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.
tetrahedra have a different size rochlore (BP) lattice, for which A– and B–sublattice turbded by weak DM interactions, on a “breathing” py-
described by a tensor field to show how the spins in a frustrated magnet can be
its electric field B, which in turn implies the form of the associated magnetic
These constraints determine the symmetry of the R2–U1 gauge field
\[ A_{ij} \rightarrow A_{ij} + \partial_i \lambda_j + \partial_j \lambda_i + \gamma \delta_{ij}, \]
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).

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 Definitions of the bond–dependent vectors \( \hat{d}_{ij} \) [53–56] are given in the Supplemental Material [cf. Fig. 1a]. 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 m_{\alpha} which transform as irreducible representations of the lattice symmetry [29, 57, 58]. In this basis [59], the Hamiltonian becomes
\[ H = \frac{1}{2} \sum_{\text{tet} \in A,X} a_{A,\lambda} m_{\lambda}^2 + \frac{1}{2} \sum_{\text{tet} \in B,X} a_{B,\lambda} m_{\lambda}^2, \]
where \( X \) runs over irreps of the group \( T_d \), i.e. \{A_2, E, T_2, T_{1+}, T_{1–}\}, with the fields m_{\alpha} and the coefficients a_{\alpha} defined in Table I and Table II of the Supplementary 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
\[ J_A = J_B , D_A = D_B = 0, \]
we find
\[ 0 < a_{A_2} = a_{E} = a_{T_2} = a_{T_1–} < a_{T_{1+}} . \]
It follows that the fields m_{A_2}, m_{E}, m_{T_2}, m_{T_{1–}} are all free to fluctuate in the ground state. We can conveniently collect all of these fields in the rank–2 tensor
\[ \mathbf{E}^{\text{HAF}} = \mathbf{E}^{\text{HAF}}_{\text{sym}} + \mathbf{E}^{\text{HAF}}_{\text{antisym}} + \mathbf{E}^{\text{HAF}}_{\text{trace}} \]
where
\[ \mathbf{E}^{\text{HAF}}_{\text{sym}} = \begin{pmatrix} \frac{2}{\sqrt{3}} m_{E} & m_{T_{1–}} & m_{T_{1+}} \\ m_{T_{1–}} & -\frac{1}{\sqrt{3}} m_{E} & m_{T_{1+}} \\ m_{T_{1+}} & m_{T_{1–}} & \frac{1}{\sqrt{3}} (m_{E} + m_{T_{1+}}) \end{pmatrix}, \]
(11)
\[ (\mathbf{E}^{\text{HAF}}_{\text{antisym}})_{ij} = -\varepsilon_{ijk} m_{T_{2}}, \quad (\mathbf{E}^{\text{HAF}}_{\text{trace}})_{ij} = -\delta_{ij} \sqrt{\frac{2}{3}} m_{A_2}. \]
The requirement of the continuity of the fields m_{\alpha} [57] imposes the conditions
\[ \frac{2}{\sqrt{3}} \left[ \frac{1}{2} \partial_x m_{E} + \frac{1}{2} \partial_y m_{E} \right] - \left[ \frac{1}{2} \partial_x m_{T_{1–}} + \frac{1}{2} \partial_y m_{T_{1–}} \right] - \left[ \frac{1}{2} \partial_x m_{T_{1+}} + \frac{1}{2} \partial_y m_{T_{1+}} \right] \]
\[ - \sqrt{\frac{2}{3}} \nabla m_{A_2} + \nabla \times m_{T_2} = 0. \]
(13)
We obtain exactly the same constraint if we substitute \( \mathbf{E}^{\text{HAF}} \) in Eq. (4), implying that HAF automatically satisfies one of the two constraints defining the R2–U1 spin liquid [66].
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 $E_{\text{HAF}}^\text{antisym}$ and $E_{\text{HAF}}^\text{trace}$ from the ground state, something which can be accomplished by opening gaps to the fields $m_T$ and $m_A$. For the BP model, Eq. (6), this is achieved by any parameter set for which

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

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

$$a_{E,A} = a_{T,A,A} < a_{B,A} < a_{T,B,A} < a_{T,A,A}, \quad (15)$$

which implies that only the fields $m_E$ and $m_{T\perp}$ 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 $m_A$ and $m_T$ can now be set identically equal to zero. When expressed in terms of the remaining tensor field $E_{\text{HAF}}^\text{sym}$, 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]

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

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 field $E_{ij}$ [50]

$$\langle E_{ij}(q)E_{kl}(-q) \rangle \propto \frac{1}{2} \left( \delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk} \right) + \frac{q_iq_jq_kq_l}{q^4}$$

$$- \frac{1}{2} \left( \delta_{ik} \frac{q_j q_l}{q^2} + \delta_{il} \frac{q_j q_k}{q^2} + \delta_{jk} \frac{q_i q_l}{q^2} + \delta_{jl} \frac{q_i q_k}{q^2} \right)$$

$$- \frac{1}{2} \left( \delta_{ij} - \frac{q_i q_j}{q^2} \right) \left( \delta_{kl} - \frac{q_k q_l}{q^2} \right). \quad (17)$$

