“Spin-Disentangled” Exact Diagonalization of Repulsive Hubbard Systems: Superconducting Pair Propagation

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By a novel exact diagonalization technique we show that bound pairs propagate between repulsive Hubbard clusters in a superconducting fashion. The size of the matrices that must be handled depends on the number of fermion configurations per spin, which is of the order of the square root of the overall size of the Hilbert space. We use CuO₄ units connected by weak O-O links to model interplanar coupling and c-axis superconductivity in Cuprates. The numerical evidence on Cu₂O₈ and Cu₃O₁₂ prompts a new analytic scheme describing the propagation of bound pairs and also the superconducting flux quantization in a 3-d geometry.

Evidence for pairing in the repulsive Hubbard and related models has been reported by several authors. Analytic approaches [1] [2], even at strong coupling [3], generalized conserving approximation theories like FLEX [4], as well as Quantum Monte Carlo Studies on supercells [5] lead to this conclusion. However, we want more evidence about the real nature of the pairing interactions. Su and coworkers [6] have reported analytically an evidence of superconducting long-range order for several repulsive Hubbard models, including a two-dimensional cluster with an infinite stack of infinite planes, some economy is needed. However in high-Tc superconductors the coherent length is ~ a few lattice constants, and Cu-O planes can be approximately represented by clusters that are large enough to host a bound pair. The interplanar hopping does not dissolve pairs and superconducting flux quantization is clear signature. The magnetic properties of attractive Hubbard models have been studied by Canright and Girvin [7]; here we propose a gedankenexperiment very much in the spirit of Little and Parks [8], in the repulsive case.

The repulsive Hubbard Hamiltonian of fully symmetric clusters C has two-body singlet eigenstates without double occupation [9] [10] [11] [12] called W = 0 pairs. The presence of such solutions at the highest occupied level of the non-interacting (Hubbard U → 0) system is necessary to allow DC(N) < 0 where DC(N) = EC(0)(N) - E0(N) - 2(E0(N) - 1) and E0(N) is the interacting ground state energy of the cluster C with N fermions. By means of a non-perturbative canonical transformation [2] [13], it can also be shown that for all C(N) < 0 is due to an attractive pairing effective interaction and at weak coupling |DC(N)| is just the binding energy of the pair.

CuO₄ is the smallest cluster which fully preserves the point-symmetry of the Copper-Oxide planes of high-Tc materials. We have already described W = 0 pairing in great detail as a function of the one-body and interaction parameters on all sites; the study was extended to larger clusters too [1] [11]. W = 0 bound pairs in the CuO₄ cluster are found to exist in the physical region of the parameter space. However, since it is the symmetry that produces the pairing force we use the simplest working model to study bound pair propagation. Here, in order to simplify the analytical formulas, we neglect the O-O hopping term and also any distinction between Cu and O sites (except geometry, of course). The only nonvanishing hopping matrix elements are those between an Oxygen site and the central Copper site; they are all equal to t. For the sake of simplicity, we parametrize the Hubbard model in such a way that actually everything depends only on the ratio U/t; the important thing is that in this way we still have access to the part of the parameter space where pairing occurs [9]. Thus, we consider the Hubbard Hamiltonian

\[ H_{\text{CuO}_4} = t \sum_{\sigma} (d^\dagger_{\sigma} p_{\sigma} + p^\dagger_{\sigma} d_{\sigma}) + U \left( \sum_i n_i^{(p)} \hat{n}_i^{(p)} + n_i^{(d)} \hat{n}_i^{(d)} \right) \]  

