



# Seeing the light: Experimental signatures of emergent electromagnetism in a quantum spin ice

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(Received 5 April 2012; published 30 August 2012)

The “spin-ice” state found in the rare-earth pyrochlore magnets  $\text{Ho}_2\text{Ti}_2\text{O}_7$  and  $\text{Dy}_2\text{Ti}_2\text{O}_7$  offers a beautiful realization of classical magnetostatics, complete with magnetic monopole excitations. It has been suggested that in “quantum spin-ice” materials, quantum-mechanical tunneling between different ice configurations, could convert the magnetostatics of spin ice into a quantum spin liquid that realizes a fully dynamical, lattice analogue of quantum electromagnetism. Here, we explore how such a state might manifest itself in experiment, within the minimal microscopic model of a such a quantum spin ice. We develop a lattice field theory for this model, and use this to make explicit predictions for the dynamical structure factor that would be observed in neutron scattering experiments on a quantum spin ice. We find that “pinch points,” which are the signal feature of a classical spin ice, fade away as a quantum ice is cooled to its zero-temperature ground state. We also make explicit predictions for the ghostly, linearly dispersing magnetic excitations which are the “photons” of this emergent electromagnetism. The predictions of this field theory are shown to be in quantitative agreement with quantum Monte Carlo simulations at zero temperature.

DOI: [10.1103/PhysRevB.86.075154](https://doi.org/10.1103/PhysRevB.86.075154)

PACS number(s): 75.10.Jm, 75.10.Kt, 11.15.Ha

## I. INTRODUCTION

The idea that a strongly interacting quantum magnet might support a spin liquid phase that remains disordered even at zero temperature has fascinated—and frustrated—physicists ever since the seminal “resonating valence bond” (RVB) paper of Anderson in 1973.<sup>1</sup> Such a phase, it was argued, need not support the spin waves found in conventional magnets, but could instead exhibit “spinons” with fractional quantum numbers. Forty years later, the search for quantum spin liquids goes on, but with strong grounds for encouragement: a growing number of quantum magnets have been identified that *do not* order down to the lowest temperatures measured, many of which have low-temperature properties that hint at spinons.<sup>2,3</sup> At the same time, the “spin ice” materials  $\text{Ho}_2\text{Ti}_2\text{O}_7$  and  $\text{Dy}_2\text{Ti}_2\text{O}_7$  have emerged as textbook examples of classical (i.e., entropy-driven) spin liquids.<sup>4–6</sup> These highly frustrated magnetic insulators show algebraic correlations of spins over macroscopic distances<sup>7–10</sup> and support magnetic monopole excitations that provide classical analogues to the spinons envisaged by Anderson.<sup>11–17</sup>

Recently, the idea of a “quantum spin ice” has also attracted considerable interest. The family of rare-earth pyrochlores to which  $\text{Ho}_2\text{Ti}_2\text{O}_7$  and  $\text{Dy}_2\text{Ti}_2\text{O}_7$  belong includes other systems in which quantum effects play a much more important role.<sup>6</sup> Perhaps the most widely studied system of this type is  $\text{Tb}_2\text{Ti}_2\text{O}_7$ . Like the classical spin ices, the magnetism of  $\text{Tb}_2\text{Ti}_2\text{O}_7$  is controlled by the competition between strong Ising anisotropy and dipolar interactions, which are ferromagnetic on nearest-neighbour bonds, so it is expected to be an “ice.” However, in  $\text{Tb}_2\text{Ti}_2\text{O}_7$ , anisotropic exchange interactions also play an important role and endow the spins with dynamics.<sup>18–21</sup> A diffuse, liquidlike structure is observed in neutron scattering for a wide range of temperatures, with no conventional magnetic order observed down to 50 mK, despite the fact that the typical scale of interactions between spins is closer to 11 K.<sup>22,23</sup> Muon spin rotation experiments,

meanwhile, suggest that spins continue to fluctuate down to the lowest temperatures,<sup>24</sup> and the most recent quasielastic neutron scattering experiments find evidence of power-law spin correlations at 50 mK.<sup>25</sup> Taken together, these facts make  $\text{Tb}_2\text{Ti}_2\text{O}_7$  a prime example of a three-dimensional quantum spin liquid.

The magnetism of  $\text{Yb}_2\text{Ti}_2\text{O}_7$  has also proved very interesting, with neutron scattering finding no evidence of order at temperatures above 210 mK, and evidence for frustrated, anisotropic exchange interactions favoring significant dynamics within an “icelike” manifold of states.<sup>26–30</sup> Comparable studies of  $\text{Pr}_2\text{Sn}_2\text{O}_7$  suggest that it does not order down to 500 mK, but with spins continuing to fluctuate.<sup>31–33</sup> There is also reason to believe that other Pr metal oxides, including  $\text{Pr}_2\text{Zr}_2\text{O}_7$ , may prove a worthwhile hunting ground for quantum spin liquids.<sup>32–35</sup> And while the dynamics of the “classical” spin ices  $\text{Ho}_2\text{Ti}_2\text{O}_7$  and  $\text{Dy}_2\text{Ti}_2\text{O}_7$  become very slow at low temperatures, neither system has ever been observed to order, despite the fact that the dipolar interactions present in these systems are expected to favor an ordered state.<sup>36,37</sup> All of this begs the question of how the classical spin liquid found in spin ice might evolve into a quantum spin liquid as quantum effects become more important?

In fact, spin ice is just one example of a much broader class of systems that obey the “ice rules.” First introduced by Bernal and Fowler in 1933 to describe the correlations of protons in water ice,<sup>38</sup> the ice rules have since found application in models of frustrated charge order,<sup>39,40</sup> proton bonded ferroelectrics<sup>41</sup> and dense polymer melts.<sup>42</sup> All of these systems possess a local “two-in, two-out” constraint, which can most conveniently be written in terms of a zero-divergence condition on a notional magnetic field:

$$\nabla \cdot \mathbf{B} = 0. \quad (1)$$

In the case of spin ice,  $\mathbf{B}$  has the physical meaning of the local magnetization of the system, and we can associate a field  $\mathbf{B}_i$  with each spin on the lattice. For this reason, spin ice offers

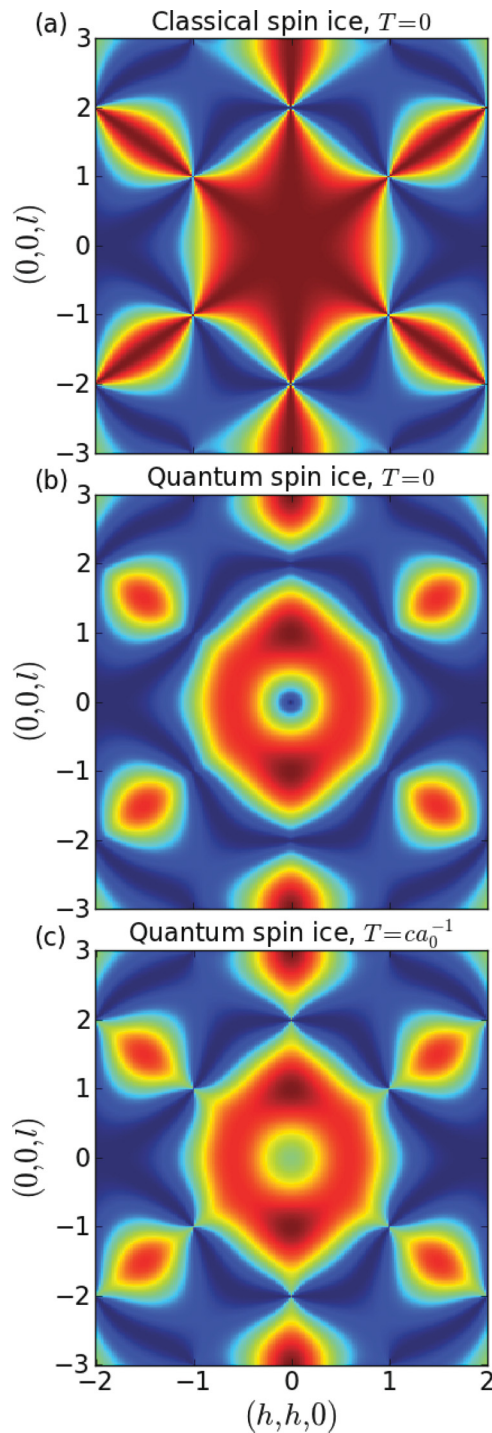


FIG. 1. (Color online) Spin correlations in a spin ice, as measured by quasielastic neutron scattering. (a) Correlations within the classical spin ice configurations, showing the characteristic pinch-point singularities at wave vector  $(1,1,1)$ , etc. (b) Correlations in a quantum ice at  $T = 0$ , showing the suppression of pinch points by quantum fluctuations. (c) Correlations in a quantum ice at an intermediate temperature  $T = ca_0^{-1}$ , showing how pinch points are progressively restored by the thermal excitation of magnetic photons. In all cases, results are shown for equal-time structure factors in the  $(h,h,0)$  plane, for a polarized neutron scattering experiment in the spin-flip channel considered by Fennell *et al.*<sup>9</sup> Temperature is measured in units where  $c$  is the speed of light associated with magnetic “photon” excitations,  $a_0$  the lattice constant, and  $\hbar = k_B = 1$ .

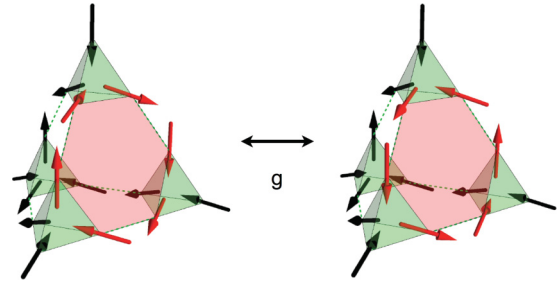


FIG. 2. (Color online) An illustration of the simplest tunneling process between different spin-ice configurations. The ice rules dictate that each tetrahedron within the lattice has two spins that point “in” and two that point “out.” Where these spins form a closed loop on a hexagonal plaquette—here shaded red—the sense of each spin within the loop can be reversed to give a new configuration that also obeys the ice rules.

a beautiful realization of classical magnetostatics, with local violations of the ice rules entering as point magnetic charges (magnetic monopoles<sup>11–17</sup>) and spin correlations that exhibit “pinch point” singularities in  $k$  space,

$$\langle S_{\mu}(-\mathbf{k})S_{\nu}(\mathbf{k}) \rangle_{\text{classical}} \propto \left( \delta_{\mu\nu} - \frac{k_{\mu}k_{\nu}}{k^2} \right), \quad (2)$$

[see Fig. 1(a)] corresponding to algebraic (dipolar) correlations in real space.<sup>7–10,41</sup> Since the ice rules can be satisfied by an exponentially large number of proton (spin, charge, polymer, etc.) configurations,<sup>43</sup> they explain the residual entropy observed in both water ice<sup>44</sup> and spin ice<sup>45</sup> at low temperatures. Given this enormous reservoir of entropy, both spin ice and water ice are natural places to look for a quantum liquid ground state.

The key ingredient needed to convert a classical ice into a quantum liquid is tunneling between different ice configurations (see Fig. 2). This opens the door to a “quantum ice”: a unique, quantum mechanical ground state, formed through the coherent superposition of an exponentially large number of classical ice configurations. Such a state could have a vanishing entropy at zero temperature, and so satisfy the third law of thermodynamics, without sacrificing the algebraic correlations and fractional excitations (magnetic monopoles) associated with the degeneracy of the ice states. If realized in a spin ice, it would provide a concrete, three-dimensional example of the long-sought quantum spin liquid.

Precisely this scenario was proposed by Moessner and Sondhi in the context of three-dimensional quantum dimer models,<sup>46</sup> by Hermele, Balents, and Fisher in a quantum, ice-type model derived from an easy-axis antiferromagnet on a pyrochlore lattice,<sup>47</sup> and by Castro-Neto, Pujol, and Fradkin in the context of a simplified model of water ice.<sup>48</sup> All of these models included tunneling between ice (or dimer) configurations of the type illustrated in Fig. 2. In a spin ice, the dominant tunneling process involves flipping loops of spins that point nose-to-tail on a hexagonal plaquette, and the resulting dynamics are described symbolically by

$$\mathcal{H}_{\text{tunneling}} = -g \sum_{\square} [|\uparrow\downarrow\rangle\langle\downarrow\uparrow| + |\downarrow\uparrow\rangle\langle\uparrow\downarrow|], \quad (3)$$

where  $g$  is the strength of the tunneling matrix element and  $\mathcal{H}_{\text{tunneling}}$  acts on the space of all possible ice (or dimer) configurations.

Both Moessner and Sondhi<sup>46</sup> and Hermele *et al.*<sup>47</sup> also introduced an additional control parameter  $\mu$  to the Hamiltonian

$$\mathcal{H}_\mu = \mathcal{H}_{\text{tunneling}} + \delta\mathcal{H}_\mu, \quad (4)$$

where

$$\delta\mathcal{H}_\mu = \mu \sum_{\square} [|\odot\rangle\langle\odot| + |\ominus\rangle\langle\ominus|]. \quad (5)$$

This makes it possible to fine tune the model to an exactly soluble Rokhsar-Kivelson (RK) point  $g = \mu$ , where the ground-state wave is an equally-weighted sum of *all* possible ice (dimer) configurations.<sup>49</sup> The authors then argued, by continuity, that a quantum liquid phase would occur for a finite range of parameters  $\mu \lesssim 1$  bordering on the RK point.<sup>46,47</sup>

The most striking feature of this quantum liquid is “light.” Attempts to construct models with “artificial light”—gapless photon excitations of an effective, low-energy  $U(1)$  gauge field—have a long history.<sup>50</sup> In recent years, it has been realized that large families of lattice models could, in principle, be described by such theories. These include abstract models of “quantum order,”<sup>51,52</sup> Bose-Hubbard models bordering on superfluidity,<sup>53</sup> systems of screened dipoles,<sup>54</sup> and suitably adapted sigma models.<sup>56</sup> Reviews of these ideas can be found in papers by Motrunich and Senthil<sup>55</sup> and Wen and Levin.<sup>57</sup>

The way in which light arises in three-dimensional quantum ice and quantum dimer models is particularly simple. The ice-rules constraint Eq. (1) is most conveniently resolved as

$$\mathcal{B}(\mathbf{r}) = \nabla \times \mathcal{A}(\mathbf{r}). \quad (6)$$

The new feature that enters where there is tunneling between ice configurations is the fluctuation in time of the gauge field  $\mathcal{A}(\mathbf{r})$ . In conventional electromagnetism, this gives rise to an electric field

$$\mathcal{E}(\mathbf{r}) = -\frac{\partial \mathcal{A}(\mathbf{r})}{\partial t}. \quad (7)$$

Then, following the heuristic arguments of Moessner and Sondhi<sup>46</sup>—or the more microscopic derivation of Hermele *et al.*<sup>47</sup>—it is reasonable to suppose that a quantum liquid found bordering the RK point  $\{\mathcal{H}_\mu$  [see Eq. (4)] with  $\mu \lesssim 1$ , would be governed by the Maxwell action

$$\mathcal{S}_{\text{Maxwell}} = \frac{1}{8\pi} \int dt d^3\mathbf{r} [\mathcal{E}(\mathbf{r})^2 - c^2 \mathcal{B}(\mathbf{r})^2]. \quad (8)$$

Any state described by  $\mathcal{S}_{\text{Maxwell}}$  [see Eq. (8)] automatically supports linearly dispersing transverse excitations of the gauge field  $\mathcal{A}(\mathbf{r})$ —photons, with a speed of light  $c$ . On the lattice, such a magnetic photon would have a dispersion  $\omega(\mathbf{k})$  of the form illustrated in Fig. 3.

Moreover, the fact that the spins now fluctuate in time as well as space introduces an additional power of  $k$  in energy-integrated (i.e., equal time) spin correlations,<sup>47,48</sup>

$$\langle S_\mu(-\mathbf{k}) S_\nu(\mathbf{k}) \rangle_{\text{quantum}} \propto k \left( \delta_{\mu\nu} - \frac{k_\mu k_\nu}{k^2} \right), \quad (9)$$

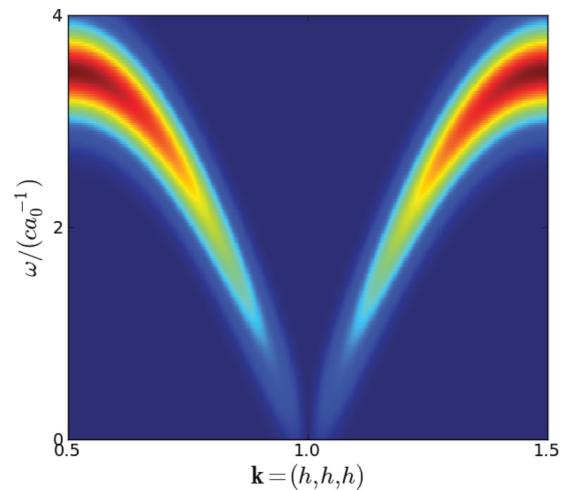


FIG. 3. (Color online) Ghostly magnetic “photon” excitation as it might appear in an inelastic neutron scattering experiment on a quantum spin ice realizing a quantum ice ground state. The photon dispersion  $\omega(\mathbf{k})$  is taken from lattice gauge theory developed in Sec. II C of this paper, convoluted with a Gaussian representing the finite energy resolution of the instrument. The intensity of scattering vanishes as  $I \propto \omega(\mathbf{k})$  at low energies.

which serves to eliminate the pinch points seen in quasielastic neutron scattering experiments [see Fig. 1(b)].<sup>58</sup> More formally, this theory is a compact, frustrated  $U(1)$  gauge theory on a diamond lattice, and we will refer to the liquid state it describes as the quantum  $U(1)$  liquid below.

The degree of fine tuning in these arguments, and the need to introduce additional parameter  $\mu$  [see Eq. (5)], might seem to render them of purely academic interest. However, the idea of a quantum  $U(1)$  liquid found strong support in finite-temperature quantum Monte Carlo simulations of an ice-type model of frustrated charge order on the pyrochlore lattice.<sup>59</sup> Subsequently, it has proved possible to determine the ground-state phase diagrams of both the quantum dimer model of Moessner and Sondhi,<sup>46</sup> and the quantum ice model of Hermele *et al.*,<sup>47</sup> from zero-temperature quantum Monte Carlo simulations.<sup>58,60,61</sup> Both models contains extended regions of a quantum liquid phase, connecting to the RK point. In both cases, this quantum liquid has low-energy excitations, which are described by a lattice analog of quantum electromagnetism.<sup>58,60,61</sup> Significantly, in the case of the quantum ice model, this quantum liquid phase encompasses the “physical” point of the model  $\mu = 0$ , and so does not require *any* fine-tuning [see Fig. 4].<sup>58</sup>

The theoretical possibility of a three-dimensional spin-liquid state with excitations described by a lattice analog of quantum electromagnetism is now well established. What remains is to connect these ideas with experiments. The purpose of this paper is therefore to set out predictions for the correlations that would be measured in neutron scattering experiments, *if* such a state were realized in a spin-ice material. For concreteness, we work with the minimal lattice model introduced by Hermele *et al.*,<sup>47</sup> transcribed to coordinates appropriate for a spin ice. More realistic generalizations of this model will be considered elsewhere.<sup>62</sup>



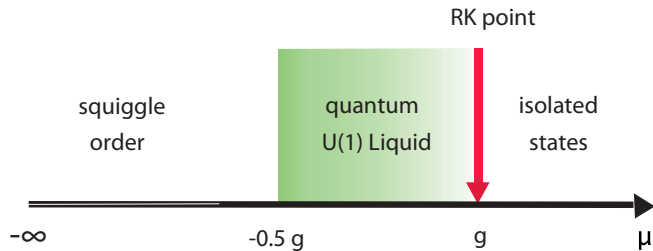


FIG. 4. (Color online) Zero-temperature phase diagram of the model of tunneling between ice states  $\mathcal{H}_\mu$  [see Eq. (16)], as determined by quantum Monte Carlo simulation in Ref. 58. The “quantum ice” point,  $\mu = 0$ , lies deep within a quantum liquid phase with low-energy excitations described by a lattice analog of quantum electromagnetism. This extends from a “squiggle” ordered phase, found for  $\mu < -0.5g$ , to the exactly-soluble RK point  $\mu = g$ . (Here,  $g$  is the strength of tunneling between ice states.)

