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Coulombic Quantum Liquids in Spin-1/2 Pyrochlores

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We develop a non-perturbative “gauge Mean Field Theory” (gMFT) method to study a general effective spin-1/2 model for magnetism in rare earth pyrochlores. gMFT is based on a novel exact slave-particle formulation, and matches both the perturbative regime near the classical spin ice limit and the semiclassical approximation far from it. We show that the full phase diagram contains two exotic phases: a quantum spin liquid and a coulombic ferromagnet, both of which support deconfined spinon excitations and emergent quantum electrodynamics. Phenomenological properties of these phases are discussed.

Amongst the celebrated exotic phases of matter, of particular recent interest are the Quantum Spin Liquids (QSLs) [1]. Behind *seemingly* innocuous defining properties –strong spin correlations, the absence of static magnetic moments, and unbroken crystalline symmetry–, QSLs display the consequences of *extreme* quantum entanglement. These include emergent gauge fields and fractional excitations, which take these states beyond the usual “mean field” paradigm of phases of matter. Not only are these phases challenging to predict and describe, they have also proven very hard to find in the laboratory, rendering their search and discovery even more tantalizing.

A consensual place to look for QSLs is among frustrated magnets [1]. Frustration allows the spins to avoid phases where they are either ordered or frozen, with relatively small fluctuations and correlations between them. Recent experiments have given compelling evidence of a QSL state in certain two-dimensional organic materials [2], but both microscopic and fully consistent phenomenological theories are lacking. By contrast, *classical* spin liquids have been conclusively seen and microscopically understood in the spin ice pyrochlores [3]. This raises the possibility, suggested experimentally [4] and theoretically [5], of QSLs in those rare earth pyrochlores in which spins are non-classical, supported by recent results on $\text{Yb}_2\text{Ti}_2\text{O}_7$ [4]. However, for any material, only detailed, quantitative theory predicting the *type(s)* and properties of QSLs that appear *and* matching experiments can take the physics to the next level.

We take up this challenge here for quantum rare earth pyrochlores. Our analysis confirms that a “ $U(1)$ ” QSL phase exists in the phase diagram (Fig. 1) of a spectrum of real materials, and is furthermore supplemented by another exotic phase, a Coulombic ferromagnet, which contains spinons, but displays non-zero magnetization. We also study the confinement transitions out of these Coulomb phases, which are analogous to “Higgs” transitions [6]. Finally, we discuss experimental signatures of the $U(1)$ QSL, and of the $U(1)$ Coulomb ferromagnet.

The most general nearest-neighbor symmetry-allowed exchange Hamiltonian for spin-1/2 spins (real or effec-

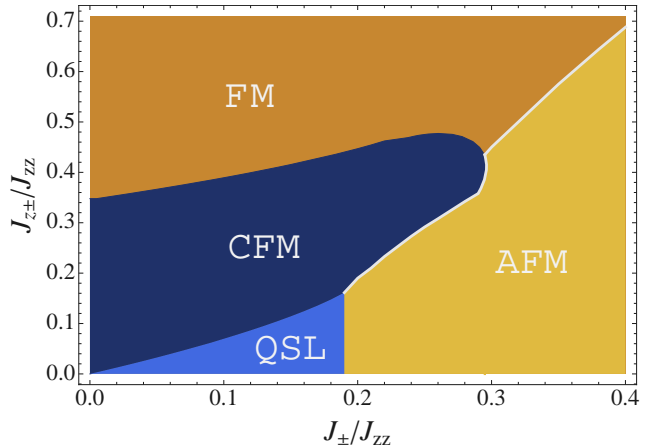


FIG. 1. Gauge mean field phase diagram obtained for $J_{\pm\pm} = 0$ and $J_{zz} > 0$. “QSL”, “CFM”, “FM” and “AFM” denote the $U(1)$ Quantum Spin Liquid, Coulomb Ferromagnet, standard ferromagnet, and standard antiferromagnet, respectively. Phase boundaries with/without white lines indicate discontinuous/continuous transitions in gMFT. Note that the diagram is symmetric in $J_{z\pm} \rightarrow -J_{z\pm}$.

