Quantum loop states in spin-orbital models on the honeycomb lattice

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We construct a physically realistic and analytically tractable model for spin-1 systems with orbital degeneracy on the honeycomb lattice, relevant to honeycomb materials with large Hund’s and weak spin-orbit couplings, and two electrons in $t_{2g}$ orbitals. This model realizes many new phases whose building blocks are orbital loops decorated by Haldane chains. These include a Haldane loop crystal, a symmetry-protected topological phase, and, notably, a regime where the decorated loops resonate. When taken to the three-dimensional hyperhoneycomb lattice, the latter regime becomes a (symmetry-enriched) $U(1)$ quantum spin-orbital liquid, “disordered” both in the spin and orbital channels. We hope this construction will pave the way for realizing many of the Haldane-chain-based phases which have been theoretically proposed in the literature.

The wide variety of proposals for exotic ground states of many-body Hamiltonians calls for physically-realistic models prone to yield such states. Among those are, for example, quantum spin liquids (QSLs) and interacting topological insulators (fermionic or bosonic), also known as “symmetry-protected topological” (SPT) phases. A very general characteristic of these phases is the existence of fractional excitations, either in the bulk or at the edge of the system. The original proposal of Anderson for QSLs involved “resonating valence bonds,” i.e. coherent superpositions of singlet coverings of the lattice [1 2]. More recently, proposals for both QSLs [3-5] and SPTs [6-8] have emerged which are now based on fluctuating chains rather than singlets. More precisely, the building blocks are Haldane-like chains [10], which are featureless in their “bulk” but host protected gapless states confined to their ends. This is clearest in the AKLT chain (a representative state of the Haldane phase) [10], where each spin one is rewritten as two spin half’s subsequently projected back onto the $S = 1$ representation at each site, and singlets form astride each bond. In this picture, two “free” $S = 1/2$ are indeed left at each end of open chains. The Haldane states are themselves one-dimensional SPTs; in the two and three-dimensional Haldane-based QSL and SPT constructions, their physical supports fluctuate and their ends act as the bulk or edge fractional excitations. While such wavefunctions and even parent Hamiltonians have been proposed, it has remained far from obvious how they could be achieved in a realistic setting, let alone an actual material.

Independently, concrete spin-orbital (“Kugel-Khomskii”) [13, 14] models, which capture single-site spin and orbital degeneracies, have been shown to host a rich spectrum of phenomena [14], notably, valence bond solids [15-23] and orbital liquids [21, 23-28]. The crucial ingredient is the modulation of the effective spin exchange strength, which allows for stronger and weaker bonds to form, owing to the relationship between effective exchange strength and orbital overlap.

Here, we show that orbital degrees of freedom provide a simple loop-forming mechanism, and allow to naturally realize the AKLT chain ground state picture. Specifically, we construct a spin-orbital model, i.e. a model with orbital degeneracy, on the honeycomb lattice for $S = 1$ and effective $L = 1$, which supports fluctuating Haldane chains (subtended by “orbital loops,” i.e. closed strings of bonds with large orbital overlap), a Haldane chain based SPT, as well as a hexagon Haldane loop crystal with “Haldane-gap wave” excitations. When taken to the three-dimensional hyperhoneycomb lattice, the model is also home to a fully-fledged symmetry-enriched $U(1)$ Coulombic spin-orbital liquid and a fractionalized antiferromagnet.

We proceed as follows. We first introduce the appropriate ingredients and mathematical formalism and derive the minimal realistic model which induces the formation of fluctuating loops. Then, we analyze in detail the pure orbital part of the Hamiltonian and show how orbital loops emerge, before introducing spin degrees of freedom. The addition of large spin exchange produces new, fluctuating, decorated loops. Along the way we derive results in a large portion of the phase diagram we set out to study.

We consider two electrons at each site of a honeycomb lattice, in degenerate $t_{2g} = \{d_{xz}, d_{yz}, d_{xy}\}$ orbitals (which we also denote for convenience $x, y$ and $z$ orbitals, respectively) [29]. We assume large Hund’s coupling $J_H$, which enforces the high-spin state $S = 1$, and large intra-orbital repulsion $U$, which imposes no more than one electron per orbital. There are then two occupied and one empty orbital at each site, and the site Hilbert space is $H =$

FIG. 1. An orbital and Haldane loop covering of a section of the honeycomb lattice.
The orbital space basis \([|x\rangle, |y\rangle, |z\rangle\) is defined such that in state \(|x\rangle\) the \(x\) orbital is empty while the other two are filled, and similarly for \(|y\rangle\) and \(|z\rangle\) (see Fig. 2). A set of nine operators acting in this space can be chosen to be \([L^\mu, P^\mu, T^\mu]\) with \(\mu = x, y, z\), such that \(L^x = i(|z\rangle\langle y| - |y\rangle\langle z|), P^x = 1 - |x\rangle\langle x| = \tilde{n}^x\) and \(T^x = -(|z\rangle\langle y| + |y\rangle\langle z|)\) and cyclic permutations. The \(L^\mu\) operators are Hermitian, obey the angular momentum algebra, and are such that \(L^\mu P^\mu = 0\). \(P^\mu\) is a projection operator which measures the occupation of the \(\mu\)-orbital, so that the two-electron-per-site constraint is written \(\sum_{\mu} P^\mu = L(L+1) = 2\). Moreover, \([P^\mu, P^\nu] = 0\).

**FIG. 2.** a) The honeycomb lattice embedded in a cubic structure. Honeycomb planes are perpendicular to \((111)\) axes, here the [111] axis. b) The \(t_{2g}\) orbitals shown in a cubic environment, surrounded by a putative octahedral cage. c) Pictorial representation of the \(|x\rangle, |y\rangle, |z\rangle\) states. In state \(|x\rangle\), the \(d_{xz}\) orbital is empty, while orbitals \(d_{xy}\) and \(d_{xz}\) each contain one electron. For clarity, only the lobes in the bond directions are shown.