In Sec. II of the paper, we develop the mathematical formalism needed to describe the spin correlations and low-energy spin excitations in a spin ice with a quantum  $U(1)$ -liquid ground state. Using this theory, we make predictions for the photon dispersion  $\omega(\mathbf{k})$  and dynamical structure factor  $S^{\alpha\beta}(\mathbf{k}, \omega)$ , which would be measured in neutron scattering experiments.

In Sec. III, we make explicit comparison of the predictions of this theory with zero-temperature quantum Monte Carlo simulations of the minimal, microscopic model of a quantum spin ice with tunneling between different ice configurations,  $\mathcal{H}_\mu$  [see Eq. (16)]. We find essentially perfect, *quantitative* agreement between simulation results and the field theory solved on a finite-size lattice, for a range of parameters  $0 \leq \mu \leq g$ , which interpolate from the minimal model of a quantum spin ice ( $\mu = 0$ ) to the classical correlations of the RK point ( $\mu = g$ ). This analysis reinforces the conclusions reached in Ref. 58 about the existence of a quantum  $U(1)$  liquid in this model, and puts the field-theory description on a quantitative footing.

In Sec. IV, we make predictions for neutron scattering experiments carried out at finite temperature. In particular, we analyze the way in which the characteristic pinch-point structure in quasielastic scattering is lost as the system is cooled towards its zero-temperature ground state. We conclude that the loss of the pinch points coincides with the progressive loss of the Pauling entropy as the system cools into a unique, quantum coherent, liquid ground state. Thus the signature features of the ice problem: pinch points and the Pauling entropy die together at low temperatures. We also give a brief discussion of the uniform magnetic susceptibility and heat capacity, in the low-temperature quantum regime.

Finally, in Sec. V, we conclude with a discussion of some of the remaining issues relating to experiment. As far as possible, each section of the paper is written so as to be self-contained. Readers uninterested in the mathematical development of the theory are therefore invited pass directly to Secs. III and IV, referring to Sec. II as required.

## II. FROM QUANTUM ICE TO QUANTUM ELECTROMAGNETISM

At first sight, an assembly of magnetic ions on a lattice does not look like a promising place to search for a gauge theory that perfectly mimics quantum electromagnetism. However, in the simplest microscopic model for quantum-mechanical tunneling between spin configurations obeying the “two in, two out” ice rule, this is exactly what happens.<sup>47,58,59</sup> In what follows, we retrace the steps that lead from a spin ice system to a theory of electromagnetism on a lattice.

In Sec. II A, we review the relevant microscopic models. In Sec. II B, we show how a lattice gauge theory resembling electromagnetism arises in these problems, recasting the earlier field-theoretical arguments of Hermele *et al.*<sup>47</sup> in terms appropriate for a spin ice. In Sec. II C, we explicitly construct the magnetic photon excitations of this lattice gauge theory. In Sec. II D, we use the mapping between spins and photons to calculate the correlations between spins in a quantum spin liquid described by this lattice gauge theory. Throughout this analysis we set  $\hbar = k_B = 1$ , restoring dimensional factors only where we quote a result for the speed of light.

### A. Spins on a pyrochlore lattice

The materials that we will seek to describe have magnetic ions that (a) have a crystal-field ground state that is a doublet, and (b) occupy the sites of the pyrochlore lattice shown in Fig. 5. In the case of the spin ices  $\text{Ho}_2\text{Ti}_2\text{O}_7$  and  $\text{Dy}_2\text{Ti}_2\text{O}_7$ , this doublet has Ising character (rare-earth moments point into, or out of, the tetrahedra that make up the lattice), and the dominant interactions between these Ising spins are dipolar.<sup>63</sup> However, since these dipolar interactions are effectively self-screened, the correlations present in spin ice are extremely well described by models with only nearest-neighbor interactions between spins.<sup>4,36,37,64,65</sup> This approximation gains further justification in quantum-spin-ice materials such as  $\text{Yb}_2\text{Ti}_2\text{O}_7$ ,

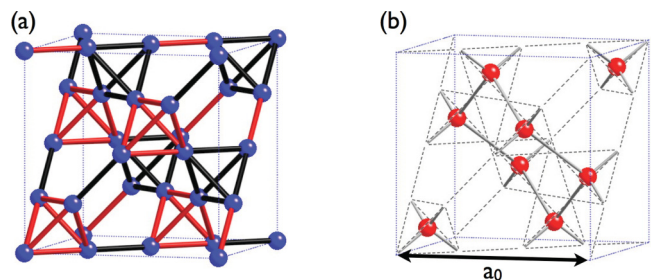


FIG. 5. (Color online) Structure of the pyrochlore lattice realized by the magnetic ions in spin-ice materials. (a) The lattice is built of corner sharing tetrahedra, and can be decomposed into a set of A-sublattice tetrahedra (here coloured red) and B-sublattice tetrahedra (here coloured black), each of which forms an fcc lattice in its own right. The primitive unit of the pyrochlore lattice consists of a single tetrahedron with four lattice sites. However, it is also possible to define a cubic unit cell, of side  $a_0$ , containing 16 lattice sites. (b) Bipartite, diamond lattice formed by the centers of tetrahedra that make up the pyrochlore lattice. The bonds of this diamond lattice define the easy axes for spins in a spin ice and play an important role in the lattice gauge theory of its excitations.

where magnetic moments are smaller than for  $\text{Ho}_2\text{Ti}_2\text{O}_7$  and  $\text{Dy}_2\text{Ti}_2\text{O}_7$ , and exchange interactions play a much larger role.

As a starting point, we can therefore consider the Hamiltonian for a (pseudo) spin-1/2 degree of freedom on a pyrochlore lattice, with the most general nearest-neighbor exchange interactions allowed by symmetry:<sup>66</sup>

$$\begin{aligned} \mathcal{H}_{S=1/2} = & \sum_{(ij)} \{ J_{zz} \mathbf{S}_i^z \mathbf{S}_j^z - J_{\pm} (\mathbf{S}_i^+ \mathbf{S}_j^- + \mathbf{S}_i^- \mathbf{S}_j^+) \\ & + J_{\pm\pm} [\gamma_{ij} \mathbf{S}_i^+ \mathbf{S}_j^+ + \gamma_{ij}^* \mathbf{S}_i^- \mathbf{S}_j^-] \\ & + J_{z\pm} [\zeta_{ij} \mathbf{S}_i^+ \mathbf{S}_j^+ + \zeta_{ij}^* \mathbf{S}_i^- \mathbf{S}_j^-] + i \leftrightarrow j \}. \end{aligned} \quad (10)$$

Here, we have followed the notation of Ross *et al.*<sup>27</sup> in which the  $\mathbf{S}_i^z$  is aligned with the local trigonal axes of the pyrochlore lattice on each site  $i$ , and  $\gamma_{ij}$  and  $\zeta_{ij}$  are  $4 \times 4$  complex unimodular matrices encoding the rotations between these local coordinate frames. In the quantum-spin-ice  $\text{Yb}_2\text{Ti}_2\text{O}_7$ , where the ground-state doublet of Yb has  $XY$  character,<sup>67–69</sup> Eq. (10) gives a good account of diffuse structure observed in neutron scattering experiments *provided* that the exchange interactions  $J_{\pm}$ ,  $J_{z\pm}$ , and  $J_{\pm\pm}$  are taken into account.<sup>26,29</sup> It also gives an excellent description of spin wave excitations about the saturated state of  $\text{Yb}_2\text{Ti}_2\text{O}_7$  in applied magnetic field, with parameters  $J_{zz} = 0.17 \pm 0.04$  meV,  $J_{\pm} = 0.05 \pm 0.01$  meV,  $J_{z\pm} = -0.14 \pm 0.01$  meV, and  $J_{\pm\pm} = 0.05 \pm 0.01$  meV obtained from fits to data.<sup>27</sup> The phase diagram associated with  $\mathcal{H}_{S=1/2}$  [see Eq. (10)] is explored in Refs. 27,35, and 70.

We can further simplify the problem by setting  $J_{\pm\pm} = 0$ , and focusing on the limit  $J_{zz} \gg J_{\pm}$ ,  $J_{z\pm} > 0$ . In this limit, the role of  $J_{zz}$  is to enforce the “ice rules” constraint, while  $J_{\pm}$  generates dynamics, and  $J_{z\pm}$  lifts the degeneracy of ice-rule obeying states. Performing degenerate perturbation theory in the basis of (spin) ice configurations, and dropping terms that lead only to a constant energy shift, leads to the effective Hamiltonian<sup>27</sup>

$$\mathcal{H}_{\text{eff}} = \mathcal{H}_{\text{tunneling}} + \mathcal{H}_{J_3} \quad (11)$$

with

$$\mathcal{H}_{\text{tunneling}} = -g \sum_{\square} [\mathbf{S}_1^+ \mathbf{S}_2^- \mathbf{S}_3^+ \mathbf{S}_4^- \mathbf{S}_5^+ \mathbf{S}_6^- + \text{H.c.}], \quad (12)$$

where  $\sum_{\square}$  runs over all hexagonal plaquettes in the pyrochlore lattice (see Fig. 2) with

$$g = \frac{12J_{\pm}^3}{J_{zz}^2}, \quad (13)$$

and

$$\mathcal{H}_{J_3} = -J_3 \sum_{(ij)_3} \mathbf{S}_i^z \mathbf{S}_j^z, \quad (14)$$

where  $\sum_{(ij)_3}$  runs over third-neighbor bonds (parallel to the nearest-neighbor bonds), with

$$J_3 = \frac{3J_{z\pm}^2}{J_{zz}} > 0. \quad (15)$$

We note that, by construction, the Hamiltonian (11) acts *only* on spin configurations satisfying the ice rules. This

implies that, in performing the degenerate perturbation theory, virtual excitations of magnetic monopoles have been projected out of the problem. This approximation will have little effect on the conclusions drawn in this paper, and could in principle be relaxed.

It is also important to note that these spin ice configurations may possess a nonzero net magnetisation  $\mathbf{M}$ . The tunneling term  $\mathcal{H}_{\text{tunneling}}$  [see Eq. (12)] generates dynamics by performing a cyclic exchange of spins on a hexagonal plaquette [see Fig. 2]. This tunneling process can be written symbolically as acting on a closed loop of spins [see Eq. (3)]. Under these dynamics, the total magnetization  $\mathbf{M}$  is a conserved quantity.

We make the final simplification of neglecting  $\mathcal{H}_{J_3}$  [Eq. (14)] and focusing exclusively on the spin-liquid favored by the tunneling term  $\mathcal{H}_{\text{tunneling}}$ —Eq. (12) or, symbolically, Eq. (3). The neglected term  $\mathcal{H}_{J_3}$  favors the six ice states with the maximum possible magnetization per site  $\mathbf{m} = (\pm 1/\sqrt{3}, 0, 0) \times S$ , etc., where  $S$  is the moment of the magnetic ion. We have confirmed through zero-temperature quantum Monte Carlo simulation of  $\mathcal{H}_{\text{eff}}$  [see Eq. (11)] that the system remains in quantum  $U(1)$  liquid ground state up to a value of  $J_3 \approx 0.27g$  at which point it undergoes first-order transition into this ordered, ferromagnetic state. These results will be discussed elsewhere.<sup>62</sup> We note that a gauge mean-field theory for  $\mathcal{H}_{S=1/2}$  [see Eq. (10)] predicts an intermediate “Coulombic ferromagnet” phase, in which the quantum  $U(1)$  liquid spontaneously acquires a finite magnetization for any finite  $J_{z\pm}$ .<sup>27,70</sup> This does not appear to be a ground state of the effective model  $\mathcal{H}_{\text{eff}}$  [see Eq. (11)].

Following Hermele *et al.*,<sup>47</sup> it is useful to augment the minimal model  $\mathcal{H}_{\text{tunneling}}$  with an additional, artificial, interaction term  $\delta\mathcal{H}_{\mu}$  [see Eq. (5)]. This renders the model exactly soluble for  $\mu = g$ . Thus the most general microscopic model we consider in this paper can be written symbolically as

$$\begin{aligned} \mathcal{H}_{\mu} = & -g \sum_{\square} [|\circ\rangle\langle\circ| + |\circ\rangle\langle\circ|] \\ & + \mu \sum_{\square} [|\circ\rangle\langle\circ| + |\circ\rangle\langle\circ|], \end{aligned} \quad (16)$$

where  $\mathcal{H}_{\mu}$  acts on the space of all possible (spin) ice configurations. This Hamiltonian is known to support a quantum  $U(1)$  liquid ground state for  $-0.5g < \mu \leq g$ .<sup>58</sup>

It is important to note that this effective description of tunneling between ice configurations might equally have been derived for the model of hardcore bosons on the pyrochlore lattice considered by Banerjee *et al.*:<sup>59</sup>

$$\begin{aligned} \mathcal{H}_{\text{charge-ice}} = & -t \sum_{(ij)} (b_i^{\dagger} b_j + b_j^{\dagger} b_i) \\ & + V \sum_{(ij)} \left( n_i - \frac{1}{2} \right) \left( n_j - \frac{1}{2} \right) \end{aligned} \quad (17)$$

with  $V \gg t$ . At 1/2-filling [ $\langle n \rangle \equiv 1/2$ ],  $V$  selects charge configurations with exactly two bosons in each tetrahedron of the lattice, and  $\mathcal{H}_{\text{charge-ice}}$  is exactly equivalent to the pseudospin-1/2 model Eq. (11), in the case where  $J_{z\pm} = J_{\pm\pm} \equiv 0$ . The leading tunneling matrix element between

different (charge) ice configurations is then

$$g = \frac{12t^3}{V^2}. \quad (18)$$

We will return to this model below in the context of predictions for experiment and simulations performed at finite temperature.<sup>59</sup>

The manifold of ice configurations on the pyrochlore lattice is equivalent to the set of possible close-packed loop coverings of the diamond lattice.<sup>71</sup> Exactly parallel arguments, leading to a formally identical Hamiltonian, can also be constructed for the closely related quantum dimer model on the diamond lattice.<sup>46,72</sup> This model also exhibits a quantum  $U(1)$  liquid ground states for a smaller—but none the less finite—range of parameters  $0.75g < \mu \leq g$  (see Refs. 60 and 61).

### B. Electromagnetism on a diamond lattice

The mappings described in Sec. II A permit us to reduce complicated interactions between magnetic ions to a problem of tunneling between spin configurations obeying the “ice rules” [see Fig. 2]. If we think of these spins as field lines of a fictitious magnetic field  $\mathbf{B}$ , these rules can conveniently be written as

$$\nabla \cdot \mathbf{B} = 0.$$

This naturally suggests an analogy with magnetostatics, with magnetic field lines constrained to lie on the bonds of a diamond lattice [see Fig. 6(a)]. And in the presence of tunneling between ice configurations, this analogy can be extended to a fully dynamical quantum electromagnetism. Here, we review the mapping from an ice with tunneling, to a compact,  $U(1)$  lattice gauge theory, before moving on to an analysis of its “photon” excitations (see Sec. II C) and spin correlations (see Sec. III C). In so doing, we follow closely the arguments of Hermele *et al.*,<sup>47</sup> but recast the discussion in terms of the magnetic fields  $\mathbf{B}$  usually associated with the spins of a spin ice.

We begin by transcribing the spin variables of  $\mathcal{H}_{\text{tunneling}}$  [see Eq. (12)] in terms of a quantum rotor variable  $\theta_i$ , and its conjugate number operator  $n_i$ :

$$\mathbf{S}_i^z = \left(n_i - \frac{1}{2}\right), \quad (19)$$

$$\mathbf{S}_i^+ = \sqrt{n_i} \exp[i\theta_i] \sqrt{1 - n_i}, \quad (20)$$

$$\mathbf{S}_i^- = \sqrt{1 - n_i} \exp[-i\theta_i] \sqrt{n_i}, \quad (21)$$

where

$$[\theta_i, n_j] = i\delta_{ij}. \quad (22)$$

The number operator  $n_i$  could equally be associated with the density of (hard-core) bosons in a charge ice, and in order to remain in the physical subspace where  $n_i = 0$  or 1, we add the term

$$\mathcal{H}_U = \frac{U}{2} \sum_i (n_i - 1/2)^2 \quad (23)$$

to the Hamiltonian, subsequently taking the limit  $U \rightarrow \infty$ . With this restriction in place, the Hamiltonian

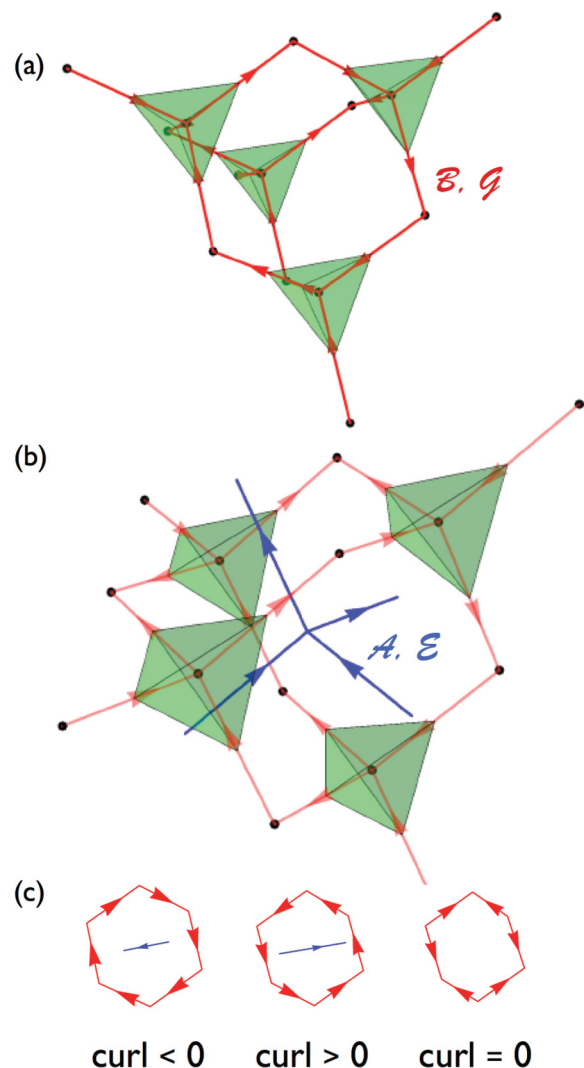


FIG. 6. (Color online) The different fields used in Sec. II B to construct lattice gauge theory of the spin liquid state, and the different lattices on which they are defined. (a) The “magnetic” field  $\mathbf{B}_{r'}$  [see Eq. (26)], and its conjugate field  $\mathcal{G}_{r'r'}$  [see Eq. (27)] are defined on the links of the diamond lattice, shown here in red. Each link of this diamond lattice corresponds to a site of the original pyrochlore lattice, and  $\mathbf{B}_{r'}$  encodes the orientation of the spin on this site. (b) The compact  $U(1)$  gauge field  $\mathcal{A}_{ss'}$  and the conjugate “electric” field  $\mathcal{E}_{ss'}$  are defined on the links of a second, dual, diamond lattice, shown here in blue. The midpoints of these bonds also form a second, dual, pyrochlore lattice, corresponding to the centers of hexagonal plaquettes in the original pyrochlore lattice. (c) An illustration of taking the lattice curl on the hexagonal plaquettes of the diamond lattice. The resulting vector lives on the links of the dual, diamond lattice.

becomes

$$\mathcal{H}_{\text{rotor}} = \frac{U}{2} \sum_i (n_i - 1/2)^2 - 2g \sum_{\square} \cos(\theta_1 - \theta_2 + \theta_3 - \theta_4 + \theta_5 - \theta_6). \quad (24)$$

It is from this rotor form of the Hamiltonian that we will make the passage to a  $U(1)$  gauge theory on the diamond lattice.