tive) on the pyrochlore lattice is

$$\begin{aligned}
 H = \sum_{\langle ij \rangle} & \left[J_{zz} S_i^z S_j^z - J_{\pm} (S_i^+ S_j^- + S_i^- S_j^+) \right. \\
 & + J_{\pm\pm} [\gamma_{ij} S_i^+ S_j^+ + \gamma_{ij}^* S_i^- S_j^-] \\
 & \left. + J_{z\pm} [S_i^z (\zeta_{ij} S_j^+ + \zeta_{ij}^* S_j^-) + i \leftrightarrow j] \right], \quad (1)
 \end{aligned}$$

where γ is a 4×4 complex unimodular matrix, and $\zeta = -\gamma^*$ [4]. The explicit expression of γ and of the local bases whose components are used in Eq. (1) are given in the Supplementary Material. The first term (we assume in this paper $J_{zz} > 0$), taken alone, gives the highly frustrated classical nearest-neighbor spin ice model, which exhibits an extensive ground state degeneracy of “two-in-two-out” states.

In fact, this model has been studied theoretically in the special case $J_{z\pm} = J_{\pm\pm} = 0$, where it reduces to an “XXZ” model with global XY spin-rotation symmetry

[7]. There, it was shown that for $J_{\pm} \ll J_{zz}$, it is perturbatively equivalent, order by order, to a lattice $U(1)$ gauge theory, with gauge fields that describe the spin configurations *constrained* to the spin ice manifold of ground states. This gauge theory was furthermore argued to exhibit a so-called ‘‘Coulomb phase’’, which corresponds to a $U(1)$ QSL phase. Subsequent numerical simulations [8, 9] verified this prediction. This Coulombic QSL is not only magnetically disordered, but also supports several exotic excitations: spinons (called magnetic monopoles in the spin ice literature), dual ‘‘electric monopoles’’, and an emergent photon. This understanding, however, was limited to the perturbative regime $J_{\pm} \ll J_{zz}$ and considered only the XXZ case. Here we develop a *non-perturbative* method to analyze the full Hamiltonian in Eq. (1).

Non-perturbative theories of QSLs based on ‘‘slave particles’’ have been developed and used extensively in $SU(2)$ invariant $S = 1/2$ Heisenberg and Hubbard models [10]. Generally these approaches work by embedding the Hilbert space on each site in some larger ‘‘spinon’’ one, with a microscopic gauge symmetry which acts to project back to the physical space. QSL phases are found when, in a mean field sense, this microscopic gauge symmetry is incompletely broken in the ground state. Here, we follow the spirit but not the letter of these approaches, by introducing redundant degrees of freedom not for each spin but for each tetrahedron of the pyrochlore lattice. This new slave particle representation is, like the aforementioned standard ones, formally exact, but additionally naturally describes the Coulombic QSL found before in the perturbative analysis, when that limit is taken. It also has the added advantage that, unlike in standard approaches, the gauge fields appear explicitly in the slave particle Hamiltonian, rendering the analogy to lattice gauge theory more direct and transparent.

By dint of the theory developed in Refs. 4, 7, and 8, we define our slave particles on the centers of the ‘‘up’’ and ‘‘down’’ tetrahedra of the pyrochlore lattice, which comprise two FCC sublattices (I/II, with $\eta_{\mathbf{r}} = \pm 1$) of sites, denoted with boldface characters \mathbf{r} , of a dual dia-

mond lattice. The sites of the original pyrochlore lattice are bonds of the dual lattice. The perturbative analysis of Ref. 7 identified the low energy states of H as the spin ice ones, supplemented by spinons corresponding to defect tetrahedra. As mentioned above, this inspires us to enlarge the Hilbert space and define ‘‘spinon’’ slave operators, which in turn can be seen as particles in a fluctuating vacuum (the two-in-two-out manifold dear to the spin ice community). We consider $\mathcal{H}_{big} = \mathcal{H}_{spin} \otimes \mathcal{H}_Q$, where $\mathcal{H}_{spin} = \bigotimes_N \mathcal{H}_{1/2}$ is the Hilbert space of Eq. (1) and \mathcal{H}_Q is the Hilbert space of a field $Q_{\mathbf{r}} \in \mathbb{Z}$. $Q_{\mathbf{r}}$ is defined on all the sites of the dual diamond lattice and, at this stage, is free and unphysical. We further define the real and compact operator $\varphi_{\mathbf{r}}$ to be the canonically conjugate variable to $Q_{\mathbf{r}}$, $[\varphi_{\mathbf{r}}, Q_{\mathbf{r}}] = i$. In \mathcal{H}_Q , the bosonic operators $\Phi_{\mathbf{r}}^{\dagger} = e^{i\varphi_{\mathbf{r}}}$ and $\Phi_{\mathbf{r}} = e^{-i\varphi_{\mathbf{r}}}$ thus act as raising and lowering operators, respectively, for $Q_{\mathbf{r}}$. Note that, by construction, $|\Phi_{\mathbf{r}}| = 1$. We now take the restriction of \mathcal{H}_{big} to the subspace \mathcal{H} , in which

