Magnetic monopole condensation in pyrochlore ice quantum spin liquid: application to Pr$_2$Ir$_2$O$_7$ and Yb$_2$Ti$_2$O$_7$

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Pyrochlore iridates and pyrochlore ices are two families of materials where novel quantum phenomena are intertwined with strong spin-orbit coupling, substantial electron correlation and geometrical frustration. Motivated by the puzzling experiments on two pyrochlore systems Pr$_2$Ir$_2$O$_7$ and Yb$_2$Ti$_2$O$_7$, we study the proximate Ising orders and the quantum phase transition out of quantum spin ice U(1) quantum spin liquid (QSL). We apply the electromagnetic duality of the compact quantum electrodynamics to analyze the “magnetic monopoles” condensation for U(1) QSL. The monopole condensation transition represents an unconventional quantum criticality with unusual scaling laws. It naturally leads to the Ising orders that belong to the “2-in 2-out” spin ice manifold and generically have an enlarged magnetic unit cell. We demonstrate that the antiferromagnetic Ising state with the ordering wavevector $Q = 2\pi(001)$ is proximate to U(1) QSL while the ferromagnetic Ising state with $Q = (000)$ is not proximate to U(1) QSL. This implies that if there exists a direct transition from U(1) QSL to the ferromagnetic Ising state, the transition must be strongly first order. We apply the theory to Pr$_2$Ir$_2$O$_7$ and Yb$_2$Ti$_2$O$_7$.

Pyrochlore iridates (R$_2$Ir$_2$O$_7$) have stimulated a wide interest in recent years, and many interesting results, including topological Mott insulators, quadratic band touching, Weyl semimetals, non-Fermi liquid, and so on, have been proposed. Among these materials, Pr$_2$Ir$_2$O$_7$ is of particular interest. In Pr$_2$Ir$_2$O$_7$, the Ir system remains metallic at low temperatures. More intriguingly, no magnetic order was found except a partial spin freezing of the Pr moments due to disorder at very low temperatures in the early experiments. A recent experiment on different Pr$_2$Ir$_2$O$_7$ samples, however, discovered an antiferromagnetic long-range order for the Pr moments. While most theory works on pyrochlore iridates focused on the Ir pyrochlores and explored the interplay between the electron correlation and the strong spin-orbit coupling of the Ir 5d electrons, very few works considered the influence of the physics of the local moments from the rare-earth sites that also form a pyrochlore lattice. In this paper, we address the local moment physics in Pr$_2$Ir$_2$O$_7$ and propose that the disordered state of the Pr moments is likely to be in the quantum spin ice (QSI) U(1) quantum spin liquid state. We explore the proximate Ising order and the confinement transition of QSI and argue that Pr$_2$Ir$_2$O$_7$ could be located near such a confinement transition.

The QSI U(1) QSL is an exotic quantum phase of matter and is described by emergent compact quantum electrodynamics, or equivalently, by the compact U(1) lattice gauge theory (LGT) with a gapless U(1) gauge photon and deconfined spinon excitations. Recently several rare-earth pyrochlores with 4f electron local moments are proposed as candidates for QSI U(1) QSLs. In these systems, the predominant antiferromagnetic exchange interaction between the Ising components of the local moments favors an extensively degenerate “2-in 2-out” spin ice manifold on the pyrochlore lattice. The transverse spin interaction allows the system to tunnel quantum mechanically within the ice manifold, giving rise to a U(1) QSL ground state. Like Pr$_2$Ir$_2$O$_7$, the experimental results on these QSL candidate materials depend sensitively on the stoichiometry and the sample preparation. In particular, for the pyrochlore ice system Yb$_2$Ti$_2$O$_7$, while some samples remain disordered down to the lowest temperature and the neutron scattering shows a diffusive scattering, others develop a ferromagnetic order. This suggests that both the Yb moments in Yb$_2$Ti$_2$O$_7$ and the Pr moments in Pr$_2$Ir$_2$O$_7$ could be located near a phase transition between a disordered state (that might be a QSI U(1) QSL) and the magnetic orders.

On the theoretical side, the instability of the QSI U(1) QSL...
This page contains a detailed discussion on the properties of the pyrochlore lattice and its connection to the U(1) quantum spin liquid (QSL) state. The authors explore the compact U(1) Landau-Ginzburg-Taylor (LGT) model for the U(1) QSL and explain the nature of the phase transition. They introduce an integer-valued dual U(1) gauge field to describe the magnetic transition from U(1) QSL via the magnetic monopoles. The results show that the compact U(1) LGT can be used to study the instability of the spin-1/2 moments on the pyrochlore lattice.

Compact QED and Electromagnetic Duality. Even more complicated realistic Hamiltonians are available for effective spin-1/2 moments on the pyrochlore lattice.

Results. Complex QED and electromagnetic duality. Even more complicated realistic Hamiltonians are available for effective spin-1/2 moments on the pyrochlore lattice. We determine the phase transition and the nature of the phase transition of the compact U(1) LGT for the U(1) QSL and explain the nature of the phase transition from the QSI U(1) QSL to the Ising order.