The three–dimensional structure of the correlation $\langle E_{xy}(q)E_{xy}(-q) \rangle$ is illustrated in Fig. 1. In the $[hk0]$ plane, correlations exhibit a conventional 2–fold pinch point, comparable to that found in spin ice [Fig. 1b]. However in the perpendicular $[h0k]$ plane, we observe a 4–fold pinch point (4FPP) [Fig. S1], which unambiguously distinguishes R2–U1 electrodynamics 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 $q \rightarrow 0$, 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 $\eta \sim |D_A|/k_BT$, 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 $q = W$ (i.e. corners of the Brillouin zone) [69]. In contrast, a finite value of $D_A > 0$ leads to a continuous phase transition into a
state with $q = 0$, 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 $S^{\alpha \beta}(q) = \langle S^\alpha(q)S^\beta(-q) \rangle$. On general grounds [50], $S^{\alpha \beta}(q)$ is expected to bear witness to the singularity in Eq. (17). But exactly how 4FPs would manifest themselves in experiment remains an open question. In Fig. 3 we present simulation results for $S^{\alpha \beta}(q)$ 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. S1].

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 Ba$_3$Yb$_2$Zn$_5$O$_{11}$ [79, 80, 86, 88], where A–tetrahedra are estimated to have the coupling parameters $J_A \approx 0.57$ meV, $D_B \approx -0.16$ 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 Ba$_3$Yb$_2$Zn$_5$O$_{11}$, while less well understood, appear to be orders of magnitude smaller [80, 86]. Thus, while it seems plausible that Ba$_3$Yb$_2$Zn$_5$O$_{11}$ could realise a R2–U1 spin liquid, this may occur at temperatures too low to measure.

The encouraging example of Ba$_3$Yb$_2$Zn$_5$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 concreteness, we consider a parameter set:

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

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 $B$, and the result, at $T = 0$, is a QSL described by 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 $S(q)$ 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 $B_{ij}$

FIG. 3. Spin structure factor found in MC simulation of the BP model, Eq. (6), showing 4-fold pinch points (4FPs) characteristic of a R2–U1 spin liquid. (a) Correlations in the [100] 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 Ba$_3$Yb$_2$Zn$_5$O$_{11}$, Eq. (19), $T = 252$ mK.
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. S1]. 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 fractionalizer 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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* han.yan@oist.jp