where \( p^\dagger_{\sigma} \) and \( p_{\sigma} \) are the creation and annihilation operators on the Oxygen \( i = 1, ..., 4 \) with spin \( \sigma = \uparrow, \downarrow \), \( d_{\sigma} \) and \( d^\dagger_{\sigma} \) are the creation and annihilation operators onto the Copper site, while \( \hat{n}_i^{(p)} = p^\dagger_{\sigma} p_{\sigma} \) and \( \hat{n}_i^{(d)} = d^\dagger_{\sigma} d_{\sigma} \) are the corresponding number operators. \( H_{\text{CuO}_4} \) is invariant under the permutation Group \( S(4) \), which has the irreducible representations \( \text{(irreps)} \) \( A_1 \) (total-symmetric), \( B_2 \) (total-antisymmetric), \( E \) (self-dual), \( T_{\sigma} \) and its dual \( T_{\overline{\sigma}} \), of dimensions 1, 1, 2, 3 and 3, respectively. The ground state of CuO₄[2] (i.e. CuO₄ with 2 fermions)
belongs to $1^1A_1$ and that of CuO$_4$[4] is in $1^1E$; both are singlets, as the notation implies. The ground state of CuO$_4$[3] is a $^2T_1$ doublet. $\Delta_{\text{CuO}_4}(4) < 0$ for this model when $0 < U \lesssim 34.77$ $t$, as shown in Fig.(1).

$$
\Delta_{\text{CuO}_4}(4)
$$

![FIG. 1. Trend of $\Delta_{\text{CuO}_4}(4)$ in $t$ units versus log[$U/t$].](image)

Thus, we introduce a graph $\Lambda$ with CuO$_4$ units as nodes. The total Hamiltonian is

$$
H_{\text{tot}} = H_0 + H_\tau.
$$

with

$$
H_0 = \sum_{\alpha \in \Lambda} \left( \sum_{\sigma} \left( d_{\alpha,\sigma}^\dagger p_{\alpha,\sigma} + p_{\alpha,\sigma} d_{\alpha,\sigma} \right) + U \sum_{i} \tilde{n}_{\alpha,i\uparrow} \tilde{n}_{\alpha,i\downarrow} + \frac{U}{2} (\tilde{n}_{\alpha,i\uparrow} + \tilde{n}_{\alpha,i\downarrow})^2 \right),
$$

where $p_{\alpha,i,\sigma}$ is the creation operator onto the Oxygen $i = 1, ..., 4$ of the $\alpha$-th cell and so on. Hence, the point symmetry Group of $H_0$ is $S[4]|^\Lambda|$, with $|\Lambda|$ the number of nodes. There are many different ways to model an inter-planar hopping. Nevertheless, to preserve the symmetry that produces the $\Delta_{\text{CuO}_4}(4) < 0$ property, $H_\tau$ must be invariant under the $S[4]$ diagonal subgroup of $S[4]|^\Lambda|$. In the following we shall consider a hopping term allowing to a particle in the $i$-th Oxygen site of the $\alpha$-th unit to move towards the $i$-th Oxygen site of the $\beta$-th unit with hopping integral $\tau_{\alpha\beta}$:

$$
H_\tau = \sum_{\alpha,\beta \in \Lambda} \sum_{i,\sigma} \left[ \tau_{\alpha\beta} p_{\alpha,i,\sigma} p_{\beta,i,\sigma} + \text{h.c.} \right].
$$

For $N = 2|\Lambda|$ and $\tau_{\alpha\beta} \equiv 0$, the unique ground state consists of 2 fermions in each CuO$_4$ unit. This paper is devoted to the inter-planar hopping produced by small $\tau_{\alpha\beta} \ll |\Delta_{\text{CuO}_4}(4)|$ with a total number of particles $N = 2|\Lambda| + 2p$; $p$ represents the number of added pairs. When $U/t$ is such that $\Delta_{\text{CuO}_4}(4) < 0$, each pair prefers to lie on a single CuO$_4$ and for $N = 2|\Lambda| + 2p$ the unperturbed ground state is $2^p \times {|\Lambda| \choose p}$ times degenerate (since $1^1E$ has dimension 2).

By this sort of models one can study the interaction of several fermion pairs in the same system. The simplest topologically non-trivial graph is the ring, with a set $\Lambda = \{1, 2, ... , |\Lambda|\}$ and

$$
\tau_{\alpha\beta} = \begin{cases} 
\tau & \text{if } \beta = \alpha + 1, \\
\tau^* & \text{if } \beta = \alpha - 1, \\
0 & \text{otherwise}.
\end{cases}
$$

where $\phi$ is the magnetic flux concatenated by the ring and $\phi_0 = \frac{hc}{e}$. In the absence of magnetic field, $\tau$ will be taken to be real.

