Confinement, reduced entanglement, and spin-glass order in a random quantum spin-ice model

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We study an effective spin model derived perturbatively from random transverse-field Ising model on the pyrochlore lattice. The model consists of spin-configurations on the pyrochlore lattice, restricted to the spin-ice subspace, with spins interacting with random Ising exchange couplings as well as ring exchanges along the hexagons of the lattice. This model is studied by exact diagonalization up to N=64 site systems. We calculate spin-glass correlation functions and local entanglement entropy $S_T$ between spins in a single tetrahedron and the rest of the system. We find that the model undergoes two phase transitions. At weak randomness the model is in a quantum spin-ice phase where $S_T = \ln 6$. Increasing randomness first leads to a frozen phase, with long-range spin-glass order and $S_T = \ln 2$ corresponding to the Cat states associated with Ising order. Further increase in randomness leads to a random resonating-hexagon phase with a frozen backbone of spins and a broad distribution of entanglement entropies. The implications of these studies for non-Kramers rare-earth pyrochlores are discussed.

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INTRODUCTION

The study of quantum spin-liquids has emerged as one of the key directions in condensed matter physics [1–9]. It is driven in part by a desire to understand emergent quantum phases that defy conventional description and in part by a hope that at some time in future one might be able to exploit topological excitations in next generation quantum devices. Indeed, on the theoretical end, there has been substantial progress. Exactly soluble models, such as the Kitaev model [6], dispel any doubts that emergent phenomena, with fractionalized quasi-particles and emergent gauge theories can arise in a system of interacting spins. Computational approaches, most notably, area-law entanglement inspired density matrix renormalization group (DMRG) and tensor-network methods have greatly improved our ability to simulate the behavior of such systems, while methods of quantum field-theory, including perton constructions and topological field-theory have helped to catalog and systematize the range of relevant phenomena [2].

In contrast, progress has been relatively slow in convincingly demonstrating quantum spin-liquid behavior in real materials. This is in spite of the fact that an abundance of candidate quantum spin-liquid materials exist. These include the triangular and Kagome families of geometrically frustrated two-dimensional materials [3–4], the highly anisotropic spin-orbit coupling dominated Kitaev materials [5–8] and the spin-ice materials mostly magnetic rare-earth pyrochlores [10–14]. Despite a few decades of concerted effort, it has been difficult to unambiguously establish fractionalization and emergence in these systems. Part of the difficulty is simply that emergent fractionalized quasiparticles do not directly couple with external probes, making their detection difficult. But, part of the difficulty is also associated with the existence of quenched impurities, which can strongly modify the behavior as well as obscure or mimic signatures of emergence [15–22]. How much impurities can a quantum spin-liquid tolerate and what kind of behavior results from quenched impurities, remains an important question in the field.

In this work we study an effective model derived perturbatively from a random transverse-field Ising model (RTFIM) on the pyrochlore lattice [23–24]. RTFIM has been shown recently to be relevant for non-Kramers family of rare-earth pyrochlores [23–25]. Because two-fold degeneracy for eigenstates of non-Kramers ions is not guaranteed by time reversal symmetry, effective two-level models for these systems have random transverse-fields coming from quenched disorder. We had recently studied such a random transverse-field Ising models by exact diagonalization (ED) and numerical linked cluster expansion (NLC) methods [26], finding a phase diagram consisting of quantum spin-liquid (QSL), paramagnetic (PM), Ising (I) and Griffiths-McCoy (GM) phases. The systems studied were too small to establish whether the Ising phases had long-range spin-glass order.

Here, we use perturbation theory to derive an effective model starting from RTFIM, where spins are restricted to the spin-ice configurations [23–24]. In the spin-ice subspace there are two types of random interactions. There are Ising exchanges along the bonds and there are cyclic ring-exchanges along the hexagonal loops of the model. Both these interactions are dependent on the configuration of random fields. The effective model can be studied up to larger systems (N=64 spins) allowing us to clearly establish the nature of the confined phases. We
study a two-parameter family of models characterized by a mean value $h$ and a width $w$ for transverse-fields. As the width is increased at small $h$ values, one observes two phase transitions. First, into a confined phase with long range spin-glass order and then into a partially confi-
dined phase, which we call a random resonating-hexagon (RRH) phase. In the latter, the resonating hexagons are randomly placed and leave a fraction of spins forming a frozen spinglass backbone. We believe the latter phase is a variant of cluster glass phases such as random sin-glet phase \[15, 21, 27, 28\] that arise naturally in random quantum spin models. The implication of these findings to the rare earth pyrochlores will be discussed.

