Disorder-Induced Quantum Spin Liquid in Spin Ice Pyrochlores

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(Received 15 April 2016; revised manuscript received 2 November 2016; published 23 February 2017)

We propose that in a certain class of magnetic materials, known as non-Kramers "spin ice," disorder induces quantum entanglement. Instead of driving glassy behavior, disorder provokes quantum superpositions of spins throughout the system and engenders an associated emergent gauge structure and set of fractional excitations. More precisely, disorder transforms a classical phase governed by a large entropy, classical spin ice, into a quantum spin liquid governed by entanglement. As the degree of disorder is increased, the system transitions between (i) a "regular" Coulombic spin liquid, (ii) a phase known as "Mott glass," which contains rare gapless regions in real space, but whose behavior on long length scales is only modified quantitatively, and (iii) a true glassy phase for random distributions with large width or large mean amplitude.

Entanglement defines the essential nonclassical features of quantum mechanics. While entanglement has been achieved and controlled for small numbers of quantum bits ("qubits"), *many-body* entanglement of a thermodynamically large system is an exciting frontier [1,2]. Long range entanglement engenders exotic phenomena such as fractional quantum numbers and emergent topological excitations, and is important not only in the realm of materials, but even in the theory of fundamental forces [3]. Theoretically, the exemplars of such massive "long range" entanglement are quantum spin liquids (QSLs), states of quantum magnets in which electronic spins reside in macroscopic superpositions of infinitely many microstates [4].

QSLs have been elusive experimentally, in part because disorder induces competing glassy states instead of entangled ones. However, here we show that this need not be the case, and propose to *use* the disorder itself to generate long-range entanglement. Because disorder is not intrinsic, it can be readily tuned so that serendipity is no longer required to find the QSL state. The essential ingredients are present in spin ice materials [5,6] such as $Ho_2Ti_2O_7$ and $Pr_2Zr_2O_7$ with *non-Kramers* magnetic ions. In the classical limit—an excellent approximation for $Ho_2Ti_2O_7$—and without disorder, these materials have Ising spins on a pyrochlore lattice of corner-sharing tetrahedra, with a frustrated interaction that selects an extensive set of ground states: those with two spins in and two spins out on each tetrahedron. We construct a model for disorder in these materials which naturally introduces the quantum fluctuations sufficient to generate a QSL from a massive superposition of the two-in-two-out states. We show that this model indeed supports not one but two QSL phases, one of which is a long range entangled analog of the Mott glass phase of disordered bosons [7–9]. On application of a physical magnetic field, we obtain an even more glassy "Bose glass" QSL phase [10]. We

emphasize these are true QSLs with long range entanglement, emergent gauge structure, and exotic nonlocal excitations. The glassy QSLs differ from the pure QSL by having additional gapless but localized excitations at low energy. To our knowledge, this is the first proven example where true QSL states are engendered by disorder. Our model applies to the archetypal *classical* spin ice material $Ho_2Ti_2O_7$, and predicts that it can be tuned to a *quantum* spin liquid by controlled introduction of disorder. The full phase diagram is shown in Fig. 1.

Our analysis begins with the atomic physics of trivalent rare earth ions in the spin ice pyrochlores [11]. In many, e.g., Pr^{3+} and Ho^{3+}, the low energy states of the magnetic ion comprise a non-Kramers doublet with degeneracy protected by the D_{3d} point group symmetry, described by a pseudo-spin $1/2$ operator $\vec{S}_i$. Under time-reversal symmetry, in the local basis aligned with the $\langle 111 \rangle$ axis of
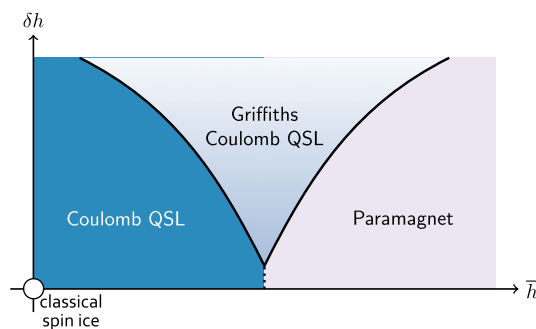

