Hidden phases born of a quantum spin liquid: Application to pyrochlore spin ice

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Quantum spin liquids (QSL) have generated considerable excitement as phases of matter with emergent gauge structures and fractionalized excitations. In this context, phase transitions out of QSLs have been widely discussed as Higgs transitions from deconfined to confined phases of a lattice gauge theory. However the possibility of a wider range of novel phases, occurring between these two limits, has yet to be systematically explored. In this Letter, we develop a formalism which allows for interactions between fractionalised quasiparticles coming from the constraint on the physical Hilbert space, and can be used to search for exotic, hidden phases. Taking pyrochlore spin ice as a starting point, we show how a U(1) QSL can give birth to abundant daughter phases, without need for fine-tuning of parameters. These include a (charged–) Z2 QSL, and a supersolid. We discuss implications for experiment, and numerical results which support our analysis. These results are of broad relevance to QSL subject to a parton description, and offer a new perspective for searching exotic hidden phases in quantum magnets.

Introduction — One of the most intriguing features of frustration in magnets is the possibility of finding phases of matter which lie outside the usual Landau paradigm of symmetry–broken states [1, 2]. A prominent example is the “quantum spin liquid” (QSL), a state where spins continue to fluctuate at low temperatures, achieving a massively-entangled state with fractionalised excitations [3–5]. Long a subject of theoretical conjecture, in the past decade QSL have also become an intense focus for experimental research [6–8]. One of their defining properties is non-locality, frequently encoded in an emergent gauge degree of freedom. And in this respect, the parton approach has proved a powerful tool for describing both the fractionalised excitations of QSL, and their (non-local) interactions [9].

Besides being interesting in their own right, QSL also give rise to “daughter” phases which may have an unconventional, or hidden, character. QSL described by the deconfined phase of a lattice gauge theory [10–13], prove surprisingly stable against weak perturbations [14–20]. None the less, at strong coupling, they can undergo confinement through a Higgs transition [21, 22], into a magnetically–ordered phase which breaks the emergent gauge symmetry [13, 19, 23]. What happens at intermediate coupling, where usual perturbation theory breaks down, remains an open question. In particular, the possibility of finding new, intermediate phases between the QSL and Higgs phases at strong coupling, has yet to be systematically explored.

In this Letter, we argue that novel phases, intermediate between the confined and deconfined limits of a pure lattice gauge theory, may be a generic feature of models supporting QSL. These intermediate phases are driven by interactions between the collective excitations of the QSL, which are obscured in the perturbative limit of the problem. In particular, fluctuations of the (gauge–)charge, absent in a pure gauge theory, generate new effective interactions between the fractionalised excitations of the QSL. As a result, the lifting of gauge symmetry can become a two–step process, with a new intermediate phase, also with QSL character, occurring between the orginal QSL and its fully–confined, Higgs phase.

We develop these ideas in the context of a parton theory of pyrochlore quantum spin ice, where the existence of a deconfined U(1) QSL and its corresponding, magnetically–ordered Higgs phase, are already well established. Starting from a standard Bosonic parton prescription, we develop a formalism which takes into account the fact that charge fluctuations are bounded by the finite Hilbert space of the underlying spins. Applying this to an extended XXZ model, we find that the U(1) QSL can give rise to a plethora of exotic phases, including a Z2 QSL; a “charged” Z2 QSL with broken inversion symmetry; and a “spinon supersolid” which breaks both inversion and time–reversal symmetries. Possible experimental signatures of these phases are discussed. While this particular hierarchy of phases is specific to the pyrochlore lattice, the formalism developed is quite general, and should help to guide the search for hidden phases in a wide range of quantum magnets.

Pyrochlore spin ice — Pyrochlore oxide materials with a chemical formula R2TM2O7 (R: rare earth, TM transition metal) [24, 25] have proved a rich source of candidates for QSL and related forms of order [26–34]. In many of these materials, localized f–electrons form a (non–)magnetic doublet described by a pseudospin–1/2, with the minimal model taking the form [35, 36]

\[ H_{XXZ} = H_0 + H_1 = \sum_{\langle ij \rangle} \left[ J_z S_i^z S_j^z - J_\pm (S_i^+ S_j^- + h.c.) \right], \] (1)

where the sum (\langle ij \rangle) runs over the first–neighbour bonds of a pyrochlore lattice. In the perturbative limit \( J_\pm \gg |J_z| \) (\( J_\pm > 0 \)), the dominant Ising term \( H_0 \) favors an extensively–degenerate set of classical spin ice states [37–39], while the spin–flip term \( H_1 \) causes mixing of these states, leading to a QSL ground state [16, 19, 20, 40–44].

This spin liquid can be elegantly described in terms of a compact, frustrated, U(1) lattice gauge theory [16, 41, 45, 46]. This is defined on the sites \( r, r' \) of a (bipartite) diamond lat-
tice, with spin operators expressed in terms of an emergent gauge field $A_{rr'}^z \pmod{2\pi}$, electric field $E_{rr'}$ (half–integer), and matter field $\phi_r = e^{-i\phi_r}$ (a Bosonic spinon), conjugate to a gauge charge $Q_r$, such that

$$Q_r = \eta_r \sum_{\mu=0}^3 S_{rr'+\eta_r}^z, \quad S_{rr'}^z = \eta_r E_{rr'};$$

$$S_{rr'}^+ = \phi_{rr'}^\dagger \phi_{rr'} (s_{rr'}^+ = \frac{1}{2} e^{iA_{rr'}} \quad \text{for} \quad r \in A),$$

where $\eta_{\in A(B)} = 1(-1)$ distinguishes sites belong to the $A(B)$ sublattice. For an $S = 1/2$ (pseudo–)spin doublet, the gauge charge takes on integer values

$$Q_r = 0, \pm 1, \pm 2 \ldots \pm 4S,$$

with spin fluctuations acting as ladder operators for this tower of states. [See supplementary material for details].

Following a standard prescription [45, 46], we further map the $U(1)$ lattice gauge theory onto a quantum rotor model of spinons coupled to a (static) $U(1)$ gauge field. In doing so we consider the length of the spin $S$ in Eq. (3) to be a formal control parameter, initially taking the limit $S \to \infty$. Within these approximations

$$Z = \int D\phi_r^\dagger D\phi_r DQ_r |Q_r \in (-\infty, \infty) e^{-S_{\text{eff}}[\phi_r, \phi_r, Q_r]},$$

$$S_{\text{eff}} = \int_0^\beta d\tau \left( \sum_r (iQ_r \partial_\tau \phi_r + \lambda (\phi_r^2 - 1)) + H_{\text{rotor}} \right),$$

where the Lagrange multiplier $\lambda$ enforces the rotor constraint $|\phi_r| = 1$ in a soft manner $\frac{1}{N} \sum_{r \in A} \phi_r^2 \phi_r = 1$, and

$$H_{\text{rotor}} = \frac{J_+}{2} \sum_r Q_r^2 - \frac{J_+}{4} \sum_{\langle rr' \rangle} \phi_r e^{i(A_{rr'} + A_{rr''})} \phi_{rr''} + \text{h.c.},$$

with spinons constrained to move on either the $A$ or $B$ sublattice of sites ($\langle rr' \rangle$), cf. Fig. 1.

Within the framework of this rotor model, transitions out of $U(1)$ QSL occur through the Higgs mechanism, and are associated with the Bose–Einstein condensation (BEC) of either electric charges (spinons) [4, 5], or the corresponding, dual, magnetic monopole [47]. Condensation of spinons leads to confined states with easy–plane magnetic order, and QMC simulations of Eq. (1) find easy–plane magnetic order with $q = 0$ for $J_+/J_z \gtrsim 0.05$, confirming the expected Higgs phase as the strong–coupling ground state [19, 42, 44]. A number of other forms easy–plane order have also been identified as Higgs phases in generic models of pyrochlore magnets [45, 46]. However, to explore the possibility of new phases at intermediate coupling, a more general approach is needed.

Heuristically, we reason as follows: In the limit $J_+/J_z \to 0$, fluctuations of the charge $Q_r \approx 0$ are negligible, and have no effect on the propagation of spinons. However as $J_+$ increases, spinons begin to interact with a dilute cloud of charge fluctuations, which modify their dynamics, cf. Fig. 1. The simplest, gauge–invariant form of interaction capturing this effect is

$$\delta H_{\text{rotor}} = g Q_r^2 \phi_r^\dagger e^{i(A_{rr'} + A_{rr''})} \phi_{rr''} + \mathcal{O}(Q_r^4),$$

where the coupling constant $g$ increases with $|J_\pm|$. Physically, this describes the interplay of two competing tendencies, charge fluctuations, mediated by the motion of spinons, and the constraint on the maximum charge on a single site, reflected in the ladder termination

$$S_{rr'}^+ | S_{rr'}^z = S \rangle = S_{rr'}^- | S_{rr'}^z = -S \rangle = 0.$$

And it is this competition, absent in the usual rotor formulation, Eq. (6), which has the potential to change the nature of the QSL.

