Effective Hamiltonians and dilution effects in kagomé and related antiferromagnets

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What is the zero-temperature ordering pattern of a Heisenberg antiferromagnet with large spin length \( S \) (and possibly small dilution), on the kagomé lattice, or others built from corner-sharing triangles and tetrahedra? First, I summarize the uses of effective Hamiltonians to resolve the large ground-state degeneracy, leading to long-range order of the usual kind. Secondly, I discuss the effects of dilution, in particular to non-frustration of classical ground states, in that every simplex of spins is optimally satisfied. Of three explanations for this, the most complete is Moessner-Chalker constraint-counting. Quantum zero-point energy may compete with classical exchange energy in a diluted system, creating frustration and enabling a spin-glass state. I suggest that the regime of over 97% occupation is qualitatively different from the more strongly diluted regime.

I. INTRODUCTION

This paper addresses the isotropic antiferromagnet with quantum Heisenberg spins \( S_i \), on kagomé and analogous lattices (“bisimplex” lattices, to be defined shortly.) Everything is restricted to the large-\( S \) and low-\( T \) limit, where \( S \) is the spin quantum number and \( T \) is the temperature. Thus, to lowest order we may visualize \( S \approx |\mathbf{S}_i| \), where \( \mathbf{S}_i \) is a classical vector of unit length.

The theme of Sec. II is the usefulness of effective Hamiltonians, in which some degrees of freedom are eliminated, in favor of new terms involving the remaining degrees of freedom. The reader is also reminded that the classical picture can be qualitatively wrong, at the temperatures of interest, which are well below the quantum spinwave energies. For example, the quantum pyrochlore lattice is seen to be less degenerate than the kagomé case, in contrast to classical results; and the effective interactions favoring collinear/coplanar and other forms of order are much more powerful in the quantum case.

The rest of the paper concerns the effects of dilution. Does it produce a spin glass, or generate an effective Hamiltonian favoring long-range order, or preserve the exceptional degeneracy of the pure lattice? Sec. II reviews the effect of dilution in ordinary frustrated systems, to contrast with its effect in bisimplex lattices (Sec. IV), in which the simplex units are all satisfied. Sec. V, the bisimplex lattice linking layer \( \mathbb{P} \). The three-dimensional magnetic lattice of \( \text{e.g. gallium gadolinium garnet} \) is appropriately dubbed “hyperkagomé” \( \mathbb{K} \), since it too consists of corner-sharing triangles. The crossed-square lattice is a pyrochlore slab normal to \{100\}, with its top and bottom surfaces identified, and serves as a two-dimensional toy model for the pyrochlore. (See Fig. 2 of Ref. \( \mathbb{K} \).)

The site-percolation threshold \( p_c \) of the bisimplex lattice is obviously the bond-percolation \( p_c \) of its parent bipartite network. It is listed in Table \( \mathbb{C} \) because one expects qualitative changes of behavior at \( p_c \), if the spin system orders in any fashion. \( \mathbb{B} \) The sandwich lattice \( p_c \) is published here for the first time, to my knowledge. (See Appendix \( \mathbb{A} \))

The Hamiltonian couples nearest neighbors,

\[ \mathcal{H} = J \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j \]  

(1.1)

or classically, with a magnetic field \( \mathbf{B} \) included,

\[ \mathcal{H} = J \sum_{\langle ij \rangle} \mathbf{s}_i \cdot \mathbf{s}_j - \mathbf{B} \cdot \sum_i \mathbf{s}_i = \sum_\alpha \frac{J}{2} |\mathbf{L}_\alpha| - \frac{\lambda}{J} |\mathbf{B}|^2 + E_0 \]  

(1.2)

where the exchange constant is \( J \equiv j S^2 > 0 \), \( \lambda = 1/2 \), and the total spin of simplex \( \alpha \) is

\[ \mathbf{L}_\alpha = \sum_{i \in \alpha} \mathbf{s}_i. \]  

(1.3)

