Orbital Resonant Valence Bond States:
Emergence of Superconductivity in Doped Multiorbital Chains

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We introduce an “Orbital Resonant Valence Bond” (ORVB) state for two-orbital Hubbard models in one dimension that explains the recent computational discovery of pairing in these systems when hole doped. Our Ansatz is based on a mathematical expression where the undoped (one electron per orbital) quantum state of two nearest-neighbor (NN) sites coupled into a global spin singlet is rewritten exactly employing only spin-1/2 singlets linking NN orbitals. Generalizing to longer chains provides an ORVB variational state that can be visualized geometrically expressing our chain as a two-leg ladder, with one orbital per leg. As in Anderson’s resonating valence-bond state, ORVB describes the undoped ground state as a “liquid” of preformed NN singlet pairs that via doping become mobile. Real materials with one-dimensional substructures, two near-degenerate orbitals, and intermediate Hubbard $U/W$ strengths – $W$ the carrier’s bandwidth – could realize the ORVB state if on-site anisotropies are small. If these anisotropies are robust, spin-triplet pairing is possible.

**Introduction** — Quantum Materials merge topological concepts [1], such as in Haldane chains with nonlocal order parameters and edge states [2, 3], with electronic correlation effects, such as in iron-based superconductors with robust Hubbard $U$ and Hund $J_H$ couplings [4–7]. The Haldane chain started the field of topological materials and physical realization were reported in CsNiCl$_3$ [8], AgVP$_2$S$_6$ [9], NENP [10], Y$_2$BaNiO$_5$ [11], and PbNi$_2$V$_2$O$_8$ [12]. These chains have a spin gap as well as protected edge states when open boundary conditions (OBC) are used [3, 13–15]. In iron- and copper-based superconductors, most efforts employ planar geometries [16, 17] but Cu-oxide two-leg ladders were widely studied when they were predicted and confirmed to have a spin gap and become superconducting [18–22]. Recently, analogous developments occurred in iron materials when ladders BaFe$_2$S$_3$ [23–25] and BaFe$_2$Se$_3$ [26, 27] were reported to become superconducting with pressure. The study of iron ladders is vast and already revealed interesting complex magnetic states [23–42]. However, similar efforts using iron chains, such as in TlFeSe$_2$ or TlFeS$_2$, have been more limited [43–45].

Within Quantum Materials, the study of quasi one-dimensional (1D) ladders and chains is attractive. In this geometry powerful computational techniques, such as the Density Matrix Renormalization Group (DMRG) [46–49] and Lanczos [16] methods, allow for the theoretical study of model Hamiltonians with considerable accuracy. This removes the veil of theoretical uncertainty in higher dimensional systems that complicates the comparison theory vs experiment. In particular, this 1D avenue may allow the accurate study of systems where both topology and correlations are simultaneously relevant, the next grand challenge in Quantum Materials.

In this context, there are only a few studies of the effects of hole doping and possible magnetic-based hole pairing on topological interacting systems. Early work in the $t–J$ limit for doped $S=1$ chains, with double occupancy removed, indicated a narrow region of pairing, suppressed by a competition with ferromagnetism [50]. In related work, triplet superconductivity was also analyzed in 1D [51, 52]. More recent DMRG efforts using multiorbital Hubbard models of interacting electrons unveiled robust tendencies to spin singlet pairing, an ex-

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**FIG. 1.** (a) Sketch of a chain with two active orbitals a and b. (b) Representation of panel (a) splitting the orbitals into legs forming a fictitious two-leg ladder, with legs only connected by the Hund coupling $J_H$. (c) ORVB state proposed in the text. Arrows indicate spin-1/2 singlets (they are oriented objects) linking NN sites. The full ORVB state is a linear combination of all possible arrangements of these singlets. (d) Doped ORVB state: holes “h” are effectively paired when a preformed electron singlet is removed.
there are preformed triplets are variational, not exact, but they capture the essence of the results \[53, 54\]. The undoped and doped ORVB states pair by pairing, \(1(d)\), in agreement with the computational results but now in the enlarged space spanned by the real orbital ladder, \(1(b)\). We rely on a hereby proposed ORVB state: the orbital generalization of the much discussed resonating valence-bond concepts \[55, 56\]. We employ preformed spin-1/2 singlets as in the original formulation but now in the enlarged space spanned by the real chain in one direction and the orbital index in another, \(1(c)\). Doping this state with two holes in principle could break two singlets. But if these holes are close to one another, they can break only one singlet minimizing the energy and leading to an effective singlet hole pairing, \(1(d)\), in agreement with the computational results \[53, 54\]. The undoped and doped ORVB states are variational, not exact, but they capture the essence of the problem, as shown below.