**Projected rotor representation** — Within the canonical rotor formalism, gauge charge takes on all real values $-\infty < Q_r < \infty$ [Eq. (4)]. The consequences of the physical constraint $|Q_r| \leq 4S$ [Eq. (3)], can be explored through the new terms generated by restriction to a physical Hilbert space, viz

$$Z' = \int D\phi_r^\dagger D\phi_r DQ_r |Q_r \in (-4S, 4S) e^{-S_{\text{eff}}},$$

$$\approx \int D\phi_r^\dagger D\phi_r DQ_r |Q_r \in (-\infty, \infty) e^{-S_{\text{eff}}},$$

where the effective action $S_{\text{eff}}'$ [cf. Eq. (5)], is modified through the projection of spinon and charge fields within the rotor Hamiltonian

$$H_{\text{rotor}}' = H_{\text{rotor}}[\bar{Q}_r^\prime, \tilde{\phi}_r^\dagger, \bar{A}_{rr'}', E_{rr'}'] \quad (\bar{Q}_r \equiv P_r O_r P_r).$$

We require that the projection operator $P_r$ satisfies

$$P_r [Q_r] = 1 \forall Q_r \in \{0, \pm 1, \ldots, \pm 4S\},$$

$$P_r [Q_r = \pm (4S + 1)] = 0.$$

ensuring that no matrix element of Eq. (10) connects to an unphysical state. Resolving $P_r$ as a polynomial and expanding
to leading order

\[ P_r(Q_\gamma) \equiv 1 - \frac{1}{(4S + 1)(8S + 1)!} \prod_{k=0}^{4S} (Q_\gamma^2 - k^2) \]

\[ = 1 - \alpha S Q_\gamma^2 + \cdots \quad (\alpha S = \frac{(4S)!^2}{(4S + 1)(8S + 1)!}) \]

(13)

we find,

\[ H'_{\text{rotor}} = H_{\text{rotor}} + \delta H_{\text{rotor}} \]

(14)

where \( \delta H_{\text{rotor}} \) is given by Eq. (7), with \( g = \alpha S J_{\pm} \). Finally, integrating out \( Q_\gamma \) in Eq. (14), we arrive at an effective model with interaction between spinons

\[ H_{\text{rotor}}' = H_{\text{rotor}} + H_u \]

(15)

\[ H_u = -u \sum_{\langle \mathbf{r}, \mathbf{r}' \rangle} \left( \phi^\dagger e^{i (A_{\mathbf{r}'} - A_{\mathbf{r}}) + i \phi_{\mathbf{r}'}} \phi^\gamma \right)^2 + \text{h.c.} + \cdots \]

(16)

\[ u \sim \alpha S J_{z}^2 / J_z \]

(17)

We note that an approach based on canonical transformations [48] also leads to an attractive interaction between spinons, with the same form of vertex, at the same order in \( J_{\pm} / J_z \). The role of longer-range interactions, omitted in Eq. (16) will be discussed below. [See supplementary material for details].

The projection method developed above has much in common with the expansion of interactions in spin–wave theory. And just as in that case, where practitioners must choose between the prescriptions of Holstein and Primakoff [49], and those of Dyson and Maleev [50, 51], the form of projection, Eq. (13), is not uniquely determined. None the less, since the structure of the vertex in Eq. (16) is constrained by symmetry, it is not sensitive to the precise choice of projection operator. With this in mind, we now proceed to examine the consequences of interactions between spinons.

**Spinon interaction and hidden phases** — For large \( u \), the implication of \( H_u \) [Eq. (16)], can be understood by direct analogy with earlier work on QSL described by a quantum rotor model [14]. Here, since \( u > 0 \), the interaction mediates pairing between spinons, favouring a charge-2 condensate \( \Delta \equiv \langle \phi^\dagger \phi^\gamma \rangle \neq 0 \), which minimizes both terms in \( H_{\text{rotor}} \) [Eq. (15)]. Since this pairing occurs in the absence of a single–spinon condensate, \( \langle \phi \rangle = 0 \), the state retains its fractionalised, QSL character, but with the gauge group broken from \( U(1) \) down to \( \mathbb{Z}_2 \). From Eq. (17), we see that this new “large–u” phase is most likely to be realised for intermediate to large \( J_{\pm} / J_z \), and for small values of \( S \), i.e. in the physical limit of \( S \approx 1/2 \).

In Fig. 2 we show a schematic phase diagram for the extended rotor model, Eq. (14), based on these expectations. Known results for the rotor model, Eq. (6), are shown for \( u = 0 \); here \( U(1) \) QSLs with flux \( 0(\pi) \) give way to Higgs phases with ordering wave vectors \( \mathbf{k}_{\pi(\pi')} \). For \( J_{\pm} \ll J_z \), these QSL become unstable at large \( u \) against a “hidden” phase with finite spinon pairing, \( \Delta \neq 0 \). The behaviour expected of the XXZ model, Eq. (1), is shown through red parabola with \( u \sim J_{z}^2 / S \). Where fluctuations of charge are small (large \( S \)), the system passes directly from the \( U(1) \) QSL to its associated Higgs phase. However where charge fluctuations are significant (small \( S \)) this trajectory can pass through the “hidden” phase, giving a range of \( J_{\pm} / J_z \) for which the ground state is a \( \mathbb{Z}_2 \) QSL. Specific estimates of the phase boundaries for \( S \approx 1/2 \) and \( u = 0.04 \) are shown in Fig. 2b.

This intermediate “hidden” regime is expected to be a rich source of other novel phases, particularly once higher–order corrections in Eq. (14) are taken into account. This is particularly true for frustrated exchange \( J_{\pm} < 0 \), where the \( U(1) \) QSL occurs with \( \pi \)–flux [46], and the Higgs transition is postponed to much larger values of \( J_{\pm} \), allowing for larger fluctuations of charge \( Q \) [Fig. 2b]. In particular, where the pairing of spinons includes an offsite component, \( \Delta_{\mu - \nu} = \langle \phi^\mu \phi_{\mathbf{r}+ \mathbf{e}_\mu - \mathbf{e}_\nu} \rangle \), the \( \mathbb{Z}_2 \) QSL will develop a quadrupole moment on bonds \( \langle S^\mu \mathbb{S}^\nu \rangle \sim \Delta_{\mu - \nu}^2 \Delta_{\mu - \nu} \), leading to a state with spin–nematic order [52–57].

**Supersolid phases** — So far, we have shown that \( \delta H_{\text{rotor}} \) qualitatively modifies the spinon action \( H_{\text{rotor}} \rightarrow H_{\text{rotor}} + H_u \), leading to the phase diagram Fig. 2. Still more new phases...
TABLE I. Phases found in extended XXZ model $H_{XXZ+}$ [Eq. (18)], as shown in phase diagram Fig. 3b. Order parameters are listed, as well as the associated experimental signatures in heat capacity $c(T)$, and spin structure factor $S(q)$.

| $H(1)_0$ QSL | $Z_2$ QSL | Higgs$_0$ | Charged $Z_2$ QSL | Supersolid |
|---------------|------------|-----------|-------------------|------------|
| $0$ | $0$ | $0$ | Power-law | Diffuse |
| $0$ | $0$ | $\neq 0$ | Expo-decay | Diffuse |
| $\neq 0$ | $0$ | $0$ | Power-law | Bragg Peaks |
| $0$ | $0$ | $\neq 0$ | Expo-decay | Bragg Peaks |
| $\neq 0$ | $0$ | $0$ | Power-law | Bragg Peaks |

FIG. 3. Phase diagram of extended XXZ model, illustrating the possibility of charge-ordered phases. (a) Convention for labelling sites in further–neighbor Ising interaction $H_{zz}$ [Eq. (19)]. Charge polarization on diamond–lattice sites $\langle Q_r \rangle$, can coexist with a transverse magnetization on pyrochlore lattice sites, $\langle S_{rr}^+ \rangle$. (b) Phase diagram, showing phases listed in Table I. At small $J_{zz}$, the formation of a charge–2 condensate (dashed grey line) converts the $U(1)_0$ QSL into a $Z_2$ QSL (‘hidden’ phase of Fig. 2). A further charge–1 condensation (dashed red line) separates this from the strong–coupling Higgs phase (easy–plane AF). At larger $J_{zz}$ and small $J_{\pm}$ there is a 1st–order phase transition (solid red line) into phases where $\langle Q_r \rangle \neq 0$. This creates a $2^{nd}$–order transition at larger $J_{zz}$ (dashed red line). These comprise a charged version of the $Z_2$ QSL, and supersolid which is the charged version of the Higgs phase. All phase boundaries were estimated within gauge mean-field theory (gMFT) for $H_{XXZ+}$ [Eq. (18)] with ferromagnetic Ising interactions $J_{zz} > 0$, $J_{\pm} > 0$, $\alpha = 0.04J_z$ [cf. Fig. 2b], as described in the Supplementary material.

Within a mean–field theory for the pure rotor model, Eq. (6), in the static limit $\delta Q(\omega_n = 0)$, spinon and gauge charge degrees of freedom are completely decoupled. In this case, as in the classical limit $J_{zz} = 0$ [58, 59], the polarisation is either zero ($\delta Q = 0$), or takes on its maximum possible value ($\delta Q \approx \delta Q_{\text{max}}$). However introducing an interaction between the gauge charge and spinons, $\delta H_{\text{rotor}}$ [Eq. (7)] changes this, allowing for intermediate values $0 < \delta Q < \delta Q_{\text{max}}$, and permitting different forms of order to coexist. The driving force for this is a gain in the kinetic energy of the spinons [60]. As a result, the charge polarization is dressed with spinons, which can condense as pairs to give a charged version of the $Z_2$ QSL, or individually, to give a state with supersolid character, cf. Fig. 3b.

These phases could be distinguished in experiment through differences in heat capacity, and the fact that the charge polarisation $\delta Q$ induces a dipole moment, which could be detected as a Bragg peak in polarized neutron scattering [61, 62] — cf. Table I. Meanwhile, inelastic scattering would reveal a continuum of excitations associated with the remaining spinon degrees of freedom. While this analysis has been developed for a specific form of interaction, Eq. (19), the route outline from a spin liquid to coexisting orders is far more general, and is a compelling manifestation of the quantum nature of the problem.
Discussion — In this Letter, we have developed a systematic method of unveiling the unusual, “hidden” phases which descend from QSLs described by a lattice gauge theory. The mechanism we identify is the effective interactions between fractionalised quasi–particles (partons), which are generated by the physical constraint on the Hilbert space of the parent spin Hamiltonian. While earlier theoretical works [15, 16, 18] typically used perturbation theory to address the domain near to a soluble point, this approach makes it possible to connect the confined and deconfined limits of the lattice gauge theory, and to explore the new phases which arise at intermediate coupling. Generically, we find that these effective interactions can lead to a partial lifting of gauge symmetry, prior to the onset of a fully–confined, Higgs phase. In the specific case of pyrochlore quantum spin ice, this takes the form of a $\mathbb{Z}_2$ QSL, intermediate between a $U(1)$ QSL, and an easy–plane antiferromagnet, in which gauge fluctuations are fully confined.

We anticipate that this approach will make it possible to explore potential new phases in models which are difficult to solve by other methods. These include quantum spin ice models with frustrated interactions, where a $U(1)$ QSL with $\pi$–flux is expected at small $|J_\perp|/J_z$ [46], but quantum Monte Carlo (QMC) simulation fails, leaving the properties of this state relatively unexplored. Here we take encouragement from recent numerical results: variational calculations identify both the $\pi$–flux QSL, and a quantum spin nematic descended from it, at larger values of $|J_\perp|/J_z$ [63]. Moreover, a $\mathbb{Z}_2$ QSL, of the type we predict, has also been identified in QMC simulations of an extended model of unfrustrated quantum spin ice, occurring intermediate between a $U(1)$ QSL and its fully–confined Higgs phase [44].