We now write the minimal physically realistic Hamiltonian acting in \(H\), including only nearest-neighbor interactions, which realizes a resonating chain regime. We assume isotropy in spin space (no spin-orbit couplings) and a local cubic environment (necessary for \(t_{2g}\) orbitals). This Hamiltonian is:

\[
H = \sum_{\langle ij \rangle} \left( P_i^{\gamma_{ij}} P_j^{\gamma_{ij}} \left[ -\zeta + J (S_i \cdot S_j + \beta (S_i \cdot S_j)^2) \right] - \nu \left[ T_i^{\gamma_{ij}-1} T_j^{\gamma_{ij}+1} + \text{h.c.} \right] \right). \tag{1}
\]

Except where otherwise noted, we take \(\zeta, J \geq 0\) and \(-1 \leq \beta \leq 1\). \(\nu\) can always be chosen positive, up to a gauge transformation (see Supp. Mat.). \(\gamma_{ij} = x, y, z\) denotes the bond type of bond \((ij)\) (in a cubic environment each of the three types of honeycomb bonds is orthogonal to a different cubic axis \(x, y, z\) and may be thereby labeled, see Fig. 2), and \(x \pm 1 = y, z\) etc.. For example, if \((ij)\) is a \(z\)-type bond,

\[
H_{(ij)\in z} = P_i^z P_j^z \left[ -\zeta + J (S_i \cdot S_j + \beta (S_i \cdot S_j)^2) \right] - \nu \left[ T_i^z T_j^z + \text{h.c.} \right]. \tag{2}
\]

The physical relevance of the \(\zeta, J, \beta, \nu\) parameters is rooted in the relative geometry of the \(t_{2g}\) orbitals and honeycomb bonds. Indeed the, e.g., \(d_{xy}\) (or “z”-) orbitals at each end of a \(z\) bond have a large overlap while all other overlaps are weak (see Fig. 2): the first term in Eq. (1) enforces precisely this concomitance of bond and orbital filling types across a bond. All terms in Eq. (1) arise from standard orbital-dependent superexchange mechanisms \([20, 21]\), and Eq. (1) with \(\nu = 0\) and \(\zeta < 0\) was studied in detail in Ref. \([20, 21]\).

We now proceed to the analysis of this model.

**Orbital sector: fluctuating orbital loops.**—First, we set \(J = 0\), and investigate the orbital part of the Hamiltonian, i.e.,

\[
H_{\text{orb}} = \sum_{\langle ij \rangle} \left( -\zeta P_i^{\gamma_{ij}} P_j^{\gamma_{ij}} - \nu T_i^{\gamma_{ij}-1} T_j^{\gamma_{ij}+1} + \text{h.c.} \right). \tag{3}
\]

**Static loops.**—To begin, we also focus on \(\nu = 0\), in which case the Hamiltonian is exactly soluble. Indeed, \(H_{\text{orb}}\) then reduces to a (classical) Potts model \(H_0 = -\zeta \sum_{\langle ij \rangle} P_i^{\gamma_{ij}} P_j^{\gamma_{ij}} \) \([20]\), with \([P^\mu, P^\nu] = 0\) and \([H_0, P_i^\mu] = 0\ \forall i, \mu, \nu\). For \(\zeta > 0\), because \(P_i^\mu\) measures the occupation of the \(\gamma\) orbital at site \(i\), on each bond \((ij)\) the energy is minimized when both orbitals \(\gamma_i\) at each end are filled, in which case we say the bond is “covered.” Because there are two electrons per site, \(\sum_i P_i^\mu = 2\), the configuration where two covered bonds stem out of every site (forming a two-bond string) is favorable energetically (see Fig. 4). The (fully-packed) loop coverings of the lattice implement this condition throughout the lattice and constitute the highly-degenerate ground state manifold of \(H_0\) \([25]\). Owing to the orthogonality of \(t_{2g}\) orbitals at the same site, all loop coverings are strictly orthogonal to one another \([19]\). This is in contrast to many dimer models where the dimers are two-spin-1/2 singlets. The elementary excitations of \(H_0\) (which take one out of the loop covering manifold) are loop “cuts”: a loop is cut open, which creates nearby two (“defect”) bonds covered by one orbital rather than two or zero (see Fig. 4c)).

For \(H_0\), while a loop cut costs an energy \(\zeta\) locally creates two defect bonds, once created the two defect bonds may travel infinitely apart at no further energy cost. This is of course reminiscent of the classical spin ice problem, where a spin flip creates two monopoles which can (quasi-)freely separate.

**Orbital fluctuations.**—We now consider a non-zero but small \(\nu \ll \zeta\). This gives dynamics to the loops, since now \([H, P_i^\mu] \neq 0\). In degenerate perturbation theory in the \(\nu = 0\) manifold, the lowest-order effective Hamiltonian is

\[
H_{\text{eff}} = -\frac{12\nu^3}{\zeta^2} \sum_{\text{O}} W_{\text{O}}, \tag{4}
\]

where the sum is taken over all hexagons (or “plaquettes” of the lattice), and corresponds to the “flip” terms given pictorially by:

\[
H_{\text{flip}} = -t \sum_{\text{O}} \left( |\circlearrowright\rangle \langle \circlearrowright| + |\circlearrowleft\rangle \langle \circlearrowleft| \right), \tag{5}
\]

(microscopically, \(t = 12\nu^3/\zeta^2\)). In Eq. (4), in terms of
FIG. 3. Phase diagram in 2d in the $V/t - J/t$ plane ($t = 12v^3/\zeta^2$), for $J \geq 0$ and $\zeta > 0$. The phase diagram of the pure plain loop model (as obtained in Ref. 36) is shown below the horizontal axis. In the intermediate $J/t$ region, the location and nature of the phase transitions are speculative. The solid and dashed lines and the white dot represent putative second and first order transitions, and critical end point, respectively. Thick blue lines represent orbital overlaps, yellow contours Haldane chains, and red and blue circles up and down spins.