Our overall conclusions are not obvious: at first sight there are preformed triplets at each rung due to the robust \(J_H/U\). In fact, we predict that triplet pairing—a rarity among superconductors \[57–59\]—should be stable when easy-plane anisotropies are not negligible. However, doping special quasi-1D materials—with two active fairly equivalent orbitals and weak spin anisotropy—should lead instead to singlet pairing.

Methods:

**Model**.— We use a canonical two-orbital Hubbard model composed of kinetic energy and interaction terms written as \(H = H_K + H_I + H_D\). The tight-binding portion of the Hamiltonian is

\[
H_K = \sum_{i,\gamma,\gamma',\sigma} t^{\gamma\gamma'} (c^\dagger_{i,\gamma\sigma} c_{i+1,\gamma'\sigma} + \text{H.c.}),
\]

where \(c^\dagger_{i,\gamma\sigma}\) \((c_{i,\gamma\sigma})\) creates (destroys) an electron at site \(i\) of a chain, orbital \(\gamma\) \((a\text{ and } b\text{ in our case, although our Hamiltonian notation is generic for arbitrary number of orbitals}), \text{ and spin projection } \sigma\). The nearest-neighbor electron hopping is chosen as a 2x2 orbital-space unit-matrix, i.e. \(t^{\gamma\gamma'} = t_{\delta\gamma\gamma'}\), with \(t\) the energy unit throughout the publication. The non-interacting bandwidth is \(W = 4.0t\). Note that the hopping symmetry between the two orbitals, and the absence of crystal-field splitting, prevents the appearance of the orbital-selective Mott physics recently studied in related multi-orbital models \[60–62\].

The electronic interaction is standard:

\[
H_I = U \sum_{i,\gamma} n_{i,\gamma\uparrow} n_{i,\gamma\downarrow} + (U' - J_H/2) \sum_{i,\gamma < \gamma'} n_{i,\gamma\uparrow} n_{i,\gamma\downarrow} - 2J_H \sum_{i,\gamma < \gamma'} \mathbf{S}_{\gamma\uparrow} \cdot \mathbf{S}_{\gamma'\uparrow} + J_H \sum_{i,\gamma < \gamma'} (P_{i,\gamma\uparrow} P_{i,\gamma'\downarrow} + \text{H.c.}).
\]  

(2)

The first term is the intraorbital Hubbard repulsion \(U\). The second contains the interorbital repulsion at different orbitals, with the usual relation \(U' = U - 2J_H\) due to rotational invariance. The third term involves the Hund’s coupling \(J_H\), and the last term represents the on-site interorbital electron-pair hopping \((P_{i,\gamma\uparrow} = c_{i,\gamma\uparrow}^\dagger c_{i,\gamma'\downarrow})\).

Finally, as shown below, it will also be important to incorporate an easy-plane anisotropy component \((D > 0)\)

\[
H_D = D \sum_i (S^x_i + S^y_i)^2.
\]  

(3)

The spin-1/2 operators \(S^x, S^y, S^z\) are defined using Pauli matrices as \(S^\dagger_{\gamma\sigma} = (1/2) \sum_{\sigma'\sigma} c^\dagger_{i,\gamma\sigma'\sigma} c_{i,\gamma\sigma'\sigma}\). For our results we used DMRG, with up to \(m = 1800\) states and typical truncation errors below \(10^{-5}\) as in previous investigations \[53\], as well as the Lanczos method.

Results:

**Undoped two-orbital Hubbard model at intermediate \(U/W\) vs Haldane state**.— Our focus on multi-orbital models originates in iron-based superconductors where ladders and chains can be synthesized, but our results are valid for other transition metal compounds. Iron superconductors are widely believed to be “intermediate” between weak and strong coupling, and \(U/W \sim 1\) is considered realistic \[4–7\]. Because the iron family is not at \(U/W \gg 1\), a pure spin model is not appropriate and interacting itinerant fermions must be used.

