Cluster-Glass Phase in Pyrochlore *XY* Antiferromagnets with Quenched Disorder

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We study the impact of quenched disorder (random exchange couplings or site dilution) on easy-plane pyrochlore antiferromagnets. In the clean system, order by disorder selects a magnetically ordered state from a classically degenerate manifold. In the presence of randomness, however, different orders can be chosen locally depending on details of the disorder configuration. Using a combination of analytical considerations and classical Monte Carlo simulations, we argue that any long-range-ordered magnetic state is destroyed beyond a critical level of randomness where the system breaks into magnetic domains due to random exchange anisotropies, becoming, therefore, a glass of spin clusters, in accordance with the available experimental data. These random anisotropies originate from off-diagonal exchange couplings in the microscopic Hamiltonian, establishing their relevance to other magnets with strong spin-orbit coupling.

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Rare-earth pyrochlores form one of the most interesting families of frustrated magnets. A lattice of corner-sharing tetrahedra combined with a multiplicity of crystal-field effects for rare-earth ions [1] gives rise to a plethora of novel states [2]. Among them are disordered spin ices [3–6] and quantum spin liquids [7–9], found in materials with magnetic easy-axis anisotropy. In contrast, compounds exhibiting an easy-plane (or *XY*) anisotropy tend to order antiferromagnetically [10–16]. A number of them realize an "order-by-disorder" mechanism where a long-range-ordered state is selected, via either thermal or quantum fluctuations, from a classically degenerate manifold resulting from strong frustration [12,17–19]. In the parameter regime relevant to the paradigmatic example $Er_2Ti_2O_7$, both classical and quantum fluctuations select the non-coplanar state dubbed ψ_2 from a one-parameter manifold, in a remarkable agreement with experiments [11,12].

Quenched disorder provides a different route for lifting the classical degeneracy by locally relieving the frustration [20]. Previous theoretical studies showed that both bond randomness and site dilution tend to stabilize, for small disorder, the coplanar state dubbed ψ_3 [21,22]. This insight motivated a series of experiments in inhomogeneous *XY* pyrochlore magnets. In $Er_{2-x}Y_xTi_2O_7$ [23] magnetic Er^{3+} is substituted by nonmagnetic Y^{3+}, corresponding to site dilution. For $NaCaCo_2F_7$ [24,25] and $NaSrCo_2F_7$ [26] quenched disorder arises from site mixing on the pyrochlore *A* sites (Na/Ca and Na/Sr, respectively), with the leading effect on magnetism being bond randomness. However, all experiments find either the ψ_2 state, for small disorder, or short-range magnetic correlations below a freezing temperature T_f at stronger disorder, suggesting a spin-glass state.

In this Letter, we solve this puzzle. We develop a more general theory, valid in the experimentally relevant regime, showing that disorder-induced random anisotropies destabilize magnetic long-range order (LRO) of easy-plane pyrochlores. We derive a semiquantitative stability criterion which indicates that LRO is destroyed beyond a critical level of quenched disorder. Using extensive Monte Carlo (MC) simulations for the relevant classical model, we verify the tendency towards ψ_3 order in the weakly diluted regime [21,22], and provide compelling evidence that the low-T state at stronger disorder is a cluster spin-glass (CSG) phase where the system breaks into domains (spins

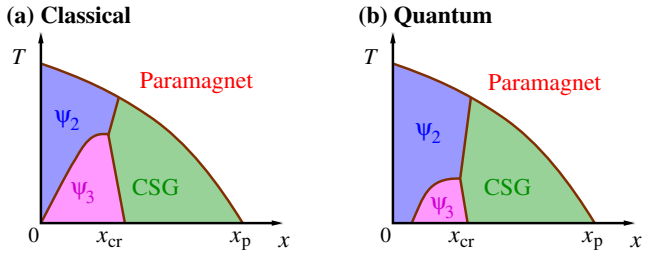

FIG. 1. Schematic phase diagram for the easy-plane pyrochlore antiferromagnet (1) as a function of temperature T and site dilution x, for parameters with $0 < J_{\pm\pm}/J_{\pm} \leq 2$ where order-by-disorder selects the ψ_2 state in the clean limit, $x = 0$. Quenched disorder tends to select the ψ_3 state at low T, but both $\psi_{2,3}$ are destroyed beyond a critical level of disorder x_{cr} where randomness breaks the system into magnetic domains resulting in a cluster spin glass (CSG) phase which survives up to the percolation threshold x_p. (a) Without quantum fluctuations, i.e., in the classical limit. (b) With quantum fluctuations. Here ψ_3 may disappear completely; for details see text.

clusters) exhibiting a variety of *local* ordering patterns besides the ψ_2 and ψ_3 ones. For the quantum case, we argue that the tendency towards ψ_3 order is diminished, such that a significant portion of the phase diagram is dominated by the CSG phase, Fig. 1, in agreement with available experiments.