FIG. 1. Phase diagram in the mean strength of disorder $\bar{h}$—disorder δh plane. The dotted line indicates a first order transition, while the solid lines represent second order transitions or crossover (between the Coulomb QSL and Griffiths Coulomb QSL). In the disordered boson language, the paramagnet is a "superfluid" (Higgs) phase, the Griffiths phase is a Mott glass, and the Coulomb QSL is a "Mott insulator." The clean spin ice point sits at $\bar{h} = \delta h = 0$ and is represented by a white circle.

the site, the "up" and "down" spin levels interchange: i.e., $\hat{\Theta}|\pm\frac{1}{2}\rangle = |\mp\frac{1}{2}\rangle$, where $\hat{\Theta}$ is the anti-unitary time-reversal operator. Note that $\hat{\Theta}^2 = +1$, which defines the non-Kramers case. It follows that $S_i^z \to -S_i^z$ under time-reversal, while S_i^x and S_i^y are time-reversal invariant.

In clean spin ice systems, an excellent first approximation to the Hamiltonian is given by the nearest-neighbor spin ice Hamiltonian,

$$H_0 = J\sum_{\langle ij \rangle} S_i^z S_j^z - \mathbf{B} \cdot \sum_i g S_i^z \hat{\mathbf{e}}_i. \qquad (1)$$

The first term, with $J > 0$, is a frustrated Ising interaction between spins. It appears antiferromagnetic in the local basis but represents ferromagnetic coupling of the magnetic moments in a global frame. The second term is the only symmetry allowed interaction of the magnetic field with the spins in the non-Kramers case: the magnetic moment operator is, by symmetry, $\mathbf{m}_i = g S_i^z \hat{\mathbf{e}}_i$. Quantum exchanges coupling in plane components S_i^x, S_i^y on nearest-neighbor sites can also occur, but are small in Ising-like systems. For example, in Pr^{3+}, it is estimated that the probability to be in the maximal $j_z = \pm 4$ states of the $j = 4$ levels is 93% [12], while Ho^{3+}, which has $j = 8$, is even more Ising-like.

Now we examine the effect of disorder. We consider *nonmagnetic* disorder maintaining rare-earth stoichiometry, and assume there is no ordered Jahn-Teller distortion as appears to be the case in experiments. Rather, disorder generates (electrostatic) crystal fields which lower the symmetry of the rare-earth site, and hence can split the non-Kramers doublet. Because of time-reversal symmetry, these crystal fields couple directly to the in plane components of S_i. Hence disorder adds the term

$$H' = -\frac{1}{2}\sum_i (\eta_i^* S_i^- + \eta_i S_i^+), \qquad (2)$$

where η_i is a random complex number, acting as an XY "random field" (though we caution there is no true field, and H' is time-reversal invariant). In general, the problem is specified by giving the full distribution of the random fields, $P[\{\eta_i\}]$, and the statistical space group symmetry of the crystal should be respected by this distribution. We will largely focus on the simplest limit of independent, identically distributed random variables, i.e., $P[\{\eta_i\}] = \prod_i p(\eta_i)$ (but this is not essential).

The full Hamiltonian, $H = H_0 + H'$, defines a quenched random transverse field Ising model. It can be simplified by defining $\eta_i = h_i e^{i\alpha_i}$, where $h_i > 0$ is real and $0 \leq \alpha_i < 2\pi$. The phase α_i can be removed by a basis rotation around the local z axis, generated by the unitary operator $U = \prod_i e^{i\alpha_i S_i^z}$. After the transformation, we have

$$H \to U^\dagger H U = J\sum_{\langle ij \rangle} S_i^z S_j^z - \sum_i h_i S_i^x - B \cdot \sum_i g S_i^z \hat{\mathbf{e}}_i. \quad (3)$$

We see that in zero applied field, $\mathbf{B} = \mathbf{0}$, this is really the standard transverse field Ising antiferromagnetic model, with random magnitudes of the transverse field, drawn from some distribution $p(h)$. We expect that a variety of distributions can be tuned experimentally (see Supplemental Material [32]).