$$Q_{\mathbf{r}} = \eta_{\mathbf{r}} \sum_{\mu} s_{\mathbf{r},\mathbf{r}+\eta_{\mathbf{r}}\mathbf{e}_{\mu}}^z, \quad (2)$$

where the \mathbf{e}_{μ} ’s are the four nearest-neighbor vectors of an $\eta_{\mathbf{r}} = 1$ (I) diamond sublattice site. This constraint can be viewed as analogous to Gauss’ law, where now $Q_{\mathbf{r}}$ counts the number of spinons. The restriction of $Q_{\mathbf{r}}$, $\Phi_{\mathbf{r}}$ and $\Phi_{\mathbf{r}}^{\dagger}$ to \mathcal{H} exactly reproduces all matrix elements of the original \mathcal{H}_{spin} , with the replacements

$$S_{\mathbf{r},\mathbf{r}+\mathbf{e}_{\mu}}^+ = \Phi_{\mathbf{r}}^{\dagger} s_{\mathbf{r},\mathbf{r}+\mathbf{e}_{\mu}}^+ \Phi_{\mathbf{r}+\mathbf{e}_{\mu}}, \quad S_{\mathbf{r},\mathbf{r}+\mathbf{e}_{\mu}}^z = s_{\mathbf{r},\mathbf{r}+\mathbf{e}_{\mu}}^z. \quad (3)$$

Here $\mathbf{r} \in \text{I}$, and $s_{\mathbf{r}\mathbf{r}'}^{\pm}, s_{\mathbf{r}\mathbf{r}'}^z$ act within the \mathcal{H}_{spin} subspace of \mathcal{H}_{big} . Note especially that, by itself, $s_{\mathbf{r}\mathbf{r}'}^{\pm} \neq S_{\mathbf{r}\mathbf{r}'}^{\pm}$ is not the physical spin, and does not remain within \mathcal{H} .

In this paper we focus on the case where $J_{\pm\pm} = 0$ (which otherwise introduces additional complications to be dealt with in a separate publication), and the Hamiltonian then becomes

$$H = \sum_{\mathbf{r} \in \text{I,II}} \frac{J_{zz}}{2} Q_{\mathbf{r}}^2 - J_{\pm} \left\{ \sum_{\mathbf{r} \in \text{I}} \sum_{\mu, \nu \neq \mu} \Phi_{\mathbf{r}+\mathbf{e}_{\mu}}^{\dagger} \Phi_{\mathbf{r}+\mathbf{e}_{\nu}} \bar{s}_{\mathbf{r},\mathbf{r}+\mathbf{e}_{\mu}} \bar{s}_{\mathbf{r},\mathbf{r}+\mathbf{e}_{\nu}}^+ + \sum_{\mathbf{r} \in \text{II}} \sum_{\mu, \nu \neq \mu} \Phi_{\mathbf{r}-\mathbf{e}_{\mu}}^{\dagger} \Phi_{\mathbf{r}-\mathbf{e}_{\nu}} \bar{s}_{\mathbf{r},\mathbf{r}-\mathbf{e}_{\mu}}^+ \bar{s}_{\mathbf{r},\mathbf{r}-\mathbf{e}_{\nu}} \right\} \\ - J_{z\pm} \left\{ \sum_{\mathbf{r} \in \text{I}} \sum_{\mu, \nu \neq \mu} \left(\gamma_{\mu\nu}^* \Phi_{\mathbf{r}}^{\dagger} \Phi_{\mathbf{r}+\mathbf{e}_{\nu}} \bar{s}_{\mathbf{r},\mathbf{r}+\mathbf{e}_{\mu}}^z \bar{s}_{\mathbf{r},\mathbf{r}+\mathbf{e}_{\mu}}^+ + \text{h.c.} \right) + \sum_{\mathbf{r} \in \text{II}} \sum_{\mu, \nu \neq \mu} \left(\gamma_{\mu\nu}^* \Phi_{\mathbf{r}}^{\dagger} \Phi_{\mathbf{r}-\mathbf{e}_{\nu}} \bar{s}_{\mathbf{r},\mathbf{r}-\mathbf{e}_{\mu}}^z \bar{s}_{\mathbf{r},\mathbf{r}-\mathbf{e}_{\mu}}^+ + \text{h.c.} \right) \right\} + \text{const.} \quad (4)$$