**MODELS AND METHODS**

We begin with the Hamiltonian for a transverse-field Ising model on the pyrochlore lattice \[29–32\]:

$$\mathcal{H} = J \sum_{<i,j>} \sigma_i^z \sigma_j^z - \sum_i h_i \sigma_i^x,$$  \hspace{1cm} (1)

where the sigmas denote Pauli spin matrices, and $J$ is the Ising exchange constant for nearest-neighbor interactions on the pyrochlore lattice. The transverse fields $h_i$ are assumed to be independent random variables at each site. In this work we will take the distribution of $h_i$ to be Gaussian with mean $h$ and standard deviation $w$, although details of the distribution are not critical to the study. Working in the limit of $J \to \infty$, restricts us to the Hilbert space of spin-ice states. In this space, the first perturbations that lift the degeneracy of ice states are fourth order terms that give rise to an Ising coupling on a bond given by strength proportional to $h_i^2 h_j^2$ \[21\]. Then, in 6th order perturbation theory we get the ring exchange term on the hexagons that can lead to a resonating QSL state.

In the spirit of effective models in a reduced subspace, like the $t-J$ model derived from the Hubbard model, we will now study the effective model in the spin-ice subspace:

$$\mathcal{H} = \frac{1}{48} \sum_{<i,j>} h_i^2 h_j^2 \sigma_i^z \sigma_j^z - \sum_u K_u (\sigma_{x1}^+ \sigma_{x2}^+ \sigma_{x3}^+ \sigma_{x4}^+ \sigma_{x5}^+ \sigma_{x6}^+ + h.c.),$$  \hspace{1cm} (2)

where $h_i$ are quenched random variables with mean $h$ and width $w$. The second sum is over all hexagons $u$ of the lattice and $K_u = \prod_{i=1}^{6} h_i$ where $h_i$ are the random fields at the six sites of the hexagon and $\sigma_1$ through $\sigma_6$ are the spin operators on the hexagon in a cyclic order. Note that the cyclic term is only operative when the spins alternate along the hexagon, otherwise it destroys the state. From here onward we will study this effective model, which depends on two parameters $h$ and $w$, and will allow for all values of $h$ and $w$.

For $w = 0$, the Ising couplings do not cause any dispersion in the spin-ice subspace. Hence, the model reduces to the pure hexagonal ring exchange model \[33, 34\] simulated by several groups before \[35, 36\]. This model is known to have a quantum spin liquid ground state with emergent quantum electrodynamics and a collective photon excitation. Our goal is to study different phases of the model as a function of $w$ and $h$.

We compute the following quantities:

1. The many-body band-width of the system, per spin, defined as

$$B = \frac{E_{\text{max}} - E_{\text{min}}}{N},$$  \hspace{1cm} (3)

where $E_{\text{max}}$ is the energy of the highest energy state and $E_{\text{min}}$ the energy of the lowest energy state for an $N$-site cluster.

2. Entanglement entropy of a tetrahedron of spins and their distribution: In a pure state, the von-Neumann entanglement entropy for subsystem $A$ and its complement $B$ is defined as

$$S_A = S_B = - \text{Tr} \rho_A \ln \rho_A,$$  \hspace{1cm} (4)

where $\rho_A$ is the reduced density matrix for subsystem $A$. In this work, $A$ is made up of the four spins belonging to any single tetrahedron. In the uniform system, it is easy to see that the tetrahedron entanglement entropy $S_T = \ln 6$. Let us label our basis states as $|\alpha, i \rangle$, where $\alpha$ stands for spins inside the tetrahedron and $i$ stands for the spins outside. Then, a general state of the system can be expressed as