Note that for $p=0$ the concentration (number of holes per atom) is $2/5 = 0.4$; this is somewhat more than half-filling ($1/3 \approx 0.33$) but still reasonable. We are using CuO$_4$ as the unit just for the sake of simplicity, but the $W = 0$ mechanism produces bound pairs at different fillings for larger clusters [12] and the full plane [1] [2] too. By replacing CuO$_4$ by larger units one can model other ranges of the hole concentration.

We exactly diagonalize the $|\Lambda| = 2$ and $|\Lambda| = 3$ ring Hamiltonian; to this end we introduce the Spin-Disentangled technique. We let $M_U + M_L = N$ where $M_\sigma$ is the number of particles of spin $\sigma$; $\{|\phi_{\alpha\sigma}\rangle\}$ is a real orthonormal basis, that is, each vector is a homogeneous polynomial in the $p^\dagger$ and $d^\dagger$ of degree $M_\sigma$ acting on the vacuum. We write the ground state wave function in the form

$$
|\Psi\rangle = \sum_{\alpha,\beta} L_{\alpha\beta} |\phi_{\alpha\uparrow}\rangle \otimes |\phi_{\beta\downarrow}\rangle
$$

which shows how the $\uparrow$ and $\downarrow$ configurations are entangled. The electrons of one spin are treated as the “bath” for those of the opposite spin: this form also enters the proof of a famous theorem by Lieb [14]. In Eq.(6) $L_{\alpha\beta}$
is a $m_+ \times m_-$ rectangular matrix with $m_\sigma = \langle \psi | M_\sigma | \psi \rangle$. We let $K_\sigma$ denote the kinetic energy $m_\sigma \times m_\sigma$ square matrix of $H_{tot}$ in the basis $\{ | \phi_{\alpha \sigma} \rangle \}$, and $N_s^{(\sigma)}$ the spin-$\sigma$ occupation number matrix at site $s$ in the same basis ($N_s^{(\sigma)}$ is a symmetric matrix since the $| \phi_{\alpha \sigma} \rangle$’s are real). Then, $L$ is acted upon by the Hamiltonian $H_{tot}$ according to the rule

$$H_{tot}[L] = [K_\sigma + L K_1] + U \sum_s N_s^{(\uparrow)} L N_s^{(\downarrow)}. \tag{7}$$

In particular for $M_+ = M_-$ ($S_z = 0$ sector) it holds $K_\uparrow = K_\downarrow$ and $N_s^{(\uparrow)} = N_s^{(\downarrow)}$. Thus, the action of $H$ is obtained in a spin-disentangled way. In the $S_z = 0$ sector for $| \Lambda | = 3$ the size of the problem is 1863225 and the storage of the Hamiltonian matrix requires much space; by this device, we can work with matrices whose dimensions is the square root of those of the Hilbert space: $1365 \times 1365$ matrices solve the 1863225 × 1863225 problem, and are not even required to be sparse. We believe that this approach will be generally useful for the many-fermion problem. Since we are mainly interested in getting the low-lying part of the spectrum as fast as possible we opted for the Lanczos method, taking advantage from our knowledge of the $S[4]$ irrep of the $\tau = 0$ ground state; the scalar product is given by $\langle \Psi_2 | \Psi_2 \rangle = \text{Tr} \{ L | L \}$. In this way, the Hamiltonian matrix takes the tri-diagonal form; however a numerical instability sets in well before convergence is achieved if one uses chains longer than a few tens of sites. Therefore we use repeated two-site chains alternated with moderate-size ones.

The two-CuO$_4$ ring (14,400 configurations in the $S_z = 0$ sector) is readily solved by a Mathematica code on the personal computer; however, this cluster is not adequate for studying the quantization (superconducting or otherwise) of a magnetic flux by the bound pair. The reason is that the two units are each at the left and at the right of each other; any vector potential perpendicular to the CuO$_4$’s can always be gauged away. However, we have verified that the ground state energy with 6 holes is $E_{\text{CuO}_4}^{(0)}(6) = E_{\text{CuO}_4}^{(0)}(4) + E_{\text{CuO}_4}^{(0)}(2)$ for $\tau = 0$ and it receives a negative correction $\propto \tau^2/|\Delta_{\text{CuO}_4}(4)|$ for small $\tau$, which is consistent with the presence of a bound pair.