Research into QSL continues to flourish, as new discoveries follow in both experiment and theory. The results in this Letter suggest that each new spin liquid discovered represents not only an opportunity in itself, but also a gateway to other new phases, which may have properties as exotic and interesting as the QSL they descend from. The approach developed here is a applicable to a wide range of spin liquids, and as such it should provide a valuable guide in the search for new quantum phases of matter.

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[63] O. Benton, L. D. C. Jaubert, R. R. P. Singh, J. Oitmaa, and N. Shannon, Phys. Rev. Lett. 121, 067201 (2018).
I. REVIEW OF U(1) LATTICE GAUGE THEORY

Here we briefly review the compact U(1) lattice gauge theory of quantum spin ice, following Refs. [4, 5, 45, 46]. We concentrate on the low–energy physics of the U(1) QSL, which is central to our analysis. We consider a pseudospin-1/2 model

\[ H_{\text{XXZ}} = H_0 + H_1, \]  

[Eq. (1) of main text], consisting of a dominant Ising exchange term

\[ H_0 = J_z \sum_{\langle ij \rangle} S_i^z S_j^z \]  

(S2)

and a quantum fluctuation term

\[ H_1 = -J_\pm \sum_{\langle ij \rangle} (S_i^+ S_j^- + \text{h.c.}). \]  

(S3)

Classical spin ice configurations satisfying a two–in, two–out condition

\[ \sum_{r' \in \langle rr' \rangle} S_{rr'}^z = 3 \sum_{\mu=0}^3 S_{r,r+e_\mu}^z = 0, \]  

(S4)

minimize

\[ H_0 = J_z \sum_{\langle ij \rangle} S_i^z S_j^z = \frac{J_z}{2} \sum_r \left( \sum_{r' \in \langle rr' \rangle} S_{rr'}^z \right)^2 + \text{const.}, \]  

(S5)

where \( r, r' \) are dual lattice (diamond) sites whose bond center is the pyrochlore site \( i \). We introduce a rotor representation,

\[ Q_r = \eta_r \sum_{\mu=0}^3 S_{r,r+e_\mu}^z, \quad S_{rr'}^z = \eta_r \epsilon_{rr'}, \]  

\[ S_{rr'}^+ = \phi_r^\dagger s_{rr'}^+ \phi_r \quad (s_{rr'}^+ = \frac{1}{2} e^{iA_{rr'}} \text{ for } r \in A), \]  

(S6)

[Eq. (2) of main text] in which the local constraint is mapped to the charge-free condition \( Q_r = 0 \) for all \( r \) by the Gauss law. Similarly, Eq. (S6) express the spin operators to be gauge-invariant under the local transformation

\[ \phi_r \rightarrow \phi_r e^{i\Lambda_r}, \quad A_{rr'} \rightarrow A_{rr'} + (\Lambda_r - \Lambda_{r'}). \]  

(S7)

From now on, we distinguish the Hamiltonian in mathcal font when it is represented in terms of the rotor variables Eq. (S6) while the original spin Hamiltonian Eqs. (S1)-(S3) is written in capital letter.

The divergenceless condition is violated by the spin flipping term \( H_1 \), which creates and annihilates a pair of gapped spinon excitations. In Eq. (S6), the spin flip \( S_{rr'}^+ \) raises (lowers) the gauge charge at the diamond sites \( r (r') \) by \( \pm 1 \), in other words

\[ [\phi_r, Q_r] = \phi_r \text{ or } [\phi_r, Q_r] = i. \]  

(S8)
where $\phi_r = e^{-i \phi r}$. The virtual excitation by spin flip term $H_1$ lifts the extensive degeneracy of classical spin ices and gives rise to the electromagnetic energy in the low-energy sector.

$$H_{EM} = \frac{U}{2} \sum_{\langle rr' \rangle} E_{rr'}^2 - \sum_{O} g_p \cos(B_p), \quad (S9)$$

$$B_p = \sum_{\langle rr' \rangle \in O} A_{rr'}, \quad g_p = 3J_z^3/2J_z^2, \quad (S10)$$

where the first term in Eq. (S9) enforces the electric field to be

$$E_{rr'} = g_p S_{rr'} = \pm 1/2 \quad \text{for large } U > 0,$$ \quad (S11)

and the symbol $O$ denotes the hexagonal plaquette which the lattice curl of the gauge field $A_{rr'}$ is defined on. For the unfrustrated $J_\perp > 0$, the coupling constant $g_p$ stabilizes 0-flux, $B_p = 0$. Meanwhile the frustrated exchange $J_\perp < 0$ stabilizes the $\pi$-flux, $B_p = \pi$, which doubles the unit cell.

Along with the gapped spinon, there are two more excitations in Eq. (S9). Since the gauge field $A_{rr'}$ (mod $2\pi$) is compact, the magnetic energy allows the topological defect, a gapped magnetic monopole excitation. Also the quantum theory of Eq. (S9) quantizes the field variables,

$$[A_{rr'}, E_{rr'}] = -i \quad \text{on the same link } rr'.$$ \quad (S12)

In the continuum limit, Eq. (S9) becomes

$$H_{EM} \sim E_{rr'}^2 + B_p^2,$$ \quad (S13)

which is analogous to the harmonic oscillator whose quanta corresponds to the gapless photon.

The total partition function, taking into account both partons and gauge fields, is then defined as

$$Z_{\text{total}} = \int D\phi^*_r D\phi_r DQ_r DA_{rr'} \delta(3 \sum_{\mu=0} E_{r,r+\eta,\tau} - Q_r) \exp \left\{ -S_{\text{eff}}[\phi^*_r, \phi_r, Q_r] - S_{\text{EM}}[A_{rr'}] \right\}, \quad (S14)$$

where $S_{\text{EM}}[A_{rr'}]$ is the pure electromagnetic action defined in Eq. (S9), and $S_{\text{eff}}$ describes a quantum rotor model

$$S_{\text{eff}} = \int_0^\beta d\tau \left( \sum_r \left( iQ_r \partial_r \phi^*_r + \lambda(\phi^*_r \phi_r - 1) \right) \right), \quad (S15)$$

[Eq. (5) of the main text], where

$$H_{\text{rotor}} = \frac{J_z}{2} \sum_r Q_r^2 - \frac{J_\perp}{4} \sum_{\langle rr' \rangle} \phi^*_r e^{i(\phi_{rr'} + A_{rr'})} \phi_{rr'} + \text{h.c.}, \quad (S16)$$

[Eq. (6) of the main text]. We note that the delta function in Eq. (S14) implements the Gauss law associated with the gauge charge. This restricts fluctuations to the physical Hilbert space $|Q_r| < 4S$, as pointed out in the main text.

In the limit of weak gauge field fluctuations, one can fix the specific gauge in Eqs. (S9) and (S10). Doing so, while allowing the gauge charge to take on values $Q \in (-\infty, \infty)$, we arrive the effective parton action for a pure quantum rotor model,

$$Z = \int D\phi^*_r D\phi_r DQ_r |\phi \in (-\infty, \infty)\rangle e^{-S_{\text{eff}}[\phi^*_r, \phi_r, Q_r]}, \quad (S17)$$

as defined in Eq. (4)–(6) of the main text.

This quantum rotor formalism also encompasses the emergence of magnetic order through a Higgs transition. Within the U(1) QSL which occurs $J_z \gg |J_\perp|$, spinons are both gapped and deconfined. As $|J_\perp|$ is increased, the spinon bandwidth increases, and the gap to spinon eventually closes, signalling the spinon condensation $\langle \phi_r \rangle \neq 0$. In the fully–confined limit $|J_\perp| \gg J_z$, the U(1) gauge structure is broken and the artificial photon is gapped out by Higgs mechanism. The ground state no longer possesses long–range entanglement, and is adiabatically connected to a product state with in–plane magnetization,

$$\langle S_{rr'}^+ \rangle = \langle \phi^*_r \rangle \langle s_{rr'}^+ \rangle \langle \phi_r \rangle \neq 0.$$ \quad (S18)

For the minimal model, Eq. (S1), these ground states are conventional–magnetically ordered states with ordering wave vector $k_0 = (0, 0, 0)$ for unfrustrated exchanges, $J_\perp > 0$, and $k_0 = (k, 0, 0)$ for frustrated exchange, $J_\perp < 0$ [46].

Once more general (anisotropic) exchange models are considered, a more diverse set of possible magnetic orders results. For example, terms of the form $\sim J_{\pm z} S_{i}^+ S_{j}^+$ can stabilize the coexistence of the deconfined spinons and the Ising orders, while terms of the form $\sim J_{\pm z} S_{i}^+ S_{j}^+$ tend to condense the spinons at $k = (2\pi, 0, 0)$ [46]. In this work, however, we concentrate instead on the possibility of phase transitions which are not driven by the condensation of a single spinon, and so lead to other, more exotic, phases of matter.
II. PROJECTED ROTOR REPRESENTATION

We now discuss the consequences of restricting fluctuation of charge to the physical domain \( Q_r \in (-4S, 4S) \), within the rotor formalism, deriving the effective interaction between spinons given in Eq. (17) of the main text. The recipe follows the two steps, (i) the restriction of the enlarged Hilbert space to be the physical one \( \mathbb{H}_S \) based on Eq. (2), (ii) then the Hamiltonian is modified \( H_{\text{rotor}}' \rightarrow H_{\text{rotor}} \) to take account of finiteness of the Hilbert space.

