})$ is related with the probability of finding one hole at \mathbf{j} when there is one at $\mathbf{0}$.

Figures 1(a) and (b) correspond to $V=0, 2J$, and $4J$. Here the results change only slightly as the lattice grows, and the two holes remain close to each other indicating the existence of a bound state. Apparently a nearest-neighbor (NN) repulsion already larger than J is *not* enough to destroy the bound state, although it weakens it. On the other hand, Figs.1(c) and (d) show similar results but now for $V=8J$ and $10J$ where a substantial change in the hole distribution is observed as the cluster grows. The spreading of the hole over the entire lattice suggests either the breaking of the pair or a weak bound state. In the large- V regime one of the holes acts as a sharp “wall” to the other, which spreads its wave function in an effective square-well potential. Figure 2(a) shows an average hole-hole distance defined as $\langle d \rangle = \sum_{\mathbf{j} \neq \mathbf{0}} d_{\mathbf{j}} \langle n_{\mathbf{0}} n_{\mathbf{j}} \rangle / \sum_{\mathbf{j} \neq \mathbf{0}} \langle n_{\mathbf{0}} n_{\mathbf{j}} \rangle$, where $d_{\mathbf{j}}$ is the $\mathbf{0}$ - \mathbf{j} distance. The convergence of the results for $V \leq 4J$ as the size grows is clear. However, for $V \geq 6J$, there is no obvious convergence. The curvature change of the 2×30 results at $V \sim 5J$ also favors the interpretation that the bound state exists up to

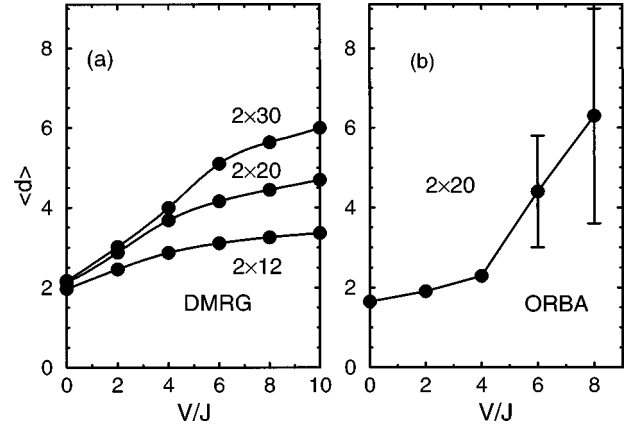


FIG. 2. (a) Mean distance between holes $\langle d \rangle$, defined in the text, vs V/J obtained with the DMRG method at $J/t=0.4$. Results for three lattice sizes are shown. The range of the Coulomb repulsion is $R=1$; (b) same as (a) but now using the ORBA technique with $\sim 10^6$ states, and a 2×20 cluster with periodic-boundary conditions.

that coupling, since in the bulk $\langle d \rangle$ vs V/J will grow with positive curvature, diverging when the pair-breaking critical coupling is reached.

To further support these conclusions, the ORBA technique has been implemented on a 2×20 cluster with periodic-boundary conditions (PBC's). The basis selected to optimize the convergence as the Hilbert space grows is the rung basis, which contains nine states for the t - J model.¹⁶ A convergence in the energy with up to three significant figures is achieved in this cluster by using $\sim 10^6$ states in the subspace of zero momentum. While this is not as precise as the DMRG method for this model, the accuracy is enough for our purposes and ORBA has the advantage that it is performed with PBC's. Figure 2(b) shows $\langle d \rangle$ vs V/J . Here $\mathbf{0}$ is any site of the lattice since there is translational invariance. For $V/J \leq 4$ it was observed that the hole-hole correlations change by a very small percentage when the space grows from 10^5 to 10^6 states, using as a starting configuration two holes in the same rung. In this regime the hole-pair bound state is tight. However, for $V/J \geq 6$ the changes in the correlations as the number of states grows are large. [Actually the hole-hole distances reported in Fig. 2(b) for large V/J are not stabilized (thus they carry large error bars)]. Nevertheless, as V/J increases the qualitative behavior of $\langle d \rangle$ is in good agreement with the DMRG data.

Let us now analyze what occurs when the range of the repulsion grows. Figure 3 contains DMRG results for a repulsion of strength V and $V/\sqrt{2}$ when holes are at distances 1 and $\sqrt{2}$ lattice spacings, respectively, and zero otherwise. Although qualitatively similar to Fig. 1, now the region where the bound state is stable is restricted to $V/J \leq 2$, while results for $V/J \geq 4$ suggest the absence of a bound state due to the spreading of the second hole when one is at the center of the ladder. Figure 4(a) shows $\langle d \rangle$ for the same interaction used in Fig. 3. In agreement with the previous discussion, the critical region is $V/J \sim 3$. For smaller (larger) couplings $\langle d \rangle$ converges (diverges) as the cluster size grows. ORBA results also suggest a critical coupling $V/J \sim 3$.