[1] P. W. Anderson, Science 177, 393 (1972).
[2] P. Anderson, Materials Research Bulletin 8, 153 (1973).
[3] L. Balents, Nature (London) 464, 199 (2010).
[4] L. Savary and L. Balents, Reports on Progress in Physics 80, 016502 (2017).
[5] Y. Zhou, K. Kanoda, and T.-K. Ng, Rev. Mod. Phys. 89, 025003 (2017).
[6] M. Hermele, M. P. A. Fisher, and L. Balents, Phys. Rev. B 69, 064404 (2004).
[7] A. Banerjee, S. V. Isakov, K. Danile, and Y. B. Kim, Phys. Rev. Lett. 100, 047208 (2008).
[8] O. Benton, O. Sikora, and N. Shannon, Phys. Rev. B 86, 075154 (2012).
[9] L. Savary and L. Balents, Phys. Rev. Lett. 108, 037202 (2012).
[10] N. Shannon, O. Sikora, F. Pollmann, K. Penc, and P. Fulde, Phys. Rev. Lett. 108, 067204 (2012).
[11] Z. Hao, A. G. R. Day, and M. J. P. Gingras, Phys. Rev. B 90, 214430 (2014).
[12] M. J. P. Gingras and P. A. McClarty, Reports on Progress in Physics 77, 056501 (2014).
[13] Y. Kato and S. Onoda, Phys. Rev. Lett. 115, 077202 (2015).
[14] G. Chen, Phys. Rev. B 96, 195127 (2017).
[15] C.-J. Huang, Y. Deng, Y. Wan, and Z. Y. Meng, Phys. Rev. Lett. 120, 167202 (2018).
[16] H. D. Zhou, C. R. Wiebe, J. A. Janik, L. Balicas, Y. J. Yo, Y. Qiu, J. R. D. Copley, and J. S. Gardner, Phys. Rev. Lett. 101, 227204 (2008).
[17] K. A. Ross, L. Savary, B. D. Gaulin, and L. Balents, Phys. Rev. X 1, 021002 (2011).
[18] T. Fennell, M. Kenzelmann, B. Roessli, M. K. Haas, and R. J. Cava, Phys. Rev. Lett. 109, 017201 (2012).
[19] K. Kimura, S. Nakatsuji, J.-J. Wen, C. Broholm, M. Stone, E. Nishibori, and H. Sawa, Nature Communications 4, 134 (2013).
[20] R. Sibille, E. Lhotel, V. Pompjakushin, C. Baines, T. Fennell, and M. Kenzelmann, Phys. Rev. Lett. 115, 097202 (2015).
[21] J.-J. Wen, S. M. Koochpayeh, K. A. Ross, B. A. Trump, T. M. McQueen, K. Kimura, S. Nakatsuji, Y. Qiu, D. M. Pajerowski, J. R. D. Copley, and C. L. Broholm, Phys. Rev. Lett. 118, 107206 (2017).
[22] J. D. Thompson, P. A. McClarty, D. Prabhakaran, I. Cabrera, T. Guidi, and R. Coldea, Phys. Rev. Lett. 119, 057203 (2017).
[23] R. Sibille, N. Gauthier, H. Yan, M. Cionagia Hatnean, J. Ollivier, B. Winn, U. Fulges, G. Balakrishnan, M. Kenzelmann, N. Shannon, and T. Fennell, Nature Physics 14, 711715 (2018).
[24] B. Gao, T. Chen, D. W. Tam, C.-L. Huang, K. Sasmal, D. T. Adroja, F. Ye, H. Cao, G. Sala, M. B. Stone, C. Baines, J. A. T. Verezhak, H. Hu, J.-H. Chung, X. Xu, S.-W. Cheong, M. Nallaiyan, S. Spagna, M. B. Maple, A. H. Nevidomskyy, E. Morosan, G. Chen, and P. Dai, Nature Physics 15, 1052 (2019).
[25] C. Xu, Phys. Rev. B 74, 224433 (2006).
[26] M. Pretko, Phys. Rev. B 96, 035119 (2017).
[27] A. Rasmussen, Y.-Z. You, and C. Xu, arXiv e-prints , arXiv:1601.08235 (2016), arXiv:1601.08235 [cond-mat.str-el].
[28] M. Pretko, Phys. Rev. B 95, 115139 (2017).
[29] O. Benton, L. D. C. Jaubert, H. Yan, and N. Shannon, Nature Communications 7, 11572 (2016).
[30] M. Pretko, Phys. Rev. D 96, 024051 (2017).
[31] M. Pretko and L. Radzihovsky, Phys. Rev. Lett. 120, 195301 (2018).
[32] A. Gromov, Phys. Rev. Lett. 122, 076403 (2019).
[33] C. Chamon, Phys. Rev. Lett. 94, 040402 (2005).
This system of equations can also be viewed as three independent copies of a U(1) gauge theory [64].

G.-W. Chern, “Pyrochlore antiferromagnet with anti-symmetric exchange interactions: critical behavior and order from disorder,” (2010), arXiv:1008.3038 [cond-mat.str-el].
Supplemental Material

RANK–2 $U(1)$ GAUGE THEORY: ELECTROSTATICS

Here, following Ref. [25, 26], we derive the relationship between electric, magnetic and gauge fields within the rank–2 $U(1)$ $[R2–U1]$ electrodynamics considered in the main text. This section focuses on the classical electrostatics which is realized in our work. The next section will focus on the quantum dynamics of the theory, which is beyond the scope of this work but nevertheless essential for future developments.

Our starting point is an electric field described by a symmetric, traceless rank–2 tensor,

$$E_{ij} = E_{ji}, \quad E_{ii} = 0.$$  \hspace{1cm} (S1)

Here we do not distinguish super and subscript since we are dealing with spacial indices.

The low energy sector of the electric field has vanishing vector charge, and is traceless,

$$\partial_i E_{ij} = 0$$  \hspace{1cm} (S2)

Here we keep all indices as subscripts but still use the Einstein summation rule. The proper rank-2 tensor with the proper Gauss law as a classical spin liquid system is achieved in this work.

RANK–2 $U(1)$ GAUGE THEORY: DYNAMICS

A quantum spin liquid requires quantum dynamics in addition to the emergent Gauss law. Broadly speaking, the dynamics play the role of $B^2$ term in electrodynamics. They are to tunnel different classical spin liquid states between each other, leading to a long-range entangled quantum ground state and gapless photon excitations. This section explains how to derive such terms and also their implication on mobility of electric charges (fractons).