Perturbative regime: $h_i \ll J$.—When all or nearly all the $h_i \ll J$, (i.e., the probability that $h > fJ$, with f a small fraction of 1, is small: $\int_{fJ}^\infty p(h)dh \ll 1$) we may apply perturbation theory. We obtain the effective Hamiltonian at *sixth* order in the transverse fields within the degenerate manifold of classical spin ice states (Supplemental Material [32]):

$$H_{\text{eff}} = -\sum_{\bigcirc} (K_{ijklmn} S_i^+ S_j^- S_k^+ S_l^- S_m^+ S_n^- + \text{H.c.}), \quad (4)$$

where $K_{ijklmn} = (63 h_i h_j h_k h_l h_m h_n / 16 J^5)$. Equation (4) defines a "random ring exchange" model. As shown first by Hermele *et al.* [13], when K is constant, the ring exchange model has the structure of a compact U(1) gauge theory, in which $S_i^\pm$ plays the role of a U(1) gauge connection (exponential of a gauge field) on the links of the dual diamond lattice formed from the tetrahedron centers, and S_i^z acts as the conjugate "electric" field. On general grounds, such a theory can support a trivial "confined" phase which is short range entangled and a deconfined Coulomb phase, which is long-range entangled [14]. In the latter, the compactness is unimportant and the low energy physics is an emergent quantum electro-dynamics, with a gapless photon and gapped electric and magnetic charged quasiparticles. This is a U(1) QSL phase. Numerical studies have shown that the ground state of this specific model for constant K is in the U(1) QSL phase [15–17].

Weak randomness: $\delta h \ll \bar{h}$.—Let us now consider first "weak" randomness, i.e., a distribution $p(h)$ peaked around $\bar{h}$ with small width $\delta h \ll \bar{h}$. The obvious potential insta-bilities of the U(1) QSL phase are due to vanishing gaps for electric and magnetic charges. The electric charges (in standard quantum conventions) correspond to tetrahedra violating the ice rules, and in the perturbative limit have a gap of order $J \gg K$, and hence remain gapped regardless of the distribution $p(h)$. The magnetic charges have a gap of order $\bar{K} \sim \bar{h}^6/J^5$, which is still much larger than the random perturbation to H_{eff} which is of order $\delta K \sim (\bar{h}/J)^5 \delta h$. Thus the gap to magnetic charges is also robust.

What of the photon? The absence of magnetic charges justifies the continuum limit, for which symmetry implies

$$H_{\text{photon}} = \int d^3 x \left\{ \frac{\epsilon}{2}(1 + v_E[x])|E|^2 + \frac{1}{2\mu}(1 + v_B[x])|B|^2 \right\}, \quad (5)$$

where $v_E(x)$ and $v_B(x)$ are zero-mean random functions of space, and ϵ, μ are the effective dielectric constant and

magnetic permeability, respectively. Simple power counting shows that both random terms are strongly *irrelevant* at low energy and long distances (with short-range correlations, $[v] = L^{-3/2}$ in three dimensions). The key point is that gauge invariance forces disorder only to couple to E and gradients of the vector potential A, so that, even if we relax the constraint of time-reversal symmetry in Eq. (5), the photon remains stable. This is similar to the suppression of the scattering of acoustic phonons at low energy in a disordered crystal [18], and the lack of localization of light in a disordered photonic material at low frequency [19].

Larger disorder: $\delta h \sim \bar{h}$.—We have established the stability of the U(1) QSL with weak disorder. Now let us consider increasing the disorder, still within the perturbative regime, i.e., the random ring model with $\delta h \sim \bar{h}$. In general, the ground state depends now on the full distribution $p(h)$ [or the induced distribution $p(K)$]. The gap to electric charges remains robust, but the magnetic gap may close, leading to confinement. The physical mechanism whereby this might occur is order by disorder [20,21]. The ring Hamiltonian, Eq. (4), is a kind of "hopping" in the high dimensional manifold of classical spin ice states. In the uniform case, the ground state is delocalized across an extensive subset of this manifold: this is the QSL state, which obtains the same energy for each ring term. We can also imagine a different state which gets a lower energy for some "strongly resonating" ring terms (better than the delocalized state) but sacrifices energy for other rings—in the nonrandom case this necessarily breaks lattice symmetries. Above some threshold width of the distribution $p(h)$—which is *a priori not* small—such a confined "order by disorder" state may occur. The confinement transition to such a state has a dual interpretation as condensation of the magnetic charged excitations of the QSL phase.