$$|\psi \rangle = \sum_{\alpha,i} C_{\alpha,i} |\alpha, i \rangle.$$  \hspace{1cm} Reduced density matrix for the tetrahedron

$$\rho_{\alpha,\beta}^T = \sum_i C_{\alpha,i}^* C_{\beta,i},$$

must be diagonal because for two state $|\alpha, i \rangle$ and $|\beta, i \rangle$ to both lie in the spin-ice subspace, one must have $\alpha = \beta$. Furthermore, the uniform system has sublattice symmetry and all 6 states are related by a permutation of sublattices. Hence, as long as there is a non-degenerate ground state, all 6 diagonal matrix-elements of the reduced density matrix must be equal implying $S_T = \ln 6$. Our earlier exact diagonalization studies of 16 and 32 site systems \[26\] showed that, in the full random traneverse-field model, this entropy remains very close to $\ln 6$, despite virtual dressing of the spin-ice states due to perturbative fluctuations. Only when one is near the confinement transition one sees deviations from this value.

3. Ising correlation function and correlation sum

$$C_{ij} = \langle [\sigma_i^z \sigma_j^z]^2 \rangle,$$  \hspace{1cm} (5)
\[ C_{zz} = \left[ \frac{1}{N} \sum_{i,j} \langle \sigma_i^z \sigma_j^z \rangle^2 \right], \]  \hspace{1cm} (6)

where the angular brackets refer to ground state averages and square brackets to an average over the distribution of random fields. Note that we have normalized the correlation sum such that in a system with long-range (random) order the correlation sum \( C_{zz} \) should scale as \( N \).

4. Inverse participation ratio in the many-body space defined as

\[ IPR = \frac{1}{\sum_i a_i^4}, \]  \hspace{1cm} (7)

where \( a_i \) are the coefficients of the ground state wavefunction in the Ising basis. In the quantum spin-ice phase this quantity should be order \( D \), where \( D \) is the dimension of the spin-ice space, whereas in an equal admixture Cat state of two Ising configurations, it should equal 2.

**NUMERICAL RESULTS**

We study finite clusters with periodic boundary conditions. We have looked at clusters of size 16, 32, 40, 48 and 64. To carry out the calculations we first pick a configuration of random fields for each site from a Gaussian random distribution with mean \( h \) and uncertainty \( w \). This allows us to determine the Ising couplings for all the bonds as well as strength of the ring exchange for each hexagon. The ground state is then obtained by the Lanczos method. We typically include 100 to 400 different field configurations to average over random configurations. Variations from different field configurations allow us to determine the statistical error bars.

With only the ring exchanges as the off-diagonal terms, this model is known to partition into disjoint subspaces. We first determine all the disjoint parts of the Hilbert space and then obtain the ground state in each subspace. The overall ground state is obtained by comparing energies in different subspaces. In the QSL phase, the ground state typically lies in the largest connected subspace but this is not necessarily true. When the ground state is in a subspace where a state and its time reversed partner are disconnected, there must be two degenerate ground states. Since, the original transverse field model does not have a disconnected Hilbert space, in this case, we take as our ground state the symmetrized linear superposition of the two. For \( N = 64 \) case, the total spin-ice subspace has dimensionality \( 2,249,370 \) whereas the largest disjoint sector has dimensionality \( 194,640 \).

In Fig. 1, the many-body band-width of the system per site is shown. It is independent of width \( w \) in the QSL phase and depends on \( h \) as \( h^6 \) due to the ring exchange term. In the perturbative regime, after the first phase transition the bandwidth goes as \( w^2 h^2 \) as known from previous studies using NLC [29] for RTFIM. This follows from the fact that within each tetrahedron the maximum difference in the energy between any two spin-ice states goes as \( w^2 h^2 \), which overwhelms the \( h^6 \) term. For large \( w \) the bandwidth must scale as \( w^6 \).