FIG. 2. (a) The Q = (000) ferromagnetic state. (b) The Q = (000) antiferromagnetic state.
where \( \mathbf{e}_\mu (\mu = 0, 1, 2, 3) \) are the four vectors that connect the I sublattice sites of the diamond lattice to their nearest neighbors. In terms of the dual gauge variables, \( H_{\text{LGT}} \) is transformed into

\[
H_{\text{dual}} = \sum_{\mathcal{O}^2} \frac{U}{2} (\nabla a - \bar{E})^2 - \sum_{\langle \mathbf{r}, \mathbf{r}' \rangle} K \cos B_{\mathbf{r}'},
\]

where we have explicitly replaced \( \nabla A \) with the magnetic field vector \( B_{\mathbf{r}'} \) that lives on the link \((\mathbf{r}, \mathbf{r}')\) of the dual diamond lattice and is conjugate to the dual gauge field \( a \) with \( [B_{\mathbf{r}'}, a_{\mathbf{r}}] = i \). In Eq. (10), we have introduced the electric field \( \bar{E} \) that combines both the background electric field distribution \( E^0 \) and the offset in Eq. (5) with

\[
\bar{E}_{\mathbf{r}, \mathbf{r} + \epsilon_r \mathbf{e}_\mu} = E^0_{\mathbf{r}, \mathbf{r} + \epsilon_r \mathbf{e}_\mu} - \frac{\epsilon_r}{2}.
\]

Since the dual gauge field \( a \) is integer valued, the dual Hamiltonian \( H_{\text{dual}} \) is difficult to work with. Moreover, the “magnetic monopole” is implicit in the dual gauge field configuration. To make the monopole explicit, we follow the standard procedure first relax the integer valued constraint of the dual gauge field by introducing \( \cos 2\pi a \) and then insert the monopole operators. The resulting dual theory is described by the magnetic monopoles minimally coupled with the dual U(1) gauge field on the dual diamond lattice,

\[
H_{\text{dual}} = \sum_{\mathcal{O}^2} \frac{U}{2} (\nabla a - \bar{E})^2 - \sum_{\langle \mathbf{r}, \mathbf{r}' \rangle} K \cos B_{\mathbf{r}'} - \sum_{\langle \mathbf{r}, \mathbf{r}' \rangle} t \cos (\theta_{\mathbf{r}} - \theta_{\mathbf{r}'} + 2\pi a_{\mathbf{r}' - \mathbf{r}}),
\]

where \( e^{-i\theta} \) (\( e^{i\theta} \)) creates (annihilates) the “magnetic monopole” at the dual lattice site \( \mathbf{r} \).

### Monopole condensation and proximate Ising order.

In the dual gauge Hamiltonian of Eq. (11), as the monopole hopping increases, the monopole gap decreases. When the monopole gap is closed, the monopole is condensed. In the confinement phase, the \( E \) field develops a static field on the dual diamond lattice, \( (\mathbf{r}, \mathbf{r}' = (0110) \) and \( q = 2\pi(100) \).

\[
\mathbf{E}_{\mathbf{r}, \mathbf{r} + \epsilon_r \mathbf{e}_\mu} = E^0_{\mathbf{r}, \mathbf{r} + \epsilon_r \mathbf{e}_\mu} - \frac{\epsilon_r}{2}.
\]

\[
H_{\text{dual}} = \sum_{\mathcal{O}^2} \frac{U}{2} (\nabla a - \bar{E})^2 - \sum_{\langle \mathbf{r}, \mathbf{r}' \rangle} K \cos B_{\mathbf{r}'} - \sum_{\langle \mathbf{r}, \mathbf{r}' \rangle} t \cos (\theta_{\mathbf{r}} - \theta_{\mathbf{r}'} + 2\pi a_{\mathbf{r}' - \mathbf{r}}),
\]

where we have introduced \( \Phi_\mathbf{r} \equiv e^{i\phi} \) (with \( |\Phi_\mathbf{r}|^2 = 1 \)). The dispersion of the lowest monopole band is given by

\[
\Omega_\mathbf{r} = -|t|(4 + 2(3 + c_x c_y - c_x c_z + c_y c_z)^{1/2})^{1/2},
\]

where \( c_\mu = \cos k_\mu (\mu = x, y, z) \). The degenerate minima of the lowest band form several lines of momentum points in the Brillouin zone. One such degenerate line is along the [001] direction of the Brillouin zone and the minimum energy is \(-2\sqrt{2}|t|\). Other degenerate lines are readily obtained by the symmetry operations. The line degeneracy of the band minima is a consequence of the background flux that frustrates the monopole hopping. These continuous degeneracies are accidental and are not protected by symmetry. It is expected that the further neighbor monopole hopping or monopole interactions should lift these degeneracies.