Nonperturbative case: $h_i \sim J$.—When the transverse fields are not small, the perturbative treatment no longer applies. Instead, we adopt the slave rotor representation introduced for the uniform quantum spin ice problem in Ref. [22], and discuss the full phase diagram in this framework. This is an exact rewriting of the original spin system, by introducing explicit operators to track spinons (or electric charges) on the sites $a, b, \dots$ of the diamond lattice. The charge is $Q_a = \epsilon_a \sum_{i \in a} S_i^z$, where $\epsilon_a = +1(-1)$ on the diamond A (B) sublattice. A conjugate phase φ_a is defined by $[\varphi_a, Q_b] = i\delta_{ab}$. Then the spin operators are rewritten as $S_i^z = s_{ab}^z$ and $S_i^+ = \Phi_a^\dagger s_{ab}^+ \Phi_b$, where a, b are the two tetrahedra sharing site i, on the A and B sublattices, respectively, and $\Phi_a = e^{-i\varphi_a}$. The s_{ab}^μ spins are canonical spin-1/2 degrees of freedom, and for convenience we define $s_{ba}^z = -s_{ab}^z$ and $s_{ba}^\pm = s_{ab}^\mp$. Then the Hamiltonian, Eq. (3) becomes

$$H = \frac{J}{2} \sum_a Q_a^2 - \frac{1}{2} \sum_{\langle ab \rangle} h_{ab} [\Phi_a^\dagger s_{ab}^+ \Phi_b + \text{H.c.}]. \qquad (6)$$

This Hamiltonian contains a potential term, and a kinetic term that represents electric charges (spinons) hopping on top of a fluctuating background gauge field, and which appears only for nonzero disorder. The coupling of the spinons to the gauge field leads to a strongly-interacting problem.

Gauge mean field theory: No gauge field fluctuations.—First, we discuss an approximate solution obtained by gauge mean field theory (gMFT) [22], which, in the present case essentially consists in suppressing the fluctuations of the gauge field. Namely, we perform the replacement $\Phi^\dagger s \Phi \to \Phi^\dagger \Phi \langle s \rangle + \langle \Phi^\dagger \Phi \rangle s - \langle \Phi^\dagger \Phi \rangle \langle s \rangle$. The resulting mean field Hamiltonian is composed of two decoupled parts, a "spin" $\vec{s}$ in a random field, and a quadratic spinon hopping Hamiltonian:

$$H_\Phi = \frac{J}{2} \sum_a Q_a^2 - \frac{1}{2} \sum_{\langle ab \rangle} [t_{ab} \Phi_a^\dagger \Phi_b + \text{H.c.}] \qquad (7)$$

$$= \frac{J}{2} \sum_a Q_a^2 - \sum_{\langle ab \rangle} t_{ab} \cos(\varphi_a - \varphi_b), \qquad (8)$$

with $t_{ab} = h_{ab} \langle s_{ab}^+ \rangle$, which we assumed to be real in the right-hand side expression, as is indeed the case for the gMFT solution. We recognize this as the Hamiltonian of a (three-dimensional) array of Josephson junctions, i.e., a quantum XY, or rotor, model, coupling "grains" on the diamond lattice with random Josephson coupling t_{ab}.

Uniform field.—While our primary interest is in disorder, we first consider the case of a uniform h, for which Eq. (6) is translationally invariant, and we make the ansatz that $\langle s \rangle$ (and hence t_{ab}) also be uniform. Then the quantum XY model in Eq. (7) is expected to have two phases: a "superfluid" state with $\langle e^{i\varphi_a} \rangle \neq 0$ and a Mott insulator phase with $\langle e^{i\varphi_a} \rangle = 0$ and a gap to all excitations. The superfluid state corresponds to the Higgs phase of the gauge theory—the trivial transverse polarized state of the original model. A calculation in Appendix B [32] locates the Higgs transition, where the spinons become gapless, between the Mott and superfluid states approximately at $(h/J)_c \approx 0.35$. Below $(h/J)_c$ the system is in the Coulomb phase ("Mott"), and characterized within gMFT by $\langle \Phi \rangle = 0$. In this phase, fluctuations around the mean field solution reproduce the photon Hamiltonian, cf. Eq. (5). We expect that the transition to the paramagnetic phase in the disordered case occurs at a similar magnitude of $\bar{h}/J$.