In the kagomé-sandwich case, the interlayer (kagomé to linking layer) coupling will be called \( J' \), but unless explicitly noted, I assume \( J' = J \), the kagomé-layer coupling.
Frustration means generally that, having broken up the Hamiltonian into local terms, we cannot simultaneously satisfy all of them. In the present case, each term in (1.2) is satisfied (in zero field) whenever $L_{ij} = 0$. Then each undiluted bisimplex lattice is completely unfrustrated from the simplex viewpoint, since it can be shown (by example) that every simplex can attain this minimum at the same time. Let $\mathbf{X}$ denote the classical ground state manifold (for $\mathbf{B} = 0$); it is massively degenerate on every bisimplex lattice.

II. EFFECTIVE HAMILTONIANS AND ORDER BY DISORDER

Our goal is to discover how the system breaks symmetry and orders (if it does). We assume provisionally that, for sufficiently large $S$, the quantum ground state is one of the classical ground states, dressed by some quantum zero-point spin fluctuations. (This clearly fails if the fluctuations about a classical ground state are as big as the distance to the next one, or if the system tunnels so freely between dressed classical states that the correct wavefunction is a superposition.) This assumption directly gives us the ordering pattern in a simple (e.g. triangular lattice) Heisenberg antiferromagnet, where the classical ground state is unique (modulo symmetries).

But commonly, the classical ground states have nontrivial degeneracies, so that every ground state has a different manifold of possible small spin deviations and consequently a different zero-point energy. Presumably, the true quantum ground state should be constructed around the particular (ordered!) classical ground state which has the lowest zero-point energy. When the thermodynamic state of the system has true long-range order in a particular pattern, due to this fluctuational energy, we could call it “order by disorder” [3]. (This usage of the term is broader in some ways, and narrower in others, than the “local” definition in Ref. 1.)

To transparently model the degeneracy-breaking, one may construct effective Hamiltonians in closed form (via intelligently applied perturbation theory). They are in general a harmonic zero-point energy $\mathcal{F} \sim \hbar \omega_h$, including all spin-wave frequencies $\hbar \omega_h$ (which depend implicitly on $\mathbf{X}$). This $\mathcal{F}(\mathbf{X})$ is the effective Hamiltonian, defined only on states in the ground state manifold. \(^1\) Call the local minima of $\mathcal{F}(\mathbf{X})$ the “favored states” $\{\mathbf{Y}\}$; these are a discrete set, (in all cases I know), modulo global rotation symmetry.

Let “ordinary degenerate antiferromagnet” mean one for which $\{\mathbf{X}\}$ is a finite-dimensional manifold – clearly every ground state is periodic. Examples are the FCC antiferromagnets [11,12] or the two-sublattice systems with second-neighbor interactions [3,4,12]. (A “highly frustrated” system might be defined as one in which the dimensionality of $\mathbf{X}$ is extensive, and two ground states may differ only locally.) For an ordinary degenerate system, with isotropic exchange interactions, a crude approximation for $\mathcal{F}(\mathbf{X})$ is the biquadratic effective Hamiltonian

$$\mathcal{H}_{\text{biq}} \equiv -\sum_{ij} K_{ij}(\mathbf{s}_i \cdot \mathbf{s}_j)^2,$$  \(2.1\)

This was independently posited phenomenologically [3]; it can be obtained analytically in a couple of ways using a perturbation expansion [2,10] around mean-field theory. One obtains $K_{ij} = S^2 j_{ij}/8h_{\text{loc}}$, where the local field $h_{\text{loc}}$ is $2jS$ in a bisimplex lattice. Eq. (2.1) correctly tells us that the favored states are the collinear ones. In the kagomé case, (2.1) should be replaced by a different functional form to approximately represent $\mathcal{F}(\mathbf{X})$, because (i) the criterion for favored states is that the spins are coplanar; (ii) the true function $\mathcal{F}(\mathbf{Y} + \delta \mathbf{X}) - \mathcal{F}(\mathbf{Y}) \sim |\mathbf{X} - \mathbf{Y}|$, linear in $\delta \mathbf{X}$ rather than quadratic in the deviations $\delta \mathbf{X}$.