FIG. 4. a) Orbital loop covering of the lattice. b,c) An elementary defect in the loop covering: b) at the loop cut, c) after part of the defect traveled. When the orbital loops are decorated by Haldane chains, the (purple) chain ends also carry a spin-1/2.

the $T$ operators

$$W_0 = \mathcal{P} T_1^x T_2^x T_3^x T_4^x T_5^x T_6^x \mathcal{P}$$

where $\mathcal{P}$ is the projection onto the loop-covering manifold and where the sites $1, \ldots, 6$ are defined around a hexagon as in Fig. 2). Hexagons with alternating covered and empty bonds, such as those in Eq. (5) are called “flip-pable.”

In dimer problems, it is customary to introduce a term, the Rokhsar-Kivelson potential [37], which counts the non-matching bonds obtained from loop cuts discussed above. It is a $U(1)$ quantum orbital liquid, with a gapless (quadratic) photon mode.

It is a well-known result, however, that, in 2+1 dimensions, the deconfined phase of $U(1)$ Coulombic gauge theories is unstable [39], so that, in our model, the quantum orbital liquid regime does not exist as a phase in an extended region of the phase diagram, but survives only at the RK point. Away from the RK point, the system instead releases its entanglement, breaks symmetries, and orders for both $V > t$ and $V < t$ into the phases shown below the horizontal axis in Fig. 3 and in Fig. 6a,c). At $V/t > 1$, the system immediately orders into ground states which feature static “parallel” chains which extend through the whole system. For $V/t < 1$, the system first enters an “intermediate phase” where $0 < \langle P_i^{\gamma_1} P_j^{\gamma_j} \rangle < 1$, before hitting a first-order phase transition below which the system favors one of the three “maximally flippable” “hexagon loop crystal” configurations (see figure in Supp. Mat.).

We note that the model presented here can be generalized to three dimensions (on the hyperhoneycomb lattice [40], which shares with the honeycomb lattice the same essential ingredients), where Coulombic phases of $U(1)$ gauge theories are stable [39, 41, 42]. Details are beyond the scope of this paper, but will be addressed in an upcoming publication [43]. Stable two-dimensional generalizations, such as those allowing for a $\mathbb{Z}_2$ spin liquid, are also possible.

Spins. — We now finally introduce the spins, i.e. consider $J \neq 0$. First, we note that the spin operators appear only in

$$H_J = \sum_{\langle ij \rangle} \tilde{J}_{ij} (S_i \cdot S_j + \beta (S_i \cdot S_j)^2),$$

where $\tilde{J}_{ij} = JP_i^{\gamma_1} P_j^{\gamma_j}$, which, when considered as a 1d problem with constant $\tilde{J}_{ij} > 0$ and $-1 \leq \beta \leq 1$, realizes the Haldane phase (and in particular the AKLT state described in the introduction at $\beta = 1/3$). Notably, the spin exchange is “modulated” by the operator $P_i^{\gamma_1} P_j^{\gamma_j}$, and vanishes when $P_i^{\gamma_1} P_j^{\gamma_j} = 0$. Therefore, when the system forms orbital loops, the problem in spin space reduces to a collection of purely one-dimensional periodic $S = 1$ Hamiltonians, which are minimized by entering the Haldane phase. This leads to the appearance of new structures, namely Haldane decorated...
loops, where each orbital loop subtends a Haldane chain, \(|\mathcal{L}| = |\mathcal{L}| \otimes |\psi_{\text{Hald}}|\), where \(|\mathcal{L}|\) is an orbital loop, and \(|\psi_{\text{Hald}}|\) the Haldane spin ground state. Interestingly, the decoration in general introduces a length-dependent energy density. Indeed, away from \(\beta = 1/3\), the energy density (the energy per site, or bond) of periodic AKLT chains is independent of their length, the energy density of length-six loops is always smaller than that of longer loops (see Supp. Mat. for results obtained using DMRG). In turn, this has consequences on the energetics of the loop coverings, which may become inequivalent.

**Static coverings:** \(\nu = 0\).—In the absence of orbital fluctuations, i.e. when \(\nu = 0\), the ground state manifold of the pure orbital model is that of all loop-coverings, as discussed above. In particular, all loop coverings are degenerate in energy, regardless of the distribution of their loop lengths. If we now consider \(J > 0\) and \(J \ll \zeta\), at first order in perturbation theory in \(\zeta/J\) (\(H_J\) perturbs \(H_0\)), spin states break the degeneracy of the loop coverings, following \(\langle \mathcal{C} | H_J | \mathcal{C} \rangle\) where the \(|\mathcal{C}\rangle\) are the otherwise-degenerate decorated loop coverings. At \(\beta = 1/3\), we retain an exact degeneracy between Haldane-decorated loop configurations, which all together form the ground state manifold, while, away from \(\beta = 1/3\), \(\langle \mathcal{C} | H_J | \mathcal{C} \rangle\) is only minimized when the system forms one of three equivalent “hexagon crystal” states where the lattice is covered by decorated loops of length six (see Supp. Mat.).

This static-orbital regime corresponds to the infinite \(J/t\) limit on Fig. 3.

We now introduce the orbital kinetic terms, distinguishing between different \(J/\nu\) regimes.

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**Large \(J/\nu\) limit.**—In the large \(J/t\) limit, we first consider the Hamiltonian

\[
H_{\text{stat}} = \sum_{ij} P_i^{(\tau)} P_j^{(\nu)} \left(-\zeta + J (S_i \cdot S_j + \beta (S_i \cdot S_j)^2)\right),
\]

(9)

and introduce the kinetic terms \(\nu\) in perturbation theory. Even with \(J > 0\) (and possibly \(J \sim \zeta\)), the eigenstates of \(H_{\text{stat}}\) are still eigenstates of the \(P_i^{(\nu)} = \tilde{n}_i^{(\tau)}\), and as discussed above \(\tilde{J}_{ij} = J P_i^{(\tau)} P_j^{(\nu)}\) is only nonzero when \(\tilde{n}_i^{(\tau)} = \tilde{n}_j^{(\nu)} = 1\). Therefore ground states of \(H_{\text{stat}}\) belong to the set of decorated loop covering, so long as \(-\zeta + \epsilon_{\text{Hald cov}} < 0\), where \(\epsilon_{\text{Hald cov}}\) is the energy density of the collection of all pure Haldane chains in the covering. For example, for \(\beta = 0\) (resp. \(\beta = 1/3\)), this is true for any \(\zeta > \epsilon_{L=\infty}(0) \approx -1.40J\) (resp. \(\zeta > \epsilon_{L=\infty}(1/3) = -2/3J\)).