The site  $i$  of the pyrochlore lattice can be thought of as the midpoint of the bond  $\mathbf{r} \rightarrow \mathbf{r}'$  of a diamond lattice [see Fig. 5]. Since this diamond lattice is bipartite, it is possible to define directed variables on these bonds,

$$\mathcal{B}_{\mathbf{r}\mathbf{r}'} = -\mathcal{B}_{\mathbf{r}'\mathbf{r}} \quad \mathcal{G}_{\mathbf{r}\mathbf{r}'} = -\mathcal{G}_{\mathbf{r}'\mathbf{r}}, \quad (25)$$

through the mapping

$$\mathcal{B}_{\mathbf{r}\mathbf{r}'} = \pm \left( \hat{n}_i - \frac{1}{2} \right), \quad (26)$$

$$\mathcal{G}_{\mathbf{r}\mathbf{r}'} = \pm \theta_i, \quad (27)$$

where the sign is taken to be positive if  $\mathbf{r}$  belongs to the  $A$  sublattice, and negative if  $\mathbf{r}$  belongs to the  $B$  sublattice. Taking this convention into account, we are left with a pair of canonically conjugate variables

$$[\mathcal{G}_{\mathbf{r}\mathbf{r}'}, \mathcal{B}_{\mathbf{r}''\mathbf{r}'''}] = i (\delta_{\mathbf{r}\mathbf{r}''} \delta_{\mathbf{r}'\mathbf{r}'''} - \delta_{\mathbf{r}\mathbf{r}'''} \delta_{\mathbf{r}'\mathbf{r}''}). \quad (28)$$

The field,  $\mathcal{B}_{\mathbf{r}\mathbf{r}'}$  will take on the role of a magnetic field in our lattice field theory. However, in order to recreate “electromagnetism,” we need also to discover an analogue to the electric field. The missing field,  $\mathcal{E}_{\mathbf{s}\mathbf{s}'}$ , inhabits the bonds  $\mathbf{s} \rightarrow \mathbf{s}'$  of a second diamond lattice, interpenetrating the first [see Fig. 6(b)]. It is defined through a lattice curl:

$$\mathcal{E}_{\mathbf{s}\mathbf{s}'} = (\nabla_{\circ} \times \mathcal{G})_{\mathbf{s}\mathbf{s}'} = \sum_{\circ} \mathcal{G}_{\mathbf{r}\mathbf{r}'}, \quad (29)$$

where the sum  $\sum_{\circ}$  is taken with *anticlockwise* sense around the hexagonal plaquette of pyrochlore lattice sites encircling the bond  $\mathbf{s} \rightarrow \mathbf{s}'$ . It follows that  $\mathcal{E}_{\mathbf{s}\mathbf{s}'}$  is also a directed variable:

$$\mathcal{E}_{\mathbf{s}\mathbf{s}'} = -\mathcal{E}_{\mathbf{s}'\mathbf{s}}. \quad (30)$$

We are now in a position to transcribe  $\mathcal{H}_{\text{rotor}}$  completely in terms of “electromagnetic” fields:

$$\mathcal{H}_{\text{rotor}} = \frac{U}{2} \sum_{(\mathbf{r}\mathbf{r}')} \mathcal{B}_{\mathbf{r}\mathbf{r}'}^2 - 2g \sum_{(\mathbf{s}\mathbf{s}')} \cos(\mathcal{E}_{\mathbf{s}\mathbf{s}'}), \quad (31)$$

where the sum  $\sum_{(\mathbf{r}\mathbf{r}')}$  runs over all bonds of the original diamond lattice, while the sum  $\sum_{(\mathbf{s}\mathbf{s}')}$  runs over all bonds of the second, dual diamond lattice. The fact that the Hamiltonian is invariant under the transformation  $\mathcal{E}_{\mathbf{s}\mathbf{s}'} \rightarrow \mathcal{E}_{\mathbf{s}\mathbf{s}'} + 2\pi$  makes it evident that this theory is compact. It is also important to note that each of the components of the *total magnetic field*

$$(\mathcal{B}_x, \mathcal{B}_y, \mathcal{B}_z) = \sum_{(\mathbf{r}\mathbf{r}')} \mathcal{B}_{\mathbf{r}\mathbf{r}'} \hat{\mathbf{e}}_{\mathbf{r}\mathbf{r}'}, \quad (32)$$

where  $\hat{\mathbf{e}}_{\mathbf{r}\mathbf{r}'}$  is a unit vector directed from  $\mathbf{r}$  to  $\mathbf{r}'$ , is a conserved quantity under the dynamics of  $\mathcal{H}_{\text{rotor}}$  [see Eq. (31)].

More generally, reversing the sign of  $\mathcal{B}_{\mathbf{r}\mathbf{r}'}$  on a closed loop of spins will tunnel one ice configuration to another, without changing the total magnetization of the system.

In deriving Eq. (31), we have assumed that the ice rules hold, i.e.,

$$(\nabla \cdot \mathcal{B})_{\mathbf{r}} = \sum_{(\mathbf{r}')} \mathcal{B}_{\mathbf{r}\mathbf{r}'} = 0, \quad (33)$$

where the sum  $\sum_{(\mathbf{r}')}$  runs over all sites neighboring  $\mathbf{r}$ . This condition is automatically satisfied if we write  $\mathcal{B}_{\mathbf{r}\mathbf{r}'}$  as the lattice curl of a gauge field  $\mathcal{A}_{\mathbf{s}\mathbf{s}'}$ . However, we must also respect the requirement that the field  $\mathcal{B}_{\mathbf{r}\mathbf{r}'}$  take on *half-integer* values

[see Eq. (26)]. This can be accomplished by introducing a static background field  $\mathcal{B}_{\mathbf{r}\mathbf{r}'}^0$ , taken from *any* spin configuration, which satisfies the ice rules, and writing

$$(\mathcal{B}_{\mathbf{r}\mathbf{r}'} - \mathcal{B}_{\mathbf{r}\mathbf{r}'}^0) = (\nabla_{\circ} \times \mathcal{A})_{\mathbf{r}\mathbf{r}'} \quad (34)$$

to give

$$\mathcal{H}_{\text{rotor}} = \frac{U}{2} \sum_{(\mathbf{r}\mathbf{r}')} [(\nabla_{\circ} \times \mathcal{A})_{\mathbf{r}\mathbf{r}'} + \mathcal{B}_{\mathbf{r}\mathbf{r}'}^0]^2 - 2g \sum_{(\mathbf{s}\mathbf{s}')} \cos(\mathcal{E}_{\mathbf{s}\mathbf{s}'}). \quad (35)$$

The fields  $\mathcal{E}_{\mathbf{s}\mathbf{s}'}$  and  $\mathcal{A}_{\mathbf{s}\mathbf{s}'}$  are canonically conjugate,

$$[\mathcal{E}_{\mathbf{s}\mathbf{s}'}, \mathcal{A}_{\mathbf{s}''\mathbf{s}'''}] = i (\delta_{\mathbf{s}\mathbf{s}''} \delta_{\mathbf{s}'\mathbf{s}'''} - \delta_{\mathbf{s}\mathbf{s}'''} \delta_{\mathbf{s}'\mathbf{s}''}). \quad (36)$$

Moreover, the theory has a local gauge symmetry since one can make the transformation

$$\mathcal{A}_{\mathbf{s}\mathbf{s}'} \rightarrow \mathcal{A}_{\mathbf{s}\mathbf{s}'} + \lambda_{\mathbf{s}} - \lambda_{\mathbf{s}'} \quad (37)$$

on any bond without changing the value of  $(\nabla_{\circ} \times \mathcal{A})_{\mathbf{r}\mathbf{r}'}$ —each value of  $\lambda_{\mathbf{s}}$  occurs twice, with opposite signs. The situation now bears more than a passing resemblance to quantum electromagnetism.

At this point, a subtlety enters the problem. In passing from Eq. (12) to Eq. (35), we have performed a series of changes of variable without making any new approximations. However, it still remains to take the limit  $U \rightarrow \infty$ . If the “magnetic” field  $\mathcal{B}_{\mathbf{r}\mathbf{r}'}$  were an integer variable, it could be eliminated from the problem by setting  $\mathcal{B}_{\mathbf{r}\mathbf{r}'} = 0$  on all bonds. This would be energetically favorable at large  $U/g$  and would imply a phase transition from a spin liquid phase at small  $U/g$ , into a phase in which spinon excitations (magnetic monopoles) were confined at large  $U/g$  [see Ref. 73]. However, the fact that  $\mathcal{B}_{\mathbf{r}\mathbf{r}'}$  takes on half-integer values “frustrates” the lattice theory, and makes it possible for a spin liquid phase to survive in the limit  $U \rightarrow \infty$ .

Keeping this in mind, we now follow Hermele *et al.*<sup>47</sup> in assuming that an average over fast fluctuations of the gauge field (a) softens the restriction that  $\mathcal{B}_{\mathbf{r}\mathbf{r}'}$  take on half-integer values and (b) restricts  $\mathcal{E}_{\mathbf{s}\mathbf{s}'}$  to small values. Provided that both of these assumptions hold true, we can drop the reference field  $\mathcal{B}_{\mathbf{r}\mathbf{r}'}^0$  and expand the cosine in Eq. (35), to obtain

$$\mathcal{H}_{U(1)} = \frac{U}{2} \sum_{(\mathbf{r}\mathbf{r}')} [(\nabla_{\circ} \times \mathcal{A})_{\mathbf{r}\mathbf{r}'}]^2 + \frac{\mathcal{K}}{2} \sum_{(\mathbf{s}\mathbf{s}')} \mathcal{E}_{\mathbf{s}\mathbf{s}'}^2, \quad (38)$$

where both the normalization of the field  $\mathcal{B}_{\mathbf{r}\mathbf{r}'}$ , and the parameters of  $\mathcal{H}_{U(1)}$  may be renormalized from their bare values  $|\mathcal{B}_{\mathbf{r}\mathbf{r}'}| \sim 1/2$ ,  $\mathcal{U} \sim U$ ,  $\mathcal{K} \sim g$ . This, finally, is the Hamiltonian for noncompact quantum electromagnetism on a diamond lattice.

At first sight, the final step of this derivation might seem to involve an uncomfortably large leap of faith.<sup>10</sup> However, this will be justified *a posteriori* in Sec. III B by the excellent, quantitative, agreement of the predictions of  $\mathcal{H}_{U(1)}$  [see Eq. (38)] with quantum Monte Carlo simulation of the microscopic model  $\mathcal{H}_{\mu}$  [see Eq. (16)]. In order to extend this comparison to finite values of the control parameter  $\mu$ , we will augment  $\mathcal{H}_{U(1)}$  with a term

$$\delta\mathcal{H}_{U(1)} = \frac{\mathcal{W}}{2} \sum_{(\mathbf{s}\mathbf{s}')} [(\nabla_{\circ} \times (\nabla_{\circ} \times \mathcal{A}))_{\mathbf{s}\mathbf{s}'}]^2, \quad (39)$$



which mimics the effect of the ‘‘RK’’ potential [see Eq. (5)]. Since this term is permitted by the gauge symmetry, in principle, it might also be generated dynamically by an average over fast fluctuations of  $\mathcal{A}_{ss'}$ .

### C. Constructing the photon

The lattice gauge theory described in Sec. II B supports three types of excitation: magnetic charges (point sources of  $\mathcal{B}$ ) and electric charges (point sources of  $\mathcal{E}$ ), together with photons (transverse excitations of  $\mathcal{A}$ ) that mediate Coulomb interactions between these emergent charges (see Refs. 46 and 47).

The magnetic charges are the magnetic monopoles of the classical theory,<sup>12</sup> now quantized and endowed with dynamics.<sup>40,74,75</sup> They correspond to the ‘‘spinon’’ excitations of the spin liquid. Since they involve spin configurations lying outside the ice manifold, they have an energy gap

$$2\Delta_B \sim J^{zz}$$

[see Eq (10)]. The electric charges are gapped topological excitations that can be constructed as a wave packet of ice configurations with suitably chosen phases.<sup>46,47</sup> These also have an energy gap

$$\Delta_E \sim \mathcal{K} \sim g = 12J_{\pm}^3/J_{zz}^2$$

[see Eq (13)].

However, the energy of the photons vanishes linearly at small wave vector,

$$\omega(\mathbf{k} \rightarrow 0) = c|\mathbf{k}|,$$

and being gapless, the photons will control the low-energy and low-temperature properties of the system. We therefore concentrate on exploring the consequences of the photons in this paper, leaving other excitations for future work.

In what follows, we will explicitly construct a photon basis for the lattice gauge theory developed in Sec. II B, with a view to calculating the spin-spin correlation functions of the original model of a quantum spin ice. We take as a starting point

$$\begin{aligned} \mathcal{H}'_{U(1)} = & \frac{\mathcal{U}}{2} \sum_{\mathbf{r} \in A, n} [(\nabla_{\circ} \times \mathcal{A})_{(\mathbf{r}, n)}]^2 + \frac{1}{2\mathcal{K}} \sum_{\mathbf{s} \in A', m} \left[ \frac{\partial \mathcal{A}_{(\mathbf{s}, m)}}{\partial t} \right]^2 \\ & + \frac{\mathcal{W}}{2} \sum_{\mathbf{s} \in A', m} [(\nabla_{\circ} \times \nabla_{\circ} \times \mathcal{A})_{(\mathbf{s}, m)}]^2, \end{aligned} \quad (40)$$

where we have used the fact that, in the absence of electric charges,

$$\mathcal{E}_{(\mathbf{s}, m)} = -\frac{1}{\mathcal{K}} \frac{\partial \mathcal{A}_{(\mathbf{s}, m)}}{\partial t}. \quad (41)$$

To avoid double counting of bonds, the sums over diamond lattice sites  $\{\mathbf{r}\}$  and  $\{\mathbf{s}\}$  are restricted to a single sublattice, with

$$\begin{aligned} (\nabla_{\circ} \times \mathcal{A})_{(\mathbf{r}, n)} = & \sqrt{\frac{2}{N}} \sum_{\mathbf{k}} \sum_{\lambda=1}^4 \sqrt{\frac{\mathcal{K}}{\omega_{\lambda}(\mathbf{k})}} \left\{ \exp[-i\mathbf{k} \cdot (\mathbf{r} - \mathbf{e}_n/2)] a_{\lambda}(\mathbf{k}) \sum_m [-2i \sin(\mathbf{k} \cdot \mathbf{h}_{nm})] \eta_{m\lambda}(\mathbf{k}) \right. \\ & \left. + \exp[i\mathbf{k} \cdot (\mathbf{r} - \mathbf{e}_n/2)] a_{\lambda}^{\dagger}(\mathbf{k}) \sum_m [2i \sin(\mathbf{k} \cdot \mathbf{h}_{nm})] \eta_{\lambda m}^*(\mathbf{k}) \right\}, \end{aligned} \quad (46)$$

where, by inspection,  $\mathbf{h}_{nm} \equiv 0$ .

bonds labeled

$$(\mathbf{r}, n) = (\mathbf{r}, \mathbf{r} + \mathbf{e}_n), \quad (\mathbf{s}, m) = (\mathbf{s}, \mathbf{s} + \mathbf{e}_m),$$

where

$$\begin{aligned} \mathbf{e}_0 = & \frac{a_0}{4} (1, 1, 1), \quad \mathbf{e}_1 = \frac{a_0}{4} (1, -1, -1), \\ \mathbf{e}_2 = & \frac{a_0}{4} (-1, 1, -1), \quad \mathbf{e}_3 = \frac{a_0}{4} (-1, -1, 1), \end{aligned} \quad (42)$$

and  $a_0$  is the linear dimension of the cubic unit cell of the lattice, shown in Fig. 5.

We proceed to quantize  $\mathcal{A}_{sm}$  by analogy with conventional electromagnetism, introducing a Bose operator

$$[a_{\lambda}, a_{\lambda'}^{\dagger}] = \delta_{\lambda\lambda'},$$

where the four sites of the tetrahedron in the primitive unit cell of the pyrochlore lattice translate into four bands  $\lambda = 1 \dots 4$ . We write

$$\begin{aligned} \mathcal{A}_{(\mathbf{s}, m)} = & \sqrt{\frac{2}{N}} \sum_{\mathbf{k}} \sum_{\lambda=1}^4 \sqrt{\frac{\mathcal{K}}{\omega_{\lambda}(\mathbf{k})}} \\ & \times \{ \exp[-i\mathbf{k} \cdot (\mathbf{s} + \mathbf{e}_m/2)] \eta_{m\lambda}(\mathbf{k}) a_{\lambda}(\mathbf{k}) \\ & + \exp[i\mathbf{k} \cdot (\mathbf{s} + \mathbf{e}_m/2)] \eta_{\lambda m}^*(\mathbf{k}) a_{\lambda}^{\dagger}(\mathbf{k}) \}, \end{aligned} \quad (43)$$

where the sum  $\sum_{\lambda=1}^4$  runs over all four branches of photons and  $\underline{\eta}(\mathbf{k})$  is a unitary,  $4 \times 4$  matrix whose columns,  $\eta_{\lambda}(\mathbf{k})$ , play the same role as the polarization vector in conventional electromagnetism. By obvious extension,

$$\begin{aligned} \mathcal{E}_{(\mathbf{s}, m)} = & i\sqrt{\frac{2}{N}} \sum_{\mathbf{k}} \sum_{\lambda=1}^4 \sqrt{\frac{\omega_{\lambda}(\mathbf{k})}{\mathcal{K}}} \\ & \times \{ \exp[-i\mathbf{k} \cdot (\mathbf{s} + \mathbf{e}_m/2)] \eta_{m\lambda}(\mathbf{k}) a_{\lambda}(\mathbf{k}) \\ & - \exp[i\mathbf{k} \cdot (\mathbf{s} + \mathbf{e}_m/2)] \eta_{\lambda m}^*(\mathbf{k}) a_{\lambda}^{\dagger}(\mathbf{k}) \}. \end{aligned} \quad (44)$$

The Hamiltonian [see Eq. (40)] is already quadratic in  $a_{\lambda}$ . What remains is to eliminate all terms that do not conserve photon number, by constructing a suitable matrix  $\eta_{\lambda m}^*(\mathbf{k})$ . To do this, we need to evaluate the Fourier transform of the lattice curl  $(\nabla_{\circ} \times A)_{(\mathbf{r}, n)}$ . This operator is defined on a six-bond plaquette, composed of pairs of bonds that enter with opposite signs in the directed sum around the plaquette (see Figs. 2 and 6). These bonds have midpoints located at

$$\mathbf{r} - \mathbf{e}_n/2 \pm \mathbf{h}_{nm},$$

where

$$\mathbf{h}_{nm} \equiv \frac{a_0}{\sqrt{8}} \frac{\hat{\mathbf{e}}_n \times \hat{\mathbf{e}}_m}{|\hat{\mathbf{e}}_n \times \hat{\mathbf{e}}_m|}. \quad (45)$$

Hence



We can rewrite the sum  $\sum_m$  in Eq. (46) in a more convenient form by introducing a Hermitian, antisymmetric matrix

$$\underline{\underline{Z}}(\mathbf{k}) = -2i \begin{pmatrix} 0 & \sin(\mathbf{k} \cdot \mathbf{h}_{01}) & \sin(\mathbf{k} \cdot \mathbf{h}_{02}) & \sin(\mathbf{k} \cdot \mathbf{h}_{03}) \\ -\sin(\mathbf{k} \cdot \mathbf{h}_{01}) & 0 & \sin(\mathbf{k} \cdot \mathbf{h}_{12}) & \sin(\mathbf{k} \cdot \mathbf{h}_{13}) \\ -\sin(\mathbf{k} \cdot \mathbf{h}_{02}) & -\sin(\mathbf{k} \cdot \mathbf{h}_{12}) & 0 & \sin(\mathbf{k} \cdot \mathbf{h}_{23}) \\ -\sin(\mathbf{k} \cdot \mathbf{h}_{03}) & -\sin(\mathbf{k} \cdot \mathbf{h}_{13}) & -\sin(\mathbf{k} \cdot \mathbf{h}_{23}) & 0 \end{pmatrix} \quad (47)$$

acting on the four component vectors  $\eta_\lambda(\mathbf{k})$ .

