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Emergence of a two-dimensional macrospin liquid in a highly frustrated three-dimensional quantum magnet

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The classical Ising model on the frustrated three-dimensional (3D) swedenborgite lattice has disordered spin liquid ground states for all ratios of inter- and intraplanar couplings. Quantum fluctuations due to a transverse field give rise to several exotic phenomena. In the limit of weakly coupled kagome layers we find a 3D version of disorder by disorder degeneracy lifting. For large out-of-plane couplings one-dimensional macrospins are formed, which realize a disordered macrospin liquid phase on an emerging two-dimensional triangular lattice. We speculate about a possibly exotic version of quantum criticality that connects the polarized phase to the macrospin liquid.

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Introduction. Geometrical frustration in magnetic systems can give rise to a multitude of exotic classical and quantum phases [1]. An analysis of the classical limit often reveals a large degeneracy in the ground state manifold that can be lifted by quantum or thermal fluctuations, thereby selecting an ordered state. This phenomenon is conventionally referred to as "order by disorder" [2–4]. A more exotic version of degeneracy lifting is called "disorder by disorder" [5–9]: Out of multiple classical ground states, a disordered state is selected. Paradigmatic examples for these two phenomena can be found in two dimensions (2D) in the transverse field Ising model (TFIM), respectively on the triangular and the kagome lattice. In both cases a polarized phase is found at high transverse fields. On the triangular lattice an infinitesimal transverse field is sufficient to select an ordered state out of the degenerate classical manifold, thereby providing an example of order by disorder. This selected state is the $\sqrt{3}\times\sqrt{3}$ state that maximizes the number of flippable spins. The two phases are connected via a second-order phase transition in the three-dimensional (3D) XY universality class [5,8,10]. In contrast, on the kagome lattice any finite transverse field selects the disordered polarized phase, which does not break any symmetry, in an instance of disorder by disorder [5,8].

Recently, it was found that interesting effects of frustrated magnetism can also be encountered on the three-dimensional swedenborgite lattice (inset of Fig. 1). Experimental efforts have shown that the class of compounds with this lattice structure has both magnetically ordered and disordered representatives [11–18]. Theoretically, it was shown that classical $O(3)$ Heisenberg spins exhibit a wide spin liquid regime and can undergo a fluctuation-driven order by disorder transition to a nematic phase at very low temperatures [19]. The Ising model with a longitudinal magnetic field boasts different phases with extensive and subextensive ground state degeneracy [20]. It was also observed that the bare swedenborgite Ising model corresponds to either a stack of weakly coupled kagome layers or an emergent triangular lattice of macrospins, depending on the choice of parameters [20]. As a result, one can speculate that the order by disorder of the triangular lattice TFIM and the disorder by disorder of the kagome TFIM enter into competition with one another once a transverse field is added.

In this Rapid Communication we investigate how the degeneracies in the classical Ising model on the swedenborgite lattice are lifted due to quantum fluctuations. This is summarized in the phase diagram shown in Fig. 1. For large transverse field values the system will assume the polarized state for all couplings. For $J_1 > J_2$, this phase is stable for all transverse field values in an instance of disorder by disorder, which was also observed on the two-dimensional kagome lattice [8]. For $J_2 > J_1$ and small transverse field, we can map the three-dimensional quantum model to an effective two-dimensional classical model at zero temperature, which is formulated in terms of macrospins on an emerging triangular lattice. This model hosts an exotic phase, in the following referred to as a macrospin liquid (MSL), which features a