As in conventional electrodynamics, the conjugate of $E$ is the rank-two gauge field $A$, which also has to be symmetric to match the degrees of freedom,

$$A_{ij} = A_{ji}. \hspace{1cm} (S3)$$

These two conditions determine the form of gauge transformation. Consider a wave-function

$$|\Psi(A)\rangle. \hspace{1cm} (S4)$$

We take a low energy configuration of $E$ obeying Eq. (S2) and construct a symmetrized operator that is identical to zero to act upon the wave-function

$$-i(\lambda_i \partial_i E_{ij} + \lambda_j \partial_j E_{ij}) |\Psi(A)\rangle = 0.$$  \hspace{1cm} (S5)

By integration by parts and assuming vanishing boundary terms, we have

$$i(\partial_i \lambda_j + \partial_j \lambda_i) E_{ij} |\Psi(A)\rangle = 0.$$  \hspace{1cm} (S6)

Since $E_{ij}$ conjugates with $A_{ij}$, it generates a transformation of $A$. Thus

$$i(\partial_i \lambda_j + \partial_j \lambda_i) E_{ij} |\Psi(A)\rangle = |\Psi(A + \nabla \otimes \lambda + (\nabla \otimes \lambda)^T)\rangle - |\Psi(A)\rangle = 0.$$  \hspace{1cm} (S7)

That is, the low energy sector wave-function is invariant under gauge transformation

$$A + \nabla \otimes \lambda + (\nabla \otimes \lambda)^T, \quad \text{i.e.,} \quad A_{ij} \rightarrow A_{ij} + \partial_i \lambda_j + \partial_j \lambda_i.$$  \hspace{1cm} (S8)

Similarly, the traceless condition

$$-i\gamma \delta_{ij} E_{ij} |\Psi(A)\rangle = 0.$$  \hspace{1cm} (S9)

leads to another gauge symmetry

$$A_{ij} \rightarrow A_{ij} + \gamma \delta_{ij}.$$  \hspace{1cm} (S10)
Finally, the magnetic field is obtained by finding the simplest gauge-invariant quantity. In this case, it has to have three derivatives acting on the gauge field,
\[ B_{ij} = \frac{1}{2} [\epsilon_{jab} (\partial_a \partial_k \partial_i A_{bk} - \partial_a \partial^2 A_{bi})] + \epsilon_{iab} (\partial_a \partial_k \partial_j A_{bk} - \partial_a \partial^2 A_{bj})]. \] 
(S11)

Finally, the Gauss law, the traceless and symmetric conditions of the electric field can be used to derive:
\[ \int dv \rho = 0 \] 
(S12)

\[ \int dv \mathbf{e} \times \mathbf{E} = - \int dv \epsilon_{ijk} E_{jk} = 0 \] 
(S13)

\[ \int dv \mathbf{e} \cdot \mathbf{E} = - \int dv E_{ii} = 0 \] 
(S14)

In this case, a vector charge excitation is fully fractonic, i.e., it cannot move in any direction of the system. Further details of the phenomenology of R2–U1 phases can be found in Refs. [26, 28].

**DERIVATION OF EFFECTIVE FIELD THEORY**

We show how a rank 2 tensor electric field, satisfying the constraint required for R2–U1 electrodynamics [Eqs. (1,2)], can be derived from a breathing pyrochlore lattice model [Eq. (6)]. The pattern of this derivation closely follows Refs. [29, 57, 58].

Our starting point is the breathing pyrochlore lattice with a spin on each of its sites, and nearest neighbor interactions between the spins. “Breathing” means the lattice is bi-partitioned into A- and B-tetrahedra [Fig. (1)], and each type of tetrahedron has its own interactions.

The model that hosts a rank-2 spin liquid has breathing Heisenberg antiferromagnetic interactions on both the A- and B-tetrahedra, and negative Dzyaloshinskii-Moriya (DM) interactions on A-tetrahedra only. The Hamiltonian for the model is
\[ H = \sum_{\langle ij \rangle \in A} \left[ J_A S_i \cdot S_j + D_A \hat{d}_{ij} \cdot (S_i \times S_j) \right] + \sum_{\langle ij \rangle \in B} \left[ J_B S_i \cdot S_j + D_B \hat{d}_{ij} \cdot (S_i \times S_j) \right]. \] 
(S15)

where \( \langle ij \rangle \in \text{A(B)} \) denotes nearest neighbour bonds belonging to the A(B)-tetrahedra. The sites 0, 1, 2, 3 are at positions relative to the center of an A-tetrahedron
\[ r_0 = \frac{a}{8} (1, 1, 1), \quad r_1 = \frac{a}{8} (1, -1, -1), \quad r_2 = \frac{a}{8} (-1, 1, -1), \quad r_3 = \frac{a}{8} (-1, -1, 1), \] 
(S16)

where \( a \) is the length of the unit cell. Vectors \( \hat{d}_{ij} \) are bond dependent, defined in accordance with Ref [53, 56, 86]:
\[ \hat{d}_{01} = \frac{(0, -1, 1)}{\sqrt{2}}, \quad \hat{d}_{02} = \frac{(1, 0, -1)}{\sqrt{2}}, \quad \hat{d}_{03} = \frac{(-1, 1, 0)}{\sqrt{2}}, \] 
\[ \hat{d}_{12} = \frac{(-1, -1, 0)}{\sqrt{2}}, \quad \hat{d}_{13} = \frac{(1, 0, 1)}{\sqrt{2}}, \quad \hat{d}_{23} = \frac{(0, -1, -1)}{\sqrt{2}}. \] 
(S17)

