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Rank-2 $U(1)$ spin liquid on the breathing pyrochlore lattice

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Higher-rank generalisations of electrodynamics have recently attracted considerable attention because of their ability to host “fracton” excitations, with connections to both fracton topological order and gravity. However, the search for higher-rank gauge theories in experiment has been greatly hindered by the lack of materially-relevant microscopic models. Here we show how a spin liquid described by rank-2 $U(1)$ gauge theory can arise in a magnet on the breathing pyrochlore lattice. We identify Yb-based breathing pyrochlores as candidate systems, and make explicit predictions for how the rank-2 $U(1)$ spin liquid would manifest itself in experiment.

Introduction. It is of great intellectual interest and practical utility to discover novel effective laws of nature emerging from many-body systems. Traditionally, this enterprise has been entwined with the concept of broken symmetry [1]. However, a powerful alternative has proved to be the local constraints which arise from competing or “frustrated”, interactions. In the context of frustrated magnets, these can lead to the emergence of a local gauge symmetry, and thereby to quantum spin liquids, which defy all usual concepts of magnetic order, and instead exhibit fractionalised excitations and long-range entanglement [2–5]. A well-studied example is quantum spin ice, a realisation of a $U(1)$ gauge theory on the pyrochlore lattice, whose emergent excitations exactly mimic conventional electrodynamics: photons, electric charges and magnetic monopoles. As such, it has attracted intense theoretical [6–15] and experimental [16–24] investigation.

Recent work has highlighted the possibility of more exotic forms of emergent electrodynamics [25–28], where electric and magnetic fields have the form of rank-2 (or higher-rank) tensors. These theories have modified conservation laws and gauge symmetries, resulting in some remarkable properties. Some are argued to mimic gravity [25, 29, 30], while others are dual to elasticity theory [31, 32]. In both cases, the charged excitations, dubbed “fractons”, have constrained mobility, and characterize a new class of topological order [33–42]. Fracton models are also linked to quantum stabilizer codes [43, 44] and holography [45]. None the less, these desirable properties come at a price: the local constraint required has a tensor character. As a consequence, prototypical models of fractons require rather complicated interactions [25, 33, 35–37], with just a handful of proposals motivated by experiment [41, 46, 47]. In the case of gapless higher-rank gauge theories, only a few concrete models exist [27, 48, 49], and even less is known about how to achieve such a phase in a real material. For this reason, realizing an emergent higher-rank electrodynamics in experiment presents a significant challenge.

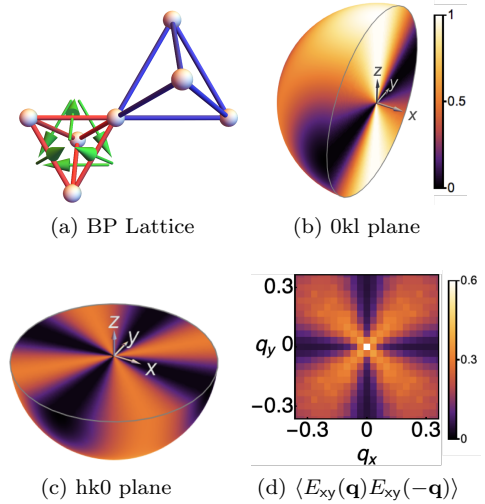


FIG. 1. Breathing pyrochlore (BP) lattice, and singular correlations characteristic of a rank-2 $U(1)$ [R2-U1] gauge theory. (a) BP lattice, with A- and B-sublattice tetrahedra of unequal size. The vectors associated with Dzyaloshinskii-Moriya (DM) interactions on the A-sublattice [Eq. (6)] are illustrated with green arrows. (b) Prediction of R2-U1 theory for the correlation function $\langle E_{xy}(\mathbf{q})E_{xy}(-\mathbf{q}) \rangle$ [Eq. (17)], showing a 2-fold pinch point in the [0kl] plane. (c) Perpendicular section, showing a 4-fold pinch point (4FPP) in the [hk0] plane. (d) Equivalent results from MC simulation of the breathing pyrochlore model [Eq. (6)].