For simplicity, we first restrict the domain \( Q_r \in (-4S, 4S) \) at a site \( r \) only and leave others unchanged \( Q_{r' \neq r} \in (-\infty, \infty) \). Our strategy is to oblige the divergent eigenenergies of \( H_{\text{rotor}}' \) to suppress the Boltzmann factors outside the domain \( Q_r \in (-4S, 4S) \). Then we look for the Hermitian operator \( P_r \) defined by

\[
H_{\text{rotor}}' = P_r H_{\text{rotor}} P_r ,
\]

ensuring Eq. (9). Since \( P_r \) diagnoses whether \( |Q_r| < 4S \) or not, it would be a function of \(|Q_r|\).

\[
|P_r(Q_r)|^2 = \Theta[4S - |Q_r|] + \Theta[-4S + |Q_r|] \cdot \infty ,
\]

\[
\Theta[x] = \begin{cases} 1 & \text{for } x > 0 \\ 0 & \text{otherwise} , \end{cases}
\]

However, the function Eq. (S20) is ill-defined due to the ambiguities in the second term and at \(|Q_r| = 4S\). From the mapping Eq. (2), the gauge charge \( Q_r \) actually takes the integer values \( 0, \pm 1, \ldots, \pm 4S \) only. Instead of Eq. (S20), we consider the function \( P_r|Q_r| \) of to real \( Q_r \in (-\infty, \infty) \). Then we can define

\[
P_r(Q_r) = \begin{cases} 1 & \text{for } Q_r = 0, \pm 1, \ldots, \pm 4S \\ 0 & \text{for } Q_r = \pm(4S + 1) , \end{cases}
\]

The first condition for \( Q_r = 0, \pm 1, \ldots, \pm 4S \) validates the first term of Eq. (S20). The second condition \( P_r(Q_r = \pm(4S + 1)) = 0 \) is resolved as

\[
P_r(Q_r) = \begin{cases} |Q_r + 1| & \text{for } Q_r = 0, \pm 1, \ldots, \pm(4S - 1), -4S \\ 0 & \text{for } Q_r = +4S \\ |Q_r - 1| & \text{for } Q_r = 0, \pm 1, \ldots, \pm(4S - 1), +4S \\ 0 & \text{for } Q_r = -4S \\ Q_r |Q_r| & \text{for } Q_r = 0, \pm 1, \ldots, \pm 4S \\ 0 & \text{for } Q_r = \pm(4S + 1) , \end{cases}
\]

as implied by the ladder termination

\[
S_{rr}^+ |S_{rr}^z = \pm S \rangle = 0 .
\]

To obtain a closed–form expression for the projection operator, we include the integers \( Q_r = \pm(4S + 1) \) in its domain, and seek a polynomial satisfying the conditions Eq. (S22). Since Eq. (S20) is an even function of \( Q_r \), this polynomial \( P_r \) also consists of even powers of \( Q_r \). Defined in this way, there is one and only one solution for \( P_r \) to Eq. (S21), a polynomial of order \( 2(4S + 1) \)

\[
P_r(Q_r) = 1 - \frac{1}{(4S + 1)(8S + 1)!} \prod_{k=0}^{4S}(Q_r^2 - k^2) = 1 - \alpha_S Q_r^2 + \ldots - \frac{1}{(4S + 1)(8S + 1)!} Q_r^{8S+2} \quad (\alpha_S = \frac{(4S)!}{(4S + 1)(8S + 1)!}) .
\]

From this series, we single out the correction with the lowest power, \( \alpha_S Q_r^2 \). This is a rapidly decreasing function of the control parameter \( S \); by application of the Stirling approximation \( \log(N!) \approx N \log N \) we find

\[
\alpha_S \approx \frac{1}{S^{2S+2}} .
\]

In the physical limit, \( S = \frac{1}{2} \), it has the value

\[
\alpha_S = \frac{1}{90} .
\]
Most significant is the sign of this term, $\alpha_S > 0$. This is fixed by the condition Eq. (S21). Adding additional constraints of the form $P[|Q_r| \geq 4S + 2] = 0$, for integer $Q_r$ outside the physical range introduce higher-order terms $O(Q_r^{8S+2})$ in Eq. (S24), but leaves the sign of $\alpha_S$ unchanged.

In Fig. S1a we plot the polynomial form of the projection operator $P_r[Q_r]$ [Eq. (S24)] as a function of $Q_r$, for $S = 1/2, 1, 3/2$. Meanwhile, in Fig. S1b we show its square $P_r[Q_r]^2$. It is the square of the projection operator which determines the matrix elements entering into the projected rotor Hamiltonian [Eq. (10) of the main text]. We see that $P_r[Q_r]^2 \approx 1$ on the physical interval $(-4S, 4S)$, but diverges rapidly for $|Q_r| > 4S + 1$, with the asymptotic behaviour $P_r[Q_r]^2 \sim Q_r^{16S+4}$ for $|Q_r| \to \infty$. This, combined with the $Q_r^2$ term in the original rotor Hamiltonian [Eq. (6) of the main text], will eliminate unphysical states at large $|Q_r|$ from the path integral determining the partition function [Eq. (9) of the main text].

We now generalize the mapping $(Q_r, \phi_r, \phi_r^\dagger) \to (\tilde{Q}_r, \tilde{\phi}_r, \tilde{\phi}_r^\dagger)$ for all $r$,

\begin{equation}
H_{\text{rotor}}' = H_{\text{rotor}}[\tilde{Q}_r, \tilde{\phi}_r, \tilde{\phi}_r^\dagger] = (\tilde{O}_r \equiv P_r O_r P_r),
= H_{\text{rotor}}[Q_r, \phi_r, \phi_r^\dagger] + \delta H_{\text{rotor}}[Q_r, \phi_r, \phi_r^\dagger],
\end{equation}

where the new matter fields $\tilde{Q}_r, \tilde{\phi}_r$ project out the unphysical portions outside the domain $Q_r \in (-4S, 4S)$. This projection generates new effective interactions between the spinons (partons) of quantum spin ice.

In Eq. (S27), we ignore the non-local interactions coming from $P_r \phi_r^{(1)} P_r$ since $\phi_r^{(1)}$ doesn’t affect the gauge charge $Q_r$ at $r$. As quantum fluctuation $H_1$ rises, it monotonically increases $Q_r^2$, implying that the corrections $P_r = 1 - \alpha_S Q_r^2 + \ldots$ in Eq. (S24) contribute to $\delta H_{\text{rotor}}$ in order of degrees. The coefficient $\alpha_S$ in the lowest correction properly recognizes that the effective interaction $\delta H_{\text{rotor}}$ is significant for the small spin $S$.

Integration of $\delta H_{\text{rotor}}$ with respect to the charge field $Q_r$ contributes to an effective spinon interaction in two different ways

(1) Non-zero commutator Eqs. (S8), (S30) during the normal-ordering.

(2) Non-trivial Jacobian transformation in the path integral.

It turns out that dominant contribution comes from (2), and that the lowest order, the effective parton interaction $\delta H_{\text{rotor}}$ is given by

\begin{equation}
S_{\text{rt}}^+ S_{\text{rt}}^{-} \approx \frac{1}{3} \left(1 - \alpha_S (Q_r^2 + Q_r^{2\prime})\right) \phi_r^\dagger e^{iA_r} \phi_r \left(1 - \alpha_S (Q_r^2 + 2Q_r^2 + Q_r^{2\prime})\right) \phi_r^\dagger e^{iA^{r\prime}} \phi_r \left(1 - \alpha_S (Q_r^2 + Q_r^{2\prime})\right).
\end{equation}

Physically, the effective parton interaction in Eq. (S29) reflects that both gauge charges $Q_r = \pm 4S$ block the propagation $\sim \phi_r^\dagger \phi_r \phi_r^\dagger \phi_r$. Thus it is natural to consider the parton interaction affected by the size of the gauge charge $|Q_r|$ rather than the gauge charge $Q_r$ itself. To evaluate Eq. (S29) in the path integral formalism, it is required to normally order them. Applying Eq. (S8) $n$-times, we obtain

\begin{equation}
\phi_r Q_r^n = (Q_r + 1)^n \phi_r, \quad \phi_r^\dagger Q_r^n = (Q_r - 1)^n \phi_r^\dagger.
\end{equation}
Before proceeding, we note that the new mapping \( S^+_{rr} = \frac{1}{2}\phi_1^+ e^{iA_{rr}} \phi_{r'} \rightarrow \frac{1}{2}\phi_1^{+*} e^{A_{rr}'} \phi_{r'} \) now not only raises/lowers the gauge charge at \( r/r' \) but also diagnoses whether the gauge charge is within the physical domain \((-4S, 4S)\) or not. Thus, the spin exchange Eq. (S29) depends on the sequences of spin operators \( S^+_{rr'}, S^{-}_{r'r} \neq S^+_{r'r}, S^-_{rr'} \). To address this problem, we evaluate the both contribution to replace \( S^+_{rr'}, S^-_{r'r} \) in Eq. (S29) by the symmetric summation

\[
S^+_{rr}, S^-_{r'r} \rightarrow \frac{1}{2}(S^+_{rr}, S^-_{r'r} + S^-_{r'r}, S^+_{rr}) .
\] (S31)

As a result, this leads to,

\[
\mathcal{H}_1 + \delta \mathcal{H}_{\text{rotor}} = -\frac{J_{\pm}}{4} \sum_{\langle(rr')\rangle} \frac{1}{2} \left( P_r |Q_r| P_{r'} |Q_{r'}| - 1 \right) P_r |Q_{r'}| \left( [P_r |Q_{r'}| + 1]^2 P_r |Q_{r'}| P_{r'} |Q_{r'}| + 1 \right) \phi_1^+ e^{i(A_{rr} + A_{rr'})} \phi_{r'} + \text{h.c.}
\]

\[
+ P_r |Q_{r'}| P_{r'} |Q_{r'}| - 1 \right) P_r |Q_{r'}| \left( [P_r |Q_{r'}| - 1]^2 P_r |Q_{r'}| P_{r'} |Q_{r'}| + 1 \right) \phi_1^+ e^{i(A_{rr} + A_{rr'})} \phi_{r'} + \text{h.c.}
\]

\[
= -\frac{J_{\pm}}{4} \sum_{\langle(rr')\rangle} \left[ 1 - \alpha_S \left( Q_r^2 + 2Q_{r'}^2 + Q_{r'} - 1 \right) \right] \phi_1^+ e^{i(A_{rr} + A_{rr'})} \phi_{r'} + \text{h.c.} + O(Q_r^4) .
\] (S32)

On the first and the second lines, we mark the underlines to emphasize the different corrections from Eq. (S31). Integrating out the gauge charge \( Q_r \), Eq. (S32) gives rise to several powers of \( J_{\pm} \phi_{r'}^1 e^{i(A_{rr} + A_{rr'})} \phi_{r'} \) in the spin action. Due to the commutator Eq. (S30), the spinon band width is trivially rescaled as

\[
J_{\pm} \rightarrow J_{\pm} (1 - 4\alpha_S) ,
\] (S33)

in Eq. (S32). The lowest correction signalling non-trivial instabilities out of U(1) QSL comes from the quartic corrections in spinons. Performing the Gaussian integral \( \mathcal{H}_0 + \delta \mathcal{H}_{\text{rotor}} \) over the charges \( Q_r \) and \( Q_{r'} \),

\[
\mathcal{H}_r[\phi_r, \phi_r^+] = 4\alpha_S \frac{1}{2} \sum_{r' \in \langle(rr')\rangle} \left( \phi_1^+ e^{i(A_{rr'} + A_{rr'})} \phi_{r'} + \text{h.c.} \right) + \sum_{r \in \langle(rr')\rangle} \left( \phi_1^+ e^{i(A_{rr} + A_{rr'})} \phi_{r'} + \text{h.c.} \right) ,
\]

\[
\mathcal{H}_1 = -\frac{J_{\pm}}{4} \sum_{r} \mathcal{H}_r[\phi_r, \phi_r^+] \mathcal{H}_{\text{rotor}}[\phi_r, \phi_r^+] ,
\] (S35)