Let us address first whether the model discussed here— with mobile electrons, intermediate \(U/W\), and hopping unit matrix—is smoothly connected to the Haldane limit. At one particle per orbital and \(U/W \gg 1\) with concomitant growth of the Hund coupling fixed at the often used ratio \(J_H/U = 1/4 \[6, 63, 64\]—our model certainly develops \(S=1\) states at every site, antiferromagnetically Heisenberg coupled (we found numerically that the biquadratic coupling, possible in \(S=1\) spin systems, is negligible). To analyze if intermediate \(U/W \sim 1\) and strong coupling \(U/W \gg 1\) (with \(S=1\) spins onsite) are qualitatively similar, we compute with DMRG the entanglement spectra ES \[66\]. For example, working at \(U/W = 1.6\), where hole-binding is maximized as shown below, Figs. 2(a,b) indicate that with increasing \(J_H/U\) the Hubbard ES clearly resembles the \(S=1\) chain ES \[67\]. For example, our model is not merely a \(S=1\) chain. For example, the inset of Fig. 2(c) indicates that the von Neumann entropy \[68–70\] \(S_{VN}\) (see Appendix A.1) converges to \(\ln(2)\) \((S=1\) chain result\) only at \(U/W \sim 5\) and beyond. At typical couplings of iron compounds, \(S_{VN}\) is approximately double the \(U/W \gg 1\) limit. Thus, the
two-orbital Hubbard model qualitatively does resemble the Haldane chain, but at $U/W \sim 1$ there are quantitative differences likely caused by non-negligible charge fluctuations.

Consider now how the model evolves with increasing anisotropy $D/t$. Recent work found a transition between the gapped Haldane region and a gapped state with trivial topology [54]. In Fig. 2(c) indeed $S_{VN}$ at fixed $U/W = 1.6$ and $J_H/U = 0.25$ does not evolve smoothly from $D/t = 0$ – connected to the Haldane limit at large $U/W$ – to the anisotropic large $D/t$ “XY” limit. The ground state in this limit has a spin triplet with $S^z = 0$ projection at every site, and no edge states. At $0.1 < D/t < 0.2$, an abrupt change occurs and eventually $S_{VN} \rightarrow \ln(1)$ as $D/t$ grows, compatible with a product state of $S^z = 0$ triplets [see discussion later, Eq.(4)].

In summary, although with quantitative differences, the undoped Hubbard model qualitatively resembles the Haldane chain. This occurs as long as $D/t$ does not cross a threshold beyond which edge states disappear and a topologically trivial regime develops.

**FIG. 3.** Binding energy $\Delta E/t$ vs $U/W$ for various values of (a) $J_H/U$, at fixed $D/t = 0$, and (b) $D/t$, at fixed $J_H/U = 0.25$. In (a,b), a 16-sites OBC chain was used and DMRG. (c) Spin-singlet $\Delta_S$ real-space pair-pair correlations $P(R)$ vs distance $R$, varying $J_H/U$, at fixed $U/W = 1.6$ and $D/t = 0$. $\Delta_S$ involves NN sites and different orbitals. (d) Same as (c) but using the rung triplet operator $\Delta_T$, varying $D/t$, at fixed $U/W = 1.6$ and $J_H/U = 0.25$. In (c,d), a 48-sites OBC chain was used and DMRG, neglecting 8 sites at each end to avoid edge effects. Correlations are normalized to the result at distance 2, $P(2)$, to better focus on the large $R$ behavior. For definition of $\Delta_S$ and $\Delta_T$, see Appendix A.2. At (c) the hole doping is $x = 0.042$ corresponding to 4 holes, while at (d) $x = 0.083$ corresponding to 8 holes. $x$ is defined as number of holes divided by 96 (48 sites, 2 orbitals).