In this Letter we show how a canonical rank-2 $U(1)$ [R2-U1] spin liquid can arise in a realistic model of a frustrated magnet. The model we consider is the Heisenberg antiferromagnet (HAF) on a breathing-pyrochlore (BP) lattice, perturbed by weak Dzyaloshinskii-Moriya (DM) interactions [Fig. 1a]. Working in the classical limit, relevant to a spin liquid at finite temperatures, we establish that fluctuations can be described using a tensor field satisfying the constraints required for a R2-U1 gauge theory. We use classical Monte Carlo (MC) simulation to confirm this scenario, and to explore how a R2-U1 spin

liquid could be identified in experiment. We find that 4-fold pinch points (4FPP), characteristic of the R2-U1 state [50], become visible in polarised neutron scattering. We discuss the application of these ideas to real materials, identifying Yb-based breathing pyrochlores as potential candidates for an R2-U1 spin liquid state. These results complement earlier work exploring gapped, fraction topological order, in models with bilinear interactions [41, 46, 47], providing an example of an R2-U1 state, in an experimentally-motivated context.

Review of R2-U1 theory. Conventional, $U(1)$, electrostatics is built around a vector field \mathbf{E} , subject to a Gauss law $\partial_i E_i = \rho$ so that, in the absence of charges,

$$\partial_i E_i = 0. \quad (1)$$

The key which unlocked the effective electrostatics of spin ice was the realisation that, at low temperatures, in a classical limit, spins satisfied a local constraint of precisely the form of Eq. (1) [51, 52].

Here we consider instead an R2-U1 electrostatics, in its self-dual, vector-charged, traceless form [26–28], and seek to show that, in an equivalent classical limit, spins satisfy the appropriate generalisation of Eq. (1). The R2-U1 theory is built around a rank-2 tensor electric field \mathbf{E} that is symmetric and traceless,

$$E_{ji} = E_{ij} \quad \text{Tr } \mathbf{E} = 0, \quad (2)$$

subject to a generalised Gauss' law for a vector charge

$$\partial_i E_{ij} = \rho_j. \quad (3)$$

In the low-energy sector, the theory is charge free, i.e.

$$\partial_i E_{ij} = 0. \quad (4)$$

These constraints determine the symmetry of the R2-U1 gauge field

$$A_{ij} \rightarrow A_{ij} + \partial_i \lambda_j + \partial_j \lambda_i + \gamma \delta_{ij}, \quad (5)$$

which in turn implies the form of the associated magnetic field, B_{ij} [26, 28]. However the key observable properties of an R2-U1 spin liquid follow from the correlations of its electric field E_{ij} [50], and our goal will therefore be to show how the spins in a frustrated magnet can be described by a tensor field E_{ij} , satisfying the constraints Eqs. (2, 4). In the Supplemental Material a more detailed review is provided, which includes Refs. [25, 26]

The model. To this end, we consider a HAF, perturbed by weak DM interactions, on a “breathing” pyrochlore (BP) lattice, for which A- and B-sublattice tetrahedra have a different size

$$\begin{aligned} \mathcal{H}_{\text{BP}} = & \sum_{\langle ij \rangle \in \text{A}} \left[J_A \mathbf{S}_i \cdot \mathbf{S}_j + D_A \hat{\mathbf{d}}_{ij} \cdot (\mathbf{S}_i \times \mathbf{S}_j) \right] \\ & + \sum_{\langle ij \rangle \in \text{B}} \left[J_B \mathbf{S}_i \cdot \mathbf{S}_j + D_B \hat{\mathbf{d}}_{ij} \cdot (\mathbf{S}_i \times \mathbf{S}_j) \right]. \end{aligned} \quad (6)$$

Definitions of the bond-dependent vectors $\hat{\mathbf{d}}_{ij}$ [cf. Fig. 1a] are given in the Supplemental Material, which includes Refs. [17, 53–56]. This model finds experimental motivation in Yb-based breathing pyrochlores, discussed below.