Model.—The low-temperature properties of many rare-earth insulating pyrochlore oxides are well described by an effective spin-1/2 model with anisotropic exchange interactions due to the combination of spin-orbit and crystalline electric-field effects [1]. In $Er_2Ti_2O_7$, for instance, the spins have a dominant planar nature, and the associated nearest-neighbor anisotropic XY model [8,11,12,27,28] can be written as

$$\mathcal{H} = -\sum_{\langle jk \rangle} [J^{xx}_{jk} S^x_j S^x_k + J^{yy}_{jk} S^y_j S^y_k + J^{xy}_{jk} (S^x_j S^y_k + S^y_j S^x_k)]. \quad (1)$$

Here, the sum runs over pairs of nearest-neighbor (NN) sites on a cubic pyrochlore lattice, the couplings are

$$J^{xx(yy)}_{jk} = 2(J^{\pm}_{jk} \mp J^{\pm\pm}_{jk} \cos\theta_{jk}), \quad J^{xy}_{jk} = 2J^{\pm\pm}_{jk} \sin\theta_{jk}, \quad (2)$$

and the spin components $S^{x,y}$ are written in the local coordinate reference frames (one for each of the four sublattices), which are confined to planes perpendicular to the local $\langle 111 \rangle$ axes. $J^{\pm}$ and $J^{\pm\pm}$ are the symmetry-allowed NN exchange couplings [8,12,29]. We adopt the choice of the local frames as in Refs. [21,25], with the corresponding angular phases (inside a tetrahedron of sites labeled from 0 to 3) $\theta_{01} = \theta_{23} = 0$, $\theta_{02} = \theta_{13} = 2\pi/3$, and $\theta_{03} = \theta_{12} = -2\pi/3$.

In the clean limit, and for $-2 < \alpha \equiv J^{\pm\pm}/J^{\pm} < 2$, the mean-field ground states of Eq. (1) are given by spins collectively pointing along any direction in the XY plane, exhibiting a continuous U(1) degeneracy with energy $E_0/J^{\pm} = -6NS^2$. The order-by-disorder mechanism selects a finite set of six states out of the degenerate manifold: for $0 < \alpha < 2$ $[-2 < \alpha < 0]$ the ψ_2 (ψ_3) state is selected, which corresponds to spins pointing along one of the $\cos[(\pi/3)n]\hat{x} + \sin[(\pi/3)n]\hat{y}$ $[\cos[(\pi/3)n + (\pi/6)]\hat{x} + \sin[(\pi/3)n + (\pi/6)]\hat{y}]$ directions, with $n = 0, \ldots, 5$, in the local reference frame [11,12].

For definiteness, we introduce quenched disorder via

$$J^{\pm}_{jk} = J^{\pm}(1 + \epsilon_{jk}), \qquad J^{\pm\pm}_{jk} = J^{\pm\pm}(1 + \epsilon_{jk}), \quad (3)$$

where ϵ_{jk} are random variables. Bond disorder corresponds to ϵ_{jk} drawn independently from some distribution, while site dilution yields $\epsilon_{jk} = -1$ if either site j or site k hosts a vacancy and $\epsilon_{jk} = 0$ otherwise. Site dilution is parametrized by the concentration x of nonmagnetic impurities; for bond randomness see Ref. [30].

Destruction of order by random-fields effects.—We adopt a transparent argument by Aharony, originally constructed to show the instability of an ordered magnetic state against weak random anisotropy [34].