The integer-valued constraint in Eq. (2) commutes with H and thereby ensures that Eq. (4) is a $U(1)$ gauge the-

ory. Explicitly, it is invariant under the transformations

$$\begin{cases} \Phi_{\mathbf{r}} \rightarrow \Phi_{\mathbf{r}} e^{-i\chi_{\mathbf{r}}} \\ s_{\mathbf{r}\mathbf{r}'}^{\pm} \rightarrow s_{\mathbf{r}\mathbf{r}'}^{\pm} e^{\pm i(\chi_{\mathbf{r}'} - \chi_{\mathbf{r}})} \end{cases}, \quad (5)$$

with arbitrary χ_r . This invariance, and the Gauss' law in Eq. (2) can be made formally identical to that in lattice electrodynamics by writing $s_{\mathbf{r}\mathbf{r}'}^z = E_{\mathbf{r}\mathbf{r}'}$ and $s_{\mathbf{r}\mathbf{r}'}^\pm = e^{\pm iA_{\mathbf{r}\mathbf{r}'}}$, where E and A are lattice electric and magnetic fields [7]. This clarifies that $s_{\mathbf{r}\mathbf{r}'}^\pm$ is to be regarded as an element of the $U(1)$ gauge group. However, the notation is unnecessary and we use it only when conceptually valuable.

Eq. (4) can be viewed as spinons hopping in the background of fluctuating gauge fields, and thereby lends itself to the application of standard mean field theory methods for lattice gauge models [11], which we call gauge Mean Field Theory (gMFT). Upon performing gMFT, we will get a Hamiltonian for spinons hopping in a fixed background. Specifically, we perform the replacement:

$$\Phi^\dagger \Phi s s \rightarrow \Phi^\dagger \Phi \langle s \rangle \langle s \rangle + \langle \Phi^\dagger \Phi \rangle s \langle s \rangle + \langle \Phi^\dagger \Phi \rangle \langle s \rangle s - 2 \langle \Phi^\dagger \Phi \rangle \langle s \rangle \langle s \rangle, \quad (6)$$

and thereby split the Hamiltonian into a spinon part H_Φ^{MF} , and a gauge part H_s^{MF} (see Supp. Mat.). Note that unlike conventional Curie-Weiss mean field theory, which entirely neglects any quantum entanglement, gMFT, while suppressing some fluctuations, still allows high correlations and entanglement.

The gMFT order parameters are closely analogous to those in $U(1)$ Higgs theory [11, 12]. A non-zero expectation value $\langle s^\pm \rangle \neq 0$ implies the phase of s^\pm is relatively well-defined, i.e. there are small fluctuations of the vector potential A . The converse case, $\langle s^\pm \rangle = 0$ would indicate confinement, but does not occur here. A non-zero scalar expectation value, $\langle \Phi \rangle \neq 0$, analogous to a Higgs phase, indicates spinon condensation and generation of a mass for the gauge field, and a conventional, non-exotic state. Combined with $\langle s^\pm \rangle \neq 0$, it also implies ‘‘XY’’ magnetic order. Conversely, $\langle \Phi \rangle = 0$ indicates the spinons have a gap, and is characteristic of the Coulomb phase. The remaining gMFT order parameter, s^z , is gauge invariant, and thus indicates only the presence ($\langle s^z \rangle \neq 0$) or absence ($\langle s^z \rangle = 0$) of ‘‘Ising’’ magnetic order, i.e. time-reversal symmetry breaking. Combining this together, the phases in gMFT are summarized in Table I. We emphasize that

TABLE I. Order parameters and phases in gMFT.