Since  $\underline{\underline{Z}}(\mathbf{k})$  is Hermitian, we are free to construct the matrix  $\underline{\underline{\eta}}(\mathbf{k})$  from the eigenvectors of  $\underline{\underline{Z}}(\mathbf{k})$ , such that

$$\underline{\underline{Z}}(\mathbf{k}) \cdot \begin{pmatrix} \eta_{\lambda 0} \\ \eta_{\lambda 1} \\ \eta_{\lambda 2} \\ \eta_{\lambda 3} \end{pmatrix} = \zeta_\lambda(\mathbf{k}) \begin{pmatrix} \eta_{\lambda 0} \\ \eta_{\lambda 1} \\ \eta_{\lambda 2} \\ \eta_{\lambda 3} \end{pmatrix}. \quad (48)$$

A specific choice of  $\underline{\underline{\eta}}(\mathbf{k})$  corresponds to a choice of gauge, since using Eq. (43), the divergence of  $\mathcal{A}_{ss'}$  is now fixed. The choice here, which is made for maximum convenience in constructing the photon dispersion, is the radiation (or Coulomb) gauge

$$\nabla \cdot \mathcal{A} = 0. \quad (49)$$

It follows from Eqs. (46) and (48) that

$$(\nabla_\circ \times \mathcal{A})_{(\mathbf{r},n)} = \sqrt{\frac{2}{N}} \sum_{\mathbf{k}} \sum_{\lambda=1}^4 \sqrt{\frac{\mathcal{K}}{\omega_\lambda(\mathbf{k})}} \{ \exp[-i\mathbf{k} \cdot (\mathbf{r} - \mathbf{e}_n/2)] a_\lambda(\mathbf{k}) \zeta_\lambda(\mathbf{k}) \eta_{n\lambda}(\mathbf{k}) + \exp[i\mathbf{k} \cdot (\mathbf{r} - \mathbf{e}_n/2)] a_\lambda^\dagger(\mathbf{k}) \zeta_\lambda(\mathbf{k}) \eta_{\lambda n}^*(\mathbf{k}) \}. \quad (50)$$

Squaring and summing over  $\mathbf{r}$  and  $n$ , we arrive at

$$\begin{aligned} \sum_{(\mathbf{r},n)} (\nabla_\circ \times \mathcal{A})_{(\mathbf{r},n)}^2 &= \frac{1}{2} \sum_{\mathbf{k}} \sum_{\lambda=1}^4 \sum_{\lambda'=1}^4 \sqrt{\frac{\mathcal{K}}{\omega_\lambda(\mathbf{k})}} \sqrt{\frac{\mathcal{K}}{\omega_{\lambda'}(\mathbf{k})}} \zeta_\lambda(\mathbf{k}) \zeta_{\lambda'}(\mathbf{k}') \left[ a_\lambda(\mathbf{k}) a_{\lambda'}(-\mathbf{k}) \zeta_\lambda(\mathbf{k}) \zeta_{\lambda'}(-\mathbf{k}) \sum_n \eta_{n\lambda}(\mathbf{k}) \eta_{n\lambda'}(-\mathbf{k}) \right. \\ &\quad + a_\lambda^\dagger(\mathbf{k}) a_{\lambda'}^\dagger(\mathbf{k}) \zeta_\lambda(\mathbf{k}) \zeta_{\lambda'}(-\mathbf{k}) \sum_n \eta_{\lambda n}^*(\mathbf{k}) \eta_{\lambda' n}^*(-\mathbf{k}) + a_\lambda(\mathbf{k}) a_{\lambda'}^\dagger(\mathbf{k}) \zeta_\lambda(\mathbf{k}) \zeta_{\lambda'}(\mathbf{k}) \sum_n \eta_{n\lambda}(\mathbf{k}) \eta_{\lambda' n}^*(\mathbf{k}) \\ &\quad \left. + a_\lambda^\dagger(\mathbf{k}) a_{\lambda'}(\mathbf{k}) \zeta_\lambda(\mathbf{k}) \zeta_{\lambda'}(\mathbf{k}) \sum_n \eta_{\lambda n}^*(\mathbf{k}) \eta_{n\lambda'}(\mathbf{k}) \right]. \end{aligned} \quad (51)$$

This rather dense expression can be simplified using the unitarity of  $\underline{\underline{\eta}}(\mathbf{k})$ :

$$\sum_n \eta_{\lambda n}^*(\mathbf{k}) \eta_{n\lambda'}(\mathbf{k}) = \delta_{\lambda\lambda'} \quad (52)$$

and the fact that

$$\underline{\underline{Z}}(-\mathbf{k}) = \underline{\underline{Z}}(\mathbf{k})^*,$$

from which it follows that

$$\eta_\lambda(-\mathbf{k}) = \eta_\lambda^*(\mathbf{k}), \quad (53)$$

$$\zeta_\lambda(\mathbf{k}) = \zeta_\lambda(-\mathbf{k}). \quad (54)$$

Whence

$$\sum_{(\mathbf{r},n)} (\nabla_\circ \times \mathcal{A})_{(\mathbf{r},n)}^2 = \frac{\mathcal{K}}{2} \sum_{\mathbf{k}} \sum_{\lambda=1}^4 \frac{\zeta_\lambda(\mathbf{k})^2}{\omega_\lambda(\mathbf{k})} [a_\lambda(\mathbf{k}) a_\lambda(-\mathbf{k}) + a_\lambda^\dagger(\mathbf{k}) a_\lambda^\dagger(-\mathbf{k}) + a_\lambda^\dagger(\mathbf{k}) a_\lambda(\mathbf{k}) + a_\lambda(\mathbf{k}) a_\lambda^\dagger(\mathbf{k})]. \quad (55)$$

Applying the same procedure again to Eq. (50), we find

$$\sum_{(s,m)} (\nabla_\circ \times \nabla_\circ \times \mathcal{A})_{(s,m)}^2 = \frac{\mathcal{K}}{2} \sum_{\mathbf{k}} \sum_{\lambda=1}^4 \frac{\zeta_\lambda(\mathbf{k})^4}{\omega_\lambda(\mathbf{k})} [a_\lambda(\mathbf{k}) a_\lambda(-\mathbf{k}) + a_\lambda^\dagger(\mathbf{k}) a_\lambda^\dagger(-\mathbf{k}) + a_\lambda^\dagger(\mathbf{k}) a_\lambda(\mathbf{k}) + a_\lambda(\mathbf{k}) a_\lambda^\dagger(\mathbf{k})]. \quad (56)$$

The remaining, electric field, term in  $\mathcal{H}_0$  [see Eq. (38)] yields

$$\sum_{(s,m)} \left( \frac{\partial \mathcal{A}_{(s,m)}}{\partial t} \right)^2 = \frac{1}{2} \sum_{\mathbf{k}} \sum_{\lambda=1}^4 \omega_{\lambda}(\mathbf{k}) [-a_{\lambda}(\mathbf{k})a_{\lambda}(-\mathbf{k}) - a_{\lambda}^{\dagger}(\mathbf{k})a_{\lambda}^{\dagger}(-\mathbf{k}) + a_{\lambda}^{\dagger}(\mathbf{k})a_{\lambda}(\mathbf{k}) + a_{\lambda}(\mathbf{k})a_{\lambda}^{\dagger}(\mathbf{k})]. \quad (57)$$

Inserting all of this into the Hamiltonian (40) gives

$$\begin{aligned} \mathcal{H}_{U(1)} = & \sum_{\mathbf{k}} \sum_{\lambda=1}^4 \left\{ \left[ \frac{\mathcal{U}\mathcal{K}\zeta_{\lambda}(\mathbf{k})^2}{4\omega_{\lambda}(\mathbf{k})} + \frac{\mathcal{W}\mathcal{K}\zeta_{\lambda}(\mathbf{k})^4}{4\omega_{\lambda}(\mathbf{k})} + \frac{\omega_{\lambda}(\mathbf{k})}{4} \right] [a_{\lambda}(\mathbf{k})a_{\lambda}^{\dagger}(\mathbf{k}) + a_{\lambda}^{\dagger}(\mathbf{k})a_{\lambda}(\mathbf{k})] \right. \\ & \left. + \left[ \frac{\mathcal{U}\mathcal{K}\zeta_{\lambda}(\mathbf{k})^2}{4\omega_{\lambda}(\mathbf{k})} + \frac{\mathcal{W}\mathcal{K}\zeta_{\lambda}(\mathbf{k})^4}{4\omega_{\lambda}(\mathbf{k})} - \frac{\omega_{\lambda}(\mathbf{k})}{4} \right] [a_{\lambda}(\mathbf{k})a_{\lambda}(-\mathbf{k}) + a_{\lambda}^{\dagger}(\mathbf{k})a_{\lambda}^{\dagger}(-\mathbf{k})] \right\}. \end{aligned} \quad (58)$$

To diagonalize the Hamiltonian, we require

$$\frac{\mathcal{U}\mathcal{K}\zeta_{\lambda}(\mathbf{k})^2}{4\omega_{\lambda}(\mathbf{k})} + \frac{\mathcal{W}\mathcal{K}\zeta_{\lambda}(\mathbf{k})^4}{4\omega_{\lambda}(\mathbf{k})} = \frac{\omega_{\lambda}(\mathbf{k})}{4}, \quad (59)$$

which implies

$$\mathcal{H}_{U(1)} = \sum_{\mathbf{k}} \sum_{\lambda=1}^4 \omega_{\lambda}(\mathbf{k}) \left[ a_{\lambda}^{\dagger}(\mathbf{k})a_{\lambda}(\mathbf{k}) + \frac{1}{2} \right] \quad (60)$$

with dispersion relation fixed by Eq. (59):

$$\omega_{\lambda}(\mathbf{k}) = \mathcal{K} \sqrt{\frac{\mathcal{U}}{\mathcal{K}} \zeta_{\lambda}(\mathbf{k})^2 + \frac{\mathcal{W}}{\mathcal{K}} \zeta_{\lambda}(\mathbf{k})^4}. \quad (61)$$

All that now remains is to determine the eigenvalues of the matrix  $Z(\mathbf{k})$ ,  $\zeta_{\lambda}(\mathbf{k})$ . We find

$$\zeta_1(\mathbf{k}) = +\sqrt{2} \sqrt{\sum_{mn} \sin(\mathbf{k} \cdot \mathbf{h}_{mn})^2}, \quad (62)$$

$$\zeta_2(\mathbf{k}) = -\sqrt{2} \sqrt{\sum_{mn} \sin(\mathbf{k} \cdot \mathbf{h}_{mn})^2}, \quad (63)$$

$$\zeta_3(\mathbf{k}) = 0, \quad (64)$$

$$\zeta_4(\mathbf{k}) = 0. \quad (65)$$

It follows that the four bands of excitations  $\zeta_{\lambda}(\mathbf{k})$  correspond to two, degenerate, physical photon modes, and two unphysical, zero-energy modes. The unphysical modes arise because of the gauge redundancy in  $\mathcal{A}$  and make no contribution to either the Hamiltonian or to any gauge invariant correlation functions.

Keeping only the physical photon modes from Eq. (60), we finally arrive at

$$\mathcal{H}_{U(1)} = \sum_{\mathbf{k}} \sum_{\lambda=1}^2 \omega(\mathbf{k}) \left[ a_{\lambda}^{\dagger}(\mathbf{k})a_{\lambda}(\mathbf{k}) + \frac{1}{2} \right], \quad (66)$$

where  $\lambda$  now has the interpretation of the polarization of the photon. The photon dispersion  $\omega(\mathbf{k})$  is independent of polarization and can be written

$$\omega(\mathbf{k}) = \mathcal{K} \sqrt{\frac{\mathcal{U}}{\mathcal{K}} \zeta(\mathbf{k})^2 + \frac{\mathcal{W}}{\mathcal{K}} \zeta(\mathbf{k})^4}, \quad (67)$$

where

$$\zeta(\mathbf{k}) = \zeta_1(\mathbf{k}) = -\zeta_2(\mathbf{k}) = \sqrt{2} \sqrt{\sum_{mn} \sin(\mathbf{k} \cdot \mathbf{h}_{mn})^2} \quad (68)$$

with  $\mathbf{h}_{mn}$  defined by Eq. (45).

For all  $\mathcal{U}/\mathcal{K} > 0$ , the photon dispersion is linear in the long-wavelength limit:

$$\omega(\mathbf{k} \approx \mathbf{0}) \approx \sqrt{\mathcal{U}\mathcal{K}} a_0 |\mathbf{k}|. \quad (69)$$

This means that there is a well-defined speed of light:

$$c = \sqrt{\mathcal{U}\mathcal{K}} a_0 \hbar^{-1}, \quad (70)$$

where we have restored the dimensional factor of  $\hbar$ .

However, in the limiting case  $\mathcal{U}/\mathcal{K} \rightarrow 0$ ,  $c \rightarrow 0$ , and the dispersion of the photon becomes quadratic in the long-wavelength limit:

$$\omega(\mathbf{k}) \approx \sqrt{\mathcal{W}\mathcal{K}} a_0^2 |\mathbf{k}|^2. \quad (71)$$

Precisely this limit is realized at the RK point  $\mu = g$  of the quantum ice model  $\mathcal{H}_{\mu}$  [see Eq. (16)] and defines the boundary of the quantum  $U(1)$  liquid phase.<sup>47,58</sup> The photon dispersion relations in the two extreme cases  $\mathcal{U}/\mathcal{K} = 0$  and  $\mathcal{W}/\mathcal{K} = 0$  are plotted in Figs. 7 and 8.

#### D. From photons to structure factors

Spin correlations in real materials can be measured directly by neutron scattering. Here, we convert the analysis of photons in Sec. II C into concrete predictions for the dynamical structure factors measured in such an experiment. We also consider the structure factors that might be measured in, e.g., x-ray scattering experiments on a charge ice of the type

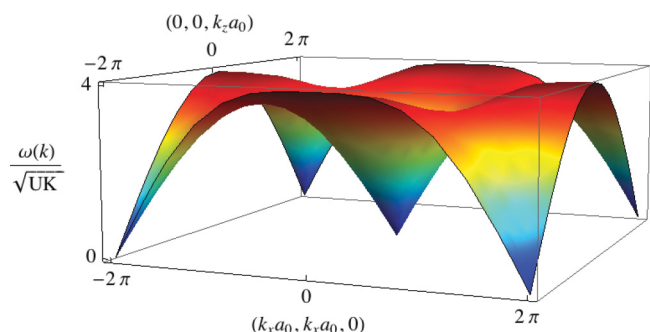


FIG. 7. (Color online) Dispersion  $\omega(\mathbf{k})$  of magnetic photon excitations, calculated for the lattice field theory Eq. (40) in the “quantum-ice” limit  $\mathcal{W}/\mathcal{K} \rightarrow 0$ . The dispersion is plotted in the  $(h, h, l)$  plane, following Eq. (67). The dispersion is linear in  $|\mathbf{k}|$  in the long-wavelength limit, with a speed of light  $c = \sqrt{\mathcal{U}\mathcal{K}} a_0$ .

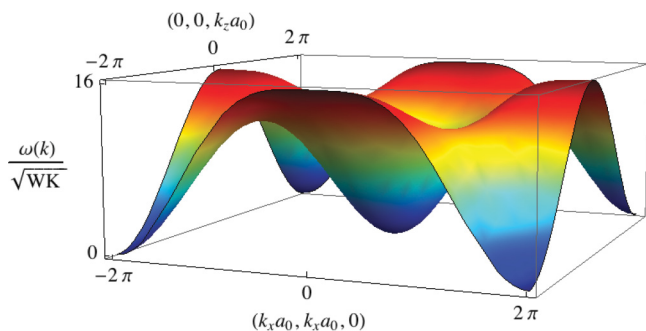


FIG. 8. (Color online) Dispersion  $\omega(\mathbf{k})$  of magnetic photon excitations, calculated for the lattice field theory Eq. (40) in the limit  $\mathcal{U}/\mathcal{K} \rightarrow 0$ . The dispersion is plotted in the  $(h, h, l)$  plane, following Eq. (67). The dispersion is quadratic in  $\mathbf{k}$  the long wavelength limit. This situation is realised in the microscopic “quantum ice” model  $\mathcal{H}_\mu$  [Eq. (16)], at the RK point,  $\mu = g$ .

considered by Banerjee *et al.*<sup>59</sup> Specifically, we will consider

$$S_{\text{spin}}^{\alpha\beta}(\mathbf{k}, \omega) = \int dt e^{-i\omega t} \langle \mathcal{S}^\alpha(-\mathbf{k}, t) \mathcal{S}^\beta(\mathbf{k}, 0) \rangle \quad (72)$$

and

$$S_{\text{charge}}(\mathbf{k}, \omega) = \int dt e^{-i\omega t} \langle n(-\mathbf{k}, t) n(\mathbf{k}, 0) \rangle. \quad (73)$$

Possessing the full photon wave function [see Eq. (43)] permits us to calculate these dynamical structure factors on a lattice, passing directly from the correlations of  $\mathcal{A}_{(s,m)}$  to those of  $\mathcal{S}_{(r,n)}$  or  $n_{(r,n)}$ .

We first consider the charge ice and, following Ref. 59, introduce an additional (dimensionless) scale factor  $\kappa \lesssim 1$  to take account of any renormalization of the field  $\mathcal{B}$  when an average is taken over fast fluctuations of  $\mathcal{A}_{(s,m)}$  [see Eq. (35) to Eq. (38)]:

$$S_{\text{charge}}(\mathbf{k}, t) = \kappa^2 \sum_{mn} \langle \tilde{\mathcal{B}}_n(-\mathbf{k}, t) \tilde{\mathcal{B}}_m(\mathbf{k}, 0) \rangle, \quad (74)$$

where

$$\tilde{\mathcal{B}}_n(\mathbf{k}, t) = \frac{1}{\sqrt{N}} \sum_{\mathbf{r}} \exp[-i\mathbf{k} \cdot (\mathbf{r} + \mathbf{e}_n/2)] \mathcal{B}_n(\mathbf{r}, t) \quad (75)$$

with

$$\mathcal{B}_n(\mathbf{r}) \equiv \mathcal{B}(\mathbf{r} - \mathbf{e}_n/2) = (\nabla_{\circ} \times \mathcal{A})_{(r,n)}.$$

The time evolution of  $\mathcal{A}_{(s,m)}$  follows directly from  $\mathcal{H}'_{U(1)}$  [see Eq. (66)]:

$$\begin{aligned} \tilde{\mathcal{B}}_n(\mathbf{k}, t) = & \frac{\sqrt{2}}{4} \sum_{\lambda=1}^4 \sqrt{\frac{\mathcal{K}}{\omega_\lambda(\mathbf{k})}} \zeta_\lambda(\mathbf{k}) [\eta_{n\lambda}(-\mathbf{k}) a_\lambda(-\mathbf{k}) e^{-i\omega_\lambda(\mathbf{k})t} \\ & + \eta_{\lambda n}^*(\mathbf{k}) a_\lambda^\dagger(\mathbf{k}) e^{i\omega_\lambda(\mathbf{k})t}], \end{aligned} \quad (76)$$

such that

$$\begin{aligned} S_{\text{charge}}(\mathbf{k}, \omega) = & \frac{\kappa^2}{8} \sum_{mn} \sum_{\lambda=1}^4 \frac{\mathcal{K}}{\omega_\lambda(\mathbf{k})} \zeta_\lambda(\mathbf{k})^2 \eta_{m\lambda}(\mathbf{k}) \eta_{\lambda n}^* \\ & \times \langle a_\lambda(\mathbf{k}) a_\lambda^\dagger(\mathbf{k}) + a_\lambda^\dagger(\mathbf{k}) a_\lambda(\mathbf{k}) \rangle \delta[\omega - \omega_\lambda(\mathbf{k})], \end{aligned} \quad (77)$$

where we have dropped all terms that fail to preserve photon number or polarization.

The unphysical photon polarisations  $\lambda = 3$  and 4 do *not* contribute to Eq. (77), since

$$\zeta_\lambda(\mathbf{k})^2 / \omega_\lambda(\mathbf{k})|_{\lambda=3,4} \equiv 0. \quad (78)$$

For the physical polarizations  $\lambda = 1$  and 2,

$$\langle a_\lambda^\dagger(\mathbf{k}) a_\lambda(\mathbf{k}) \rangle = \frac{1}{e^{\frac{\omega(\mathbf{k})}{T}} - 1} \equiv n_B(\mathbf{k}) \quad (79)$$

since the photons are bosons. Noting that

$$\sum_{\lambda=1}^4 \zeta_\lambda(\mathbf{k})^2 \eta_{m\lambda}(\mathbf{k}) \eta_{\lambda n}^* = \sum_{mn} [\underline{Z}(\mathbf{k})^2]_{mn}, \quad (80)$$

we arrive at a result for the dynamical structure factor of a quantum charge ice:

$$\begin{aligned} S_{\text{charge}}(\mathbf{k}, \omega) = & \frac{\kappa^2}{2} \frac{\mathcal{K}}{\omega(\mathbf{k})} \sum_{mn} \sum_l \sin(\mathbf{k} \cdot \mathbf{h}_{ml}) \sin(\mathbf{k} \cdot \mathbf{h}_{nl}) \\ & \times \{ \delta[\omega - \omega_\lambda(\mathbf{k})] [1 + n_B(\mathbf{k})] \\ & + \delta[\omega + \omega_\lambda(\mathbf{k})] n_B(\mathbf{k}) \}, \end{aligned} \quad (81)$$

where the vectors  $\mathbf{h}_{nm}$  are defined by Eq. (45).