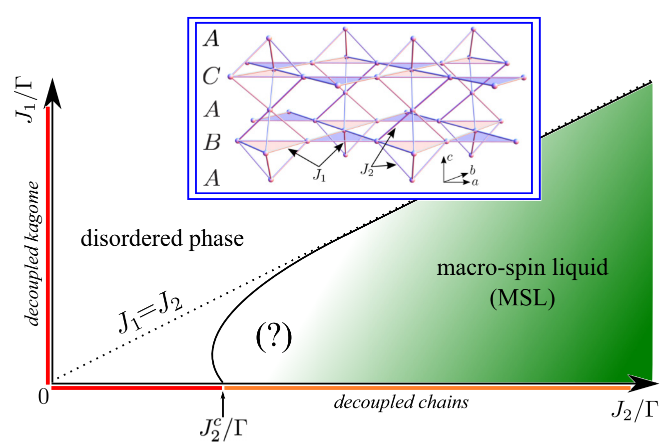


FIG. 1. Schematic phase diagram of the 3D swedenborgite TFIM. Dashed line: Classical phase transition line between two different types of spin liquids. Red lines: Disordered phase in kagome and chain limits. Orange line: Ordered phase in the chain limit, encountered on the other end of a second-order phase transition at $(0, J_2^c/\Gamma)$. Inset: The structure and spin interactions of the swedenborgite lattice.

subextensively degenerate ground state manifold. We find that these phases survive up to intermediate couplings J_2/Γ, where there is no small parameter handle to learn how they relate to one another. Within our approach, the transition out of the polarized phase reflects a reduced dimensionality in its critical exponents. However, the momentum location of the minimal gap in the polarized phase is inconsistent with all degenerate ground states in the MSL. Thus, two scenarios are possible: (a) an exotic type of continuous phase transition, where an intermediate phase occurs that is consistent with the gap closing momentum or (b) a first-order transition directly between the disordered phase and the MSL. This uncertainty is indicated by a question mark in Fig. 1.

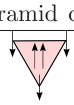
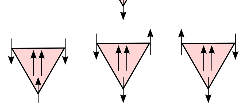
Swedenborgite TFIM. The swedenborgite lattice (inset of Fig. 1) consists of a stack in the c direction of alternating layers of inequivalent kagome planes (B,C) and triangular spins (A) in an ABACA ... pattern. The structure can alternatively be regarded as a collection of bipyramidal clusters that form columns along the c direction and are connected by intermediate triangles in the ab plane (also see Figs. S1 and S2 in the Supplemental Material [21]). We consider a nearest-neighbor Ising model with two distinct antiferromagnetic interactions: J_1 inside the kagome layers and J_2 between the kagome and triangular layers,

$$\mathcal{H} = J_1 \sum_{\substack{\langle i,j\rangle\in \\ \text{same layer}}} \sigma_i^x\sigma_j^x + J_2 \sum_{\substack{\langle i,j\rangle\in \\ \text{diff. layer}}} \sigma_i^x\sigma_j^x + \Gamma\sum_i \sigma_i^z, \qquad (1)$$

with Ising spins $\sigma_i = \pm 1$ and a transverse magnetic field term controlled by Γ. We emphasize that this model is not intended to describe a currently existing material. Instead, we investigate it on the basis of conceptional interest within the framework of spin liquids and magnetic frustration.

Classical limit. For $\Gamma = 0$, there are two distinct "spin liquid" phases (separated by a first-order transition), whose elementary building blocks are the basal units in Table I. These have distinct degrees of degeneracy [20]. For $J_1 > J_2$ the ground state manifold is extensively degenerate. For $J_2 > J_1$ the single bipyramid ground state stacks in the c direction inside each column, forming macroscopically stiff spin structures called macrospins with which the degeneracy of the ground state manifold becomes subextensive (see Fig. S1 and the discussion [21]). With the remaining binary spin degree of freedom of these macrospins, the problem reduces to a classical Ising model of these macrospins on a triangular lattice (see Fig. S2 for more details [21]). On this lattice, ground states are characterized locally by having two

TABLE I. Optimal bipyramid configurations in the different parameter regimes. The center spin pair in each triangle denotes the prefixed apical spins of a bipyramid. For $J_1 = J_2$, the two indicated manifolds combine.