To complement the analysis using hole-hole correlations, Fig. 4(b) contains the binding energy defined as $\Delta_B = E(2)$

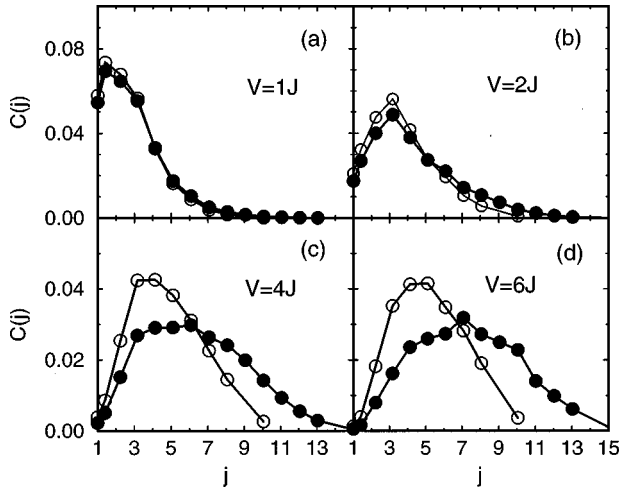


FIG. 3. Same as Fig. 1 but with a longer ranged repulsion of strength V ($V/\sqrt{2}$) at distance of one ($\sqrt{2}$) lattice spacings.

$+E(0)-2E(1)$, where $E(n)$ is the lowest energy in the subspace of n holes. A negative Δ_B implies that a bound state exists. At $J/t=0.4$ and $V=0$ there is pairing of holes on two-leg ladders.^{2,4} The figure shows that for a NN repulsion, the binding effect continues even up to large values of V/J . Actually the region $6 \leq V/J \leq 10$ corresponds to a weak bound state. Figure 4(b) also shows results for range- $\sqrt{2}$ repulsion. In this case the critical coupling $V/J|_c$ at which the bound state is lost is between 3 and 4. Both results are in agreement with the estimations based on Figs. 2(a) and 4(a).

The previous analysis indicates that the stability of the two-hole bound states in t - J -like models depends on the value and range of the electrostatic Coulomb interaction between holes. When the Coulombic term is restricted to a realistic range $R=1$, a repulsion as large as $0.6\text{eV} \sim 4J$ weakens but does not destroy the pair, implying that the effective range of attraction caused by spin polarization is larger than one lattice spacing. Retardation effects (fully considered in the present calculation) due to the different energy scales of spin and charge excitations (J vs t) likely contribute to the strong stability of the bound states.¹⁸ Note also that using, e.g., $V/J=4$, the pair size is $\sim 4-5a$ [Fig. 2(a)]. This result is close to estimations of the Cooper pair size in the 2D cuprates which report a coherence length $\xi_c/a \sim 4$ and ~ 3 , for optimally doped La-214 and Y-Ba-Cu-O, respectively^{20,21} (using $a=3.8\text{\AA}$). To the extent that ξ_c/a are similar on planes and ladders, apparently a realistic NN hole-hole repulsion can actually improve quantitatively the predictions of the t - J model, without destroying the pairs.

The results discussed thus far have been restricted to the two-holes sector. Since the pairs are very tight it is reasonable to assume that even with a finite (small) hole density the bound states will remain stable upon the addition of a NN repulsion V . This assumption can be checked explicitly using a 2×20 cluster [shown to be already close to the bulk limit, see Fig. 4(b)]. In Fig. 5(a) the binding energy defined as $\Delta_B = E(n) + E(n-2) - 2E(n-1)$ vs the hole concentration $x = n/40$ is shown for three values of V . As expected, the results are qualitatively similar to those of the two-holes problem. While for $V=0$ the pairs remain stable even up to 20% doping and beyond, using a repulsion as large as V

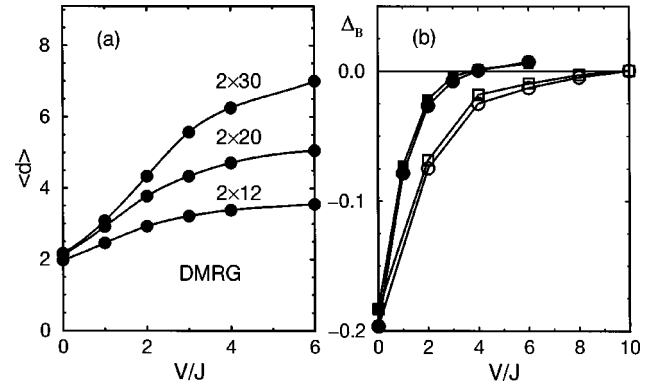


FIG. 4. (a) Same as Fig. 2(a) but with a longer ranged repulsion of strength V ($V/\sqrt{2}$) at distance of one ($\sqrt{2}$) lattice spacings; (b) DMRG binding energy of two holes obtained for $J/t=0.4$. Open circles and squares correspond to 2×20 and 2×30 clusters, respectively, both with a Coulomb repulsion of range $R=1$. Full circles and squares also correspond to 2×20 and 2×30 clusters, respectively, but now with a Coulomb repulsion of range $R=\sqrt{2}$.