Up to the quartic terms in the spinon equations, Eq. (S34) reads \( \sim -\left( \phi_1^+ e^{i(A_{rr'} + A_{rr'})} \phi_{r'} \right) (\partial_r \phi_r) \) and \( \sim -\left( \phi_1^+ e^{i(A_{rr} + A_{rr'})} \phi_{r'} \right)^2 \). The first of these terms originates in the Berry phase. For purposes of a mean field theory, in the static limit, this term can be neglected [cf. main text]. The second term, meanwhile, originates in the non-zero commutator for the normal-ordered path integral in Eqs. (S30) and (S32). Approximating the denominator in Eq. (S34) as \( 2J_{\pm} \), it becomes,

\[
\frac{1}{2J_{\pm}} \left( \left( \partial_r \phi_r \right)^2 + \left( \partial_{r'} \phi_{r'} \right)^2 \right) - \frac{J_{\pm}}{2} \left( \left( \phi_1^+ e^{i(A_{rr'} + A_{rr'})} \phi_{r'} \right)^2 + \left( \phi_1 e^{-i(A_{rr} + A_{rr'})} \phi_{r'} \right)^2 \right)
\]

\[
- i J_{\pm} \alpha_S \left( \partial_r \phi_r - \partial_{r'} \phi_{r'} \right) \left( \phi_1^+ e^{i(A_{rr'} + A_{rr'})} \phi_{r'} - \phi_1 e^{-i(A_{rr} + A_{rr'})} \phi_{r'} \right) \rightarrow 0 .
\] (S36)

Where, since \( \phi_r = e^{-i\phi_r} \), we have used

\[
(\partial_r \phi_r)^2 = \partial_r \phi_r \partial_r \phi_r ,
\]

\[
i(\partial_r \phi_r - \partial_{r'} \phi_{r'}) \left( \phi_1^+ e^{i(A_{rr'} + A_{rr'})} \phi_{r'} - \phi_1 e^{-i(A_{rr} + A_{rr'})} \phi_{r'} \right) = \partial_r \left( \phi_1^+ e^{i(A_{rr} + A_{rr'})} \phi_{r'} - \phi_1 e^{-i(A_{rr} + A_{rr'})} \phi_{r'} \right) \rightarrow 0 .
\] (S37)
where $S$ pairing; instead it will contribute to the multipolar character of the resulting condensate, discussed below.

Finally, we consider the Jacobian contribution from the case (2) in Eq. (S28). While performing the Gaussian integration in Eq. (S35),

$$
\int D\phi_r^*D\phi_rDQ_r\exp\left[-\int_0^{\beta} \! d\tau \left\{ \sum_r \left( \frac{J_r}{2} + \frac{J_r}{4} \mathcal{H}_r \right) Q_r^2 + iQ_r \partial_r \phi_r + \ldots \right\} \right],
$$

with the variable transformation $\tilde{Q}_r = Q_r \sqrt{\frac{J_r}{2} + \frac{J_r}{4} \mathcal{H}_r}$. While the integrand in the exponential are already taken into account in Eqs. (S34)-(S36), the Jacobian of spinon fields also gives rise to non-trivial contributions to the spinon action. The variable transformation is given by,

$$
\bar{\phi}_r = \frac{\phi_r^*}{\left( 1 + \frac{J_r}{2 \mathcal{H}_r} \right)^{1/4}}, \quad \bar{\phi}_r^* = \frac{\phi_r}{\left( 1 + \frac{J_r}{2 \mathcal{H}_r} \right)^{1/4}}.
$$

Thus, the inverse transformation,

$$
\phi_r \simeq \frac{\bar{\phi}_r}{\left( 1 - \frac{J_r}{2 \mathcal{H}_r} \right)^{1/4}}, \quad \phi_r^* \simeq \frac{\bar{\phi}_r^*}{\left( 1 - \frac{J_r}{2 \mathcal{H}_r} \right)^{1/4}},
$$

enable us to rewrite the partition function in terms of Eq. (S38). Assuming the correlation of $\hat{\phi}_r$ is similar to that of $\bar{\phi}_r$ in the intermediate coupling regime, the quartic spinon interaction $\sim (J_r^2/\alpha_S J_z) \mathcal{H}_r$. $\mathcal{H}_r$ is determined by,

$$
Z' = \int D\bar{\phi}_r^*D\bar{\phi}_r D\left( \frac{\tilde{Q}_r}{\left( 1 - \frac{J_r}{2 \mathcal{H}_r} \right)^{1/2}} \right) e^{-\mathcal{S}_0[\phi_r^*, \phi_r]} \int D\bar{\phi}_r^* D\bar{\phi}_r e^{-\mathcal{S}[\phi_r^*, \phi_r]} \approx \int D\bar{\phi}_r^* D\bar{\phi}_r e^{-\mathcal{S}[\phi_r^*, \phi_r]},
$$

where $S$ can be approximated by substituting Eq. (S40). Doing so, we obtain,

$$
\mathcal{H}_1 = -\frac{J_r}{4} \sum_{\langle rr' \rangle} (\phi_r^* e^{i(A_{rr'} + A_{rr'})} \phi_{rr'} + \text{h.c.})
$$

$$
\rightarrow \mathcal{H}_1 + \mathcal{H}_u = -\frac{J_r}{4} \sum_{\langle rr' \rangle} (\phi_r^* e^{i(A_{rr'} + A_{rr'})} \phi_{rr'} + \text{h.c.}) - \frac{3J_r^2 \alpha_S}{8J_z} \sum_{\langle rr' \rangle} \left( (\phi_r^* e^{i(A_{rr'} + A_{rr'})} \phi_{rr'})^2 + \text{h.c.} \right) + \ldots
$$

Combining Eqs. (S36) and (S42), we find that the rotor model, Eq. (6), is modified as follows

$$
S[\phi_r, \phi_r^*] = \int_0^{\beta} \! d\tau \left[ \sum_r \left( \frac{1}{2J_z} \partial_r \phi_r^* \partial_r \phi_r + \lambda (\phi_r^* \phi_r - 1) \right) + \mathcal{H}_1 + \mathcal{H}_u \right],
$$

$$
\mathcal{H}_u = -u_0 \sum_{\langle rr' \rangle} \left( \phi_r^* e^{i(A_{rr'} + A_{rr'})} \phi_{rr'} \right)^2 - u_1 \sum_{r_1 \neq r_2, r_3} \left( \sum_{r_1' \neq r_2'} \phi_r^* e^{i(A_{rr'} + A_{rr'})} \phi_{rr'} \right) \left( \sum_{r_3' \neq r_2'} \phi_r^* e^{i(A_{rr'} + A_{rr'})} \phi_{rr'} + \text{h.c.} \right)
$$

$$
-u_2 \sum_{r_1' \neq r_2'} \left( \sum_{r_3' \neq r_2'} \phi_r^* e^{i(A_{rr'} + A_{rr'})} \phi_{rr'} \right) \left( \sum_{r_3' \neq r_2'} \phi_r^* e^{i(A_{rr'} + A_{rr'})} \phi_{rr'} + \text{h.c.} \right) + \text{h.c.},
$$

where the coefficients

$$
u_0 = \frac{J_r^2}{8J_z} \alpha_S (3 + 4\alpha_S), \quad u_1 = \frac{J_r^2 \alpha_S}{2J_z}, \quad u_2 = \frac{J_r^2 \alpha_S}{8J_z}, \quad u_3 = \frac{J_r^2 \alpha_S}{4J_z},
$$

are all of order $\sim J_r^2/j_z$, and can be found by enumerating terms in Eq. (S35). For small $|J_r|/|J_z|$, the effective interaction $\mathcal{H}_u$ is irrelevant. However, at finite $|J_r|/|J_z|$, quantum fluctuations can be substantial, and have the potential to qualitatively change the nature of the U(1) QSL, by pairing spinons. For simplicity, the discussion of this point in the main text is restricted to the on–site term $\mathcal{H}_u$. However the presence of longer–range attractive interactions in Eq. (S44) is not expected to suppress spinon pairing; instead it will contribute to the multipolar character of the resulting condensate, discussed below.
III. PURE CHARGE-2 PROPAGATION

Multipolar moment

Here we explore the connection between spinon pairing and phases which exhibit a finite multipole moment on bonds. Since the spin operator [Eq. (S6)] is bilinear in spinons, the transverse dipole moment vanishes in the absence of a single–spinon condensate, i.e.

\[ \langle S^+_i S'^+_j \rangle = 0 \quad \text{if} \quad \langle \phi_r \rangle = 0 . \]  

(S46)

Spinons are expected to condense for large value of \( |J_{\pm}|/J_z \), in the fully–confined Higgs phase. However, for intermediate values of coupling, where there is no single–spinon condensate, multipole moments can still take on finite values on the bonds of the lattice, without leading to a confinement of spinons.

As a concrete example, we consider a quadrupole moment formed from transverse spin components,

\[ Q = \frac{1}{N} \sum_{ij} \left( Q^{ij}_{ij} \right) = \frac{1}{N} \sum_{ij} \left( S^x_i S^x_j - S^y_i S^y_j \right) , \]  

(S47)

where \( i, j \) are pyrochlore lattice sites. Although a finite \( Q \) preserves the time–reversal symmetry for both Kramer/non-Kramer materials, it breaks spin–rotation symmetry in the same way as the director in a nematic liquid crystal breaks rotation symmetry. And for this reason states with finite \( Q \) (but vanishing dipole order) are commonly referred to as a “spin nematics”.

