Letter

# Coulombic antiferromagnet in spin-1/2 pyrochlores with dipole-octupole doublets

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An exotic state of matter could coexist with conventional orders such that the gauge fields and fractionalized excitations could prevail in a seemingly ordered state. We explore the Coulombic antiferromagnet in the spin-1/2 pyrochlores with dipole-octupole doublets. This fractionalized antiferromagnetic state carries both antiferromagnetic order and emergent quantum electrodynamics with the gapless gauge photon and fractionalized quasiparticles. We explain the characteristic physical properties, including the thermodynamics and the dynamics of this exotic state. This includes, for example, the anomalously large $T^3$ specific heat and the broad spinon continuum in the inelastic neutron scattering. We discuss the experimental and material's relevance and expect this work to inspire interests in the search of exotic physics among the ordered magnets.

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Exotic quantum states of matter with long-range quantum entanglement, such as fractional quantum Hall effects and quantum spin liquids, are characterized by emergent gauge fields and fractionalized excitations [1]. Due to the emergent nonlocal gauge structures, these states are usually robust against weak local perturbations. Moreover, the emergent gauge field and fractionalization could survive even in the presence of long-range orders. This happens, for example, when the residual interactions between the fractionalized quasiparticles induce the condensation of composite objects without completely destroying the internal gauge structures. Such a scenario was theoretically proposed about 20 years ago for the superconducting cuprates where the hosting exotic state was suggested to be $\mathbb{Z}_2$ topological order [2–5]. Over there, the bilinear of the fermionic spinons was condensed to generate the antiferromagnetic long-range order while the fractionalization and the $\mathbb{Z}_2$ gauge structure persist. To distinguish it from the conventional antiferromagnet, this fractionalized antiferromagnet was dubbed an "AF* state." Although its connection to the cuprates remains illusive, such an exotically ordered state points to the important possibility of emergent exotic physics in the seemingly ordered systems, i.e., the coexistence of long-range order with the long-range quantum entanglement.

Despite the possibility of exoticity in the long-range ordered systems, the popular goal of exotic physics is to examine spin liquid candidates among disordered magnets with frustration such that the frustration enhances the quantum fluctuations and suppresses the magnetic orders down to zero temperature [6,7]. In this Letter we consider the possibility of fractionalized antiferromagnet in the spin-1/2 quantum pyrochlore antiferromagnet with dipole-octupole doublets where the emergent U(1) gauge field and the fractionalization coexist with the antiferromagnetic order. The pyrochlore magnets have been an active topic in the modern research of quantum magnetism and have attracted quite some attention in last two decades [8,9]. The classical spin ice has been observed in the pyrochlore spin ice magnets and understood based on the interacting Ising spins [10–13]. The quantum counterpart, referred to as pyrochlore quantum spin ice or pyrochlore ice U(1) spin liquid, is less conclusive [6,14–17]. There have been two kinds of relevant models [6,15,16,18–21]. Due to the low energy scales of the emergent excitations, such as the gauge photons and the fractionalized excitations, the experimental side is less conclusive compared to the classical case. So far, the existing candidates that remain disordered and promising are the Tb-based [22], Pr-based pyrochlores [23,24] with the non-Kramers doublets, and more recently, the Ce-based pyrochlores, $Ce_2Sn_2O_7$ and $Ce_2Zr_2O_7$, with the dipole-octupole (DO) doublets [25–34]. No such fractionalized antiferromagnet was previously obtained for the conventional Kramers doublets, such as $Yb^{3+}$ ions or the non-Kramers doublets like $Pr^{3+}$, and thus we turn to the dipole-octupole doublets and explore their properties and the possible existence. We show the system could stabilize another exotic quantum state, namely, the Coulombic antiferromagnet, in addition to the pyrochlore U(1) spin liquid. This fractionalized antiferromagnet is analogous to the AF* state for the cuprates, except that the gauge sector is U(1) in three dimensions (3D) and all the fractionalized quasiparticles are gapped. Thus we refer this exotic state as the "pyrochlore AF* state."