Equivalently, this model can be written in a standard matrix-exchange form for a breathing-pyrochlore lattice model as
\[ H = \sum_{\langle ij \rangle \in A} S_i^x J_A^{x} S_j^x + \sum_{\langle ij \rangle \in B} S_i^x J_B^{x} S_j^y \] 
(S18)

where \( J_{A,B} \) is a three-by-three matrix that couples spins on sub-lattice sites \( i,j \) whose bond belongs to A-tetrahedra, and \( J_B \) is the coupling matrix for B-tetrahedra. In the case of \( D_B = 0 \) that we are interested in, \( J_B \) is identical for any pair of \( i,j \),
\[ J_B = \begin{bmatrix} J_B & 0 & 0 \\ 0 & J_B & 0 \\ 0 & 0 & J_B \end{bmatrix}. \] 
(S19)
Matrices $J_{A,ij}$ are bond dependent and related to each other by the lattice symmetry. Their values are

\[
\begin{align*}
J_{A,01} & = \begin{bmatrix}
J_A & D_A/\sqrt{2} & D_A/\sqrt{2} \\
-D_A/\sqrt{2} & J_A & 0 \\
-D_A/\sqrt{2} & 0 & J_A
\end{bmatrix}, \\
J_{A,02} & = \begin{bmatrix}
J_A & -D_A/\sqrt{2} & 0 \\
D_A/\sqrt{2} & J_A & D_A/\sqrt{2} \\
0 & -D_A/\sqrt{2} & J_A
\end{bmatrix}, \\
J_{A,03} & = \begin{bmatrix}
J_A & 0 & -D_A/\sqrt{2} \\
0 & J_A & -D_A/\sqrt{2} \\
D_A/\sqrt{2} & D_A/\sqrt{2} & J_A
\end{bmatrix}, \\
J_{A,12} & = \begin{bmatrix}
J_A & 0 & D_A/\sqrt{2} \\
0 & J_A & -D_A/\sqrt{2} \\
-D_A/\sqrt{2} & D_A/\sqrt{2} & J_A
\end{bmatrix}, \\
J_{A,13} & = \begin{bmatrix}
J_A & D_A/\sqrt{2} & 0 \\
-D_A/\sqrt{2} & J_A & D_A/\sqrt{2} \\
0 & -D_A/\sqrt{2} & J_A
\end{bmatrix}, \\
J_{A,23} & = \begin{bmatrix}
J_A & -D_A/\sqrt{2} & D_A/\sqrt{2} \\
D_A/\sqrt{2} & J_A & 0 \\
-D_A/\sqrt{2} & 0 & J_A
\end{bmatrix}.
\end{align*}
\]

(S20)

The spin degrees of freedom on each tetrahedron can be rewritten in terms of fields forming the irreducible representations of the lattice symmetry,

\[
m_{A_2}, \quad m_E, \quad m_T, \quad m_{T_+}, \quad m_{T_-},
\]

(S21)

whose definition can be found in Table I. They are linear combinations of the spin degrees of freedom, allowing for a fully quadratic Hamiltonian:

\[
\mathcal{H} = \frac{1}{2} \sum_X a_{X,A} m_{X,A}^2 + \frac{1}{2} \sum_X a_{X,B} m_{X,B}^2.
\]

(S22)

where $X$ runs over irreps of the group $T_d$, i.e. $\{A_2, E, T_2, T_1, T_{1-}\}$ as listed in Eq. (S21), and the subscript $A,B$ denotes on which type of tetrahedra they are defined. The coefficients $a_X$ are listed in Table II.

For the couplings in Eq. (S15), we have on A-tetrahedra

\[
a_{A_2} = -J_A - 4D_A/\sqrt{2},
\]

(S23)

\[
a_{T_2} = -J_A - 2D_A/\sqrt{2},
\]

(S24)

\[
a_{T_+} = 3J_A,
\]

(S25)

\[
a_{T_-} = a_{E} = -J_A + 2D_A/\sqrt{2},
\]

(S26)
such that Eq. (S33) becomes

\[ \sum_x m_X^2 = 1 \]  \hspace{1cm} (S31)

and on B-tetrahedra

\[ a_{A_2,B} = a_{E,B} = a_{T_2,B} = a_{T_1-,B} = -J_B, \]
\[ a_{T_4,B} = 3J. \]  \hspace{1cm} (S27)

For \( J_A, J_B > 0 \) and \( D_A < 0 \), these parameters are in order

on A-tetrahedra: \( a_{E,A} = a_{T_1-,A} < a_{A_2,A}, a_{T_2,A}, a_{T_4,A} \), \hspace{1cm} (S29)
on B-tetrahedra: \( a_{A_2,B} = a_{E,B} = a_{T_2,B} = a_{T_1-,B} < a_{T_4,B} \), \hspace{1cm} (S30)

which plays the central role of dictating the low energy physics.