In Fig. 2, we show the average entanglement entropy \( S_T \) for a tetrahedron. It is averaged over the tetrahedron in a cluster as well as over the distribution of random fields. Two phase transitions are clearly seen in the plot. It equals \( S_T = \ln 6 \) in the QSL phase. In the spin-glass phase it approaches \( \ln 2 \) implying that the state is reduced to a cat state with only global \( \ln 2 \) entropy and there is negligible quantum fluctuation around that state. At very large \( w \) it approaches a value somewhat below \( 2 \ln 2 \). Fig. 3 shows a distribution of entanglement entropies. In the transition regions as well as in the random resonating-hexagon phase at large \( w \) the entropy distribution becomes very broad.

In Fig. 4, the Ising correlation sum is shown. The correlation sum is extensive in both the confined phases as shown in Fig. 5. In the intermediate phase, which we identify as the Ising spin-glass, it approaches the maximal value, again showing that quantum fluctuations become negligible as the Ising couplings dominate over the ring exchanges.

In Fig. 6, the inverse participation ratio is plotted. Apart from the intermediate phase where it is only slightly large than 2, it is size dependent in the other two phases. In the QSL phase it must scale with the dimension of the Hilbert space. In the random resonating-hexagon phase also it grows with the size of the system.
DISCUSSIONS: PHASE DIAGRAM

Our numerical results clearly establish three phases in the model separated by phase transitions. As shown in the phase diagram in Fig. 7 these are the quantum spin-liquid (QSL), the Ising spin-glass (ISG) and the random resonating-hexagon (RRH) phases. These phases are separated by sharp transition regions. When there is no disorder, that is $w = 0$, the model is in the QSL phase. This must be separated from other phases by a thermodynamic phase transition as the other phases have long range Ising order and hence a broken $Z_2$ symmetry. At small $h$ and $w$, the Ising Spinglass phase has negligible quantum fluctuations. This is not surprising as the Ising couplings arise in lower order of perturbation theory than the ring exchange term \[24, 26\]. Hence they totally dominate and as argued in our previous paper \[26\] all the ring terms are frozen out by disorder. The transition is not a simple level crossing as clearly the transition region is broad. However, we do not see evidence for crossing of physical properties for different sizes as expected at a critical transition. Instead there is a weak size dependence of the transition point. Yet, the transition region is characterized by substantial fluctuations, and the entanglement entropy develops a broad distribution there. Hence, we conclude that the transition is weakly first order. Savary and Balents \[23\] suggested a weakly first order confinement transition due to randomness even when no spin-glass order develops on the other side. The development of an additional order is even more likely to make it first order.

The most interesting phase is the random resonating-hexagon (RRH) phase. This phase has a broad distribution of entanglement entropy with a peak near $2 \ln 2$. The many-body inverse participation ratio suggests that it still grows exponentially with system size, although very slowly $1.04^N$. And, on top of that this phase has Ising correlation sum which scales as $N$ implying long-range order. At large random-fields the Ising exchange terms become negligible and the model becomes a random ring-exchange term. We believe the ground state of this model can be understood along the lines of real-space renormalization group (RSRG) approaches to understanding random quantum spin problems \[15, 21, 27, 28\]. As the distribution of couplings becomes very broad (being a product of 6 field terms), one can first pick out the strongest hexagon and make it into a resonating cluster. The strong resonance of one hexagon makes these six spins ineligible for additional entanglement and renders neighboring spins frozen into certain configurations as otherwise they would interfere with this resonance \[20\]. This resonating cluster is like a singlet formation in a random-singlet phase, except it involves six spins and is unrelated to any SU(2) symmetry. Then one can pick out the next strongest resonating hexagon. When this process is continued, it will lead to a random configuration of hexagons placed in the lattice and it can leave a backbone of spins that must be fixed in order to maintain

FIG. 3: Distribution of entanglement entropies for different values of $w$ for $h = 0.06$.

FIG. 4: Ising correlation sum in different parameter regions.

FIG. 5: Scaling of Ising correlation sum with $N$ the size of the system.

FIG. 6: Inverse Participation Ratio for different parameters.
compatibility with the resonance in the hexagons and be in the spin-ice subspace. Our numerical results suggest that the backbone is percolating and has a finite fraction of spins.