In comparing with quantum Monte Carlo simulation, we will also make extensive use of the zero-temperature, equal-time (i.e., energy-integrated) structure factor:

$$S_{\text{charge}}(\mathbf{k}, t=0)_{T=0} = \int d\omega S_{\text{charge}}(\mathbf{k}, \omega)_{T=0}. \quad (82)$$

This can be written as a function of just two, dimensionless, ratios of parameters  $\bar{U}$  and  $\bar{W}$ :

$$S_{\text{charge}}(\mathbf{k}, t=0)_{T=0} = \frac{\bar{S}_0(\mathbf{k})}{\sqrt{\bar{U}\zeta(\mathbf{k})^2 + \bar{W}\zeta(\mathbf{k})^4}}, \quad (83)$$

where  $\zeta(\mathbf{k})$  is defined by Eq. (68),

$$\bar{S}_0(\mathbf{k}) = \sum_{mn} \sum_l \sin(\mathbf{k} \cdot \mathbf{h}_{ml}) \sin(\mathbf{k} \cdot \mathbf{h}_{nl}), \quad (84)$$

and the dimensionless ratios of parameters are given by

$$\bar{U} = \frac{\mathcal{U}}{\mathcal{K}\kappa^4}, \quad \bar{W} = \frac{\mathcal{W}}{\mathcal{K}\kappa^4}. \quad (85)$$

It is this form of the result, evaluated at the discrete set of wave vectors  $\{\mathbf{k}\}$  appropriate for a finite-size cluster with given boundary conditions, which we will fit to simulation results in Sec. III B.

Calculating the dynamical structure factor  $S_{\text{spin}}^{\alpha\beta}(\mathbf{k}, \omega)$  for a spin ice means generalizing Eq. (81) to take account of neutron polarization, and the local easy axes of spins in a spin ice.<sup>8</sup> However, the underlying field-theoretical description of the problem  $\mathcal{H}'_{U(1)}$  [see Eq. (40)] is unchanged, and the two results differ only in the way in which the contraction of fields on different sublattices  $\langle \mathcal{B}_n(-\mathbf{k}) \mathcal{B}_m(\mathbf{k}) \rangle$  contribute to correlation functions. In a charge ice, we simply sum over  $m, n$  as in Eq. (74). In a spin ice, we must account for the easy axes that lie along the vectors  $\hat{\mathbf{e}}_n$  [see Eq. (42)] and then calculate the projection of the spin along the axis of interest.<sup>8</sup> Thus the equal

time structure factor is

$$S_{\text{spin}}^{\alpha\beta}(\mathbf{k}, t=0) = \kappa^2 \sum_{mn} (\hat{\mathbf{e}}_m \cdot \hat{\alpha})(\hat{\mathbf{e}}_n \cdot \hat{\beta}) \langle \mathcal{B}_n(-\mathbf{k}) \mathcal{B}_m(\mathbf{k}) \rangle, \quad (86)$$

where  $\hat{\alpha}$  and  $\hat{\beta}$  are unit vectors in the  $\alpha$  and  $\beta$  directions.

Following the same procedure as described above for the charge ice, we come to the general result for the dynamical structure factor in a spin ice:

$$S_{\text{spin}}^{\alpha\beta}(\mathbf{k}, \omega) = \frac{\kappa^2}{2} \frac{\mathcal{K}}{\omega(\mathbf{k})} \sum_{mn} \sum_l \sin(\mathbf{k} \cdot \mathbf{h}_{ml}) \sin(\mathbf{k} \cdot \mathbf{h}_{nl}) \\ \times (\hat{\mathbf{e}}_m \cdot \hat{\alpha})(\hat{\mathbf{e}}_n \cdot \hat{\beta}) \{ \delta[\omega - \omega_\lambda(\mathbf{k})][1 + n_B(\mathbf{k})] \\ + \delta[\omega + \omega_\lambda(\mathbf{k})]n_B(\mathbf{k}) \}. \quad (87)$$

For concreteness, where we come to plot results, we will follow the conventions of Fennell *et al.*,<sup>9</sup> who used neutrons with polarization parallel to

$$\mathbf{n}_v = (\mathbf{1}, -\mathbf{1}, \mathbf{0})$$

to measure the energy-integrated structure factor  $S_{\text{spin}}^{\alpha\beta}(\mathbf{k}, t=0)$ , for transferred momentum  $\mathbf{k}$  in the  $(h, h, l)$  plane. We also follow the conventions of Ref. 9 in choosing a coordinate system in which

$$\mathbf{x} \parallel \mathbf{k}, \quad \mathbf{y} \parallel \mathbf{n}_v \times \mathbf{k}, \quad \mathbf{z} \parallel \mathbf{n}_v, \quad (88)$$

and consider the ‘‘spin-flip’’ channel  $S_{\text{spin}}^{yy}(\mathbf{k}, \omega)$ . In this convention, the non-spin-flip channel measures  $S_{\text{spin}}^{zz}(\mathbf{k}, \omega)$ .

It follows from Eq. (87) that the dynamical structure factor in the spin-flip channel is given by

$$S_{\text{spin}}^{yy}(\mathbf{k}, \omega) = \frac{\kappa^2}{2} \frac{\mathcal{K}}{\omega(\mathbf{k})} \sum_{mn} \sum_l \sin(\mathbf{k} \cdot \mathbf{h}_{ml}) \sin(\mathbf{k} \cdot \mathbf{h}_{nl}) \\ \times \left[ \frac{\hat{\mathbf{e}}_m \cdot (\mathbf{n}_v \times \mathbf{k})}{|\mathbf{n}_v \times \mathbf{k}|} \right] \left[ \frac{\hat{\mathbf{e}}_n \cdot (\mathbf{n}_v \times \mathbf{k})}{|\mathbf{n}_v \times \mathbf{k}|} \right] \\ \times \{ \delta[\omega - \omega_\lambda(\mathbf{k})][1 + n_B(\mathbf{k})] \\ + \delta[\omega + \omega_\lambda(\mathbf{k})]n_B(\mathbf{k}) \}$$

and the corresponding zero-temperature, energy-integrated (i.e., equal-time) structure factor is

$$S_{\text{spin}}^{yy}(\mathbf{k}, t=0)_{T=0} = \frac{\kappa^2}{2} \frac{\mathcal{K}}{\omega(\mathbf{k})} \\ \times \sum_{mn} \sum_l \sin(\mathbf{k} \cdot \mathbf{h}_{ml}) \sin(\mathbf{k} \cdot \mathbf{h}_{nl}) \\ \times \left[ \frac{\hat{\mathbf{e}}_m \cdot (\mathbf{n}_v \times \mathbf{k})}{|\mathbf{n}_v \times \mathbf{k}|} \right] \left[ \frac{\hat{\mathbf{e}}_n \cdot (\mathbf{n}_v \times \mathbf{k})}{|\mathbf{n}_v \times \mathbf{k}|} \right]. \quad (89)$$

Once again, we will make extensive use of this result when comparing with quantum Monte Carlo simulation. At finite temperature we obtain, for the energy integrated structure factor in SF channel,

$$S_{\text{spin}}^{yy}(\mathbf{k}, t=0) = \frac{\kappa^2}{2} \frac{\mathcal{K}}{\omega(\mathbf{k})} \coth \left[ \frac{\omega(\mathbf{k})}{2T} \right] \\ \times \sum_{mn} \sum_l \sin(\mathbf{k} \cdot \mathbf{h}_{ml}) \sin(\mathbf{k} \cdot \mathbf{h}_{nl}) \\ \times \left[ \frac{\hat{\mathbf{e}}_m \cdot (\mathbf{n}_v \times \mathbf{k})}{|\mathbf{n}_v \times \mathbf{k}|} \right] \left[ \frac{\hat{\mathbf{e}}_n \cdot (\mathbf{n}_v \times \mathbf{k})}{|\mathbf{n}_v \times \mathbf{k}|} \right]. \quad (90)$$

Where neutron scattering is performed with unpolarized neutrons, experiments measure an average over different components of the dynamical structure factor:

$$I(\mathbf{k}, \omega) \propto \sum_{\alpha\beta} \left( \delta_{\alpha\beta} - \frac{k_\alpha k_\beta}{k^2} \right) S_{\text{spin}}^{\alpha\beta}(\mathbf{k}, \omega). \quad (91)$$

This is the result plotted where we illustrate photon dispersions in Figs. 3 and 13. The corresponding quasielastic (energy integrated) form of Eq. (91) is given by

$$I(\mathbf{k}) \propto \sum_{\alpha\beta} \left( \delta_{\alpha\beta} - \frac{k_\alpha k_\beta}{k^2} \right) S_{\text{spin}}^{\alpha\beta}(\mathbf{k}, t=0). \quad (92)$$

It is important to note the predictions for quasielastic neutron scattering  $S_{\text{spin}}^{yy}(\mathbf{k}, \omega)$  [see Eq. (89)] and  $I(\mathbf{k}, \omega)$  [see Eq. (91)], and their energy-integrated counterparts  $S_{\text{spin}}^{yy}(\mathbf{k}, t=0)$  [see Eq. (90)] and  $I(\mathbf{k})$  [see Eq. (92)] *only* include contributions from the low-energy photon excitations of a quantum spin ice. In a real quantum-spin-ice material, their would also be contributions at higher energy from gapped ‘‘electric charges’’ and magnetic monopole excitations. These are not treated in the present theory.

### III. ‘‘ELECTROMAGNETISM’’ IN A QUANTUM SPIN ICE AT $T = 0$

The arguments presented in Sec. II B explain how a spin liquid state with correlations described by an effective electromagnetism can arise in a quantum spin ice, but stop short of offering proof that this happens in any real material or microscopic model. In what follows, we validate our use of Gaussian electromagnetism  $\mathcal{H}_{U(1)}$  [see Eq. (38)] as a description of the quantum ice model  $\mathcal{H}_\mu$  [see Eq. (16)], by making explicit comparison with the results of zero-temperature quantum Monte Carlo simulation.

However, before considering results on a lattice, it is useful to ask how correlations in quantum spin ice might differ from those in a classical spin ice, within a simple continuum field theory. This is considered in Sec. III A. We then turn to simulation of the lattice model  $\mathcal{H}_\mu$  [see Eq. (16)] in Sec. III B, demonstrating that the lattice field theory  $\mathcal{H}_{U(1)}$  [see Eq. (38)] provides an excellent *quantitative* description of the results for  $S_{\text{spin}}^{\alpha\beta}(\mathbf{k}, t=0)$ . In Sec. III C, we use the same lattice field theory to make predictions for the magnetic photon excitations which could be observed in inelastic neutron scattering experiments. Finally, in Sec. III D, we use the finite-size scaling of ground-state energies in simulation to put an absolute scale on the speed of light  $c$  associated with these magnetic photons. Throughout this analysis we set  $\hbar = 1$ , restoring dimensional factors of  $\hbar$  only where we quote results for the speed of light.

#### A. Structure factors within continuum theory

The long-wavelength properties of a quantum  $U(1)$  liquid are well described by a continuum field theory of the form considered in Ref. 46:

$$S_{\text{eff}} = \frac{1}{8\pi} \int dt d^3\mathbf{r} \{ \mathcal{E}(\mathbf{r})^2 - c^2 \mathcal{B}(\mathbf{r})^2 - \rho_c [\nabla \times \mathcal{B}(\mathbf{r})]^2 \}. \quad (93)$$



This therefore provides a convenient starting point for discussing the evolution of spin correlations in quantum spin ice. We emphasize that such a theory can be derived as a continuum limit of  $\mathcal{H}'_{U(1)}$  [see Eq. (40)].<sup>47</sup> And where we go on to make comparison with quantum Monte Carlo simulation in Sec. III B, we will use the appropriate results on a lattice, i.e., Eqs. (83) and (90).

For  $\rho_c = 0$ ,  $\mathcal{S}_{\text{eff}}$  reduces to the familiar Maxwell action of quantum electromagnetism. Crucially, this action supports photon excitations with dispersion  $\omega(\mathbf{k}) = c|\mathbf{k}|$ . The additional term  $\rho_c [\nabla \times \mathcal{B}(\mathbf{r})]^2$  is invariant under gauge transformations  $\mathcal{A}(\mathbf{r}) \rightarrow \mathcal{A}(\mathbf{r}) + \nabla\phi(\mathbf{r})$  and is an *irrelevant perturbation* in the RG sense.<sup>47</sup> However, it introduces a new length scale into the problem,

$$\lambda_c = 2\pi \frac{\sqrt{\rho_c}}{c}, \quad (94)$$

which controls the curvature of the photon dispersion

$$\omega(\mathbf{k}) = c|\mathbf{k}| \sqrt{1 + \left(\frac{\lambda_c}{2\pi}\right)^2 |\mathbf{k}|^2} \quad (95)$$

and has an important impact on how correlations evolve as a function of distance.

The role of  $\lambda_c$  can most easily be understood in the limit  $c \rightarrow 0$ , where correlations of  $\mathcal{B}(\mathbf{r})$  are controlled entirely by  $\rho_c$ . Precisely this limit is realized in the microscopic model  $\mathcal{H}_\mu$  [see Eq. (16)] at the exactly soluble ‘‘RK’’ point  $\mu = g$ . At the RK point, *all* ice configurations are degenerate, and the photons have dispersion  $\omega(\mathbf{k}) = \sqrt{\rho_c}|\mathbf{k}|^2$  (see Refs. 46 and 47). Correlations of the magnetic field,

$$C_{\mu\nu}^{\mathcal{B}}(\mathbf{k}) = \langle \mathcal{B}_\mu(-\mathbf{k}) \mathcal{B}_\nu(\mathbf{k}) \rangle, \quad (96)$$

can be calculated from Eq. (93), and for  $c = 0$ , these behave as

$$C_{\mu\nu}^{\mathcal{B}}(\mathbf{k}) \approx \frac{8\pi^4}{\sqrt{\rho_c}} \left( \delta_{\mu\nu} - \frac{k_\mu k_\nu}{k^2} \right) \quad (97)$$

exhibiting the pinch-point singularities characteristic of the ‘‘Coulombic,’’ classical  $U(1)$  liquid phase.<sup>7,8,41</sup> On Fourier transform, Eq. (97) corresponds to dipolar correlations in a three-dimensional space:

$$C_{\mu\nu}^{\mathcal{B}}(\mathbf{r}) \propto \frac{3r_\mu r_\nu / r^2 - \delta_{\mu\nu}}{r^3}. \quad (98)$$

The quantum  $U(1)$  liquid phase, with its linearly dispersing photons, is stabilized by the emergence of finite value of the speed of light  $c$  for  $\mu < g$  (see Refs. 46,47, and 58). In this case, we find

$$C_{\mu\nu}^{\mathcal{B}}(\mathbf{k}) = \frac{8\pi^4 k}{c\sqrt{1 + \left(\frac{\lambda_c k}{2\pi}\right)^2}} \left( \delta_{\mu\nu} - \frac{k_\mu k_\nu}{k^2} \right) \quad (99)$$

(see Refs. 47 and 48). For wavelengths  $\lambda \ll \lambda_c$ , Eq. (99) reduces to Eq. (97), and the system exhibits ‘‘classical’’ dipolar correlations of the form (98). However, for long wavelengths  $\lambda \gg \lambda_c$ , the additional factor of  $k$  in the numerator of Eq. (99) ‘‘hollows out’’ the pinch point singularities. In this limit,  $r \gg \lambda_c$ , Eq. (99) corresponds to dipolar correlations in a

*four-dimensional* space

$$C_{\mu\nu}^{\mathcal{B}}(\mathbf{r}) \propto \frac{2r_\mu r_\nu / r^2 - \delta_{\mu\nu}}{r^4}, \quad (100)$$

the additional dimension arising because of fluctuations in time.<sup>47,48</sup> We therefore associate  $\lambda_c$  with the length scale over which the system crosses over from ‘‘classical’’ ice correlations, decaying as  $1/r^3$ , to ‘‘quantum’’ ice correlations decaying as  $1/r^4$ .

The length-scale  $\lambda_c$  will also play an important role where we compare the predictions of field theory with simulation of microscopic model  $\mathcal{H}_\mu$  [see Eq. (16)] as a function of  $\mu$ . We can gain some insight into the  $\mu$  dependence of  $\lambda_c$ , from degenerate perturbation theory about the RK point.<sup>47,58,60,61</sup> We find that  $c^2 \sim (g - \mu)$ , while  $\rho_c \approx \text{const}$ , and it follows from Eq. (94) that  $\lambda_c$  diverges as  $\lambda_c \sim 1/\sqrt{g - \mu}$ . Exactly at the RK point, where  $g = \mu$ ,  $\lambda_c$  is infinite and correlations have the classical form Eq. (98) at *all* length scales, as expected. However, as we move away from the RK point into the quantum liquid phase for  $\mu/g < 1$ , there will be a progressive evolution of correlations from classical (pinch points) at short distances to quantum (no pinch points) at long distances. This expectation is born out by quantum Monte Carlo simulations, described below.

For the purposes of these simulations,  $\lambda_c$  also sets the minimum size of cluster that is needed to capture quantum effects at a given  $\mu$ . At  $\mu = 0$ , we find that  $\lambda_c \approx 0.8a_0$ , and hence a cluster of linear dimension  $L = 5a_0$  ( $N = 2000$ ) is comfortably big enough to observe the quantum spin liquid phase.<sup>58</sup>

## B. Comparison with quantum Monte Carlo simulation

We now turn to zero-temperature quantum Monte Carlo simulation of the microscopic model  $\mathcal{H}_\mu$  [see Eq. (16)]. We have previously argued that this model supports a quantum  $U(1)$  liquid ground state for a range of parameters  $-0.5g < \mu < g$ —see Fig. 4 and Ref. 58. In this earlier work, evidence for the ground-state phase diagram was taken from the finite-size scaling of energy spectra. Our main tool here will be the equal time structure factor  $S(\mathbf{k}, t = 0)$ , calculated from simulation, and from the lattice field theory  $\mathcal{H}'_{U(1)}$  [see Eq. (40)]. These two independent calculations are found to be in excellent, quantitative agreement, confirming the conclusions of Ref. 58. Making a direct comparison between the field theory and simulation also serves to put the field theory on a quantitative footing, providing information about the evolution of the parameters of the field theory as a function of the microscopic parameter  $\mu$ .