The irreducible representation fields are subject to constraints arising from fixed spin length

\[ \frac{\partial_x m_X^2}{\partial y m_X^2} = 0 \]  \hspace{1cm} (S32)

and on A-tetrahedra

\[ m_{A_2} = 0; \]
\[ m_{T_4} = 0; \]
\[ m_{T_1+} = 0 \]  \hspace{1cm} (S33)

\[ E_{ij} = \frac{2}{\sqrt{3}} \begin{bmatrix} m_{E}^x & m_{E}^y & m_{E}^z \\ m_{T_4}^x & m_{T_4}^y & m_{T_4}^z \\ m_{T_1-}^x & m_{T_1-}^y & m_{T_1-}^z \end{bmatrix} \]  \hspace{1cm} (S34)

such that Eq. (S33) becomes

\[ \partial_x E_{ij} = 0 \]  \hspace{1cm} (S35)
FIG. S1. Structure of the 4-fold pinch point (4FPP) associated with rank-2 $U(1)$ [R2–U1] gauge theory. (a) Prediction of R2–U1 theory for the correlation function $\langle E_{xy}(q)E_{xy}(-q) \rangle$, on a surface of fixed $|q|$ near to a Brillouin zone center. (b) Exploded view, showing a 2-fold pinch point in the $[0kl]$ plane. (c) Perpendicular cut, showing a 4FPP in the $[hk0]$ plane.

with symmetric and traceless conditions

$$E_{ij} = E_{ji}, \quad \text{Tr} \mathbf{E} = 0 \quad (S36)$$

by the definition of $E_{ij}$. Hence a rank-2, traceless, vector charged magnetic field emerges at the low-energy sector from the microscopic model of Eq. (S15).

Equation (S36) constrains the form of correlations functions of $\langle E_{ij}(q)E_{kl}(-q) \rangle$, in the same spirit as how the two-in-two-out condition constrains the spin-spin correlation of spin ice. It is, however, in a more complicated form. The explicit results for the traceful scalar-charged and vector-charged versions of R2–U1 are discussed in detail in Ref. [50]. The vector-charge field correlation is

$$\langle E_{ij}(q)E_{kl}(-q) \rangle \propto \frac{1}{2}(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}) + \frac{q_iq_jq_kq_l}{q^4}$$

(S37)

In close analogy, we derive the correlation function of our traceless vector-charged model by deducting the trace,

$$\langle E_{ij}(q)E_{kl}(-q) \rangle \propto \frac{1}{2}(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}) + \frac{q_iq_jq_kq_l}{q^4}$$

(S38)

$$- \frac{1}{2} \left( \delta_{ik} \frac{q_iq_l}{q^2} + \delta_{jl} \frac{q_iq_k}{q^2} + \delta_{il} \frac{q_jq_k}{q^2} + \delta_{jk} \frac{q_iq_l}{q^2} \right)$$

$$- \frac{1}{2} \left( \delta_{ij} - \frac{q_iq_j}{q^2} \right) \left( \delta_{kl} - \frac{q_kq_l}{q^2} \right),$$

which encodes a singularity at $q \to 0$. Different choices of the components $E_{ij}$ and $E_{kl}$ show different patterns. A few representative ones can be found in Figs. S1,S2.

FIG. S2. Different components of correlation function $\langle E_{ij}(q)E_{kl}(-q) \rangle$ in $q_x - q_y$ plane, calculated from Eq. (S38).

Fig. S2b,S2c have the four-fold pinch-point (4FPP) singularity, which differentiates the rank-2 gauge theories uniquely from the conventional $U(1)$ gauge theory. It is the key signature to be looked for in experiments.
ALTERNATIVE FORMS OF R2–U1 SPIN LIQUID

In the main text we have shown how a classical spin liquid described by a symmetric, traceless R2–U1 gauge theory, descends from the $U(1) \times U(1) \times U(1)$ spin liquid found in the classical Heisenberg Antiferromagnet (HAF) on a pyrochlore lattice. The approach taken is very versatile, and by tuning the Hamiltonian, one can also obtain other forms of R2–U1 spin liquid.

Notice that the diagonal and off-diagonal components of $E_{\text{HAF}}^{\text{sym}}$ come from different irreps $m_E$ and $m_{T_{1-}}$. In the most general Hamiltonian (Eq.S22), these two irreps have their individually tunable coefficients $a_E$ and $a_{T_{1-}}$. So the symmetric part of $E_{\text{HAF}}$ can be decomposed into three components

$$E_{\text{trace}}^\text{HAF} + E_{\text{sym-diagonal}}^\text{HAF} + E_{\text{symm-off-diagonal}}^\text{HAF},$$

and each component can be individually tuned to be active or suppressed by choosing the proper Hamiltonian. The vector-charged Gauss law is unaffected. This allows us to build a variety of rank-2 U(1) gauge theories, including a "hollow" version with vanishing diagonal terms [39].