Transcription to symmetry-based coordinates. Our next step is to seek a continuum representation of Eq. (6). To accomplish this, we consider the classical limit where individual components of spin commute, and introduce a set of coarse-grained fields $\mathbf{m}_\mathbf{X}$ which transform as irreducible representations of the lattice symmetry [29, 57, 58]. In this basis, the Hamiltonian becomes (See Supplemental Materials for a more detailed derivation)

$$\mathcal{H} = \frac{1}{2} \sum_{\text{tet} \in \text{A}, \mathbf{X}} a_{\text{A}, \mathbf{X}} m_{\mathbf{X}}^2 + \frac{1}{2} \sum_{\text{tet} \in \text{B}, \mathbf{X}} a_{\text{B}, \mathbf{X}} m_{\mathbf{X}}^2, \quad (7)$$

where \mathbf{X} runs over irreps of the group T_d , i.e. $\{\text{A}_2, \text{E}, \text{T}_2, \text{T}_{1+}, \text{T}_{1-}\}$, with the fields $m_\mathbf{X}$ and the coefficients $a_\mathbf{X}$ defined in Table I and Table II of the Supplemental Material.

Before considering the effect of DM interactions, it is helpful to explore how this approach works in the case of a known spin liquid, the HAF on a pyrochlore lattice [60–65]. Setting

$$J_A = J_B, \quad D_A = D_B = 0, \quad (8)$$

we find

$$0 < a_{\text{A}_2} = a_{\text{E}} = a_{\text{T}_2} = a_{\text{T}_{1-}} < a_{\text{T}_{1+}}. \quad (9)$$

It follows that the fields $m_{\text{A}_2}, m_{\text{E}}, m_{\text{T}_2}, m_{\text{T}_{1-}}$ are all free to fluctuate in the ground state. We can conveniently collect all of these fields in the rank-2 tensor

$$\mathbf{E}^{\text{HAF}} = \mathbf{E}_{\text{sym}}^{\text{HAF}} + \mathbf{E}_{\text{antisym}}^{\text{HAF}} + \mathbf{E}_{\text{trace}}^{\text{HAF}} \quad (10)$$

where

$$\mathbf{E}_{\text{sym}}^{\text{HAF}} = \begin{bmatrix} \frac{2}{\sqrt{3}} m_{\text{E}}^1 & m_{\text{T}_{1-}}^z & m_{\text{T}_{1-}}^y \\ m_{\text{T}_{1-}}^z & -\frac{1}{\sqrt{3}} m_{\text{E}}^1 - m_{\text{E}}^2 & m_{\text{T}_{1-}}^x \\ m_{\text{T}_{1-}}^y & m_{\text{T}_{1-}}^x & -\frac{1}{\sqrt{3}} m_{\text{E}}^1 + m_{\text{E}}^2 \end{bmatrix}, \quad (11)$$

$$(E_{\text{antisym}}^{\text{HAF}})_{ij} = -\epsilon_{ijk} m_{\text{T}_2}^k, \quad (E_{\text{trace}}^{\text{HAF}})_{ij} = -\delta_{ij} \sqrt{\frac{2}{3}} m_{\text{A}_2}. \quad (12)$$