To begin with, we consider the generic symmetry-allowed spin model for the DO doublets with

$$H = \sum_{\langle ij\rangle} \big[J_x S_i^x S_j^x + J_z S_i^z S_j^z + J_{xz}\big(S_i^x S_j^z + S_i^z S_j^x\big) + J_y S_i^y S_j^y\big] - \sum_{\langle\langle ij\rangle\rangle} J_2 S_i^z S_j^z + \cdots , \tag{1}$$

where the spin $\boldsymbol{S}_i$ is defined on the DO doublet, and $\langle ij\rangle$ ($\langle\langle ij\rangle\rangle$) refers to the nearest (second) neighbors. The terms inside "[ ]" are the symmetry-allowed interactions between the nearest neighbors, while "$\cdots$" refers to other interactions, such as the dipole-dipole interaction between the $S^z$ component and the superexchange beyond the nearest neighbors. For the unfrustrated regime of the nearest-neighbor model, the ground state can be well understood, and no fractionalized antiferromagnet (AF$^*$) phase was found [19,28]. Thus extra interactions and/or the frustrated regime are required to support its existence, and we have included a second-neighbor $S^z$-$S^z$ interaction in our study.

Microscopically [19], the $S^z$ ($S^x$ or $S^y$) component for the DO doublet carries the magnetic dipole (octupole) moment. One should keep the $S^z$-$S^z$ coupling if the dipole-dipole interaction is considered. From the symmetry analysis, however, $S^z$ and $S^x$ ($S^y$) transform identically as a magnetic dipole (octupole) under the space group. This is the symmetry reason why there exists the $S^x$-$S^z$ coupling in Eq. (1). If one restricts to the nearest-neighbor model, one can apply a rotation about the $y$ axis to eliminate the crossing term between $S^x$ and $S^z$. With the further-neighbor interactions it becomes impossible, because such a rotation immediately regenerates the crossing terms from the further neighbors. After the rotation, Eq. (1) takes a new form,

$$H = \sum_{\langle ij\rangle} \big[\tilde{J}_x \tilde{S}_i^x \tilde{S}_j^x + \tilde{J}_z \tilde{S}_i^z \tilde{S}_j^z + J_y S_i^y S_j^y\big] - \sum_{\langle\langle ij\rangle\rangle} J_2 \big[\cos^2\theta \tilde{S}_i^z \tilde{S}_j^z + \sin^2\theta \tilde{S}_i^x \tilde{S}_j^x + \cos\theta\sin\theta\big(\tilde{S}_i^x \tilde{S}_j^z + \tilde{S}_i^z \tilde{S}_j^x\big)\big] + \cdots , \tag{2}$$

where $\tilde{S}_i^x = \cos\theta S_i^x + \sin\theta S_i^z$, $\tilde{S}_i^z = \cos\theta S_i^z - \sin\theta S_i^x$, $\tilde{J}_x$ and $\tilde{J}_z$ are the rotated couplings and are related to the old ones [25], and the crossing term reappears. As we show below, the irremovable crossing term is responsible for the emergence of the pyrochlore AF$^*$ state from the U(1) spin liquid. Since other interactions in "$\cdots$" necessarily renormalize the $J_2$ interactions, to capture the qualitative physics, here we consider an alternative model with the renormalized couplings:

$$H_a = \sum_{\langle ij\rangle} \big[\tilde{J}_x \tilde{S}_i^x \tilde{S}_j^x - \tilde{J}_\perp\big(\tilde{S}_i^z \tilde{S}_j^z + S_i^y S_j^y\big)\big] - \sum_{\langle\langle ij\rangle\rangle} J_{2xz}\big(\tilde{S}_i^x \tilde{S}_j^z + \tilde{S}_i^z \tilde{S}_j^x\big). \tag{3}$$

To avoid further complication, we have set $\tilde{J}_z = J_y = -\tilde{J}_\perp$. $H_a$ is our minimal model to reveal the pyrochlore AF$^*$ state.