Because of the background flux, the lattice symmetry in \( H_m \) is realized projectively, known as projective symmetry group (PSG) \( \mathbb{F}_4 \). We use PSG to generate the further neighbor monopole hoppings, but do not find obvious degeneracy breaking. Instead, the line degeneracy immediately gets lifted if we impose the unimodular constraint of the monopole field \( |\Phi_\mathbf{r}| = 1 \). This unimodular constraint, that originates from the repulsive interaction between monopoles, suppresses the magnitude fluctuation of the monopole fields. For the degenerate minima along the [001] direction, the unimodular requirement selects the monopole configurations at two equivalent momenta

\[
\mathbf{k}_1 = (0, 0, \pi), \quad \mathbf{k}_2 = (0, 0, -\pi),
\]

and the corresponding monopole configurations are

\[
\begin{align*}
\{ \mathbf{r} \in \text{I}, & \quad \varphi_1(\mathbf{r}) = \left( \frac{1 + i}{2} + \frac{1 - i}{2} e^{i\phi} \right) e^{i\pi z}, \\
\{ \mathbf{r} \in \text{II}, & \quad \varphi_1(\mathbf{r}) = e^{i\pi z}, \\
\{ \mathbf{r} \in \text{I}, & \quad \varphi_2(\mathbf{r}) = \left( \frac{1 + i}{2} + \frac{1 - i}{2} e^{i\phi} \right) e^{-i\pi z}, \\
\{ \mathbf{r} \in \text{II}, & \quad \varphi_2(\mathbf{r}) = i e^{-i\pi z},
\end{align*}
\]
where $\varphi_a$ refers to the monopole configuration at the momentum $k_a$. From the above results, we use the PSG transformations and generate in total twelve symmetry equivalent solutions.

After the unimodular constraint is enforced, the monopoles are condensed at only one of the equivalent solutions, the spinons are confined and the system develops an Ising order. Although the Ising order is induced by the monopole condensation, as monopoles are emergent particles and are not gauge invariant, the physical property of the monopole condensate is encoded in the gauge invariant monopole bilinears. Again, symmetry is a powerful tool to establish the relation between the spin density $\tau^z$ and the monopole bilinears. The candidate monopole bilinears are the monopole density and the monopole current. Although the monopole density $(\Phi^\dagger\Phi)$ transforms in the same way as the spin density $(\tau^z)$ under the space group symmetry, they behave oppositely under the time reversal.

As for the monopole current, from the Maxwell’s equations, the loop integral of monopole current is the electric flux through the plaquette enclosed by the loop (see Fig. 2a). We have

$$\tau^z_i \sim E_{rr} \sim \sum_{\nu' \in O^*_2} J_{rr}, \quad (17)$$

where the pyrochlore site $i$ is the center of the elementary honeycomb $O^*_2$ on the dual diamond lattice, and $J_{rr} \equiv i \langle \Phi^\dagger(r) \Phi(r) \rangle e^{-i\alpha_{\nu'}} - h.c.$ defines the monopole current. Here $\langle \Phi \rangle$ is the expectation value of the monopole field that is taken with respect to one of the equivalent solutions. In the inset of Fig. [2] we depict the spin density distribution of the monopole condensate at $k_1$. The resulting Ising order in the confinement phase is an antiferromagnetic state with an ordering wavevector $Q = 2\pi r(001)$, and the four spins on each tetrahedron obey the “2-in 2-out” ice rule. This Ising state breaks the translation symmetry by doubling the crystal unit cell.

The translation symmetry breaking of the proximate magnetic state is a generic phenomenon. The background gauge flux, due to the “2-in 2-out” rule, shifts the minimum of the monopole band to finite momenta. Once the monopole is condensed at the finite momentum, the resulting proximate Ising order necessarily breaks the translation symmetry. If, however, the ferromagnetic Ising order with $Q = (000)$ in Fig. 2a, preserves the translation symmetry and borders with the QSL U(1) QSL, the transition between this ferromagnetic Ising order and U(1) QSL must be strongly first order. In the Method, we write down simple models that do not have a sign problem for quantum Monte Carlo simulation. The models can realize both the ferromagnetic and antiferromagnetic Ising orders and allow the careful numerical study of the phase transition out of the QSI U(1) QSL.

**Critical theory of monopole condensation.** The monopole interaction in the confinement phase selects twelve equivalent monopole condensates that correspond to twelve symmetry equivalent Ising orders. In the vicinity of the monopole condensation transition, the monopole condensate and the gauge fields fluctuate strongly. We thereby carry out a Landau-Ginzburg-Wilson expansion of the action in terms of the monopole condensate and gauge field in the vicinity of the phase transition. We introduce the slowly-varying monopole fields $\varphi_a$ via the expansion

$$\Phi_r = \sum_{a=1}^{12} \varphi_a(r) \phi_a, \quad (18)$$

where $\varphi_a(r)$ $(a = 1, \ldots, 12)$ are the twelve discrete monopole modes that span the ground state manifold of the monopole condensate. With the monopole PSG, we generate the symmetry allowed effective action for the monopole condensation transition,

$$L = \sum_a \left[ |(\partial_\mu - i\tilde{a}_\mu)\phi_a|^2 + m^2 |\phi_a|^2 + \frac{F_{\mu\nu}^2}{2} \right. + u_0 \sum_a |\phi_a|^2 + u_1 \sum_{a\neq b} |\phi_a|^2 |\phi_b|^2 + \cdots, \quad (19)$$