We *assume* LRO, which is uniform in the local frames of the Hamiltonian (1), with $|\alpha| < 2$, such that $\langle \mathbf{S} \rangle = \langle S^x \rangle \hat{x} + \langle S^y \rangle \hat{y} = m\hat{n}_{\parallel}$, where $\langle \cdots \rangle$ denotes the thermal average. The corresponding local exchange field is $\mathbf{h}_j = \sum_{k=1}^{z} (J^{xx}_{jk} \langle S^x \rangle + J^{xy}_{jk} \langle S^y \rangle)\hat{x} + (J^{yy}_{jk} \langle S^y \rangle + J^{xy}_{jk} \langle S^x \rangle)\hat{y}$, with the sum running over all the $z = 6$ NN sites. In the presence of random off-diagonal disorder the local exchange field $\mathbf{h}_j = h^{\parallel}_j \hat{n}_{\parallel} + h^{\perp}_j \hat{n}_{\perp}$ is not parallel to the mean magnetization. If we assume, for instance, $\langle S^x \rangle = \frac{1}{2}$ (and $\langle S^y \rangle = 0$) the local transverse component stems from the coupling J^{xy}_{jk} in Eq. (1), and is simply given by $h^{\perp}_j / J^{\pm\pm} = \sum_{k=1}^{6} \epsilon_{jk} \sin\theta_{jk}$; see also Eqs. (2) and (3).

The random transverse field $h^{\perp}_j$ [35] tips the local magnetization away from the mean direction $\hat{n}_{\parallel}$. The resulting transverse magnetization can be estimated in linear response as $\langle S^{\perp}_j \rangle = \sum_k \chi^{\perp}_{jk} h^{\perp}_k$, where $\chi^{\perp}_{jk}$ is the transverse bulk susceptibility of the clean system. The disorder-averaged transverse magnetization $\overline{\langle S^{\perp}_j \rangle}$ vanishes because $h^{\perp}_j$ has zero mean. In contrast, the averaged magnetization correlation function is nonzero: $\overline{\langle S^{\perp}_i S^{\perp}_j \rangle} \propto (\delta h)^2 \int d^d q (\chi^{\perp}(\mathbf{q}))^2 e^{i\mathbf{q}\cdot\mathbf{r}_{ij}}$, where $(\delta h)^2 \equiv \overline{h^{\perp 2}_i} > 0$ [36] and $d = 3$ the number of space dimensions. Further, $\chi^{\perp}(\mathbf{q}) \sim 1/(\lambda + \kappa_\mu q^2_\mu)$ is the Fourier-transformed bulk susceptibility, with λ being an effective anisotropy energy, such that the gap $\Delta \propto \sqrt{\lambda}$, and κ_μ parametrizing the gradient expansion [12,30].

Importantly, if the anisotropy energy λ *were* zero, we would have $\chi^{\perp}(\mathbf{q}) \propto q^{-2}$, such that the local transverse magnetization fluctuations diverge for $d \leq 4$: These fluctuations, arising from random off-diagonal exchange interactions and transmitted by long-wavelength modes, then destroy the assumed ordered state. This destruction of LRO can also be interpreted in terms of breaking the system into domains of linear size ℓ, following Imry and Ma [37]. Consider domains inside which the transverse exchange fields $\{h^{\perp}_j\}$ are atypically strong, such that the local order parameter will align with it, gaining an energy scaling as $\delta h \ell^{d/2}$ (as dictates the central limit theorem). In addition, there is a domain-wall energy cost. $\lambda \to 0$ implies an (accidental) continuous symmetry; hence, the order parameter can be continuously distorted from the $\hat{n}_{\parallel}$ to the $\hat{n}_{\perp}$ direction (in a region of fractions of ℓ), yielding a domain wall energy which scales as $J^{\pm}\ell^{d-2}$. Thus, for $d < 4$ it is favorable to break the system into domains of linear size $\ell \gtrsim (J^{\pm}/\delta h)^{2/(4-d)}$.

If, instead, the anisotropy energy λ is finite, the divergence is cured. Then the transverse spin fluctuations $\overline{\langle S^{\perp 2}_i \rangle}$

remain small for small δh; i.e., the assumed LRO is stable against weak randomness. A stability criterion can be obtained by the condition $\overline{\langle S_i^{\perp 2}\rangle} \ll 1$. This yields

$$\delta h \ll \kappa^{d/4}\lambda^{1-d/4}, \tag{4}$$

up to numerical prefactors [30], where $\kappa^2 = \sum_\mu \kappa_\mu^2/3$. The criterion (4) is consistent with the fact that small randomness destroys LRO in the limit $\lambda \to 0$ for $d < 4$, but not for $d > 4$. δh grows with increasing randomness, and, hence, we expect, for $\lambda > 0$ and $d = 3$, the destruction of LRO beyond a critical level of randomness. Below we show that the resulting phase is a CSG. Importantly, this argument is rather general, not restricted to the specific choice of Eq. (1), since it relies only on the existence of an off-diagonal exchange coupling [34].