In terms of diamond–lattice sites, Eq. (S47) can be written

\[ S^+_{rr'} S^+_{r''r''} = Q^{1}_{ij} + i Q^{2}_{ij} = \frac{1}{4} \phi^+_{r'} \phi^+_{r''} e^{i(A_{r'} + A_{r''})} \phi_{r'} \phi_{r''} , \]  

(S48)

where the nearest-neighbor diamond sites \( r, r' (r'', r''') \) define the pyrochlore site \( i (j) \). A necessary condition for spin–nematic order is therefore a finite pairing of spinons on the bonds of the diamond lattice

\[ \Delta_0 \neq 0, \quad \Delta_{\mu - \nu} \neq 0 \quad \text{where} \quad \Delta_{\mu - \nu} = \langle \phi_r \phi_{r + e_\mu} e^{A_{r'} + A_{r''}} \rangle . \]  

(S49)

Such pairing is very naturally motivated by the attractive interactions between spinons in Eq. (S44), and can be studied at mean field level, as described in the main text. As also noted in the main text, the presence of pair condensate breaks the \( U(1) \) gauge structure down to \( \mathbb{Z}_2 \). And for this reason the spin–nematic phase retains many of the characteristics of a \( \mathbb{Z}_2 \) QSL.

Phase transition

The effective model studied in this Letter

\[ \mathcal{H}'' = \mathcal{H}_{\text{rotor}} + \mathcal{H}_u \]  

(S50)

[Eq. (15) of the main text], contains both terms which mediate both the propopagation of both individual spinons, and pairs of spinons. For small \( |J_{\pm}|/J_z \) this models support a \( U(1) \) QSL, and the Higgs transition occurring for \( u = 0 \) [Fig. 2a] is already well understood in terms of a BEC of spinons, leading to a phase with conventional magnetic order [46]. We now consider the phase transition occurring as a function \( u/J_z \), for \( J_{\pm} = 0 \), which occurs through the condensation of pairs of spinons. In this case, the projected rotor model reduces to

\[ \mathcal{H}_\Delta \equiv \mathcal{H}_0 + \mathcal{H}_u = \frac{J_x}{2} \sum_r q^2_r - u \sum_{\langle rr'' \rangle} \left( \phi^*_r e^{i(A_{rr''} + A_{rr'''})} \phi_{rr''} \right)^2 + \text{h.c. } . \]  

(S51)

where sum \( \langle \langle rr'' \rangle \rangle \) runs over second–neighbour bonds of the diamond lattice, with coordination number \( z' = 2 \times (\frac{4}{2}) = 12 \). This Hamiltonian is quartic in the spinon fields, and we solve it by seeking a mean–field decoupling in terms of suitable order parameters.

The pattern for this calculation follows the well–studied example of single–spinon propagation within a rotor model [14]. In this case the term endowing the spinons with dispersion [Eq. (S3)] can be transcribed in terms of the phases of rotors \( \phi_r = e^{-i\varphi_r} \), to give

\[ \mathcal{H}_1 = -\frac{J_{\pm}}{2} \sum_{\langle rr'' \rangle} \cos(\varphi_r - \varphi_{rr''} + (A_{rr'} + A_{rr''})) , \]  

(S52)
and, once a gauge has been fixed, we can anticipate a broken–symmetry state

$$\langle \cos(\varphi_r) \rangle \neq 0 \quad \text{for} \quad |J_\pm| \to \infty .$$  \hspace{1cm} (S53)

This phase transition is analogous to the emergence of superconducting order, with associated breaking of gauge symmetry.

By analogy, the second term in Eq. (S51) can be written

$$\mathcal{H}_u = -2u \sum_r \cos(2\varphi_r - 2\varphi_r' + 2(A_{rr} + A_{rr'})) ,$$  \hspace{1cm} (S54)

and we anticipate that

$$\langle \cos(2\varphi_r) \rangle \neq 0 \quad \text{for} \quad u \to \infty .$$  \hspace{1cm} (S55)

where, in the broken symmetry state, the gauge field takes on a result, the U(1) gauge fluctuation

$$\alpha$$

and we anticipate that

$$\langle \cos(2\varphi_r) \rangle \neq 0 \quad \text{for} \quad u \to \infty .$$  \hspace{1cm} (S55)

where the energies of the (localised) spinon bands are given by

$$\omega \langle \cos(2\varphi_r) \rangle \neq 0 \quad \text{for} \quad u \to \infty .$$  \hspace{1cm} (S55)

In this case, we can fix

$$\langle e^{iA_{rr'}} \rangle^2 = 1$$

and define an order parameter

$$\Delta = \langle \phi_r \phi_r \rangle$$  \hspace{1cm} (S56)

so that Eq. (S51) reduces to

$$\mathcal{H}_\Delta = \frac{J_z}{2} \sum_r Q_r^2 - 12u \sum_r (\Delta \phi_r^+ \phi_r^+ + \text{h.c.} - |\Delta|^2) ,$$  \hspace{1cm} (S57)

The phase of the order parameter $\Delta$ can be eliminated by a further gauge transformation $\Delta \to \Delta e^{2i\lambda_r}$, so that we work with a real, positive $\Delta > 0$.

We can now integrate out the gauge charge $Q_r$, and solve for the dynamics of spinon pairs in the basis

$$[\phi_{k,\omega,\alpha}, \phi_{-k,-\omega,\alpha}]^T$$  \hspace{1cm} (S58)

where $\alpha \in A$ or B sublattice of diamond lattice sites, and $\omega_n = 2\pi n T$ is a bosonic Mastubara frequency. Doing so, we obtain an (inverse) Green’s function

$$G_{k>0,\alpha}(i\omega_n) = \begin{bmatrix} \frac{\omega_n^2}{2J_z} + \lambda & -2 \cdot 12u\Delta \\ -2 \cdot 12u\Delta & \frac{\omega_n^2}{2J_z} + \lambda \end{bmatrix} ,$$

where $\lambda$ is the Lagrange multiplier enforcing the rotor constraint, cf. Eq. (S15). Here, the additional factor of 2 in the off–diagonal component comes from the restriction on momenta

$$-12u \sum_{k,\alpha=A,B} (\Delta \phi_{k,\alpha}^+ \phi_{-k,\alpha} + \text{h.c.}) = -24u \sum_{k>0,\alpha} (\Delta \phi_{k,\alpha}^+ \phi_{-k,\alpha} + \text{h.c.}) .$$  \hspace{1cm} (S60)

Transforming to imaginary time, and considering the limit of zero temperature

$$G_{k,\alpha}(\tau = 0) = T \sum_{i\omega_n} G_{k,\alpha}(i\omega_n) e^{i0\omega_n} \Rightarrow 0k = \int_{-\infty}^{\infty} \frac{d\omega_n}{2\pi} G_{k,\alpha}(i\omega_n) e^{i0\omega_n} ,$$  \hspace{1cm} (S61)

we find

$$G_{k,\alpha,11}(\tau = 0) = G_{k,\alpha,22}(\tau = 0) = \frac{J_z}{2} (\frac{1}{\omega_1} + \frac{1}{\omega_2}) ,$$  \hspace{1cm} (S62a)

$$G_{k,\alpha,12}(\tau = 0) = G_{k,\alpha,21}(\tau = 0) = \frac{J_z}{2} (\frac{1}{\omega_1} - \frac{1}{\omega_2}) ,$$  \hspace{1cm} (S62b)

where the energies of the (localised) spinon bands are given by

$$\omega_{1,2} = \sqrt{2J_z(\lambda \mp 24u\Delta)}$$  \hspace{1cm} (S63)
The values of $\lambda$ and $\Delta$ can then be determined through the self–consistency conditions

$$\frac{1}{N} \sum_{\mathbf{r} \in A} \langle \phi_{\mathbf{r}}^{2} \phi_{\mathbf{r}} \rangle = \int_{\mathbf{k}} \langle \phi_{\mathbf{k},A}^{2} \phi_{\mathbf{k},A} \rangle (\tau = 0) = \int_{\mathbf{k}} G_{\mathbf{k},A,11} = 1,$$

$$\frac{1}{N} \sum_{\mathbf{r} \in A} \langle \phi_{\mathbf{r}}^{2} \phi_{\mathbf{r}} \rangle = \int_{\mathbf{k}} \langle \phi_{\mathbf{k},A} \phi_{\mathbf{k},A} \rangle (\tau = 0) = \int_{\mathbf{k}} G_{\mathbf{k},A,21} = \Delta,$$  \hspace{1cm} (S64)

where $\int_{\mathbf{k}} = \int \frac{d^{d}k}{(2\pi)^{d}}$ denotes the $\mathbf{k}$-space integral over the Brillouin zone. Due to the absence of single spinon propagation, no BEC (of the single spinon) occurs for any value of $u/J_z$, and the integrand in Eq. (S64) is independent of $\mathbf{k}$. It follows that

$$\omega_1 = \frac{J_z}{1+\Delta}, \quad \omega_2 = \frac{J_z}{1-\Delta},$$  \hspace{1cm} (S65)

and the ground state energy, Eq. (S57), is given by

$$\frac{1}{N} \langle H_{\Delta} \rangle = \frac{J_z}{2} \frac{1}{N} \sum_{\mathbf{r}} \langle Q_{\mathbf{r}}^{2} \rangle - 24u\Delta^2 = \frac{1}{2} \int_{\mathbf{k}} (\omega_1 + \omega_2) - 24u\Delta^2 = J_z + (J_z - 24u)\Delta^2 + J_z\Delta^4 + \cdots.$$  \hspace{1cm} (S66)

It follows that there is a 2nd-order phase transition at $u_c = J_z/24 \approx 0.0417 J_z$, between states with $\Delta = 0$ and $\Delta \neq 0$.

We conclude by commenting on the order of the phase transition predicted by Eq. (S66). In Ref. [46], a BEC driven by a similar form of spinon interaction $\sim J_{\pm\pm}S_{1\pm}S_{1\pm} \sim J_{\pm\pm}\phi_{\pm}\phi_{\pm}\phi_{\pm\pm}$ was investigated, and found to be first order. However, in this case, the BEC involved a finite single–spinon condensate, $\langle \phi_{\mathbf{r}} \rangle \neq 0$. And within the framework of gauge mean field theory (gMFT), in the limit of weak gauge fluctuations, we find that any BEC driven by spinon interactions, for which $\langle \phi_{\mathbf{r}} \rangle \neq 0$, will be first order. Conversely, any continuous transition driven by interactions (in the same limit), must necessarily have $\langle \phi_{\mathbf{r}} \rangle \equiv 0$, consistent with Eq. (S66).