where we have restored the gauge field fluctuation by coupling the $\phi_a$ fields to the fluctuating part of the dual U(1) gauge field $\tilde{a}_\mu$. $\frac{1}{2}F_{\mu\nu}^2$ is the Maxwell term with $F_{\mu\nu} = \partial_\mu \tilde{a}_\nu - \partial_\nu \tilde{a}_\mu - \cdots$ contains further anisotropic terms that are marginal for the critical properties, $m$ is the mass of the monopole and is set by the band gap of the monopole band structure. The effective action in Eq. (19) is a standard multi-component Ginzburg-Landau theory in 3+1D that is the upper critical dimension of the theory. One expects the phase transition of this theory to be governed by a Gaussian fixed point or belong to a weakly first order transition driven by fluctuations. Both possibilities suggest that the mean-field treatment of the phase transition should be sufficient for a rather wide range of length scales. In a mean-field description, the monopole field correlator at the critical point (with the monopole mass $m = 0$) is

$$\langle \phi_a^\dagger(k, \omega) \phi_b(k, \omega) \rangle \sim \frac{\delta_{ab}}{k^2 + \omega^2}. \quad (20)$$

According to Eq. (17), the spin susceptibility at the ordering wavevector $Q$ is simply given by the bubble diagram of monopole fields (see Fig. 3) and is thus logarithmically divergent at low temperatures with

$$\chi(Q) \sim \ln \frac{1}{T}. \quad (21)$$

![FIG. 4. The bubble diagram of the “magnetic monopole”.](image-url)
Such a weak divergence is a unique property of the monopole condensation transition that is a non-Landau-Ginzburg-Wilson transition. For a conventional magnetic transition, one would instead have a power-law divergence for the corresponding susceptibility. Here, the Ising order is a consequence of the monopole condensation. The condensed monopole is the primary order, and the induced Ising order is secondary and is thus a perfect example of the subsidiary order.

The monopole mass controls the phase transition and is parameterized as the parameter $g$ with $g \equiv -m^2$ in Eq. (1). In the QSI U(1) QSL phase, the monopole is massive with $m^2 > 0$. The low energy physics is then governed by the Maxwell’s field theory and the emergent gapless gauge photon. Due to the gapless photon, the heat capacity of the system behaves as $C_v \sim T^3$ at low temperatures. As the system approaches the transition from the QSL side, the monopole mass decreases. The gapless monopole at the criticality gives an extra $T^3$ contribution to the heat capacity. Therefore, one would observe an enhancement of the $T^3$ heat capacity as the system approaches the criticality. Moreover, if one raises temperatures in the U(1) QSL side, the generic argument suggests that there is no thermal phase transition except a crossover due to the thermal population of the “magnetic monopoles”. The populated monopoles simply create thermal confinement of the spinons at finite temperatures. This crossover temperature is set by the mass of the monopoles.

When $m^2 < 0$, the monopole is condensed and the system develops Ising orders. Since the system breaks time reversal symmetry on the ordered side, we should have a finite temperature phase transition above which the time reversal symmetry is restored. The ordering temperature is also set by the mass of the monopoles.

Discussion.
The transition and the Ising order in Pr$_2$Ir$_2$O$_7$. In Pr$_2$Ir$_2$O$_7$, the Pr$^{3+}$ ion has a 4f$^5$ electron configuration and form a non-Kramers’ doublet which is represented by a pseudospin-1/2 operator $\tau$ with $\tau^z$ (\tau^x, \tau^y) odd (even) under time reversal $T$,

$$T: \quad \tau^z \rightarrow -\tau^z, \quad \tau^x, \tau^y \rightarrow \tau^x, \tau^y. \quad (22)$$

$$T: \quad \tau^x, \tau^y \rightarrow \tau^x, \tau^y. \quad (23)$$

In the disordered state, a metamagnetic transition is observed only for magnetic fields along the (111) direction. This is a clear evidence that the disordered state of the Pr moments is fluctuating within the ice manifold and the metamagnetic transition is a transition from the “2-in 2-out” ice manifold to the “3-in 1-out” manifold. Since the local moments in QSI U(1) QSL are fluctuating quantum mechanically within the ice manifold, this metamagnetic transition in Pr$_2$Ir$_2$O$_7$ is consistent with our proposal that the disordered state of the Pr moments is a QSI U(1) QSL.

Given the non-Kramers’ nature of the Pr moment and its unique time reversal symmetry properties in Eqs. (22) and (23), the magnetic order of the Pr moment must be the Ising order with $\langle \tau^z \rangle \neq 0$. If a non-Kramers doublet local moment system has a QSI U(1) QSL ground state, the magnetic transition from this state must be the confinement transition of the compact U(1) LGT because a nonzero $\tau^z$ corresponds to the static electric field distribution. Remarkably, the Ising order that is found in the ordered Pr$_2$Ir$_2$O$_7$ has an ordering wavevector $\mathbf{Q} = 2\pi(001)$, and this is precisely the proximate Ising state that we predict from the confinement transition. This experimental result further supports our proposal that the disordered state of the Pr moments in Pr$_2$Ir$_2$O$_7$ is a QSI U(1) QSL.