More fundamentally, the harmonic approach fails in the kagomé case because it does not fully break the degeneracy. Every coplanar state has exactly the same harmonic-order Hamiltonian, if written using as coordinates at each site (i) the $y$ (out-of-plane) spin deviation and (ii) the rotation angle around the $y$ axis. The source of this, mathematically, is that $s_i \cdot s_j$ takes the same value (here $-1/2$) for every nearest-neighbor pair, in every favored ground state. Consequentially, on a bisimplex lattice

\[1\] Analogously, in the classical model with $T/J \ll 1$ one defines a harmonic free energy $F \sim T \ll E_0 \sim J$, by integrating out some degrees of freedom in the partition function. See Ref. [4].
with only triangles, *i.e.*, the kagomé or hyperkagomé lattices, $\mathcal{F}(\mathbf{Y})$ has exactly the same value for every coplanar state $\mathbf{Y}$. The number of such states is $O(\exp N)$, where $N$ is the number of unit cells.

To resolve the surviving degeneracy among favored ground states, we need a second effective Hamiltonian $\mathcal{G}(\mathbf{Y})$, obtained from some sort of self-consistent theory that takes account of anharmonic spin-wave interactions. Since $\{\mathbf{Y}\}$ is discrete, $\mathcal{G}(\mathbf{Y})$ is parametrized by discrete-spin variables: in the kagomé case, the “chiralities” $\tau_\alpha = \pm 1$ defined from the spins on each plaquette $\mathcal{L}$. Our own approximation [19,20] gave

$$\mathcal{G}(\{\tau_\alpha\}) = -\sum_{\alpha\beta} \mathcal{J}(R_{\alpha\beta})\tau_\alpha\tau_\beta,$$

which has the form of an antiferromagnetic Ising Hamiltonian on the honeycomb lattice, and is defined for every favored ground state $\mathbf{Y}$. The energy scale [18] is certainly $\mathcal{J} = O(\langle j S^2/3\rangle)$, i.e., down by only a factor $S^{-1/3}$ from the scale of the harmonic term $\mathcal{F}(\mathbf{X})$.

In the pyrochlore case, Eq. (2.1) is also too crude: here the favored ground states $\mathbf{Y}$ are the collinear ones, but their harmonic energies $\mathcal{F}(\mathbf{Y})$ are nondegenerate since different collinear states have different patterns of $s_i \cdot s_j = \pm 1$. It turns out that the special states that minimize $\mathcal{F}(\mathbf{Y})$ are still infinite in number, but only as $\exp(\text{const} L)$, where $L$ is the system’s diameter [21]: the kagomé-sandwich lattice seems to behave similarly. Thus, in the quantum case, the pyrochlore and sandwich lattices show more ordering tendency than the kagomé lattice (at harmonic order), whereas in the classical case it is the other way around [1].

### B. Pitfalls of classical modeling

Although large $S$ justifies visualizing each spin as a fixed-length vector, it does not justify a purely classical simulation of the system. The reason is that the interesting phenomena occur when $T \ll j S$, the scale of $\mathcal{F}(\mathbf{X})$. Thus thermal effects are only a small correction to the quantum effects. I believe there is an easy fix: a classical Monte Carlo simulation using a Hamiltonian which includes (an approximation of) $\mathcal{F}(\mathbf{X})$ and $\mathcal{G}(\mathbf{Y})$ ought to give valid physical results.