Our argument proceeds as follows. Suppose that, within a gMFT treatment of a quantum spin ice, a spinon interaction $\mathcal{H}^*_u \sim u(\phi_{\mathbf{r}_1}^{\dagger} \phi_{\mathbf{r}_2}^{\dagger} \phi_{\mathbf{r}_3} \phi_{\mathbf{r}_4})$ induces a BEC of spinons, $\langle \phi_{\mathbf{r}} \rangle \neq 0$, at some $u = u_c$. In the condensed phase, if the single spinon condensate $\langle \phi_{\mathbf{r}} \rangle \neq 0$ dominates the physical quantities, we expect $\langle \phi_{\mathbf{r}_1} \phi_{\mathbf{r}_2} \rangle \simeq \langle \phi_{\mathbf{r}_1} \rangle \langle \phi_{\mathbf{r}_2} \rangle$. In this case a mean-field decoupling of the form $\mathcal{H}^*_u \sim u(\phi_{\mathbf{r}_1}^{\dagger} \phi_{\mathbf{r}_2}^{\dagger} \phi_{\mathbf{r}_3} \phi_{\mathbf{r}_4}) + \langle \phi_{\mathbf{r}_1} \rangle \langle \phi_{\mathbf{r}_2} \rangle \phi_{\mathbf{r}_3} \phi_{\mathbf{r}_4} + \cdots$ is sufficient to describe the BEC transition. It follows that, if the BEC at $u = u_c$ is continuous (2nd-order) $\langle \phi_{\mathbf{r}} \rangle |_{u=u_c} = 0$, and the spinon interaction is not effective at the transition, i.e $\mathcal{H}^*_u |_{u=u_c} \simeq 0$. At the same time, the BEC should be signalled by an integrable singularity in bosonic statistics $\langle \phi_{\mathbf{k}}^{\dagger} \phi_{\mathbf{k}} \rangle$ in $\mathbf{k}$-space. However, at a mean–field level, $\mathcal{H}^*_u |_{u=u_c} \simeq 0$ cannot lead to any singular change in the Green function at the phase boundary $u = u_c$. A first–order transition, to a state with finite $\langle \phi_{\mathbf{r}} \rangle |_{u=u_c} \neq 0$, is therefore needed to induce a sudden divergence in the Green function as $u \rightarrow u_c$. It follows that, within gMFT, any BEC driven by interactions of the form $\mathcal{H}^*_u$ that has a continuous character must be associated with the onset of an unconventional, “hidden”, phase. Discontinuous (first–order) transitions may however occur into conventional or unconventional phases.

**IV. PHASE BOUNDARIES**

In Fig. 3b of the main text, we present phase diagram exhibiting some of the exotic phases which can be born out of a $U(1)$ QSL. The characteristics of these phases are listed in Table. I. In this Section, we describe how the phase boundaries shown in Fig. 3b were estimated.

**Higgs transition**

The “Higgs” phase at large $|J_{\pm}|/J_z$ is associated with the condensation of an individual spinon mode. This is is signalled by a singularity in the associated spinon Green’s function. We consider spinons moving on a diamond lattice, subject to an XXZ model with additional longer–range interactions

$$\mathcal{H}_{XXZ} = \mathcal{H}_Q + \mathcal{H}_1, \quad \mathcal{H}_Q = \mathcal{H}_0 + \mathcal{H}_{ZZ},$$  \hspace{1cm} (S67)

where $\mathcal{H}_0$ is defined through Eq. (S2), $\mathcal{H}_1$ through Eq. (S3), and $\mathcal{H}_{ZZ}$ through Eq. (19) of the main text. The Coulomb interaction term, $\mathcal{H}_Q$, can be transcribed using the Fourier transform,

$$Q_{\mathbf{r}a}(\tau) = \frac{1}{\sqrt{N}} \sum_{\mathbf{k}} Q_{\mathbf{k}a}(\tau) e^{i\mathbf{k} \cdot \mathbf{r}},$$  \hspace{1cm} (S68)
where $\alpha = A, B$ is a sublattice index, to give

$$
\mathcal{H}_Q = \sum_{k} (Q_{kA}^* Q_{kB}^* \left( \begin{array}{cc} J_z/2 & J_{zz} R_k/2 \\ J_{zz} R_k/2 & J_z/2 \end{array} \right) \left( \begin{array}{l} Q_{kA} \\ Q_{kB} \end{array} \right)) ,
$$  
(S69)

where

$$
R_k = \sum_{\mu=0}^{3} e^{i k e_{\mu}},
$$  
(S70)

is the structure constant for a diamond lattice defined by vectors $e_{\mu}$. Above, the additional factor 1/2 in the off-diagonal term is due to the fact that $Q_{ko}$ is real, i.e. $Q_{ko}^* = Q_{-ko}$.

$$
\mathcal{H}_{ZZ} = J_{zz} \sum_{\alpha, \alpha'} Q_{ro} Q_{r, \alpha'} = \frac{J_{zz}}{2} \left( \sum_{\alpha, \alpha'} Q_{ro} Q_{r, \alpha'} + \sum_{\alpha, \alpha'} Q_{ro} Q_{r, \alpha'} \right)
$$
(S71)

The coupling of $A$ and $B$ sublattices in Eq. (S69) causes the charge stiffness in $k$-space to split into two distinct branches,

$$
J_z \rightarrow J_z^{(\pm)} \equiv J_z \pm J_{zz} | R_k |
$$
(S72)

Consistent with this, the first term in the action Eq. (S43) must be modified, with the (inverse) charge stiffness $J_z$ in the denominator replaced by $J_z^{(\pm)}$ divided by the determinant of Eq. (S69). This modifies the phase boundary of Higgs transition associated with spinon condensation.

By integrating out the gauge charge $Q_\tau$, the (inverse) spinon Green functions of $\mathcal{H}_0 + \mathcal{H}_{ZZ} + \mathcal{H}_1$ are

$$
G_k^{-1}(i \omega_n) = \left( \begin{array}{cc} J_{s+}^{(+)}(\omega_n^2 + \lambda - \frac{J_z}{4} F_k) & -J_{s+}^{(-)} R_k \omega_n^2 \\ -J_{s+}^{(-)} R_k^* \omega_n^2 & J_{s-}^{(+)}(\omega_n^2 + \lambda - \frac{J_z}{4} F_k) \end{array} \right),
$$
(S73)

where $\omega_n$ is a bosonic Mastubara frequency and $F_k = \sum_{\mu \neq \nu} e^{i k (e_{\mu} - e_{\nu})}$ describes the spinon propagation. Here, the spinon dispersion $\omega_0 = 2 J_z (\lambda - \frac{J_z}{4} F_k)$ is split into two branches

$$
\omega_0 \rightarrow \omega_{\pm} = \sqrt{2 J_z^{(\pm)} (\lambda - \frac{J_z}{4} F_k)}.
$$
(S74)

Especially, the diagonal component of Eq. (S73) is

$$
G_{k,11}(i \omega_n) = \langle \phi_{k_{1}, A}^* \phi_{k_{2}, A} \rangle = \frac{d_s}{2 J_{s+}^{(+)}(\omega_n^2 + \lambda - \frac{J_z}{4} F_k)}
$$
(S75)

To evaluate the Mastubara sum $\sum_{\omega_n} = \int_{-\infty}^{\infty} \frac{d \omega_n}{2 \pi}$ at $T = 0K$, we employ,

$$
\int_{-\infty}^{\infty} \frac{d \omega_n}{2 \pi} \frac{1}{\omega_n^2 + \lambda} = \int_{-\infty}^{\infty} \frac{d \omega_n}{2 \pi i} \sqrt{\frac{J_z^{(\pm)}}{2 \Lambda_k}} \left( \frac{1}{\omega_n - i \frac{1}{2 J_z^{(\pm)} \Lambda_k}} - \frac{1}{\omega_n + i \frac{1}{2 J_z^{(\pm)} \Lambda_k}} \right)
$$
(S76)
from the definition of $F$ is restricted to be $\theta \in [-\pi, \pi)$. All quantities Eq. (S77) for $J_z^{(\pm)} > 0$ fall into one of $z_1 = |z_1|e^{i(-\pi + 0)}$, $z_2 = |z_2|e^{i(-0)}$, $z_3 = |z_3|e^{i(0)}$, $z_4 = |z_4|e^{i(\pi - 0)}$.

where $\Theta(x)$ is the usual step function defined in Eq. (S20). In the last step, we have used,

In case $J_z^{(\pm)} > 0$
\[
\begin{cases}
(2J_z^{(\pm)} \lambda_k)_{i \omega_n \rightarrow -\infty} = (2J_z^{(\pm)} \lambda_k)_{i \omega_n \rightarrow \infty} \rightarrow |\omega_n|e^{-i0} \\
(2J_z^{(\pm)} \lambda_k)_{i \omega_n \rightarrow -\infty} = (2J_z^{(\pm)} \lambda_k)_{i \omega_n \rightarrow \infty} \rightarrow |\omega_n|e^{-i\pi}
\end{cases}
\]

In case $J_z^{(\pm)} < 0$
\[
\begin{cases}
(2J_z^{(\pm)} \lambda_k)_{i \omega_n \rightarrow -\infty} = (2J_z^{(\pm)} \lambda_k)_{i \omega_n \rightarrow \infty} \rightarrow |\omega_n| \\
(2J_z^{(\pm)} \lambda_k)_{i \omega_n \rightarrow -\infty} = (2J_z^{(\pm)} \lambda_k)_{i \omega_n \rightarrow \infty} \rightarrow -|\omega_n|
\end{cases}
\]

for any positive real $\lambda_k = \lambda - \frac{J_z}{2} F_k > 0$. In case $J_z^{(\pm)} < 0$, Eq. (S77) is obvious since all quantities are real. In case $J_z^{(\pm)} > 0$, the calculation is illustrated on the complex plane Fig. S2. Then the equal-time Green function of Eq. (S75) is

\[
G_{k,11}(\tau = 0) = \Theta(J_z - J_{zz}|R_k|) \frac{1}{2} \sqrt{\frac{J_z - J_{zz}|R_k|}{2(\lambda - \frac{J_z}{4} F_k)}} + \sqrt{\frac{J_z + J_{zz}|R_k|}{2(\lambda - \frac{J_z}{4} F_k)}}
+ \Theta(-J_z + J_{zz}|R_k|) \frac{1}{2} \sqrt{\frac{J_z + J_{zz}|R_k|}{2(\lambda - \frac{J_z}{4} F_k)}}.
\] (S78)