Random field.—Now we return to the full problem with random h_{ab}, hence random t_{ab}. The gMFT Hamiltonian in Eq. (7) then describes a well-studied "dirty boson" problem, notably with *particle-hole symmetry* $Q_a \to -Q_a$, $\varphi_a \to -\varphi_a$. We can trace this back to the time-reversal symmetry of the original model. Because of disorder, an additional phase emerges between the Mott insulator and superfluid: a gapless insulating state which has been called a "Mott glass" [7–9]. In most respects the Mott glass is similar to the Mott insulator (the Coulomb phase in spin

language), but differs by the presence of rare regions which look locally superfluid (trivial, paramagnetic, polarized), and consequently have very small gaps controlled by their finite size. In an infinite system, arbitrarily large regions of this type can be found, leading to a "Griffiths phase" [23] with a vanishing gap in the thermodynamic sense. Because of particle-hole symmetry, the superfluid regions are exceedingly rare, and numerics suggest [9] that they are exponentially distributed in their size, i.e., the density of superfluid regions of s sites decays as $e^{-(s/s_0)^\gamma}$, with $\gamma \approx 1$ and s_0 a constant. This in turn implies that the largest superfluid cluster in a system of size L grows logarithmically, $s_{max} \sim \ln L$. Since a superfluid region of size s has a gap of order $1/s$, the finite-size gap of the Mott glass is therefore order $\Delta_L \sim 1/\ln L$, which vanishes in the thermodynamic limit.

Beyond gMFT.—These properties need to be translated into their *physical* consequences for the full problem, beyond gMFT. A picture is as follows: fluctuations convert the Mott insulating bulk with superfluid inclusions to a Coulombic bulk with Higgs inclusions—a Coulombic Griffiths phase. This is analogous simply to a dielectric medium with embedded superconducting grains [24]. The latter exclude the gauge fields and act as low energy hosts for electric charges—specifically, the "charging energy" for a grain of linear size ℓ is of order $1/\ell$. The spinon gap vanishes, with the gapless low energy spinon states localized on these grains. Modifications of the photon are like those of an electromagnetic wave in a dielectric + superconducting "metamaterial." Such waves are *insensitive* to rare regions but are dominated by typical ones. At low frequency, the system behaves as an effective medium with an enhanced dielectric constant, but at frequency and wave vector scales comparable to the gap and inverse typical size or spacing of the Higgs regions, the photon will scatter and develop an intrinsic, disorder-dominated, linewidth. Since the emergent photon continues to remain gapless and propagate, and electric and magnetic charges can still exist in isolation in the system, the Griffiths phase should still be considered a Coulombic spin liquid.

In the full treatment, the mean-field superfluid phase, becomes the confined paramagnetic phase. For weak disorder, i.e., $\delta h \ll \bar{h}$, there can be a true gapped paramagnet, but for strong disorder, we expect a zero gap state with localized low energy excitations—a Griffiths paramagnet. So the zero temperature phase diagram contains both the usual Coulombic liquid with gapped electric and magnetic charges, a Griffiths Coulomb liquid with gapless electric charges, and the thermally insulating, unentangled paramagnetic state. It is worth noting that the application of a physical magnetic field (which couples to S_i^z rather than $\mathsf{S}_i^\pm$) breaks time-reversal symmetry and hence the particle-hole symmetry of the emergent gauge theory. Consequently, it converts the Mott glass to a Bose glass, which has much stronger Griffiths effects. The

experimental ramifications would be a an excellent subject for future research. Conversely, any additional type of disorder, such as microscopic exchange randomness or lattice strains, which do not break time-reversal symmetry, cannot destabilize the Mott glass, as they preserve the emergent particle-hole symmetry between positive and negative charges.