$\Gamma = 0$	optimal bipyramid configurations
$J_2/J_1 > 1$	
$J_2/J_1 < 1$	

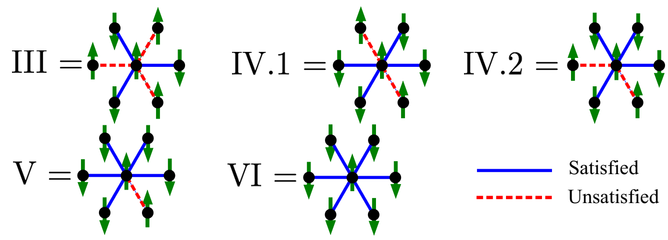


FIG. 2. The five different plaquettes allowed on the emergent triangular lattice, up to rotation. In green, the underlying macrospin configuration (unique up to global flip of macrospins).

parallel and one antiparallel macrospin on every subtriangle (2:1 rule) [22]. In a bond picture, this corresponds to two satisfied bonds and one unsatisfied bond per subtriangle. As a result, the possible configurations of each macrospin and its six neighbors are restricted to those listed in Fig. 2. Only certain combinations of these plaquettes are allowed: There needs to be an average 2:1 ratio of satisfied to unsatisfied bonds globally. There are only three combinations compatible with this ratio: (IV), (III + V), and (III + III + VI) (see Fig. S3 [21]).

Quantum-to-classical mapping. For $J_2 > J_1$ the effect of small values of Γ can be studied perturbatively by considering the induced spin-flip processes. In the ordinary 2D Ising model on the triangular lattice there are ground states in the classically degenerate manifold that are related by a single flip, for instance, at the central position of a III plaquette analog of ordinary spins. This introduces a bias to maximize the number of such plaquettes and ultimately leads to the selection of the $\sqrt{3} \times \sqrt{3}$ state as the ground state. However, such off-diagonal processes are suppressed in the case of the macrospins since they require flipping a whole macrospin. Consequently, the two degenerate states are "infinitely far apart" in the configuration space. For finite orders in Γ only diagonal processes occur. This implies there is no dynamics in play but only potential energy, so that we can map from the three-dimensional quantum model to a *two-dimensional,* $T = 0$ *classical* model on the triangular lattice with the simple Hamiltonian

$$H_{\text{dim}} = \sum_{i=\text{III,IV,V,VI}} E_i |i\rangle\langle i|, \qquad (2)$$

where we sum over the different types of plaquettes i with corresponding energy E_i. This model is still subject to the 2:1 rule, meaning it is not trivially solved by the plaquette with the lowest energy. We have determined the energy up to fourth order in perturbation theory, [21]. To second order in Γ we find that the three different plaquette combinations reduce their energy by the same amount. This behavior changes only at fourth order in Γ, when a relative shift in the energy results in the hierarchy $E_{\text{IV}} < \frac{1}{2}(E_{\text{III}} + E_{\text{V}}) < \frac{1}{3}(2E_{\text{III}} + E_{\text{VI}})$ that favors the two IV plaquettes to make up the ground states. The geometry prohibits mixing of the subtypes in one direction, so that chains form. In the remaining undetermined direction, "stripy" configurations with a varying degree of zigzagging will shape the ground states. IV.1 chains will cause straight running stripes, whereas IV.2 chains introduce bends. The two