$\langle \Phi \rangle$	$\langle s^z \rangle$	$\langle s^\pm \rangle$	phase
0	0	$\neq 0$	QSL
0	$\neq 0$	$\neq 0$	CFM
$\neq 0$	$\neq 0$	$\neq 0$	FM
$\neq 0$	0	$\neq 0$	AFM

despite the fact that $\langle \Phi \rangle$ does not appear explicitly in the decoupling in Eq. (6), the gMFT does generally allow for Higgs phases where Φ is indeed condensed. As we will

show below, the Higgs phase appears in a manner similar to Bose-Einstein condensation in an ideal Bose gas.

We now apply the following Ansatz, valid when $J_\pm > 0$ (which we assume hereafter), to $H^{\text{MF}} = H_s^{\text{MF}} + H_\Phi^{\text{MF}}$,

$$\langle s_\mu^z \rangle = \frac{1}{2} \sin \theta \varepsilon_\mu, \quad \langle s_\mu^- \rangle = \frac{1}{2} \cos \theta, \quad (7)$$

where $\mu = 0, \dots, 3$ and $\varepsilon = (1, 1, -1, -1)$, which assumes translational invariance and fully polarized ‘‘spins’’ \vec{s} , in accord with Eq. (6), and is compatible with FM polarization along the (global) x axis ($\langle s_\mu^+ \rangle = \langle s_\mu^- \rangle$). Note that Eq. (7) shows that the gMFT allows fluctuations of E and A , so long as $\theta \neq \pi/2$ and $\theta \neq 0$, respectively. Defining the dot product, and through it the vector notation, $\vec{u} \cdot \vec{v} = u^z v^z + \frac{1}{2} (u^+ v^- + u^- v^+)$, we find

$$H_s^{\text{MF}} = - \sum_{\mathbf{r} \in \mathcal{I}} \sum_{\mu} \tilde{h}_{\text{eff}, \mu}(\mathbf{r}) \cdot \vec{s}_{\mathbf{r}, \mathbf{r} + \mathbf{e}_\mu}, \quad (8)$$

where $h_{\text{eff}, \mu}^z = 4 \varepsilon_\mu J_{z\pm} I_1 \cos \theta$ and $h_{\text{eff}, \mu}^- = 4 J_{z\pm} I_1 \sin \theta + 2 J_\pm I_2 \cos \theta$, and we have defined $I_1 = \varepsilon_\mu \langle \Phi_{\mathbf{r}}^\dagger \Phi_{\mathbf{r} + \mathbf{e}_\mu} \rangle$ (no summation implied) and $I_2 = \sum_{\nu \neq \mu} \langle \Phi_{\mathbf{r}}^\dagger \Phi_{\mathbf{r} + \mathbf{e}_\mu - \mathbf{e}_\nu} \rangle$ (μ is fixed). These quantities turn out to be independent of the diamond bond μ . To treat the spinons, we relax the $|\Phi_{\mathbf{r}}| = 1$ constraint to a global one by introducing a Lagrange multiplier λ via the term $\lambda \sum_{\mathbf{r}} (|\Phi_{\mathbf{r}}|^2 - 1)$ in a path integral formulation, with free integration over Φ and Φ^* . The spinon Lagrangian is

$$\mathcal{L}_\Phi^{\text{MF}} = \frac{1}{N_{\text{u.c.}}} \sum_{\mathbf{k}} \int_{\omega_n} \Phi_{\mathbf{k}, \omega_n}^* \cdot G_{\mathbf{k}, \omega_n}^{-1} \cdot \Phi_{\mathbf{k}, \omega_n}, \quad (9)$$

where $N_{\text{u.c.}}$ is the number of unit cells, $[G_{\mathbf{k}, \omega_n}]_{ab} = \langle \Phi_b^* \Phi_a \rangle$, and we find the equal time Green's function to be

$$G_{\mathbf{k}, \tau=0} = \frac{1}{2} \sqrt{\frac{J_{zz}}{2}} \begin{pmatrix} Z_{\mathbf{k}}^+ & -\frac{M_{\mathbf{k}}}{|M_{\mathbf{k}}|} Z_{\mathbf{k}}^- \\ -\frac{M_{\mathbf{k}}^*}{|M_{\mathbf{k}}|} Z_{\mathbf{k}}^- & Z_{\mathbf{k}}^+ \end{pmatrix}, \quad (10)$$