The requirement of the continuity of the fields $m_\mathbf{X}$ [57] imposes the conditions

$$\begin{aligned} & \frac{2}{\sqrt{3}} \begin{bmatrix} \partial_x m_{\text{E}}^1 \\ -\frac{1}{2} \partial_y m_{\text{E}}^1 - \frac{\sqrt{3}}{2} \partial_y m_{\text{E}}^2 \\ -\frac{1}{2} \partial_y m_{\text{E}}^1 + \frac{\sqrt{3}}{2} \partial_y m_{\text{E}}^2 \end{bmatrix} - \begin{bmatrix} \partial_y m_{\text{T}_{1-}}^z + \partial_z m_{\text{T}_{1-}}^y \\ \partial_z m_{\text{T}_{1-}}^x + \partial_x m_{\text{T}_{1-}}^z \\ \partial_x m_{\text{T}_{1-}}^y + \partial_y m_{\text{T}_{1-}}^x \end{bmatrix} \\ & - \sqrt{\frac{2}{3}} \nabla m_{\text{A}_2} + \nabla \times \mathbf{m}_{\text{T}_2} = 0. \end{aligned} \quad (13)$$

We obtain exactly the same constraint if we substitute \mathbf{E}^{HAF} in Eq. (4), implying that HAF automatically satisfies one of the two constraints defining the R2-U1 spin liquid [59].

To convert the HAF into an R2-U1 spin liquid, we need to make the theory symmetric and traceless, and so satisfy Eq. (2). This means eliminating fluctuations of $\mathbf{E}_{\text{antisym.}}^{\text{HAF}}$ and $\mathbf{E}_{\text{trace}}^{\text{HAF}}$ from the ground state, something which can be accomplished by opening gaps to the fields m_{τ_2} and m_{A_2} . For the BP model, Eq. (6), this is achieved by any parameter set for which

$$J_A, J_B > 0, D_A < 0, D_B = 0. \quad (14)$$

In this case, the coefficients $a_{\chi,A}$ satisfy the condition

$$a_{E,A} = a_{\tau_{1-},A} < a_{A_2,A}, a_{\tau_{2,A}}, a_{\tau_{1+},A}, \quad (15)$$

which implies that only the fields \mathbf{m}_E and $\mathbf{m}_{\tau_{1-}}$ enter into the ground state of Eq. (7). Meanwhile, on the B-sublattice, we recover the condition Eq. (9), previously found for the HAF, which imposes the constraint Eq. (13), with the caveat that the fields m_{A_2} and \mathbf{m}_{τ_2} can now be set identically equal to zero. When expressed in terms of the remaining tensor field $\mathbf{E}_{\text{sym}}^{\text{HAF}}$, this is exactly Eq. (4). It follows that, in this classical limit, an R2-U1 gauge theory, satisfying both Eq. (2) and Eq. (4) emerges as the effective description at the low-energy sector of the BP model, Eq. (6).

It is worth noting that, as in the regular pyrochlore lattice [56, 67], DM interaction is only a singular perturbation in the context of the classical ground-state manifold. At finite temperature, classical spin liquids owe their stability to entropy, and a finite value of D_A will be needed to stabilise an R2-U1 spin liquid. For exactly the same reason, introducing a finite value of D_B does not immediately invalidate the mechanism driving the R2-U1 spin liquid, but will reduce the range of temperatures over which it is observed. We will see that both of these expectations are fulfilled by classical Monte Carlo simulations of Eq. (6), described below

Characteristic signatures of R2-U1 state. We now turn to the question of how the R2-U1 spin liquid can be identified, in both simulation and in experiment. The zero-divergence condition in spin ice, Eq. (1), manifests itself in a pinch-point singularity [64]

$$\langle E_i(\mathbf{q})E_j(-\mathbf{q}) \rangle \propto \delta_{ij} - \frac{q_i q_j}{q^2}, \quad (16)$$

which is observed in neutron scattering experiments [68]. In the same way, the constraints associated with an R2-U1 gauge theory, Eq. (2) and Eq. (4), lead to a characteristic singularity in correlations of the tensor