When \(\beta = 1/3\), the manifold of decorated loop coverings (which are ground states of \(H_{\text{stat}}\)) is highly degenerate, and we call \(\mathcal{C}\) the projector onto this manifold. In perturbation theory in small \(\nu/(-\zeta + \epsilon_{\text{Hald cov}})\), our analysis of the pure orbital model informs us that the lowest order contribution arises at third order, provided \(H'_{\text{eff}} = \mathcal{C} | H_{\text{flip}} \otimes \mathbb{1}_S | \mathcal{C}\) does not identically vanish. Indeed, \(H_{\text{flip}}\) is the lowest-order orbital-space Hamiltonian to take one orbital loop covering into another, and one must check that the corresponding Haldane loop coverings have non-zero overlap. Remarkably, we find that the overlap between two AKLT loop coverings (“cov”) which differ by a single “plaquette flip” is always equal to 1/4, up to exponentially-small corrections in the lengths of the (rearranged) loops, i.e.

\[
\langle \mathcal{C}_1 | (H_{\text{flip}} \otimes \mathbb{1}_S) | \mathcal{C}_2 \rangle = \langle \text{AKLT cov}_1 | \text{AKLT cov}_2 \rangle \approx \frac{1}{4},
\]

(10)

regardless of how many loops are connected by a plaquette flip \([44]\). This result is obtained using the matrix product state (MPS) formalism (see Supplemental Material). In fact, Eq. (10) is a special case of \(\langle \otimes \text{AKLT}_1 \otimes \text{AKLT}_2 \rangle \approx 1/2^{\nu_{\text{cuts}}-1}\), where \(\nu_{\text{cuts}}\) is the number of loop cuts needed to connect \(|\psi \otimes \text{AKLT}_{1,2}\rangle\) (see Fig. [5]). This momentous result is a consequence of the ultra short range entanglement of the AKLT state, which is “close” to being a product of single-site states.

Away from \(\beta = 1/3\), but within the Haldane phase (\(|\beta| \leq 1\)), we expect the same results to hold since entanglement properties are characteristic of a phase. Therefore, at large \(J/t\), and for \(\beta = 1/3\) and \(V/t = 1/4\) (“decorated RK point”), the system is “close to” \([11]\) a \(U(1)\) phase with spinful fractional excitations, in the sense that it contains large fluctuating Haldane-decorated loops. This is a state the model was designed to achieve. The lowest-energy excitations are either loop cuts (necessarily accompanied by a Haldane chain cut) or pure Haldane excitation, depending on the distribution of loop length and values of \(\zeta\) and \(\beta\) \([46]\). Local loop cuts generate two orbital chain ends, which are decorated by spin-1/2. In three dimensions, where the orbital \(U(1)\) deconfined phase survives away from the RK point, these chain ends are deconfined spinons.

**Small \(J/\nu\) limit.**—In the small \(J/t\) limit, the orbital-only model, i.e. \(H_{\text{orb}}\) from Eq. (6), is solved first and spin exchange \((J)\) is then introduced perturbatively. This results in an effective exchange pattern for the spins. More precisely, when \(J = 0\), spin space is completely degenerate, but the orbital ground state is a priori unique (or discretely degenerate due to symmetry-related states). Therefore, upon introducing \(J\), in degenerate perturba-
tion theory, we have, at first order,

$$H_{\text{eff}}'' = \sum_{(i,j)} \left[ JP_i^{\gamma_i} P_j^{\gamma_j} (S_i \cdot S_j + \beta (S_i \cdot S_j)^2) \right] p(i,j)$$

$$= \sum_{(i,j)} \langle P_i^{\gamma_i} P_j^{\gamma_j} \rangle J (S_i \cdot S_j + \beta (S_i \cdot S_j)^2),$$

where $p$ is the projector onto the $J = 0$ ground state, and $(..)$ is the expectation value taken in this ground state. We obtain the phase diagram within the pure loop model by using the results from Ref. [36] and a simple variational approach (see Supp. Mat. for a detailed derivation). Notably, the Haldane loop decoration in the extended phase at large $V/t > 0$ gives way to a two-dimensional weak SPT phase, with neutral Kramers doublet [37] edge states (the edge orbitals decorated by spin-1/2 degrees of freedom) protected by translational symmetry, provided the boundaries are appropriately chosen (and, strictly speaking, provided a weak coupling at the boundary exists). It is noteworthy that this phase is realized spontaneously, i.e. this is not an explicit “chain-stacking” construction.

To our knowledge, this is the first such example in the literature.

In this regime ($\zeta > 0$, $\zeta \gg J > v$) low-energy excitations are expected to occur in the spin in the antiferromagnetic (AFM) phase, those are simply the conventional spin flips. In the decorated chain phases, the elementary excitations are those of the (gapped) Haldane chains. Remarkably, in the length-six loop state, which is a product state of decorated hexagon loops, the excitations are local, but a weak coupling between the hexagons (e.g. when $v \neq 0$) will lead to slightly-dispersive “Haldane gap waves,” observable for example in neutron scattering.

The results derived above are summarized in the phase diagram in Fig. 6.

**Discussion.** —In summary, we have exposed a physical mechanism for the realization of fluctuating Haldane chains in spin-orbital models in two dimensions. To do so, we presented a realistic and analytically-tractable spin-orbital Hamiltonian on the honeycomb lattice, with a rich phase diagram, featuring exotic phases built out of Haldane chains. Among those are a translational-symmetry protected topological phase, with spin-1/2 edge excitations, a Haldane hexagon loop “crystal,” with “Haldane gap wave” excitations, and a regime with fluctuating Haldane chains coupled to underlying “orbital loops.” On the three-dimensional hyperhoneycomb lattice the latter becomes a Coulombic quantum spin-orbital liquid, a unique example in the spin-orbital literature of a controllable model where both the spin and orbital sectors are “disordered.” Moreover, supplementing the model with additional terms is likely to allow accessing more phases and possibly interesting phase transitions. In fact, many more avenues—in several different fields—will be worth exploring further. For example, the quantum spin liquid can be induced not only by taking the model to three dimensions but also by turning it into a $\mathbb{Z}_2$ liquid. The variation of the parameter $\zeta$ or the number of electrons per site may also lead to interesting problems and phase transitions. In general, the highlighted mechanism will hopefully be an important stepping stone for future studies to realize Haldane chains and other low-dimensional structures in higher dimensions.