The participation ratio should be exponential in number of resonating clusters, which should be of order one per ten to twenty spins. Furthermore, one would expect a large distribution of entanglement entropies in the phase with a substantial weight around $2 \ln 2$, corresponding to the four spins in a tetrahedron dividing into two groups of two and participating in two independent resonances.

Going back to the random transverse-field Ising model (RTFIM), such an RRH phase can only arise at intermediate values of disorder $w$ of order unity and $h$ not too large. We know that too large an $h$ leads to a different confining phase with a condensation of spinons and the system moves away from the spin-ice subspace altogether. Also, if the disorder $w$ becomes much larger than one, one would once again enter a local phase where the spins simply point along the local random fields and will no longer be in the spin-ice subspace. The comparison of our results with the ED study of the random-field model suggests that such a phase is possible in the RTFIM. The main difference must be that the Ising ordered backbone may be disordered due to the local fields.

It is interesting to consider this study from the perspective of rare-earth magnetic pyrochlore materials, where RTFIM has been argued to be relevant. First of all, our work implies that if $h = 0$ and one only has weak random fields, the system will be in a frozen spin-glass phase. A variety of measurements such as NMR or µSR can easily confirm that. Indeed restricted subspace generally promotes spin-glass order. This is clearly not the most interesting phase from the point of view of quantum spin-liquids. However, if disorder becomes a fraction of $J$, then one is away from the perturbative regime and the possibility of random resonating-hexagon phase becomes likely. While not a true QSL, such a phase has a lot of quantum fluctuations and local entanglement and should show interesting power-law temperature, magnetic field and frequency dependence in various responses. These can be dominated by just the behavior of single hexagons, and can be easily calculated. Indeed this physics can survive up to very large randomness.

For the material Pr$_2$Zr$_2$O$_7$, it has been argued that the width of the random-fields can be much larger than the Ising couplings. The actual distribution of random-fields was found to not be Gaussian but Lorentzian. But, that should not change the results in a significant way. It would be interesting if evidence for local resonating hexagons can be seen in these systems.

In order to obtain a true U(1) QSL phase, it is not enough to just reduce the strength of the disorder or $w$. It is important to have a uniform component of the transverse-field that is at least of order the randomness.

What kind of impurities or imperfections can lead to this, if without impurities one has two-fold degeneracy in the crystal-field states, deserves attention from a materials point of view. It is more likely that when non Kramers ions have two nearby non-degenerate crystal-field states that it can then be modeled as two level systems in a uniform transverse-field. If such a system still has spin-ice physics, it would be a good candidate for a QSL. Another possibility is that disorder is correlated over lengths much larger than the lattice constant. In that case, the system can behave effectively as having a uniform field in any region. Another possibility is that the magnitude of the random-field is nearly uniform from site to site. But, there is variation in signs or directions. This model may still have a QSL phase and deserves further attention.

**SUMMARY AND CONCLUSIONS**

In summary, in this paper we have studied an effective model derived perturbatively from the random transverse-field Ising model (RTFIM) on the pyrochlore lattice. The reduced Hilbert space of the effective model allows us to study larger system sizes and thus deduce the nature of different phases. We find three different phases. A U(1) QSL phase occurs for sufficiently small randomness. At weak transverse-fields, increased randomness leads to an Ising spin-glass (ISG) phase, with nearly frozen spins and very little quantum fluctuations. Increased random-fields can lead to a random resonating hexagon (RRH) phase, which is a kind of a cluster-glass phase where quantum fluctuations and entanglement are restricted to small clusters.

We have discussed possible relevance of this study to rare-earth pyrochlores where RTFIM have been argued to be relevant. It is clear that a broad distribution of transverse-fields, width exceeding the mean, will not lead to a QSL phase. But, it is possible for it to still be in a random resonating hexagon phase. It would be interesting if evidence for local resonating hexagons is
observed in Pr$_2$Zr$_2$O$_7$. A true U(1) QSL would need a material where the magnitude of the transverse-field is nearly uniform at least at nearby sites.

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