PREDICTIONS FOR NEUTRON SCATTERING

The 4FPP is a unique pattern that differentiates the R2–U1 from vector $U(1)$ gauge theory, which only has the conventional two-fold pinch points. The 4FPPs are most unambiguously presented in the correlation function of the irrep fields as discussed in the main text. These correlation functions are, however, not directly accessible in experiment.

In magnetism, neutron scattering is widely used to measure the spin-spin correlation of the form

$$S(q) = \sum_{\alpha,\beta,i,j} \left( \delta_{\alpha\beta} - \frac{q^\alpha q^\beta}{q^2} \right) \langle S_\alpha^i(q)S_\beta^j(-q) \rangle,$$

where $\alpha, \beta = x, y, z$ are spin-component indices and $i, j = 0, 1, 2, 3$ are sub-lattice site indices.

Furthermore, with neutrons polarized in direction of unit vector $\hat{v}$ perpendicular to the scattering plane, one can measure the spin-flip (SF) channel neutron scattering defined by

$$S(q)_{\text{SF}} = \sum_{\alpha,\beta,i,j} (v_\alpha^i v_\beta^j) \langle S_\alpha^i(q)S_\beta^j(-q) \rangle,$$

where

$$\hat{v}_\perp = \frac{\hat{v} \times q}{|\hat{v} \times q|}.$$

One can also measure the non-spin-flip (NSF) channel defined by

$$S(q)_{\text{NSF}} = \sum_{\alpha,\beta,i,j} (v^\alpha v^\beta) \langle S_\alpha^i(q)S_\beta^j(-q) \rangle$$

Here we show the spin structure factor of the $[h0k]$ and $[hhk]$ plane, with zoomed-in view of the 4FPPs.
FIG. S3. 4-Fold Pinch Points (4FPPs) in spin structure factor in the \([h0k]\) and \([hhk]\) plane of momentum space of the model [Eq. (6)] from MC simulations. The exchange parameters are from the idealized theoretical case \(J_A = J_B = 1.0, D_A = -0.01, D_B = 0.0\), at \(T = 2.5 \times 10^{-3} J_A\). (a) Total structure factor in \([h0k]\) plane. (b) Non-spin-flip (NSF) channel in \([h0k]\) plane. (c) Spin-flip (SF) channel in \([h0k]\) plane. (d) Enlarged 4FPP in \([h0k]\) plane. (a) Total structure factor in \([hhk]\) plane. (b) Non-spin-flip (NSF) channel in \([hhk]\) plane. (c) Spin-flip (SF) channel in \([hhk]\) plane. (d) Enlarged 4FPP in \([hhk]\) plane. The 4FPPs can be clearly observed in the SF channel, centered on \([0, 0, 2]\) (and points related by symmetry), but weaker than in the \([h0k]\) plane.

FIG. S4. 4-Fold Pinch Points (4FPPs) in spin structure factors in the \([h0k]\) and \([hhk]\) planes of momentum space of the model [Eq. (6)] from MC simulations. The exchange parameters are from the experimental case Eq. (19) at \(T = 252\) mK. (a) Total structure factor in \([h0k]\) plane. (b) Non-spin-flip (NSF) channel in \([h0k]\) plane. (c) Spin-flip (SF) channel in \([h0k]\) plane. (d) Enlarged 4FPP in \([h0k]\) plane. (a) Total structure factor in \([hhk]\) plane. (b) Non-spin-flip (NSF) channel in \([hhk]\) plane. (c) Spin-flip (SF) channel in \([hhk]\) plane. (d) Enlarged 4FPP in \([hhk]\) plane. The 4FPPs can be observed in the SF channel, centered on \([0, 0, 2]\) (and points related by symmetry), but weaker than in the \([h0k]\) plane.

TEMPERATURE EVOLUTION OF 4–FOLD PINCH POINT INTO A CONVENTIONAL 2–FOLD PINCH POINT

The cross-over from the HAF phase to the R2–U1 phase, with decreasing temperature, is manifested in the structure factor as a cross-over between a conventional, 2–fold, pinch point and the 4–fold pinch point (4FPP) characteristic of an R2–U1 gauge theory [50]. To provide further quantitative details of this cross-over, here we present an analysis of correlations based on a coarse-grained field theory.

As shown above, the \(U(1) \times U(1) \times U(1)\) spin liquid of the Heisenberg model can be described in terms of a non-symmetric matrix \(E\), with a Gauss’ law applied to each column. The traceless R2-U(1) spin liquid is described in the same way, but with the constraint that \(E\) be symmetric and traceless.

The following effective theory captures both cases:

\[
Z = \prod_{\mu, \nu} dE_{\mu\nu} \exp(-\beta H_{\text{eff}}[E_{\mu\nu}])
\]

\[
\beta H_{\text{eff}} = \frac{\lambda}{2} \int d^3 r \left( \sum_{\mu, \nu} E_{\mu\nu}^2 + \delta \sum_\nu \left[ \sum_\mu \partial_\mu E_{\mu\nu} \right]^2 + \eta \left[ \frac{1}{3} \text{Tr}[E]^2 + \frac{1}{2} (E_{xy} - E_{yx})^2 + \frac{1}{2} (E_{xz} - E_{zx})^2 + \frac{1}{2} (E_{yz} - E_{zy})^2 \right] \right)
\]

where the integral in Eq. \((S44)\) is taken independently over all components of the matrix \(E\) and \(\beta = 1/T\) is the inverse temperature.