Most exciting would certainly be the discovery in real materials of some of the phenomena described here. This model is relevant to insulating honeycomb materials with two electrons in degenerate $t_{2g}$ orbitals and large Hund’s coupling to enforce $S = 1$. In practice, materials need to have (at least approximate) cubic symmetry, weak spin-orbit coupling and large direct orbital overlap. Therefore, materials based on Ru, Ni, V, etc. naively appear as potential candidates. Regardless, it will be important and interesting to study the breaking of any of these constraints, through e.g. spin-orbit coupling or symmetry lowering, inevitable at some level in real materials. Magneto-elastic coupling should also be investigated. It might well play a role similar to the $V$ term in stabilizing the “extended” or “flippable” phases.

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The loop covering manifold is isomorphic to the more familiar set of 1/3-plateau states, or 2/3-filling hard-core bosons, on the kagomé lattice by identifying a covered 3-plateau states, or 2/3-filling hard-core bosons, on the kagomé lattice by identifying a covered

honeycomb bond with an “up” spin-1/2, or a hard-core boson, on the center of the bond.

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1. Construction of the states and operators

Let us consider two electrons per site, and degenerate $t_{2g}$ ($d_{xy}$, $d_{xz}$ and $d_{yz}$) orbitals at each site, and a very large intra-orbital $U$, so that there is only one electron per orbital, a large Hund’s coupling $J_H$ so that $S = 1$, and no spin orbit coupling. We define the states of the three-dimensional orbital space to be $|x\rangle$, $|y\rangle$ and $|z\rangle$, such that, if $|0\rangle$ is the Fock space vacuum at a given site $|0\rangle$.

The overlap Eq. (10) is only approximately equal to 1/4, and depends—albeit exponentially—on loop length: this could in fact bear additional interesting consequences.

For example, for $L = 6$, at $\beta = 0$ and $1/3$, $\epsilon_0 - \epsilon_0 \approx 0.120J$ and $\epsilon_0 - \epsilon_0 \approx 0.116J$, respectively, and the energy density differences between an open and a closed chain of length six are $\epsilon_0 - \epsilon_0 \approx 0.208J$ and $\epsilon_0 - \epsilon_0 = J/9$, respectively.

Time-reversal symmetry is also preserved in that phase.

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honeycomb bond with an “up” spin-1/2, or a hard-core boson, on the center of the bond.

Appendix A: Effective orbital operators
of phase was made.

We now define the operators $L^c$, $c = x, y, z$ according to:

$$ L^c = -\frac{i}{2} \sum_{a,b} \xi_{abc} \langle a \rangle \langle b \rangle - \langle b \rangle \langle a \rangle, \quad \text{(A2)} $$

which may be rewritten:

$$ L^\gamma = -i \langle |\gamma + 1\rangle \langle \gamma - 1| - |\gamma - 1\rangle \langle \gamma + 1| \rangle. \quad \text{(A3)} $$

These operators obey $L^\gamma |\gamma\rangle = 0$, and $L^2 = 2$. One may check that these operators obey the angular momentum algebra commutation relations. To form a complete basis of Hermitian operators acting in our three-dimensional space, we need six more Hermitian operators, which we choose to be $(L^a)^2 = P^a$ and $\{L^a, L^b\} = T^a, a \neq b$. Note that:

$$ P^\gamma = (L^\gamma)^2 = |\gamma + 1\rangle \langle \gamma + 1| + |\gamma - 1\rangle \langle \gamma - 1| = 1 - |\gamma\rangle \langle \gamma| \quad \text{(A4)} $$

and

$$ T^\gamma = \{L^{\gamma+1}, L^{\gamma-1}\} = - (|\gamma + 1\rangle \langle \gamma - 1| + |\gamma - 1\rangle \langle \gamma + 1|). \quad \text{(A5)} $$

2. Effective operators as “rotation” and projection operators

$L^\gamma |\gamma\rangle = 0$ so the projection operator onto the $|\gamma\rangle$ component is $P_\gamma = 1 - (L^\gamma)^2$. The projection operator in Eq. [2] is $P_\gamma = 1 - P_\gamma = (L^\gamma)^2$. In particular:

$$ P_\gamma |\gamma\rangle = 0, \quad P_\gamma |\gamma \pm 1\rangle = |\gamma \pm 1\rangle. \quad \text{(A6)} $$

Disregarding phase factors:

$$ \begin{align*}
L^\gamma |\gamma\rangle &= 0, \quad L^\gamma |\gamma \pm 1\rangle \propto |\gamma \mp 1\rangle, \\
T^\gamma |\gamma\rangle &= 0, \quad T^\gamma |\gamma \pm 1\rangle \propto |\gamma \mp 1\rangle.
\end{align*} \quad \text{(A7)} $$