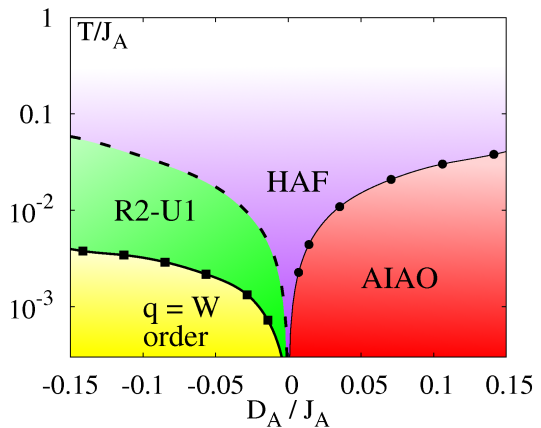


FIG. 2. Finite-temperature phase diagram of the BP model, Eq. (6), as a function of DM interaction D_A . The crossover between the R2-U1 spin liquid, and the $U(1) \times U(1) \times U(1)$ spin liquid (HAF) is shown with a dashed line. The thin solid line indicates a continuous transition into all-in all-out order (AIAO), while thick solid line denotes a first order phase transition into a state with $\mathbf{q} = W$ order. Results are taken from MC simulation with $J_A = J_B = 1, D_B = 0$.

field E_{ij} [50]

$$\begin{aligned} \langle E_{ij}(\mathbf{q})E_{kl}(-\mathbf{q}) \rangle &\propto \frac{1}{2}(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}) + \frac{q_i q_j q_k q_l}{q^4} \\ &- \frac{1}{2} \left(\delta_{ik} \frac{q_i q_l}{q^2} + \delta_{jk} \frac{q_i q_l}{q^2} + \delta_{il} \frac{q_j q_k}{q^2} + \delta_{jl} \frac{q_i q_k}{q^2} \right) \\ &- \frac{1}{2} \left(\delta_{ij} - \frac{q_i q_j}{q^2} \right) \left(\delta_{kl} - \frac{q_k q_l}{q^2} \right). \end{aligned} \quad (17)$$

The three-dimensional structure of the correlation $\langle E_{xy}(\mathbf{q})E_{xy}(-\mathbf{q}) \rangle$ is illustrated in Fig. 1. In the $[0kl]$ plane, correlations exhibit a conventional 2-fold pinch point, comparable to that found in spin ice [Fig. 1b]. However in the perpendicular $[hk0]$ plane, we observe a 4-fold pinch point (4FPP) [Fig. 1c], which unambiguously distinguishes R2-U1 electrostatics from lower-rank theories [50].

Comparison with simulation. We can use the existence of this 4FPP as a test for the R2-U1 spin liquid in simulation. We have carried out classical Monte Carlo (MC) simulations of Eq. (6), for the parameter-set

$$J_A = J_B = 1, D_A = -0.01, D_B = 0. \quad (18)$$

where the constraints Eq. (2) and Eq. (4) are expected to hold. The resulting correlations of E_{ij} , at a temperature $T = 2.5 \times 10^{-3} J_A$, are shown in Fig. 1d. For $\mathbf{q} \rightarrow 0$, these are identical to the predictions of Eq. (17), confirming that the model realizes an R2-U1 spin liquid.

Phase diagram. The results of simulations for a range of values of D_A are collected in Fig. 2. At finite temperature, a finite value of $D_A < 0$ is required to achieve a crossover from the $U(1) \times U(1) \times U(1)$ spin liquid of the pyrochlore HAF, with 2-fold pinch points, into an

R2–U1 spin liquid, with 4FPP. An analytic theory of this crossover, which is controlled by the dimensionless parameter $\eta \sim |D_A|/k_B T$, is provided in Section VI of the Supplemental Material. Meanwhile, at low temperatures, sufficiently negative values of D_A drive a first-order phase transition into an ordered state which involves the characteristic wavevector $\mathbf{q} = \mathbf{W}$ (i.e. corners of the Brillouin zone) [66]. In contrast, a finite value of $D_A > 0$ leads to a continuous phase transition into a state with $\mathbf{q} = 0$, all-out (AIAO) order.