We start from the large antiferromagnetic $\tilde{J}_x$ regime and consider the instability to the nearby phases. With the large antiferromagnetic $\tilde{J}_x$, the system prefers the "two-plus two-minus" ice configuration for $\tilde{S}^x$ and realizes a pyrochlore U(1) spin liquid with the perturbed quantum fluctuations. This is a reasonable starting point, because both the pyrochlore U(1) spin liquid and the AF$^*$ state share the same gauge structure. In this limit the system is characterized by the emergent U(1) gauge field and the fractionalized excitations. To reveal it we implement the spinon-gauge construction and express the spin operators as [16,19,20]

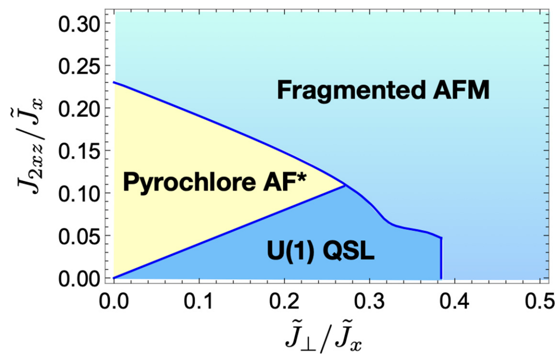


FIG. 1. Phase diagram for $H_a$ in Eq. (3) from the gauge mean-field theory. The phase boundary between the pyrochlore AF$^*$ state and the fragmented AFM is continuous via the spinon condensation. The remaining phase boundaries are all first order. U(1) QSL refers to the pyrochlore U(1) spin liquid. The AF$^*$ state is referred as the Coulombic antiferromagnet. The description of each phase within the gauge mean-field theory is listed in Table I.

$$\tilde{S}^+_{\boldsymbol{r},\boldsymbol{r}+\boldsymbol{e}_\mu} \equiv \tilde{S}^z_{\boldsymbol{r},\boldsymbol{r}+\boldsymbol{e}_\mu} - iS^y_{\boldsymbol{r},\boldsymbol{r}+\boldsymbol{e}_\mu} = \Phi^\dagger_{\boldsymbol{r}}\Phi_{\boldsymbol{r}+\boldsymbol{e}_\mu}\tilde{s}^+_{\boldsymbol{r},\boldsymbol{r}+\boldsymbol{e}_\mu}, \tag{4}$$

$$\tilde{S}^x_{\boldsymbol{r},\boldsymbol{r}+\boldsymbol{e}_\mu} = \tilde{s}^x_{\boldsymbol{r},\boldsymbol{r}+\boldsymbol{e}_\mu}, \tag{5}$$

where the pyrochlore spin is now interpreted as sitting on the link connecting the centers $\boldsymbol{r}$ and $\boldsymbol{r}+\boldsymbol{e}_\mu$ of the neighboring tetrahedra. The tetrahedral centers form a diamond lattice, and $\boldsymbol{e}_\mu$ refers to the four nearest-neighbor vectors that connect the I sublattice sites to the II sublattice sites. Here $\tilde{\boldsymbol{s}}_{\boldsymbol{r},\boldsymbol{r}+\boldsymbol{e}_\mu}$ is a spin-1/2 variable that corresponds to the emergent U(1) gauge field. The spinons carry the emergent gauge charge, and $\Phi^\dagger_{\boldsymbol{r}}$ ($\Phi_{\boldsymbol{r}}$) creates (annihilates) a spinon at the diamond site $\boldsymbol{r}$ such that the spinon particle number $Q_{\boldsymbol{r}}$ satisfies

$$[\Phi_{\boldsymbol{r}}, Q_{\boldsymbol{r}'}] = \Phi_{\boldsymbol{r}}\delta_{\boldsymbol{r}\boldsymbol{r}'}, \quad [\Phi^\dagger_{\boldsymbol{r}}, Q_{\boldsymbol{r}'}] = -\Phi^\dagger_{\boldsymbol{r}}\delta_{\boldsymbol{r}\boldsymbol{r}'}. \tag{6}$$