3. Spin-orbital model

In the orbital sector, the coupling Hamiltonian between two sites 1 and 2 connected by an $x$-type bond, as defined above, takes the form:

$$ H^\text{orb}_{12} = \left( \begin{array}{c}
\mathcal{J}_1 \mathcal{J}_4 \mathcal{J}_1 \\
\mathcal{J}_4 \mathcal{J}_2 \mathcal{J}_3 \\
\mathcal{J}_4 \mathcal{J}_3 \mathcal{J}_2
\end{array} \right) \left( \begin{array}{c}
L^2_1 \ L^y_1 \ L^2_1 \\
L^y_1 \ L^2_1 \ L^2_1 \\
L^2_1 \ L^y_1 \ L^2_1
\end{array} \right) + \left( \begin{array}{c}
\mathcal{J}_5 \mathcal{J}_6 \mathcal{J}_7 \\
\mathcal{J}_8 \mathcal{J}_6 \mathcal{J}_7 \\
\mathcal{J}_8 \mathcal{J}_7 \mathcal{J}_6
\end{array} \right) \left( \begin{array}{c}
L^2_1 \ L^y_1 \ L^2_1 \\
L^y_1 \ L^2_1 \ L^2_1 \\
L^2_1 \ L^y_1 \ L^2_1
\end{array} \right) \left( \begin{array}{c}
\mathcal{J}_9 \mathcal{J}_{10} \mathcal{J}_{11} \\
\mathcal{J}_{12} \mathcal{J}_{10} \mathcal{J}_{11} \\
-\mathcal{J}_{12} \mathcal{J}_{11} \mathcal{J}_{10}
\end{array} \right) \left( \begin{array}{c}
\{L^y_2, L^z_2\} \\
\{L^y_2, L^z_2\} \\
\{L^z_2, L^y_2\}
\end{array} \right). \quad \text{(A8)} $$

In Eq. [1], if $J = 0$, $\mathcal{J}_1 = -\zeta$, $\mathcal{J}_7 = -\nu_1$, $\mathcal{J}_{11} = -\nu_2$, and all others zero. While the number of parameters is large (12), many of them are expected to be zero, physically. For example, it is unclear whether it is possible to obtain terms which involve a single power of angular momentum at each site from standard superexchange calculations \[1, 3\].

Upon introducing the spin degrees of freedom, in principle, each independent coefficient may be a spin Hamiltonian of the form (for no spin-orbit coupling):

$$ \mathcal{J}_p = A_p + B_p S_1 \cdot S_2 + C_p (S_1 \cdot S_2)^2, \quad \text{(A9)} $$

where $p = 1, \ldots, 12$ labels the independent terms. In Eq. [1], we took $\mathcal{J}_1 = -\zeta + J (S_1 \cdot S_2 + \beta (S_1 \cdot S_2)^2)$, i.e. $A_1 = -\zeta$, $B_1 = J$, $C_1 = \beta$, and $\mathcal{J}_7 = -\nu_1$, i.e. $A_7 = -\nu_1$ and $B_7 = C_7 = 0$, and $\mathcal{J}_{11} = -\nu_2$, i.e. $A_{11} = -\nu_2$ and $B_{11} = C_{11} = 0$ and all other terms zero.

Appendix B: Details of perturbation theory

Here we focus on orbital space, i.e. set $J = 0$, and give a few details for the degenerate perturbation theory in

$$ H^\text{kin} = -v \sum_{(ij)} (T^\gamma_{ij} - 1) T^\gamma_{ij} + T^\gamma_{ij} T^\gamma_{ij} - 1 \quad \text{(B1)} $$

onto the manifold of loop coverings of the lattice, valid when $\zeta > 0$ and $\zeta \gg v$. The effective Hamiltonian is

$$ H^\text{eff} = \mathcal{P} H^\text{kin} \frac{1 - \mathcal{P}}{H - E_0} H^\text{kin} \frac{1 - \mathcal{P}}{H - E_0} \cdots \frac{1 - \mathcal{P}}{H - E_0} H^\text{kin} \mathcal{P}. \quad \text{(B2)} $$
where $H_{\text{kin}}$ appears as many times as the order in perturbation theory.

Consider a “flippable” plaquette. Acting once with $H_{\text{kin}}$ on any bond which belongs to the plaquette creates two “defect” bonds (this configuration does not belong to the loop covering manifold), with the new plaquette state looking like on Fig. 7. The energy of this configuration is that of a loop cut, i.e. $\zeta$. Acting a second time with $H_{\text{kin}}$, with the “active” bond operator one bond away from the first active bond creates another configuration of energy $\zeta$. Only at third order is the system brought back to the loop manifold. There are twelve ($= 6 \times 6$) of the $S^f$icient) onto the $S$_sector. Then, the energy is independent of chain length. Hence, at first order in perturbation theory, the spins do not lift the degeneracy of the loop coverings at the spin AKLT point.

Appendix C: Haldane chain energy

In this appendix we investigate the energy density (energy divided by the number of sites) of $S = 1$ loops in the Haldane phase as a function of their length.

1. AKLT chains

At the AKLT point, the energy density is independent of the loop length. Indeed the AKLT Hamiltonian may be rewritten as

$$H_{\text{AKLT}} = \frac{1}{4} \sum_i \left[ (S_i + S_{i+1})^2 ( (S_i + S_{i+1})^2 - 2 ) + \text{const} \right],$$

i.e. as the sum of the projectors (with equal positive coefficient) onto the $S^{tot} = 2$ sector (i.e. $(S^{tot})^2 = 2(2+1) = 6$) of the $S^{tot} = S_i + S_{i+1}$ operator. This means that the ground state will have zero components in the $S = 2$ sector. Then, the energy is independent of chain length. Hence, at first order in perturbation theory, the spins do not lift the degeneracy of the loop coverings at the spin AKLT point.

2. Numerical results away from the AKLT point

We performed exact diagonalization for the Hamiltonian in Eq. (1) on chains with periodic boundary conditions for up to length 7. The results seem to indicate that, away from the AKLT point $\beta = 1/3$, the energy density of closed even-length loops (relevant for the honeycomb and hyperhoneycomb lattices) increases with loop length. Results obtained in DMRG for longer closed loops with the use of the itensor package confirm that the energy density of loops of length 40 is always larger than that of length 6, see Fig. 8 (and Ref. 49 for Monte Carlo results at $\beta = 0$).