Predictions for neutron scattering. Neutron scattering experiments do not measure correlations of E_{ij} directly, but rather the spin structure factor $S^{\alpha\beta}(\mathbf{q}) = \langle S^\alpha(\mathbf{q})S^\beta(-\mathbf{q}) \rangle$. On general grounds [50], $S^{\alpha\beta}(\mathbf{q})$ is expected to bear witness to the singularity in Eq. (17). But exactly how 4FPPs would manifest themselves in experiment remains an open question. In Fig. 3 we present simulation results for $S^{\alpha\beta}(\mathbf{q})$ for parameters equivalent to Fig. 3a. We find that the 4FPP is *not* visible in the structure factor measured by unpolarised neutron scattering (see Supplemental Material). However the 4FPP *can* be resolved using polarised neutrons. In this case, it manifests itself in the spin-flip (SF) channel for neutrons polarised perpendicular to the scattering plane [68], [Fig. 1c].

Application to materials. Breathing-pyrochlore magnets were first studied as a tractable limit of the pyrochlore HAF [70–73], but have since been realised in materials based on both transition-metal [74–78] and rare-earth ions [79, 80]. Interesting parallels are also found in lacunar spinels [81, 82]. To date, most theoretical work has concentrated on $SU(2)$ -invariant models [70–73, 83–85]. However, in the presence of spin-orbit coupling, the symmetry of the lattice permits anisotropic exchange [80, 86–88]. And, with respect to higher-rank gauge theories, a promising line of enquiry are Yb-based materials, where the required form of interactions appear to predominate.

One concrete example is $\text{Ba}_3\text{Yb}_2\text{Zn}_5\text{O}_{11}$ [79, 80, 86, 88], where A-tetrahedra are estimated to have the coupling parameters $J_A \approx 0.57$ meV, $D_A \approx -0.16$ meV, with other interactions negligible. This is *exactly* the form of interactions needed for an R2–U1 spin liquid, a feature which is expected to be robust [88], since it holds for a wide range of Slater–Koster overlap ratios [89]. Meanwhile, exchange interactions on the larger B-tetrahedra of $\text{Ba}_3\text{Yb}_2\text{Zn}_5\text{O}_{11}$, while less well understood, appear to be orders of magnitude smaller [80, 86]. Thus, while it seems plausible that $\text{Ba}_3\text{Yb}_2\text{Zn}_5\text{O}_{11}$ could realise a R2–U1 spin liquid, this may occur at temperatures too low to measure.

The encouraging example of $\text{Ba}_3\text{Yb}_2\text{Zn}_5\text{O}_{11}$ motivates us to consider the possibility of a magnet with similar structure, but smaller B-tetrahedra, such that the interactions on the B-sublattice become non-negligible. For

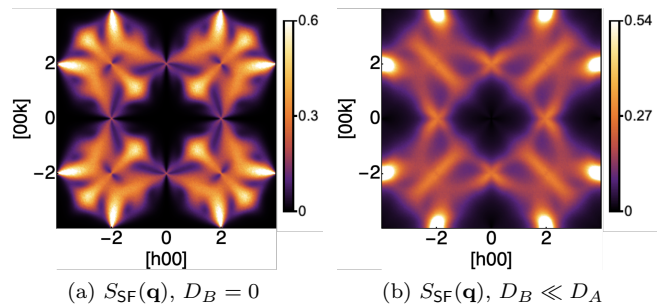


FIG. 3. Spin structure factor found in MC simulation of the BP model, Eq. (6), showing 4-fold pinch points (4FPPs) characteristic of a R2–U1 spin liquid. (a) Correlations in the $[h0k]$ plane, in the spin-flip (SF) channel measured using polarised neutrons. 4FPP are visible at $[0, 0, 2]$ and points related by symmetry. Results are for parameters Eq. (18), $T = 2.5 \times 10^{-3} J_A$. (b) Equivalent results for parameters motivated by $\text{Ba}_3\text{Yb}_2\text{Zn}_5\text{O}_{11}$, Eq. (19), $T = 252$ mK.