where $M_{\mathbf{k}} = \sum_{\mu} \varepsilon_{\mu} e^{i\mathbf{k} \cdot \mathbf{e}_{\mu}}$, $Z_{\mathbf{k}}^{\pm}(\theta, \lambda) = \frac{1}{z_{\mathbf{k}}^{\pm}} \pm \frac{1}{z_{\mathbf{k}}}$, $z_{\mathbf{k}}^{\pm}(\theta, \lambda) = \sqrt{\lambda - \ell_{\mathbf{k}}^{\pm}(\theta)}$, $\ell_{\mathbf{k}}^{\pm}(\theta) = \frac{1}{2} J_{\pm} \cos^2 \theta L_{\mathbf{k}} \mp |\frac{1}{2} J_{z\pm} \sin 2\theta M_{\mathbf{k}}|$, $L_{\mathbf{k}} = \sum_{\mu, \nu < \mu} \cos[\mathbf{k} \cdot (\mathbf{e}_{\mu} - \mathbf{e}_{\nu})]$. A couple of remarks are in order: (i) $\lambda > \max_{\mathbf{k}} \ell_{\mathbf{k}}^-$ (ii) the spinon dispersion relations are $\omega_{\mathbf{k}}^{\pm}(\theta, \lambda) = \sqrt{2 J_{zz} z_{\mathbf{k}}^{\pm}(\theta, \lambda)}$.

The gMFT consistency conditions on θ and λ (for fixed $J_{\pm}, J_{z\pm}$) arise from requiring Eqs. (7) and $\langle s_{\mu}^{\kappa} \rangle = h_{\mu}^{\kappa} / (2|\tilde{h}_{\mu}|)$, and from the normalization condition on $|\Phi|^2$, and can be written

$$\begin{cases} \tan \theta = \frac{2 J_{z\pm} I_1(\theta, \lambda)}{2 J_{z\pm} I_1(\theta, \lambda) \tan \theta + J_{\pm} I_2(\theta, \lambda)} \\ I_3(\theta, \lambda) = 1 \end{cases}, \quad (11)$$

where $I_3(\theta, \lambda) = \langle \Phi_{\mathbf{r}}^\dagger \Phi_{\mathbf{r}} \rangle$. The explicit expressions for the $I_i = N_{\text{u.c.}}^{-1} \sum_{\mathbf{k}} \mathcal{I}_{\mathbf{k}}^i$, needed to solve Eqs. (11), are readily

derived from Eq. (10), and are given in the Supplementary Material, Eq. (29). Since Eqs. (11) may allow several distinct solutions, we must choose the solution of Eq. (11) with the lowest energy. In the mean field approximation, the ground state energy can be calculated by taking the expectation value of the Hamiltonian. We find, per unit cell, $\epsilon_{GS} = \epsilon_{av} + \epsilon_{kin}$, with

$$\begin{aligned} \epsilon_{av} &= -2I_2(\theta, \lambda) \cos^2 \theta J_{\pm} - 4I_1(\theta, \lambda) \sin 2\theta J_{z\pm} \quad (12) \\ \epsilon_{kin} &= \frac{1}{2} \int_{\mathbf{k}} [\omega_{\mathbf{k}}^+(\theta, \lambda) + \omega_{\mathbf{k}}^-(\theta, \lambda)], \quad (13) \end{aligned}$$

where $\omega^{\pm} = \sqrt{2J_{zz}} z^{\pm}$. Here ϵ_{kin} measures the “kinetic” energy associated with the spinon modes, while ϵ_{av} represents the “background” energy in which the latter evolve.