Appendix D: Haldane covering overlaps

1. At the AKLT point: MPS formalism

The AKLT loop covering overlaps are calculated with the matrix product state formalism. An exact representation of the AKLT wavefunction is given as an MPS:

$$|\psi_{\text{AKLT}}\rangle = \sum_{\{\sigma_i=0,\pm1\}} \text{Tr}[M(\sigma_1) \cdots M(\sigma_N)] |\sigma_1 \cdots \sigma_N\rangle,$$

when the chain is a closed loop of length $N$ (we use the notations from Ref. 50) with the following matrices $M$:

$$\begin{cases} M(\sigma = 0) = -\sqrt{\frac{1}{3}} \sigma z \cr M(\sigma = \pm 1) = -\sqrt{\frac{2}{3}} \sigma \pm \end{cases},$$

with $\sigma^\mu$ the Pauli matrices, and with the norm $|\psi|^2$:

$$\langle \psi | \psi \rangle = \text{Tr} T^N, \quad \text{where} \quad T = \sum_{\sigma = \pm 1, 0} M^*(\sigma) \otimes M(\sigma).$$
Within the MPS formalism, these overlaps are given by:

\[
\text{plaquette of interest, before and after the plaquette flip.}
\]

The difference between those configurations lies purely in the coverings connected by a single plaquette flip. The differences between one loop and two loops, emphasizing the role of the fractional degrees of freedom near the cuts.

We then compute the overlap between different types of coverings connected by a single plaquette flip. The difference between those configurations lies purely in the number and lengths of the loops “touching” the flippable plaquette of interest, before and after the plaquette flip. Within the MPS formalism, these overlaps are given by:

- three loops connected to one:

\[
\langle \text{AKLT}_0 | \text{AKLT}_1, \text{AKLT}_2, \text{AKLT}_3 \rangle = \frac{\text{Tr}[T_1^{n_1} T_2^{n_2} T_3^{n_3}]}{\sqrt{\text{Tr}[T_1^{n_1}] \text{Tr}[T_1^{n_2} T_2^{n_2}] \text{Tr}[T_1^{n_3} T_2^{n_3} T_3^{n_3}]}} = (3 + 3^{n_1} (13 + 2\sqrt{2} 3^{n_2} + 3^{n_3} (3 - \sqrt{2} + 2 \cdot 3^{n_3})))
\]

using the configuration from Fig. 9b), where \(n_0 = n_1 + n_2 + n_3\),

\[
T_1 = \sum_{\sigma = 0, \pm 1} M^* (\sigma) \otimes M (\sigma) \otimes 1 \otimes 1
\]

\[
T_2 = \sum_{\sigma = 0, \pm 1} M^* (\sigma) \otimes 1 \otimes M (\sigma) \otimes 1
\]

\[
T_3 = \sum_{\sigma = 0, \pm 1} M^* (\sigma) \otimes 1 \otimes 1 \otimes M (\sigma)
\]

The overlaps Eqs. (D4)-(D12) take the form of a dominant 1/4 contribution and exponentially decaying terms. This (dominating) length-independent contribution is simply equal to the overlap of “neighboring” \(S = 1/2\) spins from different chains (see Fig. 9b)), without the \(S = 1\) on-site projections, and may be empirically understood from the very short range entanglement in the AKLT wavefunction. In fact, 1/4 is a special case of a more general formula according to which the overlap of two coverings where \(n_{\text{cuts}}\) loop cuts are needed to reconnect them is equal to 1/2\(^{n_{\text{cuts}}-1}\). For example, if \(|\psi_1\rangle\) and \(|\psi_2\rangle\) are as depicted on Fig. 9b), then:

\[
|\psi_1\rangle = \frac{1}{2} \left( |\uparrow_1 \downarrow_2\rangle - |\downarrow_1 \uparrow_2\rangle \right) (|\uparrow_3 \downarrow_4\rangle - |\downarrow_3 \uparrow_4\rangle)
\]

\[
T_1 = \sum_{\sigma = 0, \pm 1} M^* (\sigma) \otimes M (\sigma) \otimes 1 \otimes 1
\]

\[
T_2 = \sum_{\sigma = 0, \pm 1} M^* (\sigma) \otimes 1 \otimes M (\sigma) \otimes 1
\]

\[
T_3 = \sum_{\sigma = 0, \pm 1} M^* (\sigma) \otimes 1 \otimes 1 \otimes M (\sigma)
\]

and

\[
\tilde{T}_1 = \sum_{\sigma = 0, \pm 1} M^* (\sigma) \otimes M (\sigma) \otimes 1 \otimes 1
\]

\[
\tilde{T}_2 = \sum_{\sigma = 0, \pm 1} M^* (\sigma) \otimes 1 \otimes M (\sigma) \otimes 1
\]

\[
\tilde{T}_3 = \sum_{\sigma = 0, \pm 1} M^* (\sigma) \otimes 1 \otimes 1 \otimes M (\sigma)
\]

\[
\langle \text{AKLT}_1, \text{AKLT}_2 | \text{AKLT}_3, \text{AKLT}_4 \rangle
\]

\[
= \frac{\text{Tr}[T_1^{n_1} T_2^{n_2} T_3^{n_3} T_4^{n_4}]}{\sqrt{\text{Tr}[T_1^{n_1}] \text{Tr}[T_1^{n_2} T_2^{n_2}] \text{Tr}[T_1^{n_3} T_2^{n_3} T_3^{n_3}] \text{Tr}[T_1^{n_4} T_2^{n_4} T_3^{n_4} T_4^{n_4}]} = (3 + 3^{n_1}) (13 + 2\sqrt{2} 3^{n_2} + 3^{n_3} (3 - \sqrt{2} + 2 \cdot 3^{n_3})))
\]

\[
= 4\sqrt{2} (5 + 3^{n_2} (1 + 2 \cdot 3^{n_1}))(3 + 3^{n_3}) (3 + 3^{n_4})
\]

\[
\langle \text{AKLT}_1, \text{AKLT}_2 | \text{AKLT}_3, \text{AKLT}_4 \rangle
\]

\[
= \sum_{\sigma = 0, \pm 1} M^* (\sigma) \otimes 1 \otimes M (\sigma) \otimes 1
\]

\[
= \sum_{\sigma = 0, \pm 1} M^* (\sigma) \otimes 1 \otimes 1 \otimes M (\sigma)
\]
and so:
\[ \langle \psi_2 | \psi_1 \rangle = \frac{1}{2}, \quad (D20) \]
consistent with the fact that two loop cuts are needed to go from the upper to the lower MPS, and vice versa.