In different samples, different oxygen and Ir contents shift the Fermi energy of the Ir conduction electrons and thus modify the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction between the Pr local moments. This is likely to be the microscopic origin of the sample dependence. Usually the presence of the conduction electron Fermi surface modifies the critical properties of the local moment transition. But Pr$_2$Ir$_2$O$_7$ is very special. Due to the quadratic band touching of the Ir electron, the Fermi energy is very close to the band energy and the Fermi momentum $|k_F|$ is much smaller than the wavevector $\mathbf{Q}$ of the magnetic order. As a result, the particle-hole excitations of the Ir system actually decouple from the spin fluctuations of the Pr local moments at low energy. Therefore, the critical properties of the Pr local moments are not modified by the conduction electrons.

At this stage, it is not clear how close the existing Pr$_2$Ir$_2$O$_7$ samples are near the phase transition, therefore, it would be interesting to vary the Ir and/or oxygen contents in a continuous fashion, to drive the system between disordered and ordered phases and directly probe the phase transition. It is very useful to focus on the disordered Pr$_2$Ir$_2$O$_7$ samples and carry out the inelastic neutron scattering. Due to the unique time reversal symmetry properties in Eqs. (22) and (23), only the Ising component of Pr local moment couples with the neutron spin. As the $\tau^z$ is identified as the emergent electric field, the inelastic neutron scattering directly probes the gauge phonon excitation. Because of the quadratic band touching, the inelastic neutron spectral intensity corresponding to the particle-hole excitations of the Ir electrons concentrate near the $\Gamma$ point at low energies, and it does not mix with the gauge phonon modes that are peaked near the “pinch point” moment.

The transition and the magnetic order in Yb$_2$Ti$_2$O$_7$. The magnetic state in the ordered Yb$_2$Ti$_2$O$_7$ samples has a $\mathbf{Q} = (000)$ ferromagnetic order and preserves the translation symmetry. Though many early experiments found a disordered state, the thermal transition from the high-temperature paramagnet to the ferromagnetic one is strongly first order. Unlike the Pr$^{3+}$ moment, the Yb$^{3+}$ moment is a Kramers’ doublet with all pseudospin components odd under time reversal,
thus a direct coupling between $\tau^z$ and $\tau^{x,y}$ is allowed. The magnetic transition out of the QSI U(1) QSL for the Kramers’ doublet can be either an Anderson-Higgs’ transition or a confinement transition.

In the Higgs’ transition scenario, a predominant transverse component is induced at the first order transition, and a small Ising component is induced simultaneously via the coupling between $\tau^z$ and $\tau^{x,y}$. In the scenario of a confinement transition, however, a predominant Ising order is expected, and this seems to be case in Yb$_2$Ti$_2$O$_7$. Moreover, as we have explained, the Q = (000) Ising order is not proximate to the QSI U(1) QSL, and the direct transition between them through monopole condensation must be strongly first order. The strongly first order thermal transition in the ordered Yb$_2$Ti$_2$O$_7$ samples can thus be naturally regarded as a finite temperature extension of the zero-temperature one. To differentiate the Higgs’ and confinement scenarios in Yb$_2$Ti$_2$O$_7$, it might be helpful to numerically study the microscopic model by varying the transverse component interaction and the Ising component interaction separately and probe the nature of transition out of the QSI U(1) QSL.

Summary. To summarize, we have studied the Ising magnetic orders out of the QSI U(1) QSL via the “magnetic monopole” condensation. We find that such a confinement transition gives rise to the proximate Ising ordered state that breaks the translation symmetry. We propose that the puzzling magnetic properties of Pr$_2$Ir$_2$O$_7$ and Yb$_2$Ti$_2$O$_7$ can be understood from the “magnetic monopole” condensation. Beyond these two systems, we have argued that the magnetic transition out of the QSI U(1) QSL for a non-Kramers doublet local moments must be a confinement transition via monopole condensation. Since the Tb$^{3+}$ local moment in Tb$_2$Ti$_2$O$_7$ is a non-Kramers’ doublet, it is likely that the sample dependent magnetic order in Tb$_2$Ti$_2$O$_7$ can be understood as the monopole condensation.

Method.

Pyrochlore and dual diamond lattices. Pyrochlore lattice is a corner-shared tetrahedral structure in three dimensions. The centers of the tetrahedra in the pyrochlore lattice form a diamond lattice. The dual lattice of the diamond lattice is also a diamond lattice. For the dual diamond lattice, we choose the sites

$$d_1 = (0,0,0),$$
$$d_2 = \frac{1}{4}(1,1,1),$$

for the reference points of the I and II sublattices, respectively. The three lattice vectors of the underlying Bravais lattice are

$$a_1 = \frac{1}{2}(0,1,1),$$
$$a_2 = \frac{1}{2}(1,0,1),$$
$$a_3 = \frac{1}{2}(1,1,0),$$

where we have set the lattice constant to unity.