We now discuss how the different phases are obtained from the solutions of the gMFT equations. Condensed and uncondensed phases are distinguished by the value of λ . As in the theory of superfluidity, condensation is synonymous with off-diagonal long-range order, i.e. $\langle \Phi_{\mathbf{r}} \rangle^* \langle \Phi_{\mathbf{r}'} \rangle \equiv \lim_{|\mathbf{r}-\mathbf{r}'| \rightarrow \infty} \langle \Phi_{\mathbf{r}}^{\dagger} \Phi_{\mathbf{r}'} \rangle \neq 0$. This expectation value $\langle \Phi_{\mathbf{r}} \Phi_{\mathbf{r}'}^{\dagger} \rangle = N_{u.c.}^{-1} \sum_{\mathbf{k}} G_{\mathbf{k}} e^{i\mathbf{k} \cdot (\mathbf{r}' - \mathbf{r})}$ is non-zero in the long-distance limit if and only if the usual conversion of the sum to an integral *fails*, i.e. if there exists one \mathbf{k}_0 such that $G_{\mathbf{k}_0} = O(N_{u.c.})$. Like the chemical potential in Bose-Einstein condensation, λ in a Higgs phase differs from its minimum allowed value by a sub-extensive part. $\lambda = \lambda_{\min}(\theta) + \frac{\delta^2}{N_{u.c.}^2}$ therefore defines condensation, where δ is of order $O(1)$, and $\lambda_{\min}(\theta) = \max_{\mathbf{k}} \ell_{\mathbf{k}}^-(\theta)$. If instead $\lambda - \lambda_{\min}(\theta)$ is $O(1)$, one has a phase with deconfined gapped spinons and a Coulombic gauge structure. As we already mentioned above, these classes of phases can be further subdivided into “polarized” (i.e. with magnetization along the *local* z axis) and “unpolarized” when $\theta \neq 0$ and $\theta = 0$, respectively.

The phase diagram resulting from the gMFT solution (see Supplementary material) is shown in Fig. 1. It contains two “exotic” phases in which spinons are deconfined and uncondensed, indicated as QSL and CFM. The QSL state, with $\theta = 0$, is completely absent magnetic order, and is the phase studied in Refs. 7 and 8. Its low energy physics mimics quantum electrodynamics, and thereby contains a photonic excitation (gapless and linear near the origin) and gapped fractional monopole excitations (spinon and “electric” monopole) that interact via Coulomb interactions. In the present formalism, the photon is only obtained once quadratic fluctuations around the gMFT solution are considered, but is a universal feature of the exotic phases. The CFM, or “Coulombic Ferromagnet” phase, is a new phase of matter that can be seen as a polarized version of the $U(1)$ QSL. Despite being magnetic, its *elementary* magnetic excitations are spinons rather than spin waves, and it also supports a gapless photon mode. Indeed, in gMFT the transition from the QSL to CFM is second order, and consists of a continuous rise of magnetization from zero.

For larger $J_{z\pm}, J_{\pm}$, one obtains Higgs phases, which are conventional states of matter without exotic excitations and are continuously connected to the usual magnetically ordered states described by Curie-Weiss MFT. Interestingly, we find the exotic CFM state is considerably more stable than the “pure” QSL, occupying a much more substantial portion of the phase diagram.

How do we recognize a Coulomb phase in experiment? A generic sign of fractionalization is a two-particle continuum in inelastic neutron scattering, two spinons being excited by one neutron [4, 13]. In addition the photon can be detected directly by inelastic neutron scattering, as a linearly dispersing transverse mode. It is, however, more challenging to observe than the usual acoustic spin wave, because its scattering intensity becomes small ($\propto \omega$) at low energy (see Supp. Mat.), in contrast to the spin wave for which the intensity diverges ($\sim 1/\omega$) in the same limit. Interestingly, the pinch points in the static structure factor present for classical spin ice are absent for the quantum Coulomb phase [9], so this is not a useful measurement. Perhaps the most striking signature of the Coulomb phase is likely to be thermodynamic. Like the phonons, the photons contribute as BT^3 to the specific heat at low temperatures, but their speed is $v_{photon} \sim J \ll c$, the speed of sound. Crudely estimating $J \sim 2$ K appropriate for $\text{Yb}_2\text{Ti}_2\text{O}_7$, we obtain a coefficient $B_{photon} \approx 10^3 \text{mJ/mole-K}^4$, approximately 1000 times larger (!) than the phonon contribution $B_{phonon} \approx 0.5 \text{mJ/mole-K}^4$ measured for the isostructural material $\text{Y}_2\text{Ti}_2\text{O}_7$ [14].

With a phase diagram and a new phase of matter in hand, we take heart at discovering yet more new exciting facts in the pyrochlore lattice. Future studies should address the more frustrated case $J_{\pm} < 0$, phase transitions in applied field, and the influence of defects.

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