**Pairing in the doped two-orbital Hubbard model** — The main focus of this publication is why pairing occurs and why in the channel it occurs. However, before addressing these issues, let us first review and extend recent DMRG studies about hole pair formation and pair-pair correlations in the doped two-orbital Hubbard model. This analysis will provide hints for the intuitive explanation. In Fig. 3(a) the 2-holes binding energy vs $U/W$ is shown, parametric with $J_H/U$. This binding energy is defined as $\Delta E = E(2) - E(0) - 2[E(1) - E(0)]$, where $E(M)$ is the ground state energy with $M$ holes (zero holes refers to the half-filled state with one electron per orbital). When $\Delta E$ becomes negative, it signals a 2-holes bound state. Clearly, Fig. 3(a) indicates pair formation with maximum $|\Delta E|$ at $1 < U/W < 2$, as in [53], and growing with increasing $J_H/U$ (note $J_H/U$ should be less than $1/3$ to remain smaller than $U'/U$ due to the constraint $U' = U - 2J_H$). At $U/W \gg 1$, ferromagnetism for 1 and 2 holes – see discussion below – prevents pairing
suggesting that directly doping the S=1 chain may not be the proper theoretical approach. In Fig. 3(b) we show new results, now increasing $D/t$ at fixed $J_H/U = 0.25$. Robust pairing tendencies are observed again. However, the binding curves are almost identical at $D/t = 0.0$ and 0.2, but then they rapidly increase in magnitude. This reflects qualitative differences in the pairing properties, compatible with the von Neumann analysis with increasing $D/t$ in Fig. 2(c) that indicated a topological change in the same $D/t$ range.

FIG. 4. Qualitative phase diagram varying the easy-plane anisotropy $D/t$ and hole doping $x$, at fixed $U/W = 1.6$ and $J_H/U = 0.25$, using DMRG. Hole density $x = 0$ represents half-filling where at $D/t \lesssim 0.2$ the ground state is qualitatively connected to the S=1 Haldane phase. “ORVB” refers to the variational state introduced later in the text. At larger $D/t$, a product state of rung triplets with zero spin projection Eq. (4) is a good representation of the ground state. Upon doping at small $D/t$, first singlet pairing dominates until at $x \approx 0.1$ the spin/charge density wave (SDW/CDW) correlations – not shown – become stronger. Doping of the product state ($D/t \gtrsim 0.2$) leads to spin-triplet rung pairing over a broad range of doping because the natural “preformed” Cooper pairs are rung triplets. We used DMRG and $N = 48$ OBC chains to construct the phase diagram. Only a few points are shown with dots, but a denser grid $(x, D/t)$ was analyzed via DMRG.

The qualitative transition in Fig. 3(b) also occurs in Figs. 3(c,d) where pairing correlations are shown. At $D/t = 0$ and $J_H/U = 0.25$, when doping a region smoothly connected to the Haldane chain, spin-singlet pairing dominates (while triplet is exponentially suppressed, not shown). With increasing $D/t$ at fixed hole density $x$, a transition from singlet to triplet dominance is observed. For example, in panel (d) we observe that spin-triplet pairing, heavily suppressed at $D/t = 0$, instead becomes dominant as $D/t$ increases (while singlet is exponentially suppressed, not shown).

The “global summary” is shown in Fig. 4, where a large set of DMRG data is summarized in a phase diagram varying $D/t$ and doping $x$, with just a few representative points displayed. The red region near $x = 0$ and $D/t = 0$ is where the model resembles the Haldane state according to the entropy entanglement. Here, at light doping $x$ singlet-pairing dominates at intermediate $U/W$, but eventually other non-superconducting channels (SDW and CDW) take over at larger $x$. Increasing $D/t$, at small $x$ a transition from singlet- to triplet-dominated pairing occurs. In the singlet regime, holes are primarily located at nearest-neighbor sites and different orbitals, while in the triplet regime they are primarily at the same rung also in different orbitals.

The DMRG results unveiled a region of parameter space (small $D/t$, low hole doping, intermediate $U/W$, robust $J_H/U$) where superconducting spin-singlet correlations dominate. These results are surprising. First, the connection with the S=1 chain suggests that antiferromagnetic (AFM) fluctuations are short-range and it is unclear if they are sufficiently strong to generate pairing. Second, at every site and at intermediate-strong $U/W$ a nonzero magnetic moment develops due to $J_H$. Naively, these same-site electrons can be considered as local “preformed” triplets. Then, after hole doping the resulting ground state could be envisioned as involving these triplets that became mobile. For $D/t > 0.2$, Fig. 4, this naive perspective is confirmed numerically. Rung spin-triplet pairing dominates, and the product state wave function Eq. (4) is a good variational approximation (undoped system) which becomes exact as $D \to \infty$:

$$|TPS⟩ = \prod_{i}^{N} |1, 0⟩_{i} + \prod_{i}^{N} \frac{1}{\sqrt{2}}(|\uparrow_{ia}\downarrow_{ib}⟩ + |\downarrow_{ia}\uparrow_{ib}⟩). \quad (4)$$

Then, what originates the singlet dominance at small $D/t$? Although in a Haldane regime all triplet correlations must decay exponentially, such reasoning does not explain why singlet pairing is enhanced.Hints for the ORVB explanation presented below arise from the Affleck-Kennedy-Lieb-Tasaki (AKLT) exact solution [3], where a S=1 spin model was considered employing two auxiliary idealized S=1/2 degrees of freedom at every site. These auxiliary states form spin singlets with other S=1/2 auxiliary states at the next site. Below, we show that our two-orbital Hubbard model shares properties similar to this idea.