Each site of the dual diamond lattice is connected by four nearest neighbors. The four vectors $e_i$ that connect the neighboring sites are given as

$$e_0 = \frac{1}{4}(1,1,1),$$
$$e_1 = \frac{1}{4}(1,-1,-1),$$
$$e_2 = \frac{1}{4}(-1,-1,-1),$$
$$e_3 = \frac{1}{4}(-1,1,1).$$

Projective symmetry group. Both the pyrochlore lattice and the dual diamond lattice share the same space group symmetry Fd3m. The Fd3m space group involves three lattice translations,

$$T_i : r \rightarrow r + a_i,$$

a three-fold rotation,

$$C_3 : (x,y,z) \rightarrow (z,x,y),$$

a two-fold rotation,

$$C_2 : (x,y,z) \rightarrow (-x,-y,z),$$

a mirror reflection,

$$R : (x,y,z) \rightarrow (y,x,z),$$

and an inversion,

$$I : (x,y,z) \rightarrow \left(\frac{1}{4} - x, \frac{1}{4} - y, \frac{1}{4} - z\right).$$

The physical spin is defined on the pyrochlore lattice site, while the “magnetic monopoles” are defined on the dual diamond lattice sites. Due to the background gauge flux, the space group symmetry is realized projectively in the monopole hopping Hamiltonian $H_m$. For each symmetry operation, we need to supplement with a U(1) gauge transformation. Under the symmetry operation $O$, the monopole is transformed as

$$\hat{O} : \Phi \rightarrow e^{-i\Theta_0(r)}\Phi r,$$

where $r' = O(r)$ and $e^{-i\Theta_0(r)}$ is the associated U(1) gauge transformation. We have used $\hat{O}$ to label the generator of the projective symmetry group.
For our convenience, we introduce the unit cell index \( \mathbf{n} \) to label the monopole position and define
\[
\eta_1(\mathbf{n}) = \Phi_r, \quad \eta_2(\mathbf{n}) = \Phi_{r+e_0},
\] (39)
where \( r = \sum_{j} n_j a_j \), and \( \eta_1(\mathbf{n}) \) and \( \eta_2(\mathbf{n}) \) are monopole operators on the I and II sublattices, respectively.

Here we list the projection symmetry transformation of the monopole operators. Under the three lattice translations, the monopole operators are transformed as
\[
\hat{T}_1 : \eta_1(n_x, n_y, n_z) \rightarrow e^{-i\hat{\Theta}_{\hat{T}_1}[\mathbf{n}]}\eta_1(n_x+1, n_y, n_z), \quad (40)
\]
\[
\hat{T}_1 : \eta_2(n_x, n_y, n_z) \rightarrow e^{-i\hat{\Theta}_{\hat{T}_1}[\mathbf{n}]}\eta_2(n_x+1, n_y, n_z), \quad (41)
\]
\[
\hat{T}_2 : \eta_1(n_x, n_y, n_z) \rightarrow e^{-i\hat{\Theta}_{\hat{T}_2}[\mathbf{n}]}\eta_1(n_x, n_y+1, n_z), \quad (42)
\]
\[
\hat{T}_2 : \eta_2(n_x, n_y, n_z) \rightarrow e^{-i\hat{\Theta}_{\hat{T}_2}[\mathbf{n}]}\eta_2(n_x, n_y+1, n_z), \quad (43)
\]
\[
\hat{T}_3 : \eta_1(n_x, n_y, n_z) \rightarrow e^{-i\hat{\Theta}_{\hat{T}_3}[\mathbf{n}]}\eta_1(n_x, n_y+1, n_z+1), \quad (44)
\]
\[
\hat{T}_3 : \eta_2(n_x, n_y, n_z) \rightarrow e^{-i\hat{\Theta}_{\hat{T}_3}[\mathbf{n}]}\eta_2(n_x, n_y+1, n_z+1), \quad (45)
\]
where
\[
\Theta_{\hat{T}_1}[\mathbf{n}] = -(\epsilon \cdot \mathbf{n}) v_i
\] (46)
and \( \epsilon = (1, 1, 0), v_i = \pi(0, 1, 1) \).

Under three-fold rotation, we have
\[
\hat{C}_3 : \eta_1(n_x, n_y, n_z) \rightarrow e^{-i\hat{\Theta}_{\hat{C}_3}[\mathbf{n}]}\eta_1(n_x, n_y), \quad (47)
\]
\[
\hat{C}_3 : \eta_2(n_x, n_y, n_z) \rightarrow e^{-i\hat{\Theta}_{\hat{C}_3}[\mathbf{n}]}\eta_2(n_x, n_y), \quad (48)
\]
where
\[
\Theta_{\hat{C}_3}[\mathbf{n}] = \mathbf{n} \cdot \mathbf{B} \cdot \mathbf{n} + \delta \cdot \mathbf{n}
\] (49)
with
\[
\mathbf{B} = \frac{\pi}{2} \begin{bmatrix} 1 & 0 & 1 \\ 0 & 1 & 1 \\ 1 & 1 & 0 \end{bmatrix}
\] (50)
and \( \delta = \pi/2(1, 1, 0) \).

Under two-fold rotation, we have
\[
\hat{C}_2 : \eta_1(n_x, n_y, n_z) \rightarrow \eta_1(n_y, n_x, -n_x - n_y - n_z), \quad (51)
\]
\[
\hat{C}_2 : \eta_2(n_x, n_y, n_z) \rightarrow \eta_2(n_y, n_x, -1 - n_x - n_y - n_z). \quad (52)
\]
where \( \Theta_{\hat{C}_2}[\mathbf{n}] = 0 \).