Effective anisotropy and critical disorder.—Without quenched disorder, the system selects the ψ_2 state (for $0 < \alpha < 2$), with the spin gap Δ generated by the order-by-disorder mechanism [11,12,38]. Hence, λ is generically finite, and the ψ_2 state is stable against weak randomness, i.e., small δh. Indeed, a perturbation generated by a single defect is expected to be screened in the presence of a gap, such that a small concentration of defects does not qualitatively change the bulk state [21,22].

Importantly, in the classical limit the ψ_2 gap arises exclusively from thermal fluctuations, hence, $\Delta \to 0$ as $T \to 0$. Quenched disorder tends to stabilize the ψ_3 state instead [21,22], with the effective anisotropy energy λ scaling linearly with x [21]. Hence, the putative ψ_3 state has $\lambda \propto x$ as $T \to 0$. As we show in Ref. [30], the fluctuating transverse field follows $\delta h \propto \sqrt{x}$, such that the criterion (4) is parametrically fulfilled for small x, but can be expected to be violated at larger x. We conclude that, in the classical limit and at low T, the ψ_3 state is stable in a window $0 < x < x_{\rm cr}$, but replaced by a CSG for $x > x_{\rm cr}$. The quantum case is more involved and will be discussed later.

Classical phase diagram.—We are now in the position to discuss the classical phase diagram of the model (1). As originally explained in Refs. [21,22], the behavior at small x is governed by the competition between ψ_2 and ψ_3 LRO, favored by thermal fluctuations and weak disorder, respectively. This results in a phase boundary varying linearly with x. With increasing x, random-field effects grow, and LRO is eventually destroyed in favor of a CSG at all T. The low-T competition between ψ_3 and CSG is based on energetics, such that their phase boundary depends weakly on x at low T. Further, the effective anisotropy λ is particularly small near the $\psi_2 - \psi_3$ boundary, and this is where CSG will win first upon increasing x. Together, these considerations yield the qualitative phase diagram in Fig. 1(a), and they are well borne out by our quantitative numerical simulations, Fig. 2.

Classical MC simulations.—We turn to a detailed analysis of the state at large disorder. Previous theoretical investigations of related cases [39–41] suggest a glassy

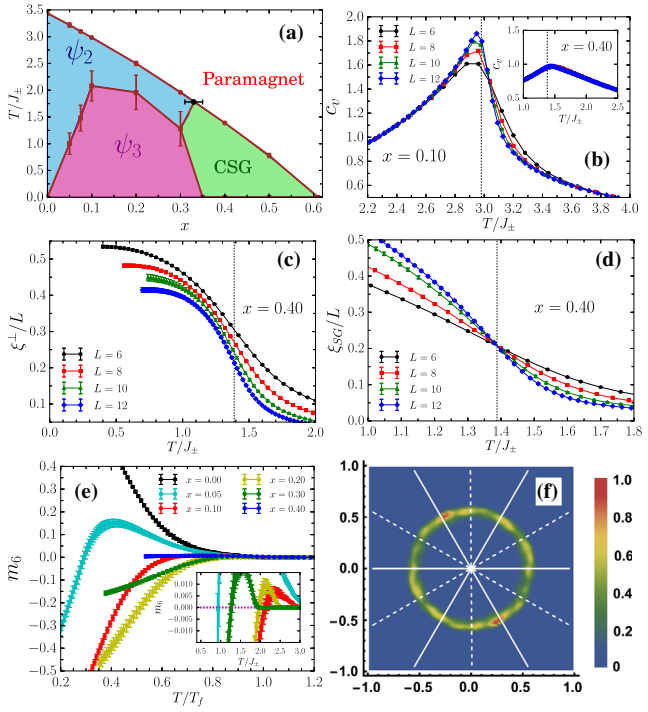

FIG. 2. Classical MC results for the Hamiltonian (1) for off-diagonal exchange ratio $\alpha = 1$. (a) Phase diagram as in Fig. 1. The freezing temperature T_f vanishes at the percolation threshold $x_p = 0.61$; the black dot marks the transition between ψ_2 and CSG; for details see text. (b) Specific heat $c_v(T)$ for $x = 0.1$ and $x = 0.4$ (inset). The vertical dashed line marks T_f. (c) Magnetic correlation length, plotted as $\xi^\perp(T)/L$, for $x = 0.4$ showing no crossing points. (d) Spin-glass correlation length, plotted as $\xi_{\rm SG}(T)/L$, for $x = 0.4$ showing a crossing point at $T_f/J_\pm = 1.39(2)$. (e) Clock order parameter m_6 as a function of T/T_f for $L = 10$ and several values of x. Inset: zoom around the region where m_6 changes sign. (f) Sample-to-sample distribution function $P(m_x, m_y)$ at $T = T_f/2$ for $x = 0.4$ and $L = 12$. The full (dashed) radial lines show the expected positions of the peaks associated to the $\psi_{2(3)}$ states.