**ORVB state for the two-orbital Hubbard chain**

Here we introduce a variational state for both the undoped and lightly doped two-orbital Hubbard chain, at intermediate and strong $U/W$. We argue that these ground states can be qualitatively described in terms of S=1/2 spin singlets involving NN sites, connecting the same or different orbitals. **Our main result is that the small $D/t$ region of the two-orbital Hubbard model has hidden “preformed” singlets that become mobile with doping.** Knowing what type of Hubbard model develops pairing, and specifically what type of hoppings, allow us to formulate predictions on what characteristics a material must display to realize this physics.
We will propose a variational state inspired by an exact equality. Consider first only 2 sites, say 1 and 2, and construct the quantum global spin-zero state using one electron per orbital. With only one orbital this has the canonical expression $|\text{Singlet}_{S=1/2 \ 2-\text{sites}}\rangle = \frac{1}{\sqrt{2}}(|1/2, 1/2\rangle_{1/1,2} - |1/2, -1/2\rangle_{1/1,2}) = \frac{1}{\sqrt{2}}(|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle)$, where, $|1/2, 1/2\rangle$ means total spin 1/2 and z-projection $\uparrow$, etc.

For two spins 1, the global spin zero state is still relatively simple

$$|\text{Singlet}_{S=1 \ 2-\text{sites}}\rangle = \frac{1}{\sqrt{3}}(|1, 1\rangle_{1,2} + |1, -1\rangle_{1,2} + |1, 0\rangle_{1,2}) \quad (5)$$

Because in our case each site $S=1$ arises from two real $S=1/2$ electrons at each orbital and the same site, we use the notation $|1, 1\rangle_{1} = |\uparrow\uparrow\rangle_{1, b}, |1, -1\rangle_{1} = |\downarrow\downarrow\rangle_{1, a}, |1, 0\rangle_{1} = \frac{1}{\sqrt{2}}(|\uparrow\downarrow\rangle_{1, a} + |\downarrow\uparrow\rangle_{1, b})$, and an analogous expression at site 2. Then, simple algebra leads to

$$|\text{Singlet}_{S=1 \ 2-\text{sites}}\rangle = \frac{1}{\sqrt{3}} (|\uparrow\uparrow\rangle_{1,2} - |\downarrow\downarrow\rangle_{1,2} - |\downarrow\uparrow\rangle_{1,2}) \quad (6)$$

What is remarkable is that this last expression can be exactly rewritten as a combination of $S=1/2$ singlets, involving either different or the same orbitals:

$$|\text{Singlet}_{S=1 \ 2-\text{sites}}\rangle = -\frac{1}{\sqrt{3}} (|\uparrow\uparrow\rangle_{1,2} - |\downarrow\downarrow\rangle_{1,2}) - \frac{1}{\sqrt{2}} (|\uparrow\uparrow\rangle_{1,2} - |\downarrow\downarrow\rangle_{1,2}) - \frac{1}{\sqrt{2}} (|\uparrow\uparrow\rangle_{1,2} - |\downarrow\uparrow\rangle_{1,2}) \quad (7)$$

Much intuition is gained when this exact mathematical result is presented visually, as in Fig. 5, where we have rewritten exactly Eq.(7) doubling the number of valence-bonds states for an easier extrapolation to more sites.

Our exact result is counterintuitive: with perfect $S=1$ states at each site, the total spin-zero state of the two-orbital two-sites Hubbard model can be represented exactly as a linear combination of $S=1/2$ singlets. This is qualitatively in agreement with the AKLT perspective [3]. Our result Eq.(7) leads to our conjecture that a “hidden” $S=1/2$ orbital-resonating-valence-bond state emerges. Figure 5 resembles the original views of Anderson, Affleck et al., but now with orbitals considered as legs of a two-leg ladder, with these legs only connected by the Hund coupling (no interleg hopping).