Under the reflection, we have
\[
\hat{R} : \eta_1(n_x, n_y, n_z) \rightarrow e^{-i\hat{\Theta}[\mathbf{n}]}\eta_1(n_x, n_y, n_z), \quad (53)
\]
\[
\hat{R} : \eta_2(n_x, n_y, n_z) \rightarrow e^{-i\hat{\Theta}[\mathbf{n}]}\eta_2(n_y, n_x, n_z), \quad (54)
\]
where
\[
\Theta_{\hat{R}}[\mathbf{n}] = \mathbf{n} \cdot \mathbf{B}' \cdot \mathbf{n} + \delta' \cdot \mathbf{n}
\] (55)
with
\[
\mathbf{B}' = \frac{\pi}{2} \begin{bmatrix} 1 & 1 & 0 \\ 1 & 1 & 0 \\ 0 & 0 & 0 \end{bmatrix}
\] (56)
and \( \delta' = \pi/2(1, 1, 0) \).

Finally, for the inversion symmetry, we have
\[
\hat{I} : \eta_1(n_x, n_y, n_z) \rightarrow e^{-i\hat{\Theta}[\mathbf{n}]}\eta_1(-n_x, -n_y, n_z), \quad (57)
\]
\[
\hat{I} : \eta_2(n_x, n_y, n_z) \rightarrow e^{-i\hat{\Theta}[\mathbf{n}]}\eta_2(-n_x, -n_y, n_z), \quad (58)
\]
where
\[
\Theta_{\hat{I}}[\mathbf{n}] = \lambda \cdot \mathbf{n}
\] (59)
and \( \lambda = \pi(0, 1, 0) \).

**Further neighbor monopole hoppings.** The general monopole hopping model should be invariant under the PSG transformation. We here give an example for the second neighbor monopole hopping to illustrate the procedure to determine the hopping parameters. The second neighbor connects the lattice sites within the same sublattice. Each site has twelve second-neighbor sites. For the sites in the I sublattice, we consider the monopole hopping Hamiltonian,
\[
H_m' = \sum_{\mathbf{n}} d_1[\mathbf{n}] \eta_1^\dagger(n_x, n_y, n_z) \eta_1(n_x+1, n_y, n_z)
\]
\[
+ d_2[\mathbf{n}] \eta_1^\dagger(n_x, n_y, n_z) \eta_1(n_x, n_y+1, n_z)
\]
\[
+ d_3[\mathbf{n}] \eta_1^\dagger(n_x, n_y, n_z) \eta_1(n_x, n_y, n_z+1)
\]
\[
+ d_4[\mathbf{n}] \eta_1^\dagger(n_x, n_y, n_z) \eta_1(n_x, n_y-1, n_z+1)
\]
\[
+ d_5[\mathbf{n}] \eta_1^\dagger(n_x, n_y, n_z) \eta_1(n_x-1, n_y, n_z+1)
\]
\[
+ d_6[\mathbf{n}] \eta_1^\dagger(n_x, n_y, n_z) \eta_1(n_x, n_y-1, n_z+1)
\]
\[
+ h.c.,
\] (60)
where \( \{d_i[\mathbf{n}]\} \) are the hopping parameters. Applying the \( \hat{T}_1 \) translation, we compare the transformed Hamiltonian with the original Hamiltonian and obtain
\[
d_1[n_x, n_y, n_z] = d_1[n_x - 1, n_y, n_z].
\] (61)

Similarly, for the \( \hat{T}_2 \) and \( \hat{T}_3 \) translations, we have
\[
d_1[n_x, n_y, n_z] = -d_1[n_x, n_y - 1, n_z],
\] (62)
\[
d_2[n_x, n_y, n_z] = -d_2[n_x, n_y - 1, n_z],
\] (63)
\[
d_3[n_x, n_y, n_z] = +d_3[n_x, n_y - 1, n_z],
\] (64)
\[
d_4[n_x, n_y, n_z] = -d_4[n_x, n_y - 1, n_z],
\] (65)
\[
d_5[n_x, n_y, n_z] = -d_5[n_x, n_y - 1, n_z],
\] (66)
\[
d_6[n_x, n_y, n_z] = +d_6[n_x, n_y - 1, n_z],
\] (67)
and
\[
d_1[n_x, n_y, n_z] = -d_1[n_x, n_y, n_z - 1],
\] (68)
\[
d_2[n_x, n_y, n_z] = -d_2[n_x, n_y, n_z - 1],
\] (69)
\[
d_3[n_x, n_y, n_z] = +d_3[n_x, n_y, n_z - 1],
\] (70)
\[
d_4[n_x, n_y, n_z] = -d_4[n_x, n_y, n_z - 1],
\] (71)
\[
d_5[n_x, n_y, n_z] = -d_5[n_x, n_y, n_z - 1],
\] (72)
\[
d_6[n_x, n_y, n_z] = +d_6[n_x, n_y, n_z - 1],
\] (73)
respectively. Applying the remaining symmetries, we obtain the following hopping parameters for the second
Focusing on the \( [001] \) direction in the momentum space, spectrum has line degeneracies in the momentum space. In Fig. 3, we specify the signs of the hopping parameters with regards to the background flux and the gauge choice, the unit cell is fictitiously doubled. In the monopole hopping model. Due to the background flux, we immediately require the monopoles to be condensed at these lowest energy momenta. To satisfy the unimodular condition for the monopoles, we require the monopoles to be condensed at \( k_z = \pm \pi \).