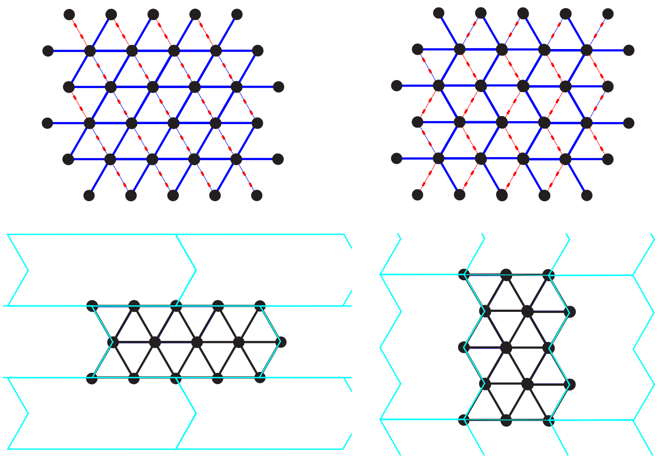


FIG. 3. Upper panel: The minimally (left) and maximally zigzagging (right) stripy structures that form the ground states on the emergent triangular lattice, with horizontal monocultural chains of respectively IV.1 and IV.2 plaquettes. Lower panel: Sketch of the finite macrospin configuration used for the ED. Gray (cyan) lines denote the unit cells and indicate the chosen periodic boundary conditions. Each circle represents a triangle in the kagome planes of the swedenborgite lattice so that each finite macrospin configuration contains 24 spins in each kagome plane. These 24-site kagome clusters are considered in the ED.

extremal cases of such configurations are depicted in Fig. 3, but any combination of the two is allowed. The result is a subextensively degenerate ground state manifold. Coincidentally, these stripy lattice coverings have also been suggested as a ground state configuration for a model with antiferromagnetic interactions between Ising spins on an elastic triangular lattice [23].

It is natural to ask whether the degeneracy is lifted further beyond order four [shown in Eq. (S4) [21]]. From a perturbative perspective the remaining degeneracy can be traced back to the topological equivalence of graphs IV.1 and IV.2. The degeneracy is lifted when virtual processes of the minimally stripy structure have no symmetric analog for the maximally zigzagging stripy structure. Similar to Ref. [8], in the situation at hand this can only occur when fluctuations on loops of the kagome lattice play a role. The relevant minimal loop on the kagome lattice is a hexagon consisting of six spins. In each order the perturbation leads to local spin flips linking two neighboring bonds of a hexagon. Consequently, a linked process that covers the hexagon and lifts the subextensive degeneracy appears at best at order 12 and inside the kagome planes. In fact, the system seems to be even more reluctant to order in either fashion, which we have checked by exact diagonalization (ED). We considered the two finite macrospin configurations shown in the lower panel of Fig. 3. Now, we freeze the spins of the three-dimensional system in one of the two configurations of unperturbed macrospins except the 24 spins of a single kagome plane. We stress that these are the only kagome clusters tractable by ED for which the associated minimally and maximally zigzagging macrospin structures fit with the same loop length. The frozen spins above and below the considered kagome plane give rise to an effective field which is included in the ED. Physically, the dominant fluctuations lifting the degeneracy take place inside the kagome plane which are therefore captured nonperturbatively on the length scales of the considered kagome clusters. Surprisingly, the ground state energy of both stripy structures is exactly degenerate on both clusters up to numerical precision. This implies that all virtual fluctuations that fit on the shown clusters, up to infinite order, are unable to fully lift the subextensive degeneracy and the system remains disordered. We consequently conclude that these systems shows evidence for the existence of a MSL.

Notice that a similar analysis cannot be carried out for $J_2 \leqslant J_1$, where some of the different spin bipyramid configurations in the degenerate ground state manifold are connected through a second-order spin-flip process. There are off-diagonal tunneling terms between the distinct classical ground states that lead to a complicated effective quantum problem.

High-field expansion. For $\Gamma \gg J_\nu$ ($\nu = 1,2$), we can understand the excitations generated by a single spin flip as quasiparticles above a vacuum provided by the fully polarized ground state. We introduce hardcore boson operators $(a^\dagger, a)$ acting on kagome plane sites and $(b^\dagger, b)$ operating in the triangular layers that generate or destroy a spin flip. The Hamiltonian (1) expressed in terms of these operators reads, for $\Gamma = \frac{1}{2}$,