**Lanczos overlaps** — How accurate is this state? Using Lanczos, we calculated the normalized exact ground state $|\text{GS}\rangle$ of the two-orbital Hubbard model in short chains and computed the overlap with a linear combination of the individual ORVB states of Fig. 6. The coefficients for each class were optimized to maximize the global overlap, arriving to a final normalized-to-one
FIG. 6. Individual ORVB states for the four-sites half-filled two-orbital Hubbard model with PBC. The color-framed states represent 4 distinct “classes”. A representative of each class is shown at the bottom with the same color convention. By applying translation and orbital exchange for each representative (reflection is not needed for the undoped case), we recover all the rest of the states shown in the upper frames. Note that the 16 ORVB states displayed are not orthogonal to one another.

state dubbed $|ORVB\rangle$. Care must be taken because the individual ORVB components do not form an orthogonal set since their overlap is finite. By this procedure, at $U/W = 20$ and 2 sites, the overlap $|\langle ORVB|GS\rangle|$ is virtually 1, because double occupancy is nearly entirely suppressed, as in Eq.(7). Using 4 sites, the binding energy is now optimized at $U/W = 4$. At this coupling and size, we found $|\langle ORVB|GS\rangle| = 0.95$ indicating that $|ORVB\rangle$ is a good representation of the ground state.

We carried out extensions to more sites using PBC. As already explained, below when “classes” are mentioned for the undoped case, they represent groups of ORVB states related by applying translations and orbital exchange to a particular representative. For 6 sites, there are 8 classes and at $U/W = 2$, where binding is maximized (not shown), the overlap is 0.79. As the lattice size grows, other configurations involving longer S=1/2 singlets and specially doubly occupied orbitals will contribute to the ground state because the optimal $U/W$ where binding is maximized is reduced reaching the intermediate range. But finding a robust overlap with 6 sites indicates that $|ORVB\rangle$ is a good variational state. The same occurs for 8 sites: here the number of classes grows to 16, and when $U/W = 1.5$ is chosen because it optimizes the binding energy, the overlap $|\langle ORVB|GS\rangle|$ for 8 sites remains robust at 0.61. As the system continues increasing in size, the optimal binding converges to intermediate $U/W$, see Fig. 3.

Doped ORVB — Now let us generalize the variational ORVB state to the doped case, to our knowledge a topic barely addressed in the resonating valence-bond context. In our DMRG studies in Fig. 3 and in previous efforts [53], we found that in the 2-holes ground state the largest-weight configuration occurs when holes are placed at NN sites and in different orbitals. Having the two holes in the same leg is not optimal because it leads to collisions: with one hole per leg they can move without obstacles, while taking advantage of the effective attraction in the ORVB arrangement. For this reason, and to reduce complexity, our proposed doped ORVB state will have only one hole per orbital and will be obtained primarily by taking the undoped ORVB and removing one diagonal singlet. This is exemplified for 4-sites PBC in Fig. 7 (left) where classes are shown. Note that the diagonal character of the $2\times2$ hopping matrix establishes that the number of holes per orbital is conserved.

However, two extra ingredients are needed for a proper physical description. First, quantum mechanically each hole in each orbital is “oscillating” (zero-point motion) via the intraorbital hopping because these are not frozen holes. Then, the configuration with 2 holes in the same rung must be included because it is generated in that oscillation within the bound state. Second, to avoid having unpaired S=1/2 electrons left and right of that 2-holes rung, a singlet across is required, as shown on the three
Classes for $N = 4$

2-holes

FIG. 7. A representative of each of the 7 ORVB classes used for a 4-sites chain with PBC and 2-holes doping. Each class state represents a linear combination involving translated, orbital $a < - > b$ exchanged, and parity-inverted states.

classes in Fig. 7 (right). Indeed it is known that a “π-shift” across-the-hole develops in the spin correlations of the two-orbital Hubbard ground state [53]. This also occurs in one-orbital $t-J$ models [71, 72]. For completeness, singlets across-the-hole were also added for diagonal hole configurations as in the bottom left class of Fig. 7.

This procedure resembles qualitatively the exact solution of the $U = \infty$ single-orbital Hubbard chain [73]: holes and spins are independent in this limit. Our mobile holes can be visualized as effectively inserted in between the original singlets of the undoped ORVB state.