Simulations were performed using a Green’s function Monte Carlo (GFMC) technique based on the statistical sampling of ice configurations. This sampling is weighted using a variational estimate of the ground-state wave function, which is optimized in a separate variational Monte Carlo (VMC) calculation. In this sense, GMFC can be thought of a systematic method of improving upon variational calculations. There is no sign problem associated with  $\mathcal{H}_\mu$ , since *all* of its off-diagonal matrix elements are equal to 0 or  $-g$ , with  $g > 0$ . Where simulations converge, the results obtained are numerically exact. Our implementation of VMC and GFMC calculations for quantum ice<sup>58</sup> exactly parallels our earlier

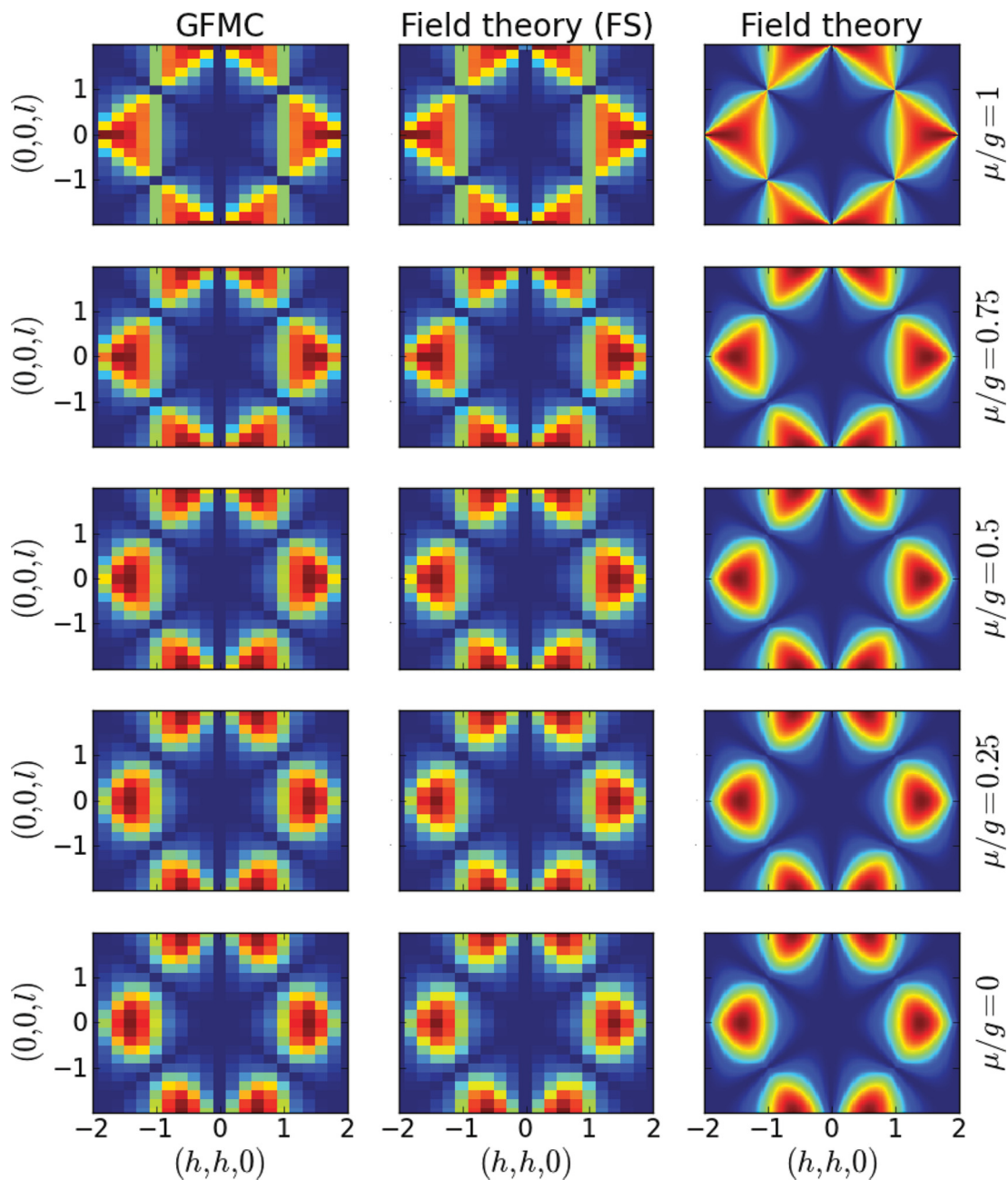


FIG. 9. (Color online) Comparison between the predictions of the lattice field theory  $\mathcal{H}'_{U(1)}$  [see Eq. (40)] and quantum Monte Carlo simulation of the microscopic model  $\mathcal{H}_\mu$  [see Eq. (16)], for a quantum charge ice at  $T = 0$ . (First column) Equal-time structure factor  $S_{\text{charge}}(\mathbf{k}, t = 0)$  calculated using Green's function Monte Carlo (GFMC) simulation of a 2000-site cubic cluster, for a range of  $\mu$  ranging from  $\mu/g = 1$  (RK point) to  $\mu/g = 0$  (quantum ice). (Second column) Best fit of the finite-size (FS) prediction of the lattice field theory to simulation, following Eq. (83). There is excellent, quantitative, agreement between theory and simulation for all values of  $\mu/g$ . (Third column) Prediction of lattice field theory in the thermodynamic limit, for parameters obtained from fits to simulation.

work on the quantum dimer model on a diamond lattice,<sup>60,61</sup> with correlation functions calculated using techniques described in Ref. 76. We refer the interested reader to these papers for further details of the method.

In the left-hand column of Fig. 9, we present GFMC simulation results for the equal-time correlations in a quantum charge ice,

$$S_{\text{charge}}(\mathbf{k}, t = 0)_{T=0} = \langle n(\mathbf{k})n(-\mathbf{k}) \rangle_{T=0}.$$

Simulations were performed for a 2000-site cubic cluster possessing the full symmetry of the lattice, for parameters  $\mu/g = 1, 0.75, 0.5, 0.25, 0$ .

The classical, dipolar correlations at the RK point  $\mu/g = 1$  are clearly visible as sharp “bow-tie” motifs in  $S_{\text{charge}}(\mathbf{k}, t = 0)$ , centered on pinch points at  $\mathbf{k} = (1, 1, 1)$ , etc. As expected, these pinch points are progressively eliminated as  $\mu/g \rightarrow 0$ , and quantum effects come to dominate the long-length scale physics of the problem. This erosion of the pinch points is

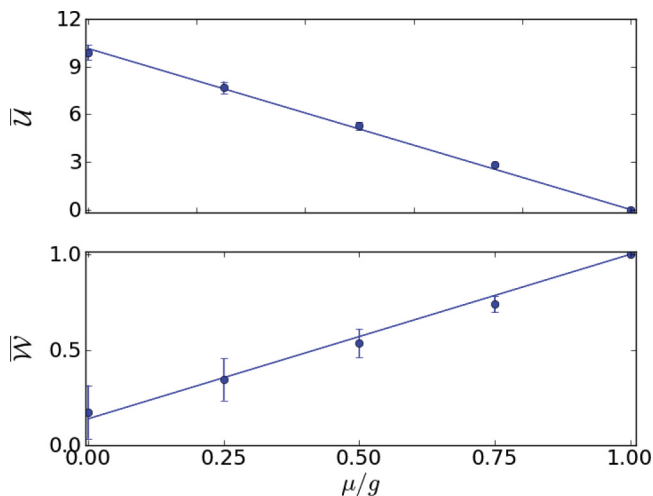


FIG. 10. (Color online) Parameters of lattice field theory  $\mathcal{H}'_{U(1)}$  [see Eq. (40)] as a function of  $\mu/g$ , from comparison with quantum Monte Carlo simulation of spin correlations in the microscopic model  $\mathcal{H}_\mu$  [see Eq. (16)] at  $T = 0$ . The dimensionless combinations of parameters  $\bar{U}$  and  $\bar{W}$  [see Eq. (85)] were obtained by fitting the predictions of the lattice field theory  $S_{\text{charge}}(\mathbf{k}, t = 0)_{T=0}$  [see Eq. (83)] to simulation results at fixed  $\mu/g$  (see Fig 9).

accompanied by a gradual redistribution of spectral weight, with high-intensity regions evolving from a triangular into an oval shape.

In the central column of Fig. 9, we present the best fit to simulation results obtained from the lattice field theory. Fits were made using the result  $S_{\text{charge}}(\mathbf{k}, t = 0)_{T=0}$  [see Eq. (83)], evaluated for the same 2000-site cluster, as a function of the two dimensionless parameters  $\bar{U}$  and  $\bar{W}$  [see Eq. (85)]. The two results are indistinguishable by eye, and differ maximally by a few percent, for values of  $\mathbf{k}$  close to the Brillouin zone boundary. The quality of these fits implies that they can be used to accurately parametrize the lattice field theory  $\mathcal{H}'_{U(1)}$  [see Eq. (40)], and the values of  $\bar{U}$  and  $\bar{W}$  obtained are shown in Fig. 10. We note that the values obtained at the RK point,  $\bar{U} = 0$  and  $\bar{W} = 1$ , are uniquely determined by the known form of correlations within the classical ice states.<sup>8</sup> A separate evaluation of the speed of light  $c \propto \sqrt{UK}$  from finite-size scaling of the ground-state energy is given in Sec. III D below.

In Fig. 11, we show equivalent results for the equal-time structure factor of a spin ice,

$$S_{\text{spin}}^{yy}(\mathbf{k}, t = 0)_{T=0} = \langle \mathbf{S}^y(\mathbf{k}) \mathbf{S}^y(-\mathbf{k}) \rangle_{T=0},$$

in the spin-flip channel considered by Fennell *et al.*<sup>9</sup> Superficially, these results look very different to those presented in Fig. 9. This is because the local easy axis is different for each of the four sublattices, leading to a staggering of correlations not present in the charge ice problem. However, the information content of the two structure factors is exactly the same.

At the RK point  $\mu/g = 1$ , correlations are classical, and  $S_{\text{spin}}^{yy}(\mathbf{k}, t = 0)$  exhibits a characteristic “snow flake” motif in the  $(h, h, l)$  plane, also seen in neutron scattering experiments on  $\text{Ho}_2\text{Ti}_2\text{O}_7$  (see Ref. 9). Pinch point singu-

larities are clearly visible at the reciprocal lattice vectors  $\mathbf{k} = (1, 1, 1)$ , etc.

Once again, these pinch points are progressively eroded as the system is tuned away from the RK point into the quantum spin liquid regime for  $\mu/g < 1$ . Probably the most striking change, however, occurs at  $\mathbf{k} = (0, 0, 0)$ . Here, for a classical spin ice,

$$S_{\text{spin}}^{yy}(\mathbf{k} \rightarrow \mathbf{0}, t = 0)_{T=0} \rightarrow \text{const.}$$

However, in a quantum spin ice,

$$S_{\text{spin}}^{yy}(\mathbf{k} = \mathbf{0}, t = 0)_{T=0} \equiv 0,$$

and spectral weight is progressively excavated from the region of reciprocal space around  $\mathbf{k} = (0, 0, 0)$  for  $\mu/g < 1$ . This has important consequences for the evolution of correlations at finite temperature, discussed in Sec. IV A and for the uniform magnetic susceptibility, discussed in Sec. IV C.

We wish to emphasize that the results shown in Fig. 11 are *not* the outcome of separate simulations of a quantum spin ice. They are taken from the same simulations of the quantum ice model  $\mathcal{H}_\mu$  [see Eq. (16)], recast in the coordinates appropriate for a spin ice. It follows that the parameters obtained from fits to field theory at finite size are exactly the same as those for a charge ice, given in Fig. 10.

### C. Seeing the light: photons and inelastic neutron scattering

Inelastic neutron scattering provides a direct method of measuring the dynamical structure factor  $S_{\text{spin}}^{\alpha\beta}(\mathbf{k}, \omega)$ , and so of resolving photon excitations in a quantum spin ice. These photons disperse linearly out of those reciprocal lattice vectors where pinch points are observed in quasielastic scattering experiments. However, since these experiments measure the energy integral of the dynamical structure factor, the suppression of pinch points in a quantum spin ice at  $T = 0$  has important implications for the observation of its photon excitations. Specifically, for noninteracting photons, the suppression of energy-integrated structure factor must imply the suppression of the weight in the photon peak itself. This is illustrated in Fig. 12.

To see how this works, we consider the result for the dynamical structure factor in a quantum spin ice  $S_{\text{spin}}^{\alpha\beta}(\mathbf{k}, \omega)$  [see Eq. (87)], in the (physically relevant) limit where  $\mathcal{W} = 0$ . In this case, weight in the photon peak is determined by the ratio

$$\frac{\bar{S}_0^{\alpha\beta}(\mathbf{k})}{\omega(\mathbf{k})},$$

where

$$\bar{S}_0^{\alpha\beta}(\mathbf{k}) = \sum_{mn} \sum_l \sin(\mathbf{k} \cdot \mathbf{h}_{ml}) \sin(\mathbf{k} \cdot \mathbf{h}_{nl}) (\hat{\mathbf{e}}_m \cdot \hat{\alpha})(\hat{\mathbf{e}}_n \cdot \hat{\beta}) \quad (101)$$

and

$$\omega_\lambda(\mathbf{k}) = \sqrt{UK} \zeta_\lambda(\mathbf{k}). \quad (102)$$



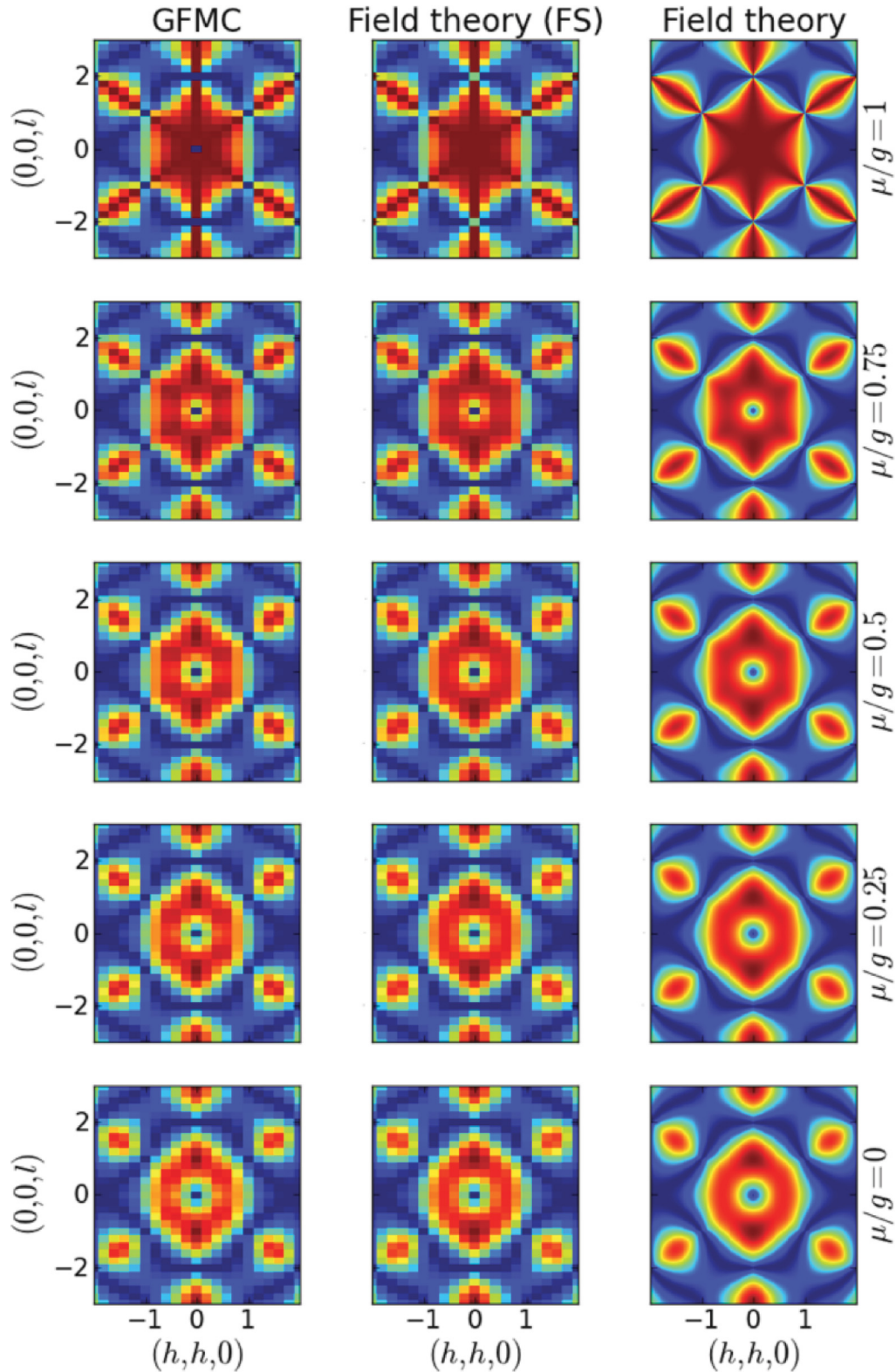


FIG. 11. (Color online) Comparison between the predictions of the lattice field theory  $\mathcal{H}_{U(1)}$  [see Eq. (40)] and quantum Monte Carlo simulation of the microscopic model  $\mathcal{H}_\mu$  [see Eq. (16)], for a quantum spin ice at  $T = 0$ . (First column) Equal-time structure factor  $S_{\text{spin}}^{yy}(\mathbf{k}, t = 0)$ , as measured in neutron scattering by Fennell *et al.*,<sup>9</sup> calculated using Green's function Monte Carlo (GFMC) simulation of a 2000-site cubic cluster for parameters ranging from  $\mu/g = 1$  (RK point) to  $\mu/g = 0$  (quantum ice). (Second column) Best fit of the finite-size (FS) prediction of the lattice field theory to simulation, following Eq. (90). There is excellent, quantitative, agreement between theory and simulation for all values of  $\mu/g$ . (Third column) Prediction of lattice field theory in the thermodynamic limit, for parameters obtained from fits to simulation.



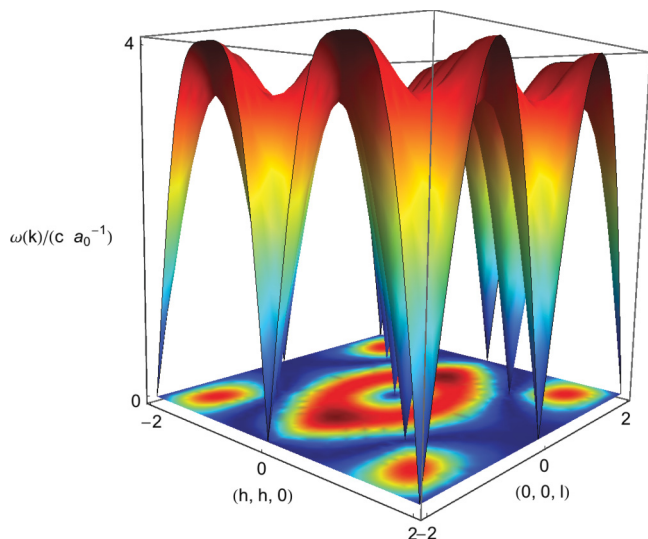


FIG. 12. (Color online) Relationship between the dispersion of the magnetic photon excitation  $\omega(\mathbf{k})$  [see Eq. (67)], and the equal time structure factor  $S_{\text{spin}}^{yy}(\mathbf{k}, t=0)$  [see Eq. (90)] in a quantum spin ice. The photon dispersion  $\omega(\mathbf{k})$  in the  $(h, h, l)$  plane is plotted above the corresponding equal-time structure factor, demonstrating how the photon disperses out of the (suppressed) pinch points at reciprocal lattice vectors. Note that the intensity of the scattering  $S_{\text{spin}}^{yy}(\mathbf{k}, t=0) \rightarrow 0$  where  $\omega(\mathbf{k}) \rightarrow 0$  [see Eq. (105)]. Results were calculated within the lattice field theory [see Eq. (40)] for  $\mathcal{W} = 0$ , with energy measured in units such that  $\hbar = 1$ .

We can use the spectral representation of  $\underline{Z}(\mathbf{k})$  [see Eq. (80)] to write

$$\sin(\mathbf{k} \cdot \mathbf{h}_{m1}) \sin(\mathbf{k} \cdot \mathbf{h}_{n1}) = \frac{1}{4} \sum_{\lambda=1}^4 \frac{\omega_{\lambda}(\mathbf{k})^2}{\mathcal{K}\mathcal{U}} \eta_{m\lambda}(\mathbf{k}) \eta_{\lambda n}^*(\mathbf{k}). \quad (103)$$

Since the only contributions to the RHS of Eq. (103) come from the two dispersing modes  $\lambda = 1, 2$ , [see Eq. (78)], Eq. (101)

simplifies to

$$\overline{S}_0^{\alpha\beta}(\mathbf{k}) = \frac{1}{4} \frac{\omega(\mathbf{k})^2}{\mathcal{K}\mathcal{U}} \sum_{\lambda=1}^2 \sum_{mn} \eta_{m\lambda}(\mathbf{k}) \eta_{\lambda n}^*(\mathbf{k}) (\hat{\mathbf{e}}_m \cdot \hat{\boldsymbol{\alpha}}) (\hat{\mathbf{e}}_n \cdot \hat{\boldsymbol{\beta}}). \quad (104)$$

Expanding in the first Brillouin zone, for  $\mathbf{k} \approx 0$ , we find

$$\sum_{mn} \eta_{m\lambda}(\mathbf{k}) \eta_{\lambda n}^*(\mathbf{k}) (\hat{\mathbf{e}}_m \cdot \hat{\boldsymbol{\alpha}}) (\hat{\mathbf{e}}_n \cdot \hat{\boldsymbol{\beta}}) \approx \frac{1}{3}$$

for  $\alpha = \beta = y, z$  and zero otherwise. It follows that

$$S_{\text{spin}}^{yy}(\mathbf{k} \approx \mathbf{0}, \omega \approx 0) = S_{\text{spin}}^{zz}(\mathbf{k} \approx \mathbf{0}, \omega \approx 0) \propto \omega(\mathbf{k}) \delta[\omega - \omega(\mathbf{k})]. \quad (105)$$

Therefore at low energies, in the first Brillouin zone, inelastic neutron scattering experiments will resolve the magnetic photon excitation as a ghostly, linearly dispersing peak, with intensity vanishing as  $I \propto \omega(\mathbf{k})$ , as noted in Ref. 70. However, at higher energies and in other Brillouin zones, the momentum dependence of  $\eta_{m\lambda}(\mathbf{k}) \eta_{\lambda n}^*(\mathbf{k})$  in Eq. (104) will lead to a significant variation in the intensity of the signal at fixed  $\omega$ . This behavior is illustrated in Fig. 13, where we have plotted the intensity of scattering  $I(\mathbf{k}, \omega)$  [see Eq. (91)] for an experiment performed using *unpolarised* neutrons. The corresponding quasielastic scattering, and the path within the  $[h, h, l]$  plane, are shown in Fig. 14.