**Monopole condensates.** We consider the nearest neighbor monopole hopping model. Due to the background flux and the gauge choice, the unit cell is fictitiously doubled. In Fig. 3, we specify the signs of the hopping parameters on the dual diamond lattice. The lowest energy spectrum has line degeneracies in the momentum space. Focusing on the [001] direction in the momentum space, we have the following eigenstates for a given \( k_z \),

\[
\begin{align*}
\mathbf{r} \in \mathbf{I}, & \quad \Phi(\mathbf{r}) = \frac{1}{\sqrt{2}} (e^{i \frac{\mathbf{r} \cdot \mathbf{z}}{4\pi}} + e^{i \frac{\mathbf{r} \cdot \mathbf{z}}{4\pi}}) e^{i k_z z}, \\
\mathbf{r} \in \mathbf{II}, & \quad \Phi(\mathbf{r}) = e^{i k_z z}.
\end{align*}
\]

The monopoles are condensed at these lowest energy momenta. To satisfy the unimodular condition for the monopoles, we require the monopoles to be condensed at \( k_z = \pm \pi \).

**A sign-problem free model for quantum Monte Carlo simulation.** Here we propose a simple exchange model that does not have a sign problem for quantum Monte Carlo (QMC) simulation. This model can realize both the \( Q = 2\pi(001) \) order and the \( Q = (000) \) order. Although both Ising orders belong to the spin ice manifold, the former is proximate to the QSI U(1) QSL via a confinement transition and the latter is not (see the main text for the detailed discussion). The model is given as

\[
H_1 = \sum_{\langle ij \rangle} J_z \tau_i^z \tau_j^z - J_{\perp} \left( \tau_i^+ \tau_j^- + h.c. \right) + \sum_{\langle\langle ij \rangle\rangle} J_{3z} \tau_i^z \tau_j^z,
\]

where \( J_{3z} \) is the third neighbor Ising exchange.

We focus our discussion on the case when \( J_{\perp} > 0 \). This is precisely the parameter regime where the sign problem for QMC is absent. To be in the spin ice regime, we keep \( J_z > 0 \). When \( J_{\perp} \ll J_z \) and \( J_{3z} \ll J_z \), the ground state is a QSI U(1) QSL. If we fix \( J_{\perp} \ll J_z \) to make the system in the QSI U(1) QSL phase, as we gradually increase \( |J_{3z}/J_z| \) from 0, the system will eventually become ordered. Since \( J_{3z} \) is the interaction between spins from the same sublattice, a ferromagnetic \( J_{3z} \) would simply favor \( Q = (000) \), even though the four spins on each tetrahedron of the pyrochlore lattice obey the "two-in two-out" ice rule (see Fig. 2a of the main text). Since this \( Q = (000) \) is not proximate to the U(1) QSL phase, we expect a strongly first order transition as we increase \( |J_{3z}/J_z| \) for a ferromagnetic \( J_{3z} \).

For an antiferromagnetic \( J_{3z} \), although the Luttinger-Tisza method gives a continuous line degeneracy for the ordering wavevector, the Ising constraint immediately select the collinear order with an ordering wavevector \( Q = 2\pi(001) \). As we show in the main text, this Ising order is proximate to the U(1) QSL via a monopole condensation transition. Therefore, we expect either a continuous transition or an extremely weakly first order transition driven by fluctuations as we increase \( |J_{3z}/J_z| \) for an antiferromagnetic \( J_{3z} \).

In the future, it would be interesting to implement a large scale QMC simulation of the model in Eq. (82) to confirm the monopole condensation transition out the QSI U(1) QSL.

Finally, we propose a perturbative version of the model in Eq. (82). The new model includes the ring exchange on the pyrochlore hexagons and the third neighbor Ising exchange and is given as

\[
H_2 = -\sum_{\mathbf{Q}_p} \frac{K}{2} (\tau_i^+ \tau_j^- \tau_3^+ \tau_4^- \tau_5^+ \tau_6^+ + h.c.) + \sum_{\langle\langle ij \rangle\rangle} J_{3z} \tau_i^z \tau_j^z,
\]

and we further restrict the Hilbert space to be the "2-in 2-out" ice manifold. Therefore, this new Hamiltonian will only act on the states in the ice manifold. This perturbative model was already proposed in one perturbative limit of the realistic spin model for Yb$_2$Ti$_2$O$_7$ in Ref. [39]. When \( |J_{3z}| \ll K \), the ground state of \( H_2 \) is the QSI U(1) QSL phase. When \( |J_{3z}| \gg K \), the system develops a \( Q = 2\pi(001) \) antiferromagnetic order for a positive \( J_{3z} \), and \( Q = (000) \) ferromagnetic order for a negative \( J_{3z} \). Again, we expect the transition from the QSI U(1) QSL to the ferromagnetic state is strongly first order, while the transition to the antiferromagnetic state is either continuous or extremely weakly first order.

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