Phase transitions.—Disorder has major effects upon the transition from the QSL to the trivial state. In mean field theory and without disorder, the transition is described by condensation of a complex field representing the spinon or Higgs field. This must be corrected by both disorder and coupling to the $U(1)$ gauge field, effects which have been considered separately before but not together in the literature. The gauge coupling alone renders this a $U(1)$ Abelian Higgs transition, governed by an effective action which has the form of a Ginzburg-Landau theory. The coupling to the gauge field is marginal in the renormalization group (RG) sense in $3 + 1$ dimensions, and it is known [25] to destabilize the continuous transition and render it weakly first order. Disorder alone is strongly relevant, and the transition becomes nontrivial: a double epsilon expansion [26] exists for the critical theory, but the extrapolation to $3 + 1$ dimensions is quantitatively poor. Nevertheless, it supports a picture of a statistically scale-invariant theory, characterized by a dynamical exponent z relating time and space, $t \sim x^z$ or frequency and wave vector, $\omega \sim k^z$, with $z > 1$, reflecting the slow-down of dynamics by disorder.

Now we consider the two effects together. For very weak disorder, the first order transition of the Abelian Higgs theory is stable according to Imry-Ma arguments [27], but it should be rapidly removed with stronger disorder. To access the resulting continuous transition, we perturb the disordered critical point, which has some nontrivial critical action S_d, by coupling to the gauge field, and show that this coupling is relevant in the RG sense. We write the action as $S = S_d + \int d^3x \, d\tau [ieA_\mu J_\mu + F_{\mu\nu}^2]$, where $F_{\mu\nu}$ is the field strength of the emergent gauge field A_μ, and $J^\mu(\mathbf{x}, \tau)$ are the $U(1)$ spacetime currents of the bosons. Integrating out the gauge fields in the Coulomb gauge $\nabla \cdot A = 0$ we obtain an effective long-range interaction between currents. For the time-components,

$$S_{00} \sim e^2 \int d^3\mathbf{x} \, d^3\mathbf{x}' \, d\tau \frac{J^0(\mathbf{x}, \tau) J^0(\mathbf{x}', \tau)}{|\mathbf{x} - \mathbf{x}'|}. \quad (9)$$

Now we use the nonrenormalization of scaling dimensions of conserved currents, even in disordered field theories [28]. This allows us to *exactly* power count Eq. (9), according to $J^0 \sim L^{-d}$, and $\tau \sim L^z$. We obtain $S_{00} \sim e^2 L^{z-1}$, which implies that the coupling e^2 is relevant for $z > 1$. Thus we predict the system flows to a new critical theory with both nonzero disorder and gauge coupling. This is a new quantum critical universality class not heretofore studied to our knowledge.

Experiments and beyond.—We argued that, remarkably, the well-studied and characterized classical spin ice $Ho_2Ti_2O_7$ may be converted to a QSL by introducing disorder. Interestingly, the dynamics are nonmonotonic with disorder: introducing weak disorder first *speeds up* the dynamics by introducing transverse processes, while strong disorder fully quenches and freezes the moments. This nonmonotonicity should be visible in the spin thermal conductivity [29,30]. Unfortunately its interpretation is typically clouded by the difficulty of separating the (sought after) contribution from the intrinsic heat conduction of the spins, from the (less interesting) heat carried by phonons but *scattered* by spins. Here the nonmonotonicity aids in a clean separation of these effects: on introducing disorder, the spin thermal conductivity grows, developing a large T^3 contribution, whose coefficient first increases, reaches a maximum, and then collapses on leaving the QSL state. Indications of the disorder-catalyzed dynamics should be visible also in many other probes, such as a NMR and NQR relaxation, μSR, and microwave conductivity. Within the QSL state, the photon mode could be observed in inelastic neutron scattering, with an intrinsic width controlled by disorder and growing with frequency. In the Griffiths QSL, the gapless localized electric excitations can also be pairwise excited, introducing a momentum independent background, which we expect scales as $S(k, \omega) \sim e^{-c/\omega^x}$, with x of order one. We expect that the dc thermal conductivity, however, is not much affected by the rare regions, as the heat flow simply avoids them, and thus the conductivity should be similar to that of the non-Griffiths QSL. A whole range of other measurements should be possible to study scaling properties at the quantum critical point terminating the QSL phase. Our results may also be applicable to $Pr_2Zr_2O_7$, in which random crystal field splittings have already been observed [12,31]. A slowly varying texture of the random fields h_i, implicated there, does not reduce the stabilization of the QSL, which, as discussed above, even occurs for constant, nonrandom $h_i = h$.

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