state: The system breaks into domains of size ℓ, exhibiting no long-range magnetism, and eventually freezes into a spin glass below a temperature T_f. This scenario is in accordance with available experimental results for the random XY pyrochlores [23–26], and we now provide numerical evidence for it in the classical limit [42].

We perform classical MC simulations of the model (1) in the presence of site dilution and bond randomness [30]. Interpreting the simulation results requires care due to the several length scales present in the problem. In the clean limit, besides the linear system size L, there is an emergent length $\Lambda(T)$ [45,46] associated to a dangerously irrelevant $\mathbb{Z}_6$ anisotropy [13]. Therefore, the ground-state selection only takes place for $L \gg \Lambda$; to observe this numerically requires either low T or large L. With quenched disorder there is yet another length scale, the domain size ℓ, and we require $L \gg \ell$ to observe domain formation.

To monitor the order in the local XY planes we compute $m_{x(y)} = N^{-1}\sum_{j=1}^{N} S_j^{x(y)}$. The magnetic order parameter is $m = (m_x^2 + m_y^2)^{1/2}$, and we define an associated correlation length $\xi^{\perp}$. To discriminate between the states ψ_2 and ψ_3 we define a clocklike order parameter $m_6 = m\cos(6\varphi)$, where $\varphi = \tan^{-1}(m_y/m_x)$, with m_6 being positive (negative) for the $\psi_{2(3)}$ states. Moreover, we also keep track of the order-parameter distribution $P(m_x, m_y)$ [13,45,46], which is obtained considering statistics from different samples. In order to study spin-glass freezing we use the spin-glass (Edwards-Anderson) order parameter $q^{\alpha,\beta}(\mathbf{p}) = N^{-1}\sum_j S_j^{\alpha(1)} S_j^{\beta(2)} e^{i\mathbf{p}\cdot\mathbf{r}_j}$, where α and β are spin components, and (1) and (2) denote two identical copies of the system ("replicas") containing the same defect configuration. We define the spin-glass correlation length ξ_{SG} from this order parameter [47], and the freezing temperature T_f is obtained locating the crossing point of $\xi_{\mathrm{SG}}(T)/L$ for different system sizes L. Note that the Edwards-Anderson parameter alone does not differentiate spin glass from LRO [30].

In the following we focus on the case of site dilution relevant, e.g., to the compound $Er_{2-x}Y_xTi_2O_7$ [23]; bond randomness is discussed in Ref. [30]. Sample results are presented in Fig. 2. We determine the transition from ψ_2 to ψ_3 as the point where the curve $m_6(T)$ changes sign for $L = 12$; see inset of Fig. 2(e). On general grounds, we expect this transition to be first order. However, we find no traces of it in the specific heat, as also reported in Refs. [21,48], a fact which contributes to the error bars in Fig. 2(a). We leave a more detailed investigation of this point for future work.

For larger $x \gtrsim 0.3$ the behavior becomes more glassy: This is seen in the specific-heat curves; i.e., the maxima for all L become broad and size independent, signaling the building up of short-range magnetic correlations above a freezing temperature T_f [49], Fig. 2(b). Moreover, the correlation-length data $\xi^{\perp}(T)/L$ do not display crossing points, Fig. 2(c), whereas $\xi_{\mathrm{SG}}(T)/L$ show well-defined crossing points, Fig. 2(d). This signals glassy freezing in the *absence* of LRO. The freezing temperature $T_f(x)$ decreases with x, but remains finite up to the percolation threshold, $x_p = 0.61$, Fig. 2(a). The fate of the system is best judged by considering m_6 and the distribution $P(m_x, m_y)$, Figs. 2(e) and 2(f). m_6 is essentially zero for $x \gtrsim 0.4$, and $P(m_x, m_y)$ is peaked along a circle (instead of displaying sharp maxima), suggesting coexisting finite domains with distinct local spin orientations. We consider these MC data as clear evidence for the breaking of the system into domains with frozen spin configurations and no LRO, i.e., a CSG—this is the central result of this Letter.