For a specific example, consider SCGO with $S = 3/2$ and a Curie-Weiss constant [24] of 515K, hence $j \approx 80K$. Then, in a kagomé lattice, the effective energy of coplanarity would be $0.14 j S$ per spin along a straight “spin fold” [17]: if we presume this also applies to the shortest rotatable loop of six sites, we get a barrier of 100K (alternatively $\frac{5}{3} j S \approx 300K$ from App. B of Ref. [1]). That is vastly larger than the analogous free energy barrier in a classical system, at the SCGO freezing temperature $\sim 3.5K$. Again, in a pyrochlore lattice with the same coupling $j$, the coefficient in $\mathcal{F}(\mathbf{Y})$ comes out to $K_{ij} = \frac{1}{15} j S \approx 7.5K$ per nearest-neighbor bond. That ought to induce a transition to long-range-ordered collinearity at a temperature of order 10K, which wouldn’t happen at any temperature in a classical system [1].

### III. DILUTION IN ORDINARY CASES

I now turn to the longer part of this paper: what effect(s) does dilution have on a highly frustrated system, specifically on a bisimplex-lattice antiferromagnet? Can it be represented by an effective Hamiltonian? Or can it turn an ordered system into a spin glass? The answers are “yes” for the “ordinary” antiferromagnets, which are reviewed in this section (in three regimes). The answers are different for highly frustrated magnets (later sections).

Starting with this section, I am considering classical ground states at $T = 0$ (unless explicitly noted).

#### A. Unfrustrated case

In an unfrustrated 2-sublattice (Néel) antiferromagnet (Fig. [1](a)), the ground state at occupied fraction $p$ (on the unique extended cluster) has exactly the same spin directions as at $p = 1$, on all the magnetic sites. The total magnetization does not cancel exactly, since the moment on the even or odd sublattice has $O(\sqrt{N})$ statistical fluctuations. An observable corollary of this observation is that the structure factor at wavevector $\mathbf{q}$,

$$S(\mathbf{q}) \equiv \frac{1}{N} \langle \sum_i e^{i\mathbf{q} \cdot \mathbf{r}_i} s_i^2 \rangle,$$

has the limiting behavior

$$\lim_{\mathbf{q} \to 0} S(\mathbf{q}) = p(1-p) > 0.$$
dependence. (It has the angular dependence of a dipole field and decays with distance as $1/|r|^d$, where $d$ is the spatial dimension.)

In the ordinary degenerate antiferromagnets, the energy reduction due to spin deviations depends on which ground state one deviates from. The average of this energy, over all the ways to place a low density $x_{\text{def}}$ of defects, defines an effective Hamiltonian function $\mathcal{H}_{\text{dil}}(X)$, proportional to $x_{\text{def}}$ and possessing the full symmetry of the ground states $X$ of the pure system. In exchange-coupled systems, $\mathcal{H}_{\text{dil}}(X)$ favors the least collinear ground states, and thus has an interesting competition with $\mathcal{H}_{\text{quant}}$.

Often, depending on how a defect’s local symmetry relates to that of the ground-state manifold $X$, each defect may prefer a particular ground state, out of the subspace $\{Y\}$ favored by $\mathcal{H}_{\text{dil}}$. In that case, dilution creates an “effective random field” varying in space and coupling to the discrete degrees of freedom $Y$. As in other random-field models, disorder wins if it is sufficiently strong or the dimension is low enough. The resulting state is a spin glass, in the sense that it lacks long-range correlations and has barriers, but has not been proven to be the same phase as a $\pm J$ spin glass.

C. Frustrated case: strong dilution

Now return to the same triangular lattice, but with strong dilution so that $p$ is just above $p_c$. It is well known that, near percolation, the typical connection between two sites (if any) is tenuous, and order is propagated over one-dimensional chains of sites, which are multiply-connected at occasional places. At $T = 0$, the spin directions alternate along such a chain, so it constrains the relative orientation of the endpoint spins to be parallel or antiparallel, depending on whether the number of bonds connecting them is even or odd. Order is propagated (at $T = 0$) as if there were a direct bond between the endpoints. But if there are two paths in parallel, they may disagree on the relation between endpoint spins; the smallest example is shown in Fig. 1(c). The ground state is a twisted spin-configuration that is found neither in the pure lattice nor in a singly-connected chain. In Fig. 1(c), the effect is to force an $80^\circ$ angle between the endpoint spin directions.