$=2J$ the stability persists roughly up to $x \sim 0.17$, and for $V=4J$ up to $x \sim 0.10$. Finding a critical coupling $V/J|_c$ decreasing with doping is reasonable since the stability of the pairs at finite x depends not only on the hole-hole attraction, but also on the mean distance between holes that introduces limitations on the pair size. As example consider $V/J=2$. From Fig. 2(a) the pair size is $\langle d \rangle \sim 3$. The density at which the hole mean distance is also ~ 3 corresponds to $x \sim 0.17$, in good agreement with the critical density at this V [Fig. 5(a)]. Then, from the two-hole problem information about finite x can be obtained.

Superconducting pair-pair correlations $C(r)$ have also been measured here. $C(r)$ is proportional to $\sum_m \langle \Delta(m) \Delta^\dagger(m+r) \rangle$, where m, r are rung indices, and $\Delta(m)$ destroys a pair of electrons in a spin singlet at rung m . An average over the whole cluster is used to calculate $C(r)$. At $V=0$ previous results were reproduced as a test.²² The new results are shown in Fig. 5(b) using hole density $x=0.1$ as example. The data are presented at three characteristic distances ($r=3, 6, \text{ and } 10$) (Ref. 23) vs V/J . The nor-

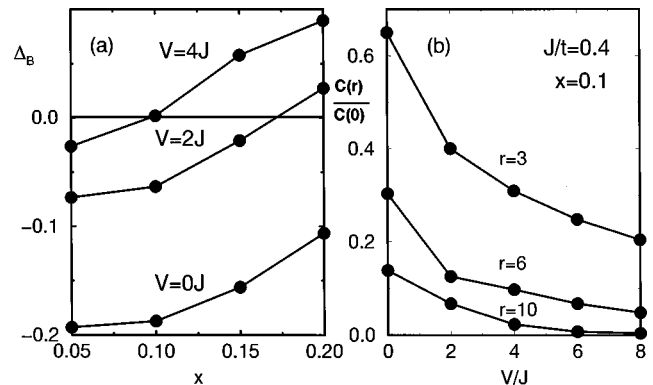


FIG. 5. (a) Binding energy of hole pairs obtained working at a finite density of holes on a 2×20 cluster. Results at $J/t=0.4$ and several V 's are shown; (b) pairing correlations $C(r)/C(0)$ (see definition in text) vs V/J at three characteristic distances r along the legs. $J/t=0.4$ and the hole density is $x=0.1$. The cluster size is 2×30 .

malization using $C(0)$ is important since the signal for superconducting correlations has an overall penalization due to the reduction of the probability of finding two holes in the same rung as V/J grows. This reduction only indicates that pairs become larger as V/J increases. Figure 5(b) shows that the strong $V=0$ superconducting correlations remain robust as V/J grows. Even for r as large as 10 the correlations are not negligible in the scale of the plot in the region $0 \leq V/J \leq 4$, which coincides with the region of hole binding according to [Fig. 5(a)]. Similar conclusions were reached at $x=0.2$ (not shown). Then, our results are compatible with a picture where hole-binding and strong superconducting correlations coexist. Note, however, that as $x \rightarrow 0$ the pair correlations must decrease due to the reduction in the density of pairs even if the binding energy is robust.

Summarizing, it has been shown that (1) the two-hole ladder bound state is more stable than expected upon the introduction of a NN hole repulsion, and that (2) this conclusion persists in the presence of a finite hole density. Hole pairs and robust superconducting correlations were found in the same region of parameter space. These results provide support to theories of ladders that predict hole pairing based

on electronic mechanisms that describe holes as immersed in spin-liquid backgrounds.² It also reinforces striped scenarios for cuprates where pairing is produced by carriers moving from the fluctuating stripes to the ladders between them.¹³ Regarding 2D systems our results show that the effect of a Coulomb interaction is a subtle quantitative problem. Actually the binding energy of two holes on a four-leg ladder with $V=0$ and $J/t=0.4$ has also been estimated here. The result is $|\Delta_B^{4L}| \sim 0.14t$ (using a 4×12 cluster, $m \sim 400$ states, and truncation error $\sim 10^{-5}$), which is similar to previous results in 2D (Fig. 24 of Ref. 3). Then, the influence of the NN Coulomb interaction on planes will be stronger than on ladders. In this respect it is imperative to obtain either experimental or theoretical information about the range and strength of the Coulomb interaction in effective one-band models for cuprates, specially in 2D.

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