$$\begin{aligned}\mathcal{H} &= -\frac{N}{2} + \sum_{i,\mu\in a,b} \hat{n}_i^{(\mu)} + J_1 \sum_{\langle i,j\rangle} (a_i^\dagger a_j^\dagger + a_i^\dagger a_j + \text{H.c.}) \\ &\quad + J_2 \sum_{\langle i,j\rangle} (a_i^\dagger b_j^\dagger + a_i^\dagger b_j + \text{H.c.}) \\ &= \mathcal{H}_0 + \sum_{\nu=1,2} J_\nu \sum_{m=0,\pm 2} T_m^{(\nu)}, \end{aligned} \tag{3}$$

where $\mathcal{H}_0 = -N/2 + \sum_{i,\mu} \hat{n}_i^{(\mu)}$ with density operator $\hat{n}_i^{(\mu)} = \mu_i^\dagger \mu_i$, N is the total number of sites, and the index $m = \{0, \pm 2\}$ refers to the change of the quasiparticle number due to the operator $T_m^{(\nu)}$.

A perturbative continuous unitary transformation (pCUT) [24,25] transforms Hamiltonians of the form Eq. (3) into effective quasiparticle conserving Hamiltonians $\mathcal{H}_{\text{eff}}$ so that $[\mathcal{H}_{\text{eff}}, \mathcal{H}_0] = 0$. This first step of the method can be done model independently, i.e., the effective Hamiltonian in the thermodynamic limit is calculated perturbatively up to high orders. The second, model-specific step is to normal order $\mathcal{H}_{\text{eff}}$, which is done most efficiently via a linked-cluster expansion. The computational effort of a linked-cluster expansion scales exponentially with the number of graphs as well as with the number of perturbative parameters, implying that the three-dimensional model under consideration with two parameters J_ν is of challenging complexity. Fortunately, the recently introduced white-graph expansion [26] is perfectly suited for this problem. This technique performs calculations on graphs without specifying the couplings, which are reintroduced only in the final embedding procedure. As a consequence, the total number of graphs is strongly reduced. Here, we concentrate on the physical properties of a single quasiparticle in order to investigate the breakdown of the polarized high-field phase. Applying a white-graph expansion, we determine the effective one-quasiparticle Hamiltonian up to order 11 in both parameters J_ν.

The effective one-particle hopping Hamiltonian $\mathcal{H}_{\text{eff}}$ can then be block diagonalized via Fourier transformation with respect to the eight-site unit cell of the lattice. One

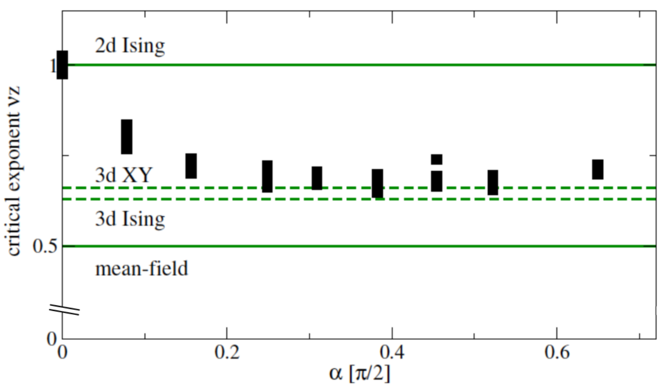


FIG. 4. Critical exponent $z\nu$ as a function of α, where $\tan(\alpha) = J_1/J_2$, obtained from various Dlog-Padé extrapolations of the high-field gap $\Delta(J_1,J_2)$. Solid and dashed horizontal lines indicate known critical exponents for certain universality classes.

obtains eight one-particle bands $\omega_n(\vec{k})$ with $n \in \{1,\ldots,8\}$ and $\vec{k} = (k_a,k_b,k_c)$. Here, k_a and k_b are the momenta in the kagome ab planes and k_c is parallel to the c direction. We find that the one-particle gap Δ is always located at $\vec{k} = (2\pi/3,-2\pi/3,0)$ (and other momenta related by symmetry). This is fully consistent with the findings for the pure kagome TFIM [8]. Note that this momentum is not compatible with any of the stripy configurations in Fig. 3 in the ground state manifold of the MSL phase. Next, we use the Dlog-Padé method [27] to extrapolate our findings for the gap $\Delta(J_1,J_2)$ to lower-field regimes. Notably, we estimate the location of quantum critical points (J_1^c,J_2^c) with $\Delta(J_1^c,J_2^c) = 0$ and the corresponding critical exponent $z\nu$. The quantum critical line is displayed in Fig. S4, corroborating Fig. 1, while the critical exponent is shown in Fig. 4.