The $|\text{ORVB}\rangle$ state generalized from Fig. 7 but now for 6 sites and 2 holes requires 23 classes [for 2 holes, to generate all states not only translational symmetry and exchange of orbitals must be used, but also reflection (parity) with respect to the middle]. The overlap with the 2-holes Lanczos exact GS at $U/W = 2$ is 0.59. For 8 sites and 2 holes, 84 classes are needed, and the overlap at $U/W = 1.5$ is 0.48. These are good numbers, but more important is how qualitatively these states capture the essence of the problem. For instance, from the 6-sites exact GS the spin-spin correlations can be measured using special operators to project [53, 74] the mobile holes to their most likely position in the state via projector operators [53, 74], and then spin-spin correlations are measured. Blue (red) lines represent AFM (FM) bonds with line-thickness proportional to the magnitude of spin correlations. (c) Total ground-state spin quantum number vs $U/W$, for 0, 1, and 2 holes. All calculations (a,b,c) are performed using Lanczos on a 6-sites PBC chain, at $U/W = 2.0$, $J_H/U = 0.25$, and $D/t = 0$.

Panel Fig. 8 (c) also shows that at very large $U/W$ the pairing and overall proposed picture breaks down. In this regime, the Hund coupling is so large that “double-exchange” physics dominates and the system turns globally ferromagnetic to improve the kinetic energy of the now unbounded holes. This shows that simply doping the S=1 Haldane chain may not be a proper path, but $U/W$ must be reduced to intermediate values to avoid the ferromagnetic competition. However, in the hole-binding range the total spin quantum numbers are qualitatively compatible with those of the ORVB picture.

Varying $D/t$ — When easy-plane anisotropies are included, the spin-triplets product state at each rung, $|\text{TPS}\rangle$ Eq.(4), becomes increasingly a better approximation as $D/t$ grows (at $D/t \to \infty$, $|\text{TPS}\rangle$ is the exact ground state). This evolution is illustrated in Fig. 9. In (a), the half-filling overlaps $|\langle \text{ORVB}|GS\rangle|$ and $|\langle \text{TPS}|GS\rangle|$ are shown. The ORVB (TPS) overlap decreases (increases) with increasing $D/t$, as expected. Note that the TPS state used here is crude: larger overlaps in the $D/t$ range shown could be obtained adding fluctuations but this is irrelevant for our main focus i.e. the origin of the spin-singlet pairing at small $D/t$. 
and the number of orbitals in the short axis. Using DMRG and Lanczos, we find excellent agreement with the ORVB predictions at intermediate $U/W$. The entanglement spectra and von Neuman entropy indicate that the undoped intermediate $U/W$ regime is analytically connected to the Haldane limit at $U/W \gg 1$. However, in the realm of spin models at $U/W \gg 1$, a strong competition with ferromagnetism upon doping prevents pairing from occurring.

Our ORVB state relies on a mathematical expression involving two $S=1$ spins at NN sites, with one electron per orbital. The global spin-zero state of these two sites is written exactly exclusively using spin-1/2 singlets linking electrons at those NN sites, involving the same or different orbitals. Extended to more sites, the ORVB state is a linear combination of spin-1/2 singlets in all possible NN arrangements. Note that same-rung singlets are excluded because of the large ferromagnetic Hund coupling. Using Lanczos on finite chains, the ORVB was shown to be a good approximation to the true ground state for small easy-plane anisotropy $D/t$, robust Hund coupling $J_H/U$, intermediate $U/W$, and light hole doping.

The preformed NN spin singlets – resembling Cooper pairs – become mobile upon hole doping, and form a coherent superconducting state, at least within the limitations of one dimensionality that only allow for power-law decays. A weak coupling among chains will render the state truly superconducting with long-range order, as in two-leg Cu- or Fe-based ladders. Our results explain the unexpected spin-singlet dominance observed computationally in a regime that naively contains preformed on-site rung triplets. These triplets indeed do condense into a triplet superconductor but only increasing $D/t$ beyond a finite threshold. This is also an exotic state since triplet superconductivity is rare.