As the Hilbert space is enlarged by the spinon-gauge construction, the constraint $Q_{\boldsymbol{r}} = \eta_{\boldsymbol{r}}\sum_\mu \tilde{s}^x_{\boldsymbol{r},\boldsymbol{r}+\eta_{\boldsymbol{r}}\boldsymbol{e}_\mu}$ is imposed. The spinon-gauge construction captures the nature of the pyrochlore U(1) spin liquid as a string-net condensed phase [35], where $\tilde{S}^\pm_i$ corresponds to the shortest open string with the spinons at the two ends. The model $H_a$ becomes

$$H_a = \sum_{\boldsymbol{r}} \frac{\tilde{J}_x}{2} Q_{\boldsymbol{r}}^2 - \sum_{\boldsymbol{r}}\sum_{\mu\neq\nu} \frac{\tilde{J}_\perp}{2} \Phi^\dagger_{\boldsymbol{r}+\eta_{\boldsymbol{r}}\boldsymbol{e}_\mu}\Phi_{\boldsymbol{r}+\eta_{\boldsymbol{r}}\boldsymbol{e}_\nu}\tilde{s}^{-\eta_{\boldsymbol{r}}}_{\boldsymbol{r},\boldsymbol{r}+\eta_{\boldsymbol{r}}\boldsymbol{e}_\mu}\tilde{s}^{+\eta_{\boldsymbol{r}}}_{\boldsymbol{r},\boldsymbol{r}+\eta_{\boldsymbol{r}}\boldsymbol{e}_\nu} - \sum_{\boldsymbol{r}\in \mathrm{I}}\sum_\mu \sum_{j\in[\boldsymbol{r},\boldsymbol{r}+\boldsymbol{e}_\mu]_2} \frac{J_{2xz}}{2}\big(\Phi^\dagger_{\boldsymbol{r}}\Phi_{\boldsymbol{r}+\boldsymbol{e}_\mu}\tilde{s}^+_{\boldsymbol{r},\boldsymbol{r}+\boldsymbol{e}_\mu} + \text{H.c.}\big)\tilde{s}^x_j, \tag{7}$$

TABLE I. Description of each phase within gauge mean-field theory.

| Phases | $\langle\Phi\rangle$ | $\langle\tilde{s}^x\rangle$ | $\langle\tilde{s}^\pm\rangle$ | $\langle S^x\rangle$ | $\langle S^z\rangle$ |
|---|---|---|---|---|---|
| U(1) QSL | $=0$ | $=0$ | $\neq 0$ | $=0$ | $=0$ |
| AF* state | $=0$ | $\neq 0$ | $\neq 0$ | $\neq 0$ | $\neq 0$ |
| Fragmented AFM | $\neq 0$ | $\neq 0$ | $\neq 0$ | $\neq 0$ | $\neq 0$ |

where $[\boldsymbol{r}, \boldsymbol{r}+\boldsymbol{e}_\mu]_2$ refers to the set of the second neighbors from this site at $\boldsymbol{r}+\boldsymbol{e}_\mu/2$. To solve $H_a$, we decouple it into the spinon sector and the gauge field sector with

$$H_{\rm spinon} = \sum_{\boldsymbol{r}} \frac{\tilde{J}_x}{2} Q_{\boldsymbol{r}}^2 - \sum_{\boldsymbol{r}} \sum_{\mu\neq\nu} \frac{1}{2} \tilde{J}_\perp \chi_1 \Phi^\dagger_{\boldsymbol{r}+\eta_r \boldsymbol{e}_\mu} \Phi_{\boldsymbol{r}+\eta_r \boldsymbol{e}_\nu} - \sum_{\boldsymbol{r}\in {\rm I}} \sum_{\mu} 6 J_{2xz} \chi_2 \big(\Phi^\dagger_{\boldsymbol{r}} \Phi_{\boldsymbol{r}+\boldsymbol{e}_\mu} + \Phi^\dagger_{\boldsymbol{r}+\boldsymbol{e}_\mu} \Phi_{\boldsymbol{r}}\big), \quad (8)$$

$$H_{\rm gauge} = -\sum_{\langle ij\rangle} \tilde{J}_\perp I_1 \big(\tilde{s}_i^z \tilde{s}_j^z + s_i^y s_j^y\big) - \sum_{\langle\langle ij\rangle\rangle} J_{2xz} I_2 \big(\tilde{s}_i^x \tilde{s}_j^z + \tilde{s}_i^z \tilde{s}_j^x\big), \quad (9)$$