FIG. 2.: Numerical results for Cu$_3$O$_{12}$ with $\tau = 0.001$. Lowest-energy eigenvalues labeled by their interplanar quasi-momentum are shown versus flux $\phi$. The pattern is periodic (a flux quantum can be gauged away). (a) $U = 0$. A paramagnetic current is excited by the field and the system is utterly normal. (b) $U = 5$. The ground state shows a clear superconducting pattern, with a minimum at $\phi = \phi_0$. All energies are in $t$ units.

The three-CuO$_4$ ring behaves similarly, but can also concatenate a flux. In Figure (2.a) and (2.b) we show the lowest eigenvalues versus $\phi$ for $U = 0$ and $U = 5\,t$, respectively; $k$ denotes the interplanar quasi-momentum quantum label. At $\tau = 0$ the ground state energy is $E_{\text{CuO}_4}^{(0)}(8) = E_{\text{CuO}_4}^{(0)}(4) + 2 E_{\text{CuO}_4}^{(0)}(2)$ and the low energy sector derives mainly from the tensor product of the ground states of three independent CuO$_4$’s with 4, 2 and 2 holes (which is the fundamental multiplet) and 3, 3 and 2 holes (which is the lowest lying excited multiplet separated by a gap $\Delta_{\text{CuO}_4}(4)$). For $U = 0$, see Fig.(2.a), there is no pairing in CuO$_4$ and indeed the ground state energy is linear in the field at small fields (normal Zeeman effect). The lowest state is $k = 2$ throughout. Interestingly, Cu$_3$O$_{12}$ concatenated with half a flux quantum would be diamagnetic, but the absence of a second minimum shows that it would be Larmor diamagnetism. By contrast, at $U = 5\,t$, when pairing in CuO$_4$ is about optimum, see Fig.(1), the $k = 2$ state is lowest in the central sector, $k = 0$ is the ground state at $\phi \rightarrow 0$ while $k = 1$ is lowest as $\phi \rightarrow \phi_0$, see Fig.(2.b); this produces level crossings and the superconducting flux quantization; there is a central minimum when the system swallows a half quantum of flux while, as we verified, $\Delta_{\text{CuO}_4} < 0$. Remarkably, one also observes superconducting quantization of a magnetic flux orthogonal to the plane [12]. With increasing $U/t$, the binding energy of the pair starts decreasing and eventually vanishes for $U \approx 34.77\,t$; we have found that at this point the flux quantization returns normal and the system behaves like a paramagnet. Even at optimal $U$, the side barriers are depressed by increasing $\tau$; at $\tau \approx 0.1\,t$ only small
Due to the minus sign, the term in \(|T_\beta|\) unit in Eq.(9), the first term in the r.h.s. describes hole pair propagation, a set obtained an effective Hamiltonian by the cell-perturbation method with \(H_0\), Eq.(3), the “cell-Hamiltonian” and \(H_r\), Eq.(4), the “inter-cell perturbation” and by taking into account only the low-energy singlet sector. We note that the cell-perturbation method was already used in Ref. [15] to support the original Anderson’s conjecture [16] on the “low-energy equivalence” between the \(d - p\) model (proposed by Emery [17]) and the single-band Hubbard model. Despite the analogies with Ref. [15] (like the same cell-Hamiltonian and weak O-O links between different cells) our inter-cell perturbation is different and, more important, it is the low-energy sector which differs (one needs to consider CuO units with 2, 3 and 4 holes to get bound pairs, in contrast with 0, 1 and 2 holes of Ref. [15]).

For a general graph \(\Lambda\), with \(2|\Lambda| + 2p\) holes, we treat \(H_r\) by a simplified second-order degenerate perturbation theory, since \(H_r\) is a one-body operator. Each degenerate unperturbed ground state \(|\Phi_0^S\rangle\) may be labelled by a set \(S \subset \Lambda\) of units occupied by four holes; \(|S| = p\). The secular problem yields the eigenvalue equation

\[
\sum_{S'} \sum_q \langle \Phi_0^S | H_r | \Phi_q \rangle \langle \Phi_q | H_r | \Phi_0^{S'} \rangle a_{S'} = \varepsilon a_S
\]