In summary, our study combines topological concepts with correlation effects. Pairing emerges with hole doping via the liberation of preformed spin-1/2 singlets already contained in the undoped ORVB state. For experimentalists to realize our model the challenge is to find quasi-1D materials with two nearly-degenerate dominant active orbitals, and with similar overlaps along the chain so that the hopping matrix is nearly the unit matrix, as in our calculations. How robust the Haldane regime is with regards to small deviations from this hopping symmetric case, as well as the introduction of an orbital small crystal-field splitting, remain to be investigated (the orbital-selective Mott phase is close in parameter space, as recently shown [60–62]). Doping the existing physical realizations of undoped Haldane chains [8–12] can provide a starting point towards the predicted superconductivity, but again we remark that intermediate $U/W$ is a more attractive parameter region than $U/W \gg 1$. For this reason the iron-superconductors family provides a natural starting point, although realizations could also be found in other multiorbital-active compounds.

![FIG. 9. Overlap $|\langle \text{ORVB}|GS \rangle|$ and $|\langle \text{TPS}|GS \rangle|$ vs $D/t$ for (a) zero hole (half-filling) and (b) two holes. The discontinuity in (b) indicates a first-order transition due to a level crossing and associated change in the ground state quantum numbers under orbital exchange. (c) Ground-state chain energies, as well as their derivatives to emphasize sudden slope changes, for two holes $E_{N-2}$. All results (a–c) obtained with Lanczos using $N = 6$ and 8 sites and PBC.](image-url)
Appendix:

A.1 Entropy — In the main text, we present calculations of the entanglement spectrum and the Von Neumann entanglement entropy. These results are obtained by creating a bipartite system, as naturally occurs in the DMRG algorithm. Labeling these partitions A and B, one can calculate the density matrix of sub-system A by tracing over the B sub-section of the full density matrix, defining the reduced density matrix \( \rho_A = \text{tr}_B(\rho_{0}) \). Finally, using the eigenvalues of \( \rho_A \), the von Neumann entanglement entropy can be defined

\[
S_{VN} = - T [\rho_A \ln(\rho_A)] = - \sum \rho_{ii}^A \ln(\rho_{ii}^A) \quad \text{in the diagonal representation.} \tag{8}
\]

It is well known that the eigenvalues of the ground-state reduced density matrix of a generic bipartition of the lattice correspond to the squares of the singular values of a Schmidt decomposition of the many-body wave-function written as a matrix product state \([47]\). These singular values represent the correlations or entanglement between sub-system matrices as determined by tracing over the B sub-section of the full density matrix, defining the reduced density matrix \( \rho_A \).

A.2 Operators — To address pairing, we defined a general pair creation operator as

\[
\Delta_{(i,j),\pm}^{\gamma \gamma'} = \frac{1}{\sqrt{2}} (c_{i,\gamma,\gamma'}^{\dagger} c_{j,\gamma',\gamma}^{\dagger} \pm c_{i,\gamma',\gamma}^{\dagger} c_{j,\gamma,\gamma'}^{\dagger}), \tag{9}
\]

where \( i, j \) are sites, \( \gamma, \gamma' \) are orbitals (a or b), and \( \pm \) sign represents a spin singlet or triplet. We only focused on two different pair operators: (1) nearest neighbor pair that is odd under spin (singlet) and under orbital exchange (\( S^z_{nn} \) below), (2) on-site inter-orbital pair that is even under spin (triplet) and under orbital exchange (\( T^z_{nn} \)). These are defined as

\[
\Delta_S = S^z_{nn}(i) = \Delta_{(i,i+1),-}^{ab} + \Delta_{(i,i+1),+}^{ba} \\
\Delta_T = T^z_{nn}(i) = \Delta_{(i,i),+}^{ab}
\]

In the main text, we often refer to \( S^z_{nn} \) as odd diagonal singlet and to \( T^z_{nn} \) as rung triplet, where “diagonal” and “rung” refers to the ladder representation of a two-orbital chain Fig. 1. Using these pair creation operators, we study the decay of the pair-pair correlations

\[
P_S(R) = \frac{1}{N_R} \sum_i (S^z_{nn}(i) S^z_{nn}(i + R)),
\]

\[
P_T(R) = \frac{1}{N_R} \sum_i (T^z_{nn}(i) T^z_{nn}(i + R)),
\]

where \( N_R \) represents the number of total neighbors at distance \( R \) with respect to site \( i \), summed over all sites.

Code and Data Availability — The computer codes used in this study are available at https://g1257.github.io/dmrgPlusPlus/. The data that support the findings of this study are available from the corresponding author upon request.

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