The phenomenology of this photon excitation stands in stark contrast to conventional antiferromagnets, whose linearly dispersing spin-wave excitations have the greatest intensity approaching the zero-energy magnetic Bragg peak associated with magnetic order. The difference between these two problems stems from the fact that the photon is a quantized excitation of  $\mathcal{A}$ , while neutron scattering measures correlations of  $\mathcal{B}$ . The lattice curl needed to relate one to the other introduces additional factors of  $\zeta_{\lambda}(\mathbf{k})$  in  $S_{\text{spin}}^{\alpha\beta}(\mathbf{k}, \omega)$  [see Eq. (87)], which leads to the suppression of spectral weight at low energies.

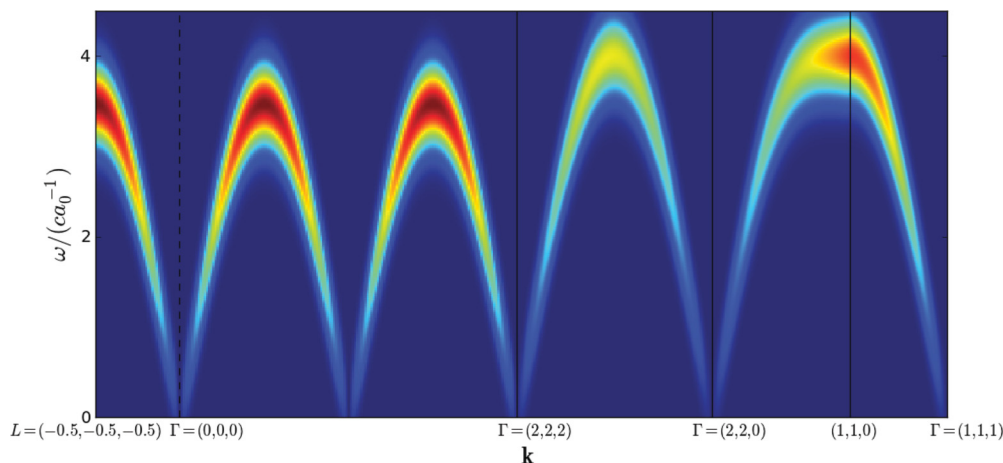


FIG. 13. (Color online) Ghostly magnetic “photon” excitation as it might appear in an inelastic neutron scattering experiment on a quantum spin ice realising a quantum ice ground state, for a series of cuts along high symmetry directions in reciprocal space. The prediction of the lattice field theory  $\mathcal{H}_{U(1)}$  [see Eq. (40)] for inelastic scattering by unpolarized neutrons,  $I(\mathbf{k}, \omega)$  [see Eq. (91)] has been convoluted with a Gaussian of variance  $0.3 c a_0^{-1}$  to represent the finite energy resolution of the instrument. The intensity of scattering vanishes as  $\omega \rightarrow 0$  and is strongest at high energies. Energy is measured in units such that  $\hbar = 1$  and the photon dispersion calculated for  $\mathcal{W} = 0$ .

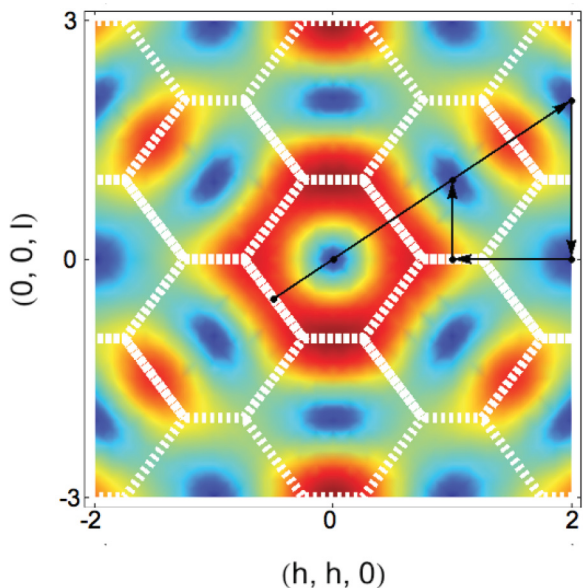


FIG. 14. (Color online) Prediction of the lattice field theory  $\mathcal{H}'_{U(1)}$  [see Eq. (40)] for quasielastic neutron scattering performed using unpolarised neutrons, for comparison with Fig. 13. Results for  $I(\mathbf{k})$  are taken from Eq. (92), and calculated for  $\mathcal{W} = 0$ . The path within the  $[h, h, l]$  plane used for plotting the photon dispersion in Fig. 13 is shown using unbroken black arrows, with Brillouin zone boundaries marked as dashed white lines.

A better point of comparison, in fact, is the scattering of neutrons by *real* photons in models of a hot early universe.<sup>77</sup> In both cases, photons are associated with a periodically fluctuating magnetic field, transverse to the direction of their propagation. And in both cases, neutrons scatter inelastically from these locally fluctuating magnetic fields. In a spin ice, this scattering can occur in both the spin-flip (SF) channel, in which case there is a transfer of angular momentum to the sample, and in the non spin-flip (NSF) channel (see Figs. 13, 14, and 16). It is also interesting to note that the same phenomenology of linearly dispersing excitations, with a vanishing spectral weight at long wavelengths, is encountered in quantum spin nematics.<sup>78</sup> In this case, low-energy spin fluctuations are controlled by a time derivative of the underlying nematic order parameter<sup>79</sup> and so vanish for  $\omega \rightarrow 0$ .

#### D. Estimating the speed of light

The signal feature of the quantum  $U(1)$  liquid is its photon excitations. One important consequence of these, so far as the simulation of finite-size systems is concerned, is a characteristic finite-size correction to the ground-state energy per site  $E_0/N$ , coming from the zero-point energy of the photons:

$$\frac{\delta E_0(L)}{N} = \frac{1}{N} [E_0(L) - E_0(\infty)] = x_1 L^{-4} + \dots, \quad (106)$$

where  $L \sim N^{\frac{1}{3}}$  is the linear dimension of the cluster, and the coefficient  $x_1$  is proportional to the speed of light  $c$  (see Ref. 61). This means that it is possible to extract the speed of light from the finite-size scaling of the ground-state energy found in simulations of  $\mathcal{H}_\mu$  [see Eq. (16)], shown in Fig. 15.

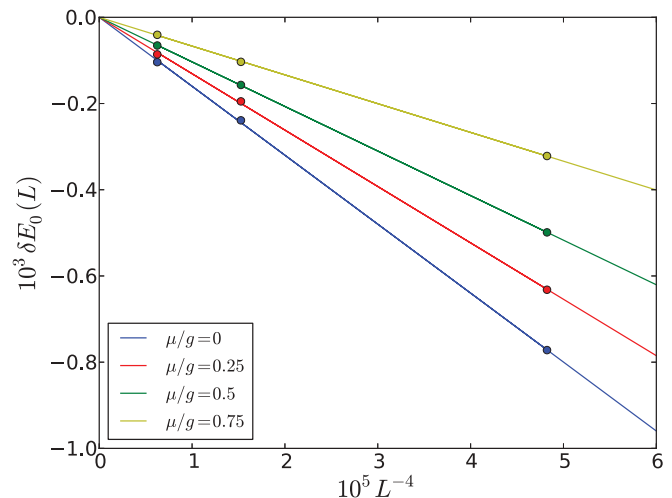


FIG. 15. (Color online) Finite-size scaling of the finite-size correction to the ground-state energy per site  $\delta E_0/N$  found in quantum Monte Carlo simulations of the quantum ice model  $\mathcal{H}_\mu$  [see Eq. (16)]. Results are shown for cubic clusters of  $N = 432, 1024$ , and 2000 sites, for parameters  $\mu/g = 0, 0.25, 0.5, 0.75$ , as function of the linear dimension of the system  $L = (a_0/2)(N/2)^{\frac{1}{3}}$ . The fact that  $\delta E_0/N \sim 1/L^4$  implies the existence of a linearly dispersing excitation—the photon of the underlying lattice gauge theory.

Approaching this problem from the lattice field theory  $\mathcal{H}'_{U(1)}$  [see Eq. (40)], we know that

$$c = \sqrt{\mathcal{U}\mathcal{K}}a_0 = \kappa^2\mathcal{K}\sqrt{\bar{\mathcal{U}}}a_0, \quad (107)$$

where the dimensionless parameter  $\bar{\mathcal{U}} = \frac{\mathcal{U}}{\mathcal{K}^2}$  can be determined separately from fits to structure factors (see Fig. 10). We also have enough information from the fits to the structure factor to evaluate the sum

$$\frac{1}{N} \sum_{\mathbf{k}} \frac{\omega(\mathbf{k})}{\kappa^2\mathcal{K}} = \frac{1}{\kappa^2\mathcal{K}} \left( \frac{E_0}{N} + \text{const} \right), \quad (108)$$

where  $E_0/N$  is the ground-state energy per site found from Monte Carlo simulations.

For  $\bar{\mathcal{U}} = 0$ , the LHS of Eq. (108) does not depend on  $L$ . This is consistent with simulations of the microscopic model at  $\mu = g$ . For  $\bar{\mathcal{U}} > 0$ , we expect a scaling law  $\sim 1/L^4$  for large  $L$ . We write

$$\epsilon(L) \equiv \frac{1}{N} \sum_{\mathbf{k}} \frac{\omega(\mathbf{k})}{\kappa^2\mathcal{K}} = \epsilon(\infty) - x_2 L^{-4}, \quad (109)$$

and it follows that

$$\frac{x_1}{x_2} = \kappa^2\mathcal{K} \quad (110)$$

with

$$c = \frac{x_2}{x_1} \sqrt{\bar{\mathcal{U}}}a_0, \quad (111)$$

where the coefficients  $x_1$  and  $x_2$  can be found from the finite-size scaling of the ground state energy in simulation (see Fig. 15), and through the numerical evaluation of the  $\sum_{\mathbf{k}}$  in Eq. (108) for a finite-size system.

We find that, for  $0 \leq \mu \leq 1$ , the evolution of the speed of light as a function of  $\mu$  is well described by

$$c^2 = \alpha\delta\bar{\mu} + \beta\delta\bar{\mu}^2 + \mathcal{O}(\delta\bar{\mu}^3), \quad (112)$$

where

$$\delta\bar{\mu} = 1 - \mu/g, \quad (113)$$

$$\alpha = 0.22 g^2 a_0^2, \quad (114)$$

$$\beta = 0.13 g^2 a_0^2. \quad (115)$$

In particular, for  $\mu = 0$ , the physical point of our model, we find

$$c = (0.6 \pm 0.1) g a_0 \hbar^{-1}, \quad (116)$$

where we have restored the dimensional factor of  $\hbar$ . We have also calculated an upper bound on  $c$  from a single mode approximation, in the spirit of Ref. 46. We find

$$c \leq (0.6 \pm 0.1) g a_0 \hbar^{-1}. \quad (117)$$

Within errors, the two numbers are indistinguishable.

It is interesting to use this result to make an order of magnitude estimate of the speed of light in a quantum-spin-ice material. Considering  $\text{Yb}_2\text{Ti}_2\text{O}_7$ , as (presently) the best-characterized material, and inserting the exchange parameters obtained by Ross *et al.*<sup>27</sup> into the expression for the tunneling matrix element [see Eq. (13)], we obtain

$$g_{\text{Yb}_2\text{Ti}_2\text{O}_7} \approx 0.05 \text{ meV}. \quad (118)$$

From Eq. (116) and the known size of the unit cell  $a_0 = 10.026 \text{ \AA}$  (see Ref. 26), we find a speed of light

$$c \sim 0.3 \text{ meV \AA} \sim 50 \text{ ms}^{-1}, \quad (119)$$

which implies a photon bandwidth

$$\Delta\omega \sim 0.1 \text{ meV}, \quad (120)$$

within the range accessible to modern inelastic neutron scattering experiments.<sup>80</sup>

The accuracy of this estimate is limited by the approximations made in setting up the minimal model of a quantum spin ice  $\mathcal{H}_{\text{tunneling}}$  [see Eq. (12)], and so it should only be regarded as a “ballpark” figure. It should also be remembered that  $\text{Yb}_2\text{Ti}_2\text{O}_7$  is believed to order ferromagnetically at the lowest temperatures.<sup>29,70</sup> However, as long as a given system remains an “ice,” the inclusion of further tunneling processes beyond  $\mathcal{H}_{\text{tunneling}}$  should only increase the speed of light.

#### IV. “ELECTROMAGNETISM” IN A QUANTUM SPIN ICE AT FINITE TEMPERATURE

In Sec. III B, we have demonstrated that the field theory  $\mathcal{H}'_{\text{U}(1)}$  [see Eq. (40)]—quantum electromagnetism on a pyrochlore lattice—gives an excellent account of the results of zero-temperature quantum Monte Carlo simulations of the minimal microscopic model of a quantum spin ice,  $\mathcal{H}_\mu$  [see Eq. (16)]. These results confirm the conjecture that this model could support a spin-liquid phase, down to  $T = 0$ . Encouraged by this, we now use the same field theory to explore how correlations in this spin liquid state develop at finite temperature.

In Sec. IV A, we assess how the thermal excitation of magnetic photons changes the temperature dependence of the energy-integrated structure factors measured in quasielastic scattering. We find that pinch points eliminated by quantum

fluctuations at zero temperature are progressively restored as the temperature of the spin liquid is raised.

In Sec. IV B, we compare the results of the lattice field theory with published results for quantum Monte Carlo simulations of quantum charge ice at finite temperature.<sup>59</sup> We find that both the form and the temperature dependence of the correlations are well described by the lattice field theory. Finally, in Sec. IV C, we conclude with a few remarks about the finite temperature behavior of the heat capacity and uniform magnetic susceptibility in a quantum spin ice. Throughout this analysis we set  $\hbar = k_B = 1$ , restoring dimensional factors of  $\hbar$  and  $k_B$  only where we quote results for the coefficient of the heat capacity associated with photons.

##### A. Temperature dependence of structure factors

The qualitative changes in correlations between spins at finite temperature can most easily be understood within the continuum field theory  $\mathcal{S}_{\text{eff}}$  [see Eq. (93)]. The thermal excitation of photons enhances correlations of the magnetic field  $\mathcal{B}$  at small  $|\mathbf{k}|$ :

$$C_{\mu\nu}^{\mathcal{B}}(\mathbf{k}) = \frac{8\pi^4 k}{c\sqrt{1 + \left(\frac{\lambda_c k}{2\pi}\right)^2}} \left( \delta_{\mu\nu} - \frac{k_\mu k_\nu}{k^2} \right) \times \coth \left[ \frac{ck\sqrt{1 + \left(\frac{\lambda_c k}{2\pi}\right)^2}}{2T} \right] \quad (121)$$

and introduces a thermal de Broglie wavelength for the photons,

$$\lambda_T = \frac{\pi c}{T}. \quad (122)$$

Over sufficiently long distances, this enhancement of correlations exactly cancels their suppression by quantum fluctuations. Assuming that  $\lambda_c \ll \lambda_T$  and expanding Eq. (121) for small wave number, we find

$$C_{\mu\nu}^{\mathcal{B}}(|\mathbf{k}| \ll 2\pi/\lambda_T) = \frac{16\pi^4 T}{c^2} \left( \delta_{\mu\nu} - \frac{k_\mu k_\nu}{k^2} \right) + \dots \quad (123)$$

This implies that, for these small wave vectors, the pinch point is restored, but with a prefactor that depends linearly on temperature.

This result has a very simple interpretation. At finite temperature, photons are only coherent quantum excitations over a length scale  $\lambda_T$ . Therefore while correlations in a quantum spin ice may decay as  $1/r^4$  over distances  $\lambda_c \ll r \ll \lambda_T$ , at long distances for  $r \gg \lambda_T$ , the classical  $1/r^3$  decay of the spin correlations is restored.

All of these arguments generalize to the lattice field theory  $\mathcal{H}'_{\text{U}(1)}$  [see Eq. (40)], and to expressions for the equal-time structure factor at finite temperatures derived from  $S_{\text{spin}}^{\alpha\beta}(\mathbf{k}, \omega)$  [see Eq. (89)]. Thus we anticipate that they will apply equally to a quantum spin ice at finite temperatures. This suggests a simple diagnostic for a quantum spin ice in quasielastic neutron scattering experiments—as the sample is cooled, and photons become coherent over longer length scales, the pinch points observed at reciprocal lattice vectors are progressively “bleached out.” This slow, cold, death of pinch points is illustrated in Fig. 16.



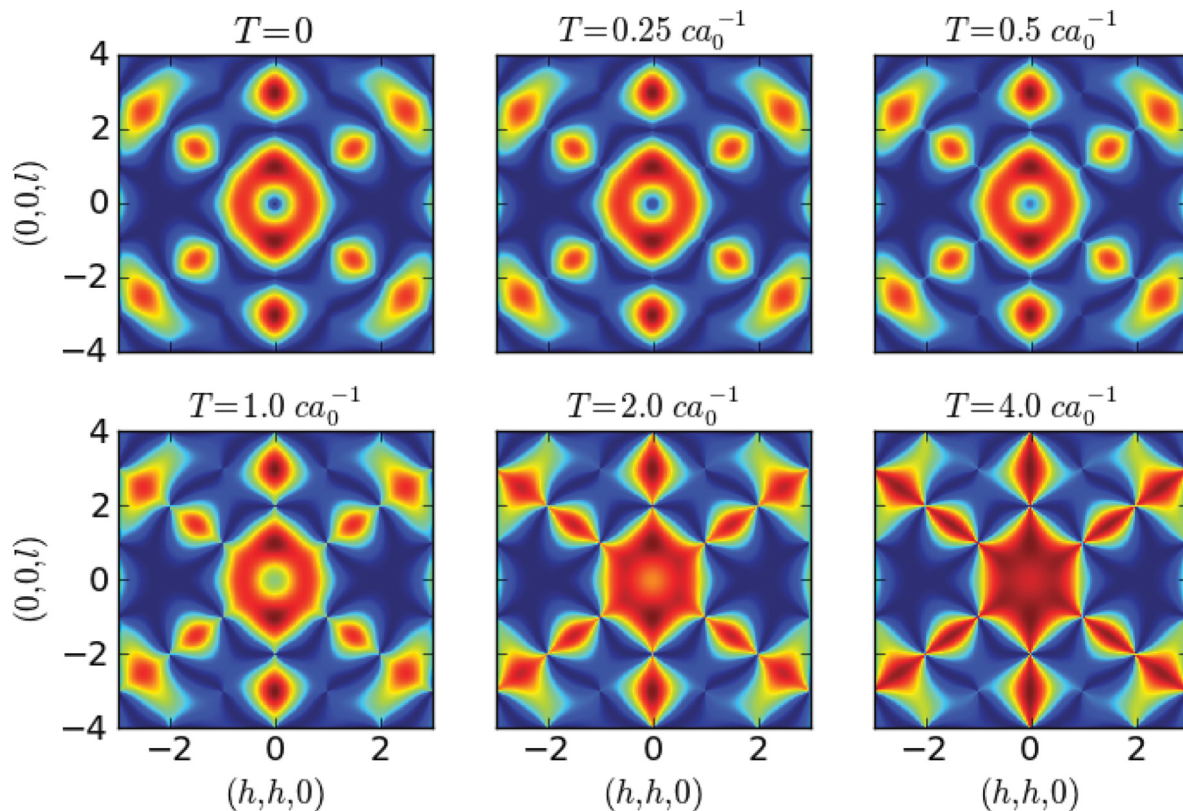


FIG. 16. (Color online) Slow, cold death of pinch points in a quantum ice. Equal-time structure factor  $S_{\text{spin}}^{yy}(\mathbf{k}, t=0)$  [see Eq. (90)] in the spin-flip channel measured by Fennell *et al.*,<sup>9</sup> calculated from the lattice field theory  $\mathcal{H}'_{\text{U}(1)}$  [see Eq. (40)], for comparison with neutron scattering experiments on a quantum spin ice. Results are plotted for temperatures ranging from  $T = 0$  to  $4.0 c a_0^{-1}$ , where  $c$  is the speed of light and  $a_0$  the linear dimension of the cubic unit cell, with temperature measured in units such that  $\hbar = k_B = 1$ . The pinch-point structure observed at finite temperature is progressively “hollowed out” as the system is cooled towards its zero-temperature ground state.