The limit \(\delta \to \infty, \eta \to 0\), captures the Heisenberg model spin liquid, with Gauss’ law enforced on every column of \(E\) and no correlations between columns. The limit \(\delta \to \infty, \eta \to \infty\) captures the R2-U(1) spin liquid, with \(E\) forced to be symmetric and traceless and still obeying Gauss’ law for each column.
FIG. S5. Evolution of the correlation function $\langle E^{yx}(q)E^{yx}(-q) \rangle$ [Eq. (S47)] from a 2-fold pinch point to a 4-fold pinch point as we tune from the Heisenberg to R2-U(1) spin liquids by increasing the parameter $\eta$ [Eq. (S45)]. In terms of our microscopic model, this may be viewed either as decreasing the temperature at fixed, small, $|D_A|$ or as increasing $|D_A|$ at fixed small temperature.

In terms of the parameters of the microscopic model:

$$\delta \sim \beta J, \quad \eta \sim \beta |D_A|.$$ (S46)

since $\delta$ is the coefficient enforcing the Gauss’ law (which is generated by $J$) and $\eta$ is the coefficient enforcing the symmetric and traceless conditions (which are generated by $D_A$).

We can study the crossover from the Heisenberg to R2-U(1) spin liquids by first taking $\delta \to \infty$ and observing the behavior as a function of $\eta$. The crossover can be illustrated by calculating the correlation function

$$\langle E^{yx}(q)E^{yx}(-q) \rangle\text{ which should have a 2-fold pinch point in the Heisenberg limit and a 4-fold pinch point in the R2-U(1) limit.}$$

Calculating the correlation function from Eqs. (S44)-(S45) and taking the limit $\delta \to \infty$, gives us

$$\langle E^{yx}(q)E^{yx}(-q) \rangle = \frac{f(q,\eta)}{2\lambda q^4(1 + \eta)(2 + \eta)(3 + 2\eta)}$$ (S47)

where

$$f(q,\eta) = 4q^2_2(1 + \eta)(3 + 2\eta) + q^2_2(q^2_2 + q^2_z)^2(2 + \eta)^2(3 + 2\eta) + 2q^2_2q^2_y(1 + \eta)(2 + \eta)(3 + \eta) + q^2_2q^2_z(3 + 2\eta)(8 + 8\eta + \eta^2).$$ (S48)

The $\eta \to 0$ limit of Eq. (S47) gives the Heisenberg limit of the correlation function (a 2-fold pinch point)

$$\lim_{\eta \to 0} \langle E^{yx}(q)E^{yx}(-q) \rangle = \frac{1}{\lambda} \left(1 - \frac{q^2_y}{q^2_z}\right).$$ (S49)

Whereas, the $\eta \to \infty$ limit gives the R2-U(1) correlation function (a 4-fold pinch point):

$$\lim_{\eta \to \infty} \langle E^{yx}(q)E^{yx}(-q) \rangle = \frac{1}{2\lambda} \frac{(q^2 - q^2_z)(q^2_z - q^2_y)}{q^4}.\text{ (S50)}$$

Since $\eta \sim \beta |D_A|$, we can see from Eq. (S47) that for a fixed $q$ at any, fixed, finite temperature the evolution as a function of $D_A$, will be smooth, although at small temperatures, a small change in $D_A$ will give a large change in $\eta$ and hence the correlation function. Only at $T = 0$ does the correlation function behave in a singular fashion as a function of $D_A$, but this is not surprising, since the Heisenberg limit has a highly degenerate ground state.

The progression of the correlation function as $\eta$ is increased is given in Fig (S5). This progression can either be seen as decreasing the temperature at fixed, small, $|D_A|$ or as increasing $|D_A|$ at fixed small temperature.
Monte Carlo simulations are performed on systems of classical O(3) spins with $16L^3$ sites, where $L^3$ is the number of cubic unit cells. The spin length is $|S| = 1/2$. To decorrelate the system, we use jointly the heatbath method, over-relaxation and parallel tempering. Thermalization is made in two steps: first a slow annealing from high temperature to the temperature of measurement $T$ during $t_c$ Monte Carlo steps (MCs) followed by $t_c$ MCS at temperature $T$. After thermalization, measurements are done every 10 MCs during $t_m = 10t_c$ MCs. All structure factors have been computed from simulations with $L = 30$ and $t_m = 5 \times 10^5$ MCs. The phase diagram of Fig. 5 has been computed from simulations with $L = 8$ and $t_m = 10^7$ MCs.