where the sum has been truncated to the low-energy excited eigenstates involving CuO\(_4\) units with \(2 \leq n \leq 4\) holes, all taken in their ground states \(|\Psi_0^{(\alpha)}(\alpha)\rangle\), \(\alpha = 1 \ldots |\Lambda|\). The amplitude \(a_S = a(\alpha_1, \ldots, \alpha_p)\) is totally symmetric with respect to permutations of the distinct indices \(\alpha_1, \ldots, \alpha_p\). Letting \(\mathcal{C}(\alpha) = \{\beta \in \Lambda : \tau_{\alpha \beta} \neq 0\}\), after some algebra Eq.(8) may be written in the form:

\[
\varepsilon a(\alpha_1, \ldots, \alpha_p) = \sum_{j=1}^{p} \sum_{\beta \in \mathcal{C}(\alpha_j)} T_{\tau, \alpha_j}^{\text{Bose}} a(\alpha_1, \ldots, \alpha_{j-1}, \beta, \alpha_{j+1}, \ldots, \alpha_p) - \sum_{i \neq j} T_{\tau, \alpha_j}^{\text{Bose}} (1 - \delta_{\beta, \alpha_i}) a(\alpha_1, \ldots, \alpha_p).
\]

This is a Schrödinger equation for \(p\) hard-core bosons hopping with an effective hopping integral \(T_{\alpha, \beta}^{\text{Bose}} \equiv (\tau_{\alpha, \beta}^{\text{eff}})^2 / \Delta_{\text{CuO}_4}(4)\), with

\[
\tau_{\alpha, \beta}^{\text{eff}} = (|\Psi_0^{(2)}(\alpha)\rangle \otimes |\Psi_0^{(4)}(\beta)\rangle |H_r| |\Psi_0^{(3)}(\alpha)\rangle \otimes |\Psi_0^{(3)}(\beta)\rangle).
\]

In Eq.(9), the first term in the r.h.s. describes hole pair propagation, e.g. from unit \(\alpha_j\) to an unoccupied unit \(\beta\); in the second sum, the system gets back to the initial state after virtually exploring unit \(\beta\); the term \(\prod_{i \neq j} (1 - \delta_{\beta, \alpha_i})\) takes into account that if \(\beta\) is one of the occupied units, the particle cannot move toward it. Due to the minus sign, the term in \(|T_{\beta, \alpha_j}^{\text{Bose}}|\) represents pair repulsion.

\[
|T_{\beta, \alpha_j}^{\text{Bose}}|^2
\]

FIG. 3.: (a) \((|T_{\beta, \alpha_j}^{\text{Bose}}|^2 / |\tau_{\alpha, \beta}|^2)\) versus \(U/t\). (b) Results of Eq.(9) for Cu\(_3\)O\(_2\) with \(\tau = 0.001\) \(t\), \(U = 5\) \(t\). Lowest-energy eigenvalues labeled by their interplanar quasi-momentum are shown versus flux \(\phi\). All energies are in \(t\) units.

In Fig.(3.b) the superconducting flux-quantization for the \(|\Lambda| = 3\) ring is reported as reproduced by solving Eq.(9); it agrees well both qualitatively and quantitatively with the numerical results of Fig.(2.b), thus confirming the above approximation. More data and a fuller account of the low-energy theory will be presented elsewhere.

In conclusion, we used a set of CuO\(_4\) units connected by weak O-O links to model interplanar coupling and c-axis superconductivity in Cuprates. The results show that the system with two holes in each unit is a

\[
\sum_{\alpha = 1}^{4}
\]
background such that inserting $2p$ holes one gets $p$ pairs, bound by the repulsive interaction. The bound pairs propagation is well described by Eq.(9). We found analytically the superconducting flux quantization in the ring-shaped systems and confirmed this finding numerically for the 3-unit ring (1,863,225 configurations). To this end, we introduced a novel exact-diagonalization technique, which reduces the size of the matrices that must be handled to the square root of the overall size of the Hilbert space. Actually, real systems contain also vertical links via the orbitals of the apical oxygens. We expect that the inclusion of these hoppings does not change qualitatively the results since they do not contribute to the propagation of the bound pair in the lowest-order approximation.

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