**Appendix E: Details of the spin model phase diagram on the non-uniform honeycomb lattice**

Here we take a simple variational approach and calculate the energy of three different states, for varying \( \beta \) and \( \lambda_p \) for the pattern depicted in Fig. 6a). The three states are the simple antiferromagnet, a valence bond solid where the singlets lie on \( \lambda_d \) bonds, and a Haldane hexagon crystal where length-six Haldane chains form along \( \lambda_p \) bonds. We consider the Hamiltonian

\[ H = \lambda_p J \sum_{(ij) \in p} (S_i \cdot S_j + \beta (S_i \cdot S_j)^2) + \lambda_d J \sum_{(ij) \in d} (S_i \cdot S_j + \beta (S_i \cdot S_j)^2). \quad (E1) \]

The approach could be readily refined to the next simplest level of variational approach by considering more general MPS network states (e.g. non-uniform along a plaquette, still with bond dimension 2 to keep it simple), but we deem it unnecessary for our purpose, as we see below.

The energy per bond in the simple antiferromagnet, where the state of a bond is of the form \( |\text{AFM} \rangle = |1\rangle - |1\rangle \), is given by:

\[ \epsilon_{\text{AFM}}(\lambda_p, \beta) = \frac{J}{3} (2\lambda_p + \lambda_d)(-1 + 2\beta) = \frac{2J}{3} (2\beta - 1). \quad (E3) \]

The energy per bond in the valence bond solid with the valence bonds across \( \lambda_d \) bonds, the state of a \( \lambda_d \) bond is \( |\text{VBS} \rangle = \frac{1}{\sqrt{2}}(|1\rangle - |1\rangle) \) is

\[ \epsilon_{\text{VBS}}(\lambda_p, \beta) = \frac{J}{3} (\lambda_d + \beta(\lambda_d + 3\lambda_p)) = \frac{2J}{3} \left( -1 + \lambda_p + \beta(1 + \frac{\lambda_d}{2}) \right). \quad (E4) \]

The energies of length-six Haldane chains at various \( \beta \)'s obtained with exact diagonalization are given Table I. To obtain the energy of the \( \lambda_d \) bonds, we use the eigenvectors, and find that it is simply proportional to \( \beta \),

\[ \epsilon_{\text{Haldane}}(\lambda_p, \beta) = \frac{J}{3} \left( 2\lambda_p \epsilon_6(\beta) + \beta \lambda_d \frac{4\beta}{3} \right) = \frac{2J}{3} \left( \frac{4\beta}{3} + \lambda_p (\epsilon_6(\beta) - \frac{4\beta}{3}) \right). \quad (E6) \]

Hence, the system is in the AFM phase if:

\[ \begin{cases} 
\lambda \geq \frac{2\beta}{4\beta + 2} & \text{if } 4\beta - 3\epsilon_6(\beta) \geq 0 \\
\lambda \leq \frac{3 - 2\beta}{4\beta - 3\epsilon_6(\beta)} & \text{if } 4\beta - 3\epsilon_6(\beta) \geq 0 
\end{cases} \quad (E7) \]

the system is in the VBS phase if:

\[ \begin{cases} 
\lambda \leq \frac{2\beta}{2 + \beta} & \text{if } 6 + 11\beta - 6\epsilon_6(\beta) \geq 0 \\
\lambda \leq \frac{2(3 + \beta)}{6 + 11\beta - 6\epsilon_6(\beta)} & \text{if } 6 + 11\beta - 6\epsilon_6(\beta) \geq 0 
\end{cases} \quad (E9) \]

and the system is in the Haldane crystal phase if:

\[ \begin{cases} 
\lambda \geq \frac{3 - 2\beta}{4\beta - 3\epsilon_6(\beta)} & \text{if } 4\beta - 3\epsilon_6(\beta) \geq 0 \\
\lambda \geq \frac{3}{6 + 11\beta - 6\epsilon_6(\beta)} & \text{if } 6 + 11\beta - 6\epsilon_6(\beta) \geq 0 
\end{cases} \quad (E10) \]

Using these relations, we draw the approximate phase diagram shown in Fig. 6 in the main text. The results are consistent with those obtained for the uniform exchange bilinear-biquadratic model in Refs. 51 and 52.

**FIG. 10.** The three different hexagon loop crystals.

**Appendix F: Ground and excited state energies for chains of length six**

| \( \beta \) | -1 | -2/3 | -1/3 | 0 | 1/12 |
|---|---|---|---|---|---|
| \( E_0^6 \) | -24.8774 | -19.2953 | -13.8305 | -8.61742 | -7.38496 |
| \( E_1^6 \) | -22.6517 | -17.6728 | -12.7378 | -7.8968 | -6.71459 |
| \( \bar{E}_0^6 \) | -21.8484 | -16.901 | -12.0325 | -7.30727 | -6.2737 |

| \( \beta \) | 1/6 | 1/4 | 1/3 | 5/12 | 1/2 |
|---|---|---|---|---|---|
| \( E_0^6 \) | -6.19548 | -5.06111 | -4 | -3.03786 | -2.1977 |
| \( E_1^6 \) | -5.50707 | -4.41121 | -3.30427 | -2.24068 | -1.27149 |
| \( \bar{E}_0^6 \) | -5.22356 | -4.2353 | -10/3 | -2.55428 | -1.91296 |

| \( \beta \) | 7/12 | 2/3 | 3/4 | 5/6 | 11/12 |
|---|---|---|---|---|---|
| \( E_0^6 \) | -1.47434 | -0.834305 | -0.244138 | 0.317211 | 0.861294 |
| \( E_1^6 \) | -0.543207 | 0.163247 | 0.848938 | 1.51519 | 2.16354 |
| \( \bar{E}_0^6 \) | -1.36832 | -0.874155 | -0.406012 | 0.047453 | 0.491835 |

**Table I.** Ground state energy, first excited state energy of periodic Haldane chains (\( E_0^6 \) and \( E_1^6 \), respectively) and ground state energy of open Haldane chains (\( \bar{E}_0^6 \)) of length six as a function of \( \beta \) (obtained in exact diagonalization), in units of \( J \).