where we have set

$$\chi_1 = \langle \tilde{s}_i^+ \tilde{s}_j^- \rangle, \quad \text{for } \langle ij\rangle, \quad (10)$$

$$\chi_2 = \big\langle \tilde{s}_i^+ \tilde{s}_j^x \big\rangle, \quad \text{for } \langle\langle ij\rangle\rangle, \quad (11)$$

and

$$I_1 = \langle \Phi^\dagger_{\boldsymbol{r}} \Phi_{\boldsymbol{r}'} \rangle, \quad \text{for } \langle\langle ij\rangle\rangle, \quad (12)$$

$$I_2 = \langle \Phi^\dagger_{\boldsymbol{r}} \Phi_{\boldsymbol{r}'} \rangle, \quad \text{for } \langle ij\rangle. \quad (13)$$

These parameters are solved self-consistently [36]. Due to the spatial uniformity of the model, these parameters are uniform throughout the system. The gauge mean-field phase diagram is depicted in Fig. 1, where the description of each phase within the gauge mean-field theory is listed in Table I. Two exotic phases, the pyrochlore U(1) spin liquid and the pyrochlore AF* state, occupy the regions with small $\tilde{J}_\perp$ and $J_{2xz}$. The transition between these two states in Fig. 1 is first order. Both states have gapped spinon and gapless U(1) gauge photon excitations, while the pyrochlore AF* state has an antiferromagnetic all-in all-out order. The fragmented antiferromagnet (AFM) in Fig. 1 is a conventional AFM with an all-in all-out order and can be obtained from the AF* state via the spinon condensation at the Γ point. Such a transition is an Anderson-Higgs' transition. The visible part of the magnetic order is in the $S^z$ component and behaves as

$$\big\langle S_i^z \big\rangle = \cos\theta \big\langle \tilde{S}_i^z \big\rangle + \sin\theta \big\langle \tilde{S}_i^x \big\rangle = \tfrac{1}{2} \cos\theta [\langle \Phi^\dagger_{\boldsymbol{r}} \Phi_{\boldsymbol{r}'} \rangle \langle \tilde{s}^\dagger_{\boldsymbol{r}\boldsymbol{r}'} \rangle + {\rm H.c.}] + \sin\theta \big\langle \tilde{s}_i^x \big\rangle. \quad (14)$$

The "fragmented AFM" is used to capture this moment fragmentation where the ordered momentum is fragmented into the gauge link piece and the spinon condensate piece. Although the $S^x$ moment also develops order, it is invisible or hidden due to its magnetic multipolar nature. Moreover, the $S^z$ moment, despite being ordered, could simultaneously function as a disordering operator to flip the $S^x$ order and generate the magnetic excitations.

From Eq. (14) it is clear that the all-in all-out ($S^z$) order appears when there exists the spinon tunneling between two diamond sublattices and/or a nonvanishing emergent gauge field. Due to the generic coupling between $S^x$ and $S^z$ or between $\tilde{S}^x$ and $\tilde{S}^z$, these two conditions are actually concomitant. In the above, we have explicitly shown that the pyrochlore AF* state appears from the irremovable coupling between $\tilde{S}^x$ and $\tilde{S}^z$ and supports both contributions in Eq. (14) nonvanishing. It is also possible that the spinon interaction leads to the condensation in the spinon particle-hole channel and results in an antiferromagnetic order. The key physical properties of the pyrochlore AF* state, however, are independent from the physical origin and are discussed below.

Despite the presence of the all-in all-out order, the quantum fluctuations of the pyrochlore AF* state are still governed by the fractionalized spinons and the emergent U(1) gauge fluctuations. Thus the gapless U(1) gauge photon is responsible for the $T^3$ specific heat at low temperatures. Due to the low energy scale of the exchange coupling, the coefficient of the $T^3$ specific heat should be quite large and visible experimentally. We further explore the spectroscopic consequences of the pyrochlore AF* state. Due to the fractionalized nature of this state, the excitation spectrum should be quite different from the spin-wave excitation for the conventional ordered antiferromagnet. This can be analyzed from a more phenomenological treatment that introduces the all-in all-out order on top of the fractionalized spin liquid state. The resulting spinon Hamiltonian will be of an identical structure as Eq. (8), and we write it down here,