Since there is also a characteristic loss of spectral weight in  $S_{\text{spin}}^{\alpha\beta}(\mathbf{k}, t=0)$  for  $\mathbf{k} \approx \mathbf{0}$ , exactly the same process could be seen in the angle integrated structure factor measured in neutron scattering experiments on powder samples. In this case, the intensity of scattering is given by

$$I(k, T) \propto \sum_{\alpha\beta} \int d\Omega \left( \delta_{\alpha\beta} - \frac{k_\alpha k_\beta}{k^2} \right) S_{\text{spin}}^{\alpha\beta}(\mathbf{k}, t=0). \quad (124)$$

For classical spin ice, or a quantum spin ice at sufficiently high temperature,

$$I(k \approx 0, T) \approx \text{const.}$$

However, as a quantum spin ice is cooled to zero temperature, the growing coherence of photons will manifest itself as a progressive loss of spectral weight at small  $k$ ,

$$I(k=0, T) \sim T,$$

until, for  $T = 0$ , spectral weight at  $k = 0$  is eliminated entirely:

$$I(k \approx 0, T=0) \propto k.$$

This progression is illustrated in Fig. 17.

### B. Comparison with quantum Monte Carlo simulation

It is also interesting to compare the predictions of the lattice field theory  $\mathcal{H}'_{\text{U}(1)}$  [see Eq. (40)], with the results of finite-

temperature quantum Monte Carlo simulations of a quantum charge ice described by  $\mathcal{H}_{t,v}$  [see Eq. (17)], as published by Banerjee *et al.*<sup>59</sup> Banerjee *et al.* performed their simulations for hard-core bosons on a pyrochlore lattice at half-filling, with hopping integral  $t = 1$ , and nearest-neighbor repulsion  $V = 19.4$ , at temperatures  $T = 1.05g$  and  $1.57g$ , where

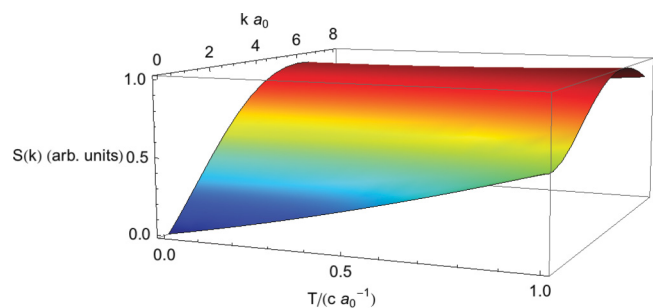


FIG. 17. (Color online) Angle-integrated scattering intensity  $I(k \approx 0, T)$  [see Eq. (124)] calculated from the lattice field theory  $\mathcal{H}'_{\text{U}(1)}$  [see Eq. (40)], for comparison with neutron scattering experiments on a powder sample of a quantum spin ice. Results are plotted for temperatures ranging from  $T = 0$  to  $1.0 c a_0^{-1}$ , where  $c$  is the speed of light and  $a_0$  the linear dimension of the cubic unit cell, with temperature measured in units such that  $\hbar = k_B = 1$ . The progressive elimination of pinch points as the sample is cooled manifests itself as a steady loss of scattering for  $|\mathbf{k}| \rightarrow 0$ .



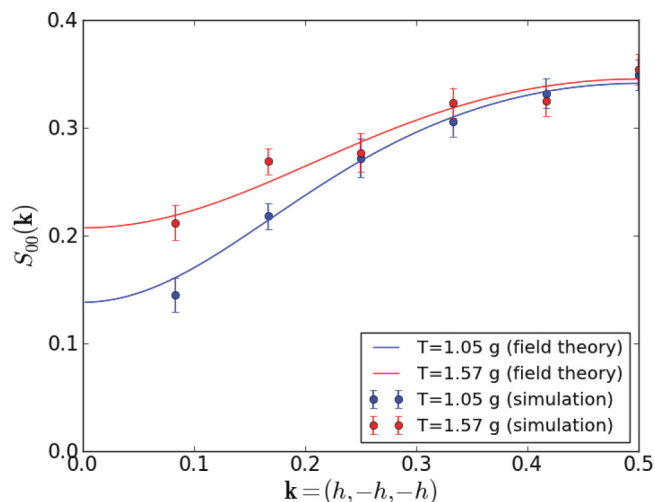


FIG. 18. (Color online) Comparison of the predictions of the lattice field theory  $\mathcal{H}'_{U(1)}$  [see Eq. (40)] with the results of finite-temperature quantum Monte Carlo simulations of a quantum charge ice described by  $\mathcal{H}_{t,v}$  [see Eq. (17)]. Results are shown for the equal time, on-sublattice structure factor  $S_{00}(\mathbf{k}) = \langle n_0(-\mathbf{k})n_0(\mathbf{k}) \rangle$ . Simulations are taken from Banerjee *et al.*<sup>59</sup> and were performed at temperatures  $T = 1.05g$  and  $1.57g$ , where  $g = 12t^3/V^2$  is the size of the leading tunneling matrix element between different charge ice configurations. The temperature dependence of the spin correlations makes it possible to estimate the speed of light  $c \approx 1.8 g a_0 \hbar^{-1}$ .

$g = 12t^3/V^2$  is the size of the leading tunneling matrix element between different charge ice configurations.

In Fig. 18, we plot simulation results for  $S_{\text{charge}}(\mathbf{k}, t = 0)$  at these temperatures, calculated within a single sublattice of pyrochlore lattice sites, together with the best fit to Eq. (83), projected onto a single sublattice. We assume that the parameters of the field theory depend relatively weakly on temperature, and attribute the temperature dependence of correlations entirely to the thermal excitations of photons. Under these assumptions, the lattice field theory gives a good account of both the form and the temperature dependence of  $S_{\text{charge}}(\mathbf{k}, t = 0)$ , within the error bars on points taken from simulation.

These fits suggest a speed of light

$$c = (1.8 \pm 0.2) g a_0 \hbar^{-1}, \quad (125)$$

which is  $\sim 3$  times larger than that found in Sec. III D from finite-size scaling of the ground-state energy of  $\mathcal{H}_\mu$  [see Eq. (16)]. This discrepancy can probably be attributed to the fact that the simulations of Banerjee *et al.* were performed close to the melting point of the charge ice,<sup>59</sup> where both interactions between photons, and tunneling processes involving more than six lattice sites, are likely to play an important role. Since all of these processes will contribute to the rate at which the gauge field fluctuates in time, they can be expected to increase the speed of light.

### C. Heat capacity and magnetic susceptibility at low temperatures

Neutron scattering experiments have the potential to give decisive information about emergent electromagnetism in a

quantum spin ice. However, these experiments are expensive and difficult to perform, and depend critically on the size and quality of available samples. We therefore conclude with a few brief remarks on potential signatures of a quantum  $U(1)$  liquid in thermodynamic quantities. The results given will hold in the low-temperature regime where the physics of a quantum spin ice can be described as a gas of photons. At higher temperatures, the thermal excitation of the gapped spinons (monopoles) and electric charges also play an important role.

We have seen in Sec. IV A how quantum fluctuations lead to an equal-time structure factor which, in the limit  $k \rightarrow 0$ , vanishes at low temperatures as

$$\lim_{k \rightarrow 0} S(k, T) \propto T. \quad (126)$$

This in turn implies a bulk magnetic susceptibility  $\chi(T)$ , which is independent of temperature at low temperatures:

$$\chi^{-1}(T \ll g) = \frac{3\mathcal{U}}{\kappa^2}, \quad (127)$$

where  $\mathcal{U}$  is the coefficient of  $\mathcal{B}^2$  in the effective Hamiltonian  $\mathcal{H}_{U(1)}$  [see Eq. (38)] and  $\kappa \approx 1$  is the dimensionless scale factor introduced in Eq. (74).

This result provides another means of parameterizing the lattice field theory. It is also a potentially useful diagnostic for experiment, since, a classical spin ice which remains in thermodynamic equilibrium at low temperatures, will exhibit an effective Curie law<sup>11,81</sup>

$$\chi^{-1}(T) \sim T. \quad (128)$$

This result follows directly from the fact that there are more spin ice configurations with vanishing magnetization  $\mathbf{M} = 0$  than with any finite magnetization  $\mathbf{M} \neq 0$ , and so, in the absence of any other considerations, a state with  $\mathbf{M} = 0$  is selected by an entropic term  $\delta\mathcal{F} = T\delta S \sim T \mathbf{M}^2$  in the free energy.<sup>8</sup> Nonetheless, any comparison with a classical spin ice should be approached with some caution, as these systems need not remain in equilibrium at low temperatures,<sup>82,83</sup> and the character of the spin fluctuations that control  $\chi(T)$  changes as a function of temperature.<sup>16,84</sup>

As noted elsewhere,<sup>47,70</sup> the fact that photons are linearly dispersing excitations implies that they must make a  $T^3$  contribution to the heat capacity at low temperatures. While this contribution has exactly the same temperature dependence as that from acoustic phonons, the large amount of entropy available in ice states, and low speed of light (see Sec. III D), mean that the heat capacity at low temperatures will be dominated by photons. The photon contribution to the heat capacity per mole is

$$C_{\text{photon(mole)}} = BT^3 \quad (129)$$

with the coefficient  $B$  given by

$$B = \left(\frac{\pi^2}{30}\right) R \left(\frac{k_B a_0}{\hbar c}\right)^3. \quad (130)$$

From the characterization of  $\text{Yb}_2\text{Ti}_2\text{O}_7$  by Ross *et al.*<sup>27</sup>, and the analysis of the speed of light in Sec. III D, we estimate

$$B \approx 65 \text{ J mol}^{-1} \text{ K}^{-4}, \quad (131)$$

which is several orders of magnitude larger than the expected phonon contribution. This should be compared with the value

$$1 \text{ J mol}^{-1} \text{ K}^{-4}$$

obtained in Ref. 70. We note that, since the photons are magnetic excitations, measurements of the heat capacity in an applied magnetic field may also prove instructive.

## V. DISCUSSION AND CONCLUSIONS

In this paper we have developed a detailed theory for the simplest microscopic model that could describe quantum tunneling between different spin ice configurations [see Eq. (12)]. The striking claim that this type of model could support a spin liquid phase that perfectly mimics quantum electromagnetism<sup>47</sup> has been verified by quantum Monte Carlo simulations.<sup>58,59</sup> Here we have explored how such a quantum spin liquid might manifest itself in experiment, parameterizing an “electromagnetic” lattice gauge theory from quantum Monte Carlo simulations at zero temperature, and using this to calculate the dynamical structure factor  $S^{\alpha\beta}(\mathbf{k}, \omega)$  [see Eq. (89)], which would be measured in neutron scattering experiments at finite temperature.

We find that a key signature of the emergent electromagnetism is the suppression of pinch points singularities in the energy-integrated structure factor  $S^{\alpha\beta}(\mathbf{k}, t=0)$  as the system is cooled to its zero-temperature ground state (see Fig. 16). This will coincide with the appearance of a gapless, linearly dispersing, mode—the photon of the lattice gauge theory—in inelastic neutron scattering (see Fig. 13). In sharp contrast with a conventional antiferromagnet, the dispersing feature associated with this photon vanishes as  $\omega \rightarrow 0$ . These photons will also strongly influence the low-temperature thermodynamic properties of the system, giving rise to a temperature-independent contribution to the magnetic susceptibility [see Eq. (127)] and an anomalously large  $T^3$  contribution to specific heat [see Eq. (129)].<sup>70</sup>

Neither the idea of “artificial light,”<sup>50–55</sup> nor the observation that there could be quantum tunneling between different spin ice configurations,<sup>85</sup> is new. However, the possibility that one might lead to another is both new and exciting, and adds to the general frisson surrounding pyrochlore magnets. Without attempting to review all of this fast-developing field—but with the possibility of observing photons in mind—it is interesting to ask whether any of the materials currently studied “fit the bill.”

The most widely studied example of a three-dimensional spin liquid is the insulating pyrochlore oxide  $\text{Tb}_2\text{Ti}_2\text{O}_7$ .<sup>6</sup>  $\text{Tb}_2\text{Ti}_2\text{O}_7$  does not order down to 50 mK (see Ref. 23), despite having a Curie-Weiss temperature  $\theta_{\text{CW}} \sim 14$  K (see Ref. 18), and a strong tendency to order under magnetic field or pressure.<sup>86,87</sup> In a series of papers, Gingras and coauthors have argued that  $\text{Tb}_2\text{Ti}_2\text{O}_7$  is a quantum spin ice in which spins fluctuate strongly about the crystallographic [111] axes. These claims were made on the basis of a characteristic checkerboard structure observed in diffuse neutron scattering experiments at high temperatures,<sup>19,88</sup> and a subsequent microscopic analysis of crystal field levels,<sup>20,89</sup> and find support in the recent

observation of partial magnetisation plateau for magnetic field applied along a [111] axis.<sup>21,90</sup>

Within this framework, the field at which the plateau is observed implies that the energy scale relevant for “quantum-spin-ice” behavior in  $\text{Tb}_2\text{Ti}_2\text{O}_7$  is  $J_{\text{eff}} \approx 0.2$  K.<sup>90</sup> Unfortunately, the interpretation of experiment at these low temperatures is muddled by questions of sample quality, with inconsistent results for spin-freezing obtained by different authors.<sup>23,24,91–93</sup> Published thermodynamic data at low temperatures is also less than conclusive, showing hints of a saturation of  $\chi(T)$  at low temperatures, but strong sample dependence.<sup>23,92–95</sup> And the picture is further complicated by strong fluctuations of the lattice,<sup>96,97</sup> with alternative theories of  $\text{Tb}_2\text{Ti}_2\text{O}_7$  building on lattice effects.<sup>98–100</sup>

At present, there is no published neutron scattering data for  $\text{Tb}_2\text{Ti}_2\text{O}_7$  with the combination of  $k$  resolution, energy resolution and low temperature needed to compare with the predictions in Secs. IID and IIIC of this paper. However, recent evidence of “pinch-point” structure in quasielastic neutron scattering on single crystals of  $\text{Tb}_2\text{Ti}_2\text{O}_7$ ,<sup>25</sup> taken together with inelastic neutron scattering experiments on powder samples,<sup>101</sup> suggest that the comparison might be interesting. The latter find evidence of a quasielastic feature evolving into two bands of excitations at temperatures  $T < 0.4$  K.<sup>101</sup> If—and it remains a big if—the behavior of  $\text{Tb}_2\text{Ti}_2\text{O}_7$  is connected with the physics of the quantum ice described in this paper, it would be tempting to identify these bands with the excitations of electromagnetism on a lattice: gapless photons, together with gapped “electric” and “magnetic” charges (spinons). But more, and more delicate, experiments will be needed to determine whether this is indeed the case. And ultimately,  $\text{Tb}_2\text{Ti}_2\text{O}_7$  will remain a fascinating system to study, regardless of whether or not it is a quantum spin ice.

Recently, there has also been intense experimental and theoretical interest in the closely-related Yb pyrochlore,  $\text{Yb}_2\text{Ti}_2\text{O}_7$ . Originally identified in the pioneering survey of Blöte *et al.*<sup>102</sup> as a ferromagnet with  $T_c = 0.21$  K and  $\theta_{\text{CW}} = 0.4$  K,  $\text{Yb}_2\text{Ti}_2\text{O}_7$  differs from the classical spin ice materials  $\text{Ho}_2\text{Ti}_2\text{O}_7$  and  $\text{Dy}_2\text{Ti}_2\text{O}_7$  in that the lowest-lying crystal-field state is a Kramers doublet with easy-plane anisotropy.<sup>32,67,68</sup> An XY ferromagnet on a pyrochlore lattice—modern estimates suggest  $\theta_{\text{CW}} \approx 0.65$  K for  $\text{Yb}_2\text{Ti}_2\text{O}_7$  (see Refs. 67 and 106)—would naturally be expected to order ferromagnetically at low temperatures. However,  $\text{Yb}_2\text{Ti}_2\text{O}_7$  exhibits a far more complicated phenomenology.

Neutron scattering experiments at temperatures below 10 K find diffuse liquidlike structure that offers evidence of anisotropic exchange interactions.<sup>26,29</sup> At temperature of order 1 K, rodlike structure emerges, reminiscent of a dimensional crossover.<sup>26,28,103–105</sup> Some authors have found evidence of a first-order transition into a ferromagnetically ordered state at  $T_c = 0.24$  K,<sup>29,106</sup> although this has been contested, and may not occur in all samples.<sup>67,103,104,107,108</sup>

That  $\text{Yb}_2\text{Ti}_2\text{O}_7$  orders ferromagnetically in applied magnetic field is, however, uncontroversial. And this has made it possible for Ross *et al.*<sup>27</sup> to accurately characterise an exchange Hamiltonian for  $\text{Yb}_2\text{Ti}_2\text{O}_7$  [see Eq. (10)] from fits to spin wave excitations in the polarised state. The parameters obtained confirm that the dominant interactions in  $\text{Yb}_2\text{Ti}_2\text{O}_7$

favor “ice” states, but that these are complimented by terms which will drive significant fluctuations at low temperatures.

Reassuringly, this description of  $\text{Yb}_2\text{Ti}_2\text{O}_7$  is also in quantitative agreement with measurements of thermodynamic properties over a wide range of temperatures.<sup>30</sup> This makes  $\text{Yb}_2\text{Ti}_2\text{O}_7$  the best-characterised quantum spin ice, and as such, it is a natural place to look for emergent electromagnetism. However, neutron scattering data with sufficient resolution to compare with the predictions of this paper are not, as yet, available.

$\text{Tb}_2\text{Ti}_2\text{O}_7$  and  $\text{Yb}_2\text{Ti}_2\text{O}_7$  are by no means the only pyrochlore systems with spin liquid properties,<sup>6</sup> and some of these other systems, notably  $\text{Pr}_2\text{Sn}_2\text{O}_7$ <sup>31,32,109</sup> and  $\text{Pr}_2\text{Zr}_2\text{O}_7$ <sup>32–34</sup> are also worth investigating as potential realisations of a quantum ice. It might also be interesting to revisit two-dimensional ice-type materials, such as the proton bonded ferroelectric copper formate tetrahydrate.<sup>41</sup> While two-dimensional quantum ice models are known to order at low temperatures,<sup>110–115</sup> they are described by the same class of lattice gauge theory, and possess the same spinon excitations as their three-dimensional counterparts.<sup>40,111,115</sup> These excitations will be confined in the ordered state, but might be visible at finite energy, and above the ordering temperature.

Although the theoretical possibility of emergent electromagnetism in quantum ice<sup>47,48,58,59</sup> and quantum

dimer<sup>46,60,61,72</sup> models is now well-established, many theoretical questions remain open. In this paper, we have considered only the simplest microscopic model of a quantum spin ice [see Eq. (16)], and fully characterized only its photon excitations. The study of more realistic models, and of other excitations, is still in its infancy.<sup>30,70,74</sup> We have also made no attempt to resolve the question of how the quantum ice state that we find at low temperatures, becomes a classical ice at high temperatures. All of these issues remain for future study. But we believe that the best motivation for studying them is experiment, and hope that the results in this paper will encourage further experiments on spin liquid materials that might realize artificial light.

## ACKNOWLEDGMENTS

The authors are pleased to acknowledge helpful conversations with Steven Bramwell, John Chalker, Peter Fulde, Bruce Gaulin, Michel Gingras, Paul McClarty, Roderich Moessner, Karlo Penc, Frank Pollmann, Lucile Savary, and Alan Tennant. We are particularly grateful to Tom Fennell and Radu Coldea for critical readings of the manuscript. This work was supported by EPSRC Grants EP/C539974/1 and EP/G031460/1. O.S. and N.S. gratefully acknowledge the hospitality of the guest program of MPI-PKS Dresden.

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