The transition from $\psi_{2,3}$ to the CSG can be determined from the absence both of crossing points in $\xi^{\perp}(T)/L$ and of a clear-cut temperature trend of m_6. The resulting quantitative phase diagram is in Fig. 2(a), in remarkable agreement with the qualitative considerations which led to Fig. 1(a).

Quantum effects.—Turning to the quantum case, we note that the main effect of quantum fluctuations is to stabilize ψ_2 even at $T = 0$. This makes ψ_2 more competitive against ψ_3 and shifts the corresponding boundary to finite x [21]. As the competition between ψ_3 and CSG is expected to be weakly affected by quantum fluctuations, the extent of the ψ_3 phase consequently shrinks, Fig. 1(b). For strong quantum effects, $S = 1/2$, we may speculate that ψ_3 disappears completely from the phase diagram, yielding a direct transition from ψ_2 to CSG for all temperatures below T_f, but this requires a more detailed and quantitative analysis of quantum effects which is beyond the scope of this Letter.

Experiments.—We now confront our theory with experimental data. Assuming that $m = \langle S^x\rangle = \frac{1}{2}$ and $\langle S^y\rangle = 0$, we obtain the surprisingly simple result $\delta h = \sqrt{3x(1-x)}J^{\pm\pm}$ for site dilution [30]. For $Er_2Ti_2O_7$, extensive investigations of the order-by-disorder mechanism estimate $J^{\pm\pm} = 4.2\times10^{-2}$ [12] and $\Delta = 5.3\times10^{-2}$ meV [38]. To discuss the disappearance of LRO, and given that Eq. (4) is valid at weak disorder only, we instead compare the strength of the fluctuations δh to the clean-limit spin gap Δ to obtain an upper bound for the critical randomness, which reads $\delta h_{\mathrm{cr}} = f\Delta$, where f is a numerical factor of order unity. Experimentally, LRO disappears in $Er_{2-x}Y_xTi_2O_7$ around $x_{\mathrm{cr}} \approx 0.15$ [23]; from which we extract $f \approx 1/2$. Evidently, a more quantitative theory is desirable for an accurate determination of x_{cr}—this is left for future work. Our results are applicable to $Er_2Pt_2O_7$ as well, a compound which shows so-called Palmer-Chalker order ($\alpha > 2$) [16], not arising from an order-by-disorder mechanism. Using $f = 1/2$ and experimentally known model parameters, we predict that a small amount of vacancies, $x_{\mathrm{cr}} \approx 4\%$, destabilizes the magnetic order in $Er_2Pt_2O_7$ [30].

Broader aspects of the theory.—Our ideas are of generic relevance to magnets with strong spin-orbit coupling. Key to our scenario is the presence of off-diagonal exchange couplings in the microscopic Hamiltonian. These break spin-rotational invariance, leading to an anisotropy gap in a clean ordered state. In the presence of inhomogeneities, the off-diagonal couplings also produce random fields [35], which compete with the gap (protecting the ordered state) and favor CSG formation instead. The critical level of disorder where LRO disappears is, of course, material specific.

For the 2D quantum-spin-liquid candidate $YbMgGaO_4$ [50,51], it has been argued that quenched disorder in the off-diagonal couplings is responsible for the destruction of LRO [52,53] due to a "pinning-field" mechanism [53]. According to our scenario, this mechanism is akin to the random-field one which leads to CSG phase. In lower

dimensions ($d < 5/2$ [54]), the CSG phase melts into a fluctuating cluster-paramagnet phase, in accordance with the observations in Ref. [53].

Conclusions.—Combining analytical arguments and large-scale MC simulations, we have shown that defects induce fluctuating random fields in XY pyrochlore antiferromagnets. These ultimately destroy magnetic LRO, leading to a CSG phase beyond a critical level of randomness. Our theory resolves the previous discrepancy between theory and experiment, and is in semiquantitative agreement with experimental data on diluted $Er_2Ti_2O_7$.

We expect our ideas to motivate further studies into the nontrivial role of randomness in magnets with strong spin-orbit coupling, where the presence of off-diagonal exchange terms triggers a nontrivial competition between anisotropy gap and random fields [35].

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