concreteness, we consider a parameter set:

$$\begin{aligned} J_A &= 0.57 \text{ meV}, \quad J_B = 0.028 \text{ meV}, \\ D_A &= -0.16 \text{ meV}, \quad D_B = -0.007 \text{ meV}, \end{aligned} \quad (19)$$

where we assume that the interactions on the B-sublattice are of the same form as on the A-sublattice, but substantially weaker, $J_A/J_B = D_A/D_B \approx 20$. To demonstrate that the R2–U1 physics persists in the presence of finite D_B we have used MC simulation to calculate the spin structure factor. Once again, the 4FPP associated with the R2–U1 spin liquid remains clearly visible for a range of temperatures [Fig. 3b]. The same will hold for a more general choice of interactions, as long as the anisotropic part of the exchange on the B-sublattice is sufficiently weak; for $D_B \sim D_A$, fluctuations are restricted to the local easy plane, and the R2–U1 physics will be lost.

Quantum effects. The theory of an R2–U1 spin liquid presented above is classical, so it is important to ask what might change once quantum effects are taken into account. A useful point of comparison is quantum spin ice (QSI), where quantum fluctuations leads to tunnelling between different spin configurations satisfying the “ice rules” constraint Eq. (1). This tunnelling, which occurs on loops of spins, introduces a fluctuating magnetic field \mathbf{B} , and the result, at $T = 0$, is a QSL described by a the deconfined phase of a $U(1)$ quantum lattice gauge theory [6–15]. However it is important to note that the temperature scale associated with this QSL is three orders of magnitude smaller than the range of temperatures over which Eq. (1) holds [15]. Moreover, since the $U(1)$ QSL is gapless, any finite temperature immediately restores classical correlations at long length scales [8]. As a consequence, the spin structure factor $S(\mathbf{q})$ continues to be dominated by pinch-point singularities of the form Eq. (16), down to the lowest temperatures studied [13].

The quantum limit of R2–U1 gauge theories has already been studied as a continuum field theory, and is qualitatively very similar to QSI [26, 28, 50]. The lowest lying excitations are gapless emergent photons which modify, but do not eliminate, the singular features observed in scattering [8, 50]. The microscopic study of quantum effects in Eq. (6) lies outside the scope of this Letter. However we anticipate that coherent gauge fluctuations will be confined to an even lower temperature scale than in QSI, by the fact that the magnetic field B_{ij} is an extended object, involving third–order derivatives of A_{ij} [26, 28]. For this reason the classical theory developed here should prove sufficient to interpret experiments searching for an R2–U1 in a BP material.

Summary and perspectives. In this Letter, we have used a combination of analytic field theory and classical Monte Carlo simulation to show how a rank–2 $U(1)$ [R2–U1] spin liquid, a state described by a higher–rank generalisation of electrodynamics, can arise in a pyrochlore magnet with breathing anisotropy and Dzyaloshinskii–Moriya interactions, Eq. (6) [cf. Fig. 1c]. These results provide a concrete starting point for the experimental search for higher–rank gauge theories, and clarify the type of neutron scattering experiment which would be needed to resolve the 4–fold pinch points (4FPP) of a R2–U1 spin liquid [cf. Fig. 3].

This work opens a number of interesting perspectives. On the experimental side, we identify Yb based breathing–pyrochlore materials as potential candidates for a R2–U1 spin liquid state. On the theoretical side, determining the quantum ground state of Eq. (6), should ultimately prove tractable, since breathing anisotropy provides a natural control parameter for both perturbative [71, 72] and variational approaches [90]. And, while the model studied here does not correspond to a fracton stabilizer code upon Higgsing [38, 39], the parital–confinement mechanism used to eliminate selected components of the tensorial electric field is very versatile, and easily adapted to generate other versions of R2–U1 theory [91].

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