In the pure kagome limit $J_2 = 0$ we recover the results of Ref. [8]. The lowest band is completely flat up to and including order seven, and a specific momentum is only chosen at order eight. Extrapolating the one-particle gap for small values of J_2 gives no indications for a gap closing at any values of (J_1,J_2) (when $J_1 < J_2$). Let us mention that the extrapolations might not be very accurate for $|(J_1,J_2)| \to \infty$ and that no complementary calculation for the limit $\Gamma \to 0$ can be done efficiently for the 3D swedenborgite lattice, in contrast to the two-dimensional counterpart on the kagome lattice [8]. Nevertheless, our findings are consistent with a remarkable disorder by disorder scenario in three dimensions for an extended parameter range that includes the pure kagome TFIM.

The physics is fundamentally different in the regime $J_2 > J_1$. In the 1D limit $J_1 = 0$ one has isolated and unfrustrated TFIMs on the bipyramidal chains along the c direction. One therefore expects a quantum phase transition in the 2D Ising universality class with $z\nu = 1$ for these one-dimensional quantum systems, which is quantitatively confirmed by our calculation. We find $(0,J_2^c) = (0,0.239 \pm 0.002)$ and $z\nu = 1.00 \pm 0.02$. Introducing a finite J_1, we deduce a nontrivial quantum critical line sprouting from the critical point of the 1D limit. For small values of J_1 the critical value of J_2 decreases, i.e., quantum fluctuations due to the frustrated Ising interactions in the kagome planes weaken the polarized phase. Interestingly, this behavior changes for larger ratios J_1/J_2, where the polarized phase is stabilized by quantum fluctuations. This quantum critical line also shows a remarkable behavior in the corresponding critical exponents: We find a $z\nu$ that deviates from conventional behavior in (3+1) dimensions. For a broad range of ratios J_1/J_2 a value $\approx$0.7 is observed, which is more typical for (2+1)-dimensional criticality. This suggests that the dimensional reduction observed in the region $J_2 > J_1$ is also displayed in the quantum critical properties.

Discussion. One may wonder how our findings for the MSL and the polarized phase concerning the nature of the quantum phase transition can be reconciled. In the simplest scenario there is a first-order phase transition between these two phases, for which the momentum of the one-particle gap need not be compatible with any ground state configuration in the MSL phase. Let us mention that a single series expansion in one phase is not able to detect first-order phase transitions. However, the lack of attractive quasiparticle interactions in the polarized phase gives no obvious tendency towards such a direct transition. In another scenario there is a second-order quantum critical line between the two phases, as suggested by the high-order series expansion. This is clearly more interesting. In this case the physics must be even more exotic, since an intermediate phase (indicated by the question mark in Fig. 1) that is compatible with the polarized phase gap momentum is expected to emerge as a breakdown of the MSL. Because this falls outside of the reach of most perturbative methods, investigating this putative intermediate phase and the attached quantum phase transitions is a formidable challenge. More research is certainly needed to elucidate which of these scenarios is realized in the swedenborgite TFIM.

Conclusion. The present study reveals the fascinating physics of highly frustrated quantum magnets in three spatial dimensions. The swedenborgite lattice is a prototypical system that displays several exotic features due to the competition between geometric frustration and quantum fluctuations, which gives rise to a rich phase diagram. In a broader perspective, the emergence of macrospins as classical entities within the context of three-dimensional frustrated quantum systems deserves more investigation in future research as it appears to give a route towards exotic quantum criticality where the critical dynamics is both strongly constrained and direction dependent.

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