The extended connected cluster is an irregular network containing loops within loops of this type. It is plausible that its global “energy landscape” is like that of a spin glass, possessing numerous low-lying, nearly degenerate energy minima, separated by energy barriers.

IV. DILUTION IN A BISIMPLEX LATTICE

Dilution affects a bisimplex lattice quite differently than an ordinary frustrated lattice: simulations find that essentially every simplex remains satisfied, i.e., $L_\alpha = 0$, even for strong dilution. (This includes the simplices with nonmagnetic sites: they are still simplices, but the number of corners $q$ is reduced.) In this section, I will discuss the evidence for, and some corollaries of, that fact.

A. Simulations of 2-layer kagomé lattice

The original evidence was that (in the kagomé lattice) the local field is exactly 2 on most sites. I carried out more extensive simulations like Ref. [24] for the diluted kagomé-sandwich lattice – relevant to experiments on SCGO. (High-quality crystals of that material can be grown only with $p < 1$.) Over 500 independent realizations of the disorder were constructed, for a $10 \times 10$ lattice with $p = 0.55$ (i.e., 385 spins); each realization was relaxed from three different random initial configurations, to a ground state by 250 sweeps, in which each site in turn was set to its local-field direction. The program flags all configurations in which $|L_\alpha| > 0.1$ on any simplex with $q > 1$. This happened only on “one-eared” loops like Fig. 3(a). Now, the “one-eared” loop connects to the rest of the world in (at most) one point. Hence it can’t induce twisted, frustrated relationships among distant spins (like Fig. 1(c)), and can’t be responsible for spin-glass behavior. Similar results were found even when $J' \neq J$ (unequal interlayer and kagomé layer exchange), as well as in a plain kagomé lattice.

B. “Half-orphan spins”

Simplex satisfaction has observable consequences. Every simplex has $L_\alpha = 0$ – except, of course, that a $q = 1$ (one-spin) simplex has $|L_\alpha| = 1$. Let $O$ be the set of spins which have $q = 1$ on one side, with frequency $x_{\text{def}} \sim (1 - p)^2$ per unit cell. Also let $O'$ be the spins which are completely isolated ($q = 1$ on both sides), with frequency $x'_{\text{def}} \sim (1 - p)^5$ in the sandwich lattice – i.e., rare for $p > p_c$. The total magnetization (in units of $\mu \equiv (2\mu_B)S$) is

$$M_{\text{tot}} = \frac{1}{2} \sum_\alpha L_\alpha = \frac{1}{2} \sum_{i \in O} s_i + \sum_{i \in O'} s_i$$ (4.1)
The prefactors of $1/2$ in (4.1) appear because each spin’s moment is divided between two simplices. Schiffer and Daruka observed a Curie law contribution to the susceptibility, ascribed to “orphan spins” by them (which suggests the isolated spins), but more plausibly to the spins in $O[27]$, which might better be called “half-orphan”, since they belong to a simplex on one side but are isolated on the other side. If these moments had independent directions (but see Sec. V C 2), they would produce the Curie susceptibility, per cell,

$$\chi(T) = \frac{\mu_0^2}{3T} \left( \frac{1}{4} x_{\text{def}} + x'_{\text{def}} \right). \tag{4.2}$$

Insofar as the system is built from satisfied simplices, its total (classical) magnetization is zero. So, in place of (4.2), the structure factor (3.2) scales as $x_{\text{def}} \ll (1-p)$. Thus even with significant dilution, one expects $\lambda$ quantum chains with a valence-bond state [28]. I speculate that the same sort of paramagnetic defect as a “half-orphan” spin.

The half-orphan site has an unfrustrated, with a configuration like (3.1), the structure factor (3.2) scales as $1$ (classical) magnetization is zero. So, in place of (3.2), the structure factor (3.2) scales as $x_{\text{def}} \ll (1-p)$. Thus even with significant dilution, one expects $S(q \to 0) \approx 0$, as seen in SCGO (according to Ref. [3]).