$$H_{\rm AF^*} = \sum_{\boldsymbol{r}} \frac{\tilde{J}_x}{2} Q_{\boldsymbol{r}}^2 - \sum_{\boldsymbol{r}} \sum_{\mu\neq\nu} t_1 \Phi^\dagger_{\boldsymbol{r}+\eta_r \boldsymbol{e}_\mu} \Phi_{\boldsymbol{r}+\eta_r \boldsymbol{e}_\nu} - \sum_{\boldsymbol{r}\in {\rm I}} \sum_{\mu} t_2 \big(\Phi^\dagger_{\boldsymbol{r}} \Phi_{\boldsymbol{r}+\boldsymbol{e}_\mu} + \Phi^\dagger_{\boldsymbol{r}+\boldsymbol{e}_\mu} \Phi_{\boldsymbol{r}}\big), \quad (15)$$

where the intersublattice hopping $t_2 \neq 0$ when the magnetic order is present. This is a bit different from the case without the sublattice mixing, where the spinon number is separately conserved on each diamond sublattice and the two spinon bands are degenerate [14]. In our case here the two spinon dispersions are not degenerate, and we have

$$\omega_\pm(\boldsymbol{k}) = \sqrt{2\tilde{J}_x} \left[ \lambda - t_1 \sum_n \cos(\boldsymbol{k}\cdot\boldsymbol{a}_n) \pm t_2 \left| \sum_\mu e^{i\boldsymbol{k}\cdot\boldsymbol{e}_\mu} \right| \right]^{\frac{1}{2}}, \quad (16)$$

where $\{\boldsymbol{a}_n\}$ refers to the twelve second-neighbor vectors on the diamond lattice, and $\lambda$ is the Lagrangian multiplier used to fix the constraint $|\Phi_{\boldsymbol{r}}^2| = 1$ and is determined self-consistently. Since the $S^z$ moment in Eq. (14) contains the spinon bilinear and is the only component that is linearly coupled to the external magnetic field, the spinon-pair excitations are included in the $S^z$-$S^z$ correlation and can be detected in the inelastic neutron-scattering measurement. From the energy-momentum conservation, the spinon-pair excitation is characterized with the energy-momentum relation,

$$\Omega(\boldsymbol{q}) = \omega_\mu(\boldsymbol{k}_1) + \omega_\nu(\boldsymbol{q}-\boldsymbol{k}_1), \quad (17)$$

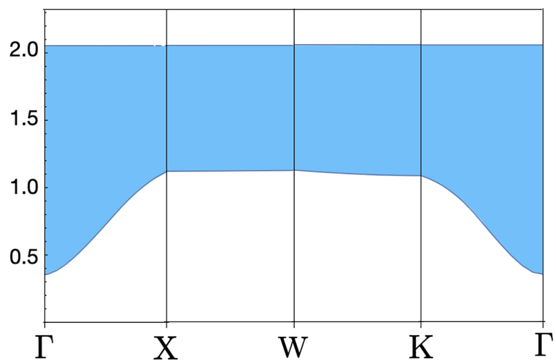


FIG. 2. Spinon continuum along the high-symmetry momentum lines of the pyrochlore AF* state. Here $t_1 = 0.025$, $t_2 = 0.02$, and $\tilde{J}_x$ is set to unity.

where $\mu, \nu = \pm$ refer to the spinon branches. Due to the freedom of the spinon momentum $\boldsymbol{k}_1$, the above excitation corresponds to a continuum in both energy and momentum domains. In Fig. 2 we plot the momentum-resolved energy range of the two-spinon continuum.