Here, the Lagrange multiplier $\lambda$ is determined by the rotor condition

\[
\frac{1}{N} \sum_{r \in A} \langle \phi^* \tau (r = 0) \phi \tau (r = 0) \rangle = \int_k G_{k,11}(\tau = 0) = 1.
\] (S79)

Then the BEC of spinons arises by the integrable divergence in the bosonic statistics $G_{k,11}$. Since $G_{k,11}$ should be real

\[
\lambda \geq \frac{J_z}{4} F_k, \quad -4 \leq F_k \leq 12 \quad \text{for all } k,
\] (S80)

from the definition of $F_k$. Thus the BEC occurs when $\lambda = 3J_z$ and Eq. (S79) becomes

\[
J_z^{(\pm)} \text{Higgs} = \left[ \int_k \{ \Theta(J_z - J_{zz}|R_k|) \frac{1}{2} \sqrt{\frac{J_z - J_{zz}|R_k|}{2(3 - \frac{J_z}{4} F_k)}} + \sqrt{\frac{J_z + J_{zz}|R_k|}{2(3 - \frac{J_z}{4} F_k)}}
+ \Theta(-J_z + J_{zz}|R_k|) \frac{1}{2} \sqrt{\frac{J_z + J_{zz}|R_k|}{2(3 - \frac{J_z}{4} F_k)}} \right]^2.
\] (S81)
Spinon pairing

We now consider a model which includes spinon pairing
\[ \mathcal{H} = \mathcal{H}_{XXZ_+} + \mathcal{H}_u \] (S82)

where \( \mathcal{H}_{XXZ_+} \) is defined through Eq. (S67), and the attractive, on-site, spinon interaction \( \mathcal{H}_u \) through Eq. (16) of the main text. In the presence of pairing, the Green’s function for spinons must be enlarged to allow for anomalous off–diagonal terms, as well two sublattices

\[
G_{k,z>0}^{-1}(i\omega_n) = \left( \begin{array}{cc} M_k & -24u\Delta I_{2\times2} \\ -24u\Delta I_{2\times2} & M_{-k} \end{array} \right),
\]

\[
M_k = M_{-k}^* = \left( \begin{array}{cc} j_z \frac{j_z}{2} \omega_n^2 + \lambda - \frac{j_z}{4} F_k & -j_z \frac{j_z}{2} R_k \omega_n^2 \\ -j_z \frac{j_z}{2} R_k^\dagger \omega_n^2 & j_z \frac{j_z}{2} \omega_n^2 + \lambda - \frac{j_z}{4} F_k \end{array} \right) \quad \text{[cf. Eq. (S73)]},
\]

in the basis of \((\phi_{k,\omega_n,A}, \phi_{k,\omega_n,B}, \phi_{-k,-\omega_n,A}, \phi_{-k,-\omega_n,B})^T\) and \(I_{2\times2}\) is the \(2 \times 2\) identity matrix. For the unitary matrix \(U_k\) diagonalizing,

\[
U_k^\dagger M_k U_k = U_k^T M_{-k} U_k^* = D_k,
\]

we rearrange Eq. (S83),

\[
\begin{pmatrix} U_k^\dagger & 0 \\ 0 & U_k^T \end{pmatrix} \begin{pmatrix} M_k & -24u\Delta I_{2\times2} \\ -24u\Delta I_{2\times2} & M_{-k} \end{pmatrix} \begin{pmatrix} U_k & 0 \\ 0 & U_k^* \end{pmatrix} = \begin{pmatrix} D_k & -24u\Delta U_k^T U_k^* \\ -24u\Delta U_k U_k^* & D_k \end{pmatrix} \equiv \begin{pmatrix} D_k & -24u\Delta I_{2\times2} \\ -24u\Delta I_{2\times2} & D_k \end{pmatrix},
\]

\[
D_k = \begin{pmatrix} \frac{\omega_n^2}{2j_z^2} & \left( \lambda - \frac{j_z}{4} F_k \right) \\ 0 & \frac{\omega_n^2}{2j_z^2} + \left( \lambda - \frac{j_z}{4} F_k \right) \end{pmatrix}.
\]

Since the Green function is influential near the origin \(k = (0, 0, 0)\), the unitary matrices \(U_k\) and \(M_k\) are almost real, \(U_k^\dagger U_k^* \approx I_{2\times2}\) which is harmless for small \(u\Delta\). As a result, Eq. (S83) is decoupled into 2 matrices.

\[
G_{k,z>0}^{-1}(i\omega_n) = \begin{pmatrix} M_k^+ & 0 \\ 0 & M_k^- \end{pmatrix} \quad \text{where} \quad M_k^\pm = \begin{pmatrix} \frac{\omega_n^2}{2j_z^2} + \left( \lambda - \frac{j_z}{4} F_k \right) & -24u\Delta \\ -24u\Delta & \frac{\omega_n^2}{2j_z^2} + \left( \lambda - \frac{j_z}{4} F_k \right) \end{pmatrix}.
\]

Likewise Eqs. (S75)-(S78), the equal-time Green functions are

\[
G_{k,11} = G_{k,22} = \frac{1}{2} \left( \begin{array}{cc} j_z^+ / \left( 2(\lambda - j_z^+ F_k - 24u\Delta) \right) & j_z^+ / \left( 2(\lambda - j_z^+ F_k + 24u\Delta) \right) \\ j_z^- / \left( 2(\lambda - j_z^- F_k - 24u\Delta) \right) & j_z^- / \left( 2(\lambda - j_z^- F_k + 24u\Delta) \right) \end{array} \right),
\]

\[
G_{k,12} = G_{k,21}^* = \frac{1}{2} \left( \begin{array}{cc} j_z^+ / \left( 2(\lambda - j_z^+ F_k - 24u\Delta) \right) & j_z^- / \left( 2(\lambda - j_z^- F_k + 24u\Delta) \right) \\ j_z^- / \left( 2(\lambda - j_z^+ F_k - 24u\Delta) \right) & j_z^+ / \left( 2(\lambda - j_z^- F_k + 24u\Delta) \right) \end{array} \right),
\]

\[
G_{k,33} = G_{k,44} = \Theta(j_z^-) \left( \begin{array}{cc} j_z^- / \left( 2(\lambda - j_z^- F_k - 24u\Delta) \right) & j_z^- / \left( 2(\lambda - j_z^- F_k + 24u\Delta) \right) \\ j_z^+ / \left( 2(\lambda - j_z^+ F_k - 24u\Delta) \right) & j_z^+ / \left( 2(\lambda - j_z^+ F_k + 24u\Delta) \right) \end{array} \right),
\]

\[
G_{k,34} = G_{k,43}^* = \Theta(j_z^-) \left( \begin{array}{cc} j_z^- / \left( 2(\lambda - j_z^- F_k - 24u\Delta) \right) & j_z^+ / \left( 2(\lambda - j_z^+ F_k - 24u\Delta) \right) \\ j_z^- / \left( 2(\lambda - j_z^- F_k + 24u\Delta) \right) & j_z^- / \left( 2(\lambda - j_z^- F_k + 24u\Delta) \right) \end{array} \right). \quad (S87)
\]

The pairing instability out of U(1) QSL is evaluated by self-consistent equations of \(\lambda, \Delta \neq 0\).

\[
\frac{1}{N} \sum_{r \in A,B} \langle \phi_r^* \phi_r \rangle = \int_k (G_{k,11} + G_{k,33}) = 2, \quad \frac{\partial}{\partial \Delta} \langle \mathcal{H}^\prime_{\text{rotor}} \rangle = 24u \left( - \int_k (G_{k,12} + G_{k,34}) + 2\Delta \right) = 0. \quad (S88)
\]
In case $J_\pm = 0$, the ground state energy per unit cell is simply

$$E[\delta Q]|_{J_\pm=0} = 2 \cdot J_z \frac{J_\pm}{2} (\delta Q)^2 - 4 J_{zz}(\delta Q)^2,$$

where the coefficients 2 and 4 count the number of sublattices and bonds connecting them respectively. The phase transition between $\delta Q = 0$ and $\delta Q \neq 0$ is strongly 1st-order since $\delta Q = 0$ for $J_{zz} < J_z/4$ and $\delta Q = \delta Q|_{\text{max}} = 2$ for $J_{zz} > J_z/4$.

Turning on the spinon kinetics $J_\pm > 0$, we desire the fourth terms $\sim (\delta Q)^4$ in $\langle H_1 + \delta H_{\text{rotor}} \rangle$ for a partial polarization $0 < \delta Q < \delta Q|_{\text{max}}$ which allows the in-plane magnetization or the spinon pairing marginally. In the static limit with $\delta Q(i\omega = 0)$, $\langle H_1 \rangle$ hardly loosen the full polarization $\delta Q|_{\text{max}}$ since the spinon and gauge charge fields are decoupled. Then we consider only the correction $\langle \delta H_{\text{rotor}} \rangle$ and fixing $s^+_{\text{rr}} = 1/2$ for simplicity. Since Eq. (S89) is the classical mean field energy of $\delta Q$, the decoupled product evaluation $\langle f[Q_r] \phi^r \phi^r' \rangle \approx f[Q_r|_{A} = -Q_r|_{B} = \delta Q] \langle \phi^r \phi^r' \rangle$ of Eq. (S32) is a plausible approximation.

$$\frac{1}{N} \langle \delta H_{\text{rotor}} \rangle \approx -\frac{J_\pm}{4} \left( \frac{7}{90} (\delta Q)^2 - \frac{78689}{1440000} (\delta Q)^4 + \ldots \right) \left( \int_k G_{k,11} F_k \right),$$

Then the quadratic term $\sim (\delta Q)^2$ corrects the critical value $J_{zz}|_{c,J_\pm=0} = J_z/4$.

$$J_{zz}|_{c} \approx \frac{J_z}{4} - \frac{7 J_\pm}{1440} \left( \int_k G_{k,11} F_k \right),$$

and the quartic correction $\sim (\delta Q)^4$ smooths out the phase transition boundary of Eq. (S89). As long as the spinon propagation $\langle \phi^r \phi^{r'} \rangle < 0$ is negative for $J_\pm/J_z < 0.043$, the phase transition is the 1st-order whereas the positive spinon propagation allows the 2nd-order transition for $J_\pm/J_z > 0.043$. (Fig. S3)