Apart from the gapped spinon continuum and the gapless gauge photon in the dynamics, there also exists the electric monopole continuum in the $\tilde{S}^x$-$\tilde{S}^x$ correlation for the pyrochlore AF* state. For the pyrochlore U(1) spin liquid, the electric monopole experiences a background dual U(1) gauge flux that is a $\pi$ flux in the unit cell, and the spectrum has an enlarged spectral period in the reciprocal space. In the pyrochlore AF* state, because $\langle \tilde{S}^x \rangle \neq 0$, the background dual U(1) gauge flux is deviated from the $\pi$ flux, and the electric monopole will have a Hofstadter band structure that can be manifested in the monopole continuum [37]. As the $S^z$-$S^z$ correlation contains both $\tilde{S}^z$-$\tilde{S}^z$ and $\tilde{S}^x$-$\tilde{S}^x$ correlations [28], the monopole continuum can be revealed. In contrast to the gapped monopoles and the gapless gauge photon that have a close and low energy scale, the spinons appear in a much higher energy and may be more accessible in the scattering measurements [38]. In general, the emergent parton-gauge dynamics for the pyrochlore AF* state is fundamentally different from the simple magnons for a conventionally ordered magnet. As a comparison, we further plot the magnon dispersion for the fragmented AFM in the Supplemental Material [36]. Among the four magnon branches, two are flat bands and the other two are dispersive.

As the pyrochlore AF* state carries an antiferromagnetic order, there should exist a finite-temperature phase transition. The antiferromagnetic order occurs in the particle-hole channel of the spinons and does not carry any emergent gauge charge. Thus, the Ginzburg-Landau theory for such a transition is absent the gauge degrees of freedom, and we expect this transition to be a 3D Ising transition. As for the material relevance, we propose $Nd_2Sn_2O_7$ as a candidate for the pyrochlore AF* state. The magnetic transition [39] in $Nd_2Sn_2O_7$ was found to be continuous [40]. It would be illuminating to examine the critical exponents at the transition.

Below the ordering transition, $Nd_2Sn_2O_7$ shows an anomalously large $T^3$ specific heat, and Ref. [40] expected a linearly dispersive mode with an excitation velocity $v_{ex} = 55$ m/s. There are two factors against the Goldstone magnon mode interpretation for this mode. First, the $Nd^{3+}$ ground state doublet is a DO doublet [19] and is well separated from the excited ones by a crystal field gap of 26 meV [40] such that the effective spin-1/2 moment from the ground states captures the magnetic properties below ~20 K. The effective model does not have any continuous symmetry [19] to support gapless Goldstone modes nor an accidental continuous degeneracy to support pseudo-Goldstone modes via quantum order by disorder [41,42]. Second, how about a fine-tuned spin model that is close to the point with a continuous spin symmetry? To obtain the all-in all-out order, the model should be a ferromagnetic model with a continuous symmetry. Little frustration would be expected for the continuous ferromagnetic case, which is incompatible with the frustrated nature of the slow paramagnetic spin dynamics from the inelastic neutron and $\mu$SR measurements [40,43]. Moreover, the ordered fraction of $Nd^{3+}$ moments is not small [40], so it is unlikely to be proximate to a quantum transition with gapless critical modes. If the pyrochlore AF* state proposal for $Nd_2Sn_2O_7$ is relevant, $v_{ex}$ would be interpreted as the speed of emergent gauge photon. To further examine the proposal, we suggest the inelastic neutron-scattering measurement to directly detect the gauge photon as well as the continuum of the spinons and the electric monopoles.

In contrast to $Nd_2Sn_2O_7$, $Nd_2Zr_2O_7$ and $Nd_2Hf_2O_7$ are deep in the "fragmented AFM" state, with an all-in all-out order in the $S^z$ component and the well-defined spin-wave modes [44–48]. As the actual order involves the hidden $S^x$ component, the $S^z$ operator is then responsible for flipping the order in $S^x$ in addition to having the order within itself. This interesting moment fragmentation [49] arises from the $J_{xz}$ crossing term [19] in Eq. (1) and has been well understood from the peculiar microscopic properties and the model for the DO doublets of the $Nd^{3+}$ ions [44,47,48]. Here we do not get into too much detail. Another Nd compound $Nd_2GaSbO_7$ with the all-in all-out order was experimentally studied and the moment fragmentation physics was absent [50]. The continuumlike feature in the ordered regime seems more compatible with the continuous excitations of the AF* state. Due to the intrinsic disorder, however, more information needs to be collected and examined.

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