The half-orphan site has an unfrustrated, with a configuration like (3.2) or (3.1). The perfect classical analog of the spin-1 moment that appears in diluted $S = 1$ quantum spin chains with a valence-bond state [28]. I speculate that quantum $S = 1$ bisimplex lattices indeed have a valence-bond state, and $S = 1/2$ defect moments.

The simplex-satisfaction concept also encompasses bond-randomness effects (see start of Sec. V C 2). The ground state of a tetrahedron with one ferromagnetic bond still has $L_\alpha = 0$ so it does not affect the near-cancellation of $M_{\text{tot}}$ or the Gauss’s law (Sec. IV D). On the other hand, a triangle with one ferromagnetic bond is unfrustrated, with a configuration like $\uparrow\downarrow\uparrow$. Then $|L_\alpha| = 1$: a ferromagnetic bond produces exactly the same sort of paramagnetic defect as a “half-orphan” spin.

### C. Satisfaction with unequal couplings

What happens to the picture of Eq. (4.2) when $J' \neq J$ (coupling to linking layer and/or field $B$ is nonzero)? It turns out (4.2) still works, provided we now take

$$L_\alpha = \sum_{i \in \alpha} w_{\alpha i} S_i \tag{4.3}$$

where $w_{\alpha i} = J'/J$ when $\alpha$ is (before dilution) a tetrahedron and $i$ is the linking-layer spin that caps it, and $w_{\alpha i} = 1$ otherwise. Also, we must replace $\lambda$ in (1.2) by $\lambda(\alpha) = J'/2J'$ when $\alpha$ is (before dilution) a triangle, and $1 - J'/2J'$ otherwise (when $\alpha$ is, before dilution, a tetrahedron). To satisfy each term, $L_\alpha = \lambda(\alpha)B$, which immediately implies (in the pure system)

$$s_i = \begin{cases} \frac{J}{\alpha' J} B, & i \text{ in kagomé layer}; \\ (1 - \frac{J}{\alpha' J}) B, & i \text{ in linking layer}. \end{cases} \tag{4.4}$$

Notice that the sum of all linking-layer spins must be exactly zero either if $B = 0$ or if $J' = J$ (even in nonzero field). Also, the total magnetization of satisfied simplices is (4.3), except the coefficients $1/2$ must be replaced by $\lambda(\alpha)$. Thus, with $J' \neq J$ the net magnetization of the satisfied simplices still must be zero.

There is a problem whenever $i$ is a linking-layer spin and $\alpha$ (after dilution) is a $q = 2$ simplex: one must set $w_{\alpha i} = 1$ again to correctly describe the satisfied bond. But then, in a magnetic field, the decomposition into terms (4.2) breaks down since there is no consistent value for $\lambda(\alpha)$. Even for $B = 0$, if $w_{\alpha i} = J'/J$ still for the other simplex that includes site $i$, the result (4.4) of the next section breaks down; in effect, this site enters (4.4) as another sort of “point charge”.

### D. Divergence theorem

I now introduce a sort of “Gauss’s Law”, which is handy for revealing the nonlocal effects of defects. Recall that in a “bisimplex lattice”, every spin belongs to one domain containing a subset of the simplices. Define a kind of “charge”,

$$Q(D) = \sum_{\beta \in D} (-1)^{\delta} L_\beta. \tag{4.5}$$

The theorem states that, assuming simplex satisfaction,

$$\sum_{i \in O} (-1)^{\delta(i)} s_i = Q(D) = \sum_{i \in \partial D} (-1)^{\delta(i)} s_i, \tag{4.6}$$

where $\partial D$ is the set of sites the domain boundary cuts through. Also, $\delta(i)$ tells which simplex has $q = 1$ (of the two containing the half-orphan site $i$), and $\delta(i)$ tells which simplex is in the interior (of the two containing boundary site $i$). The left-hand side of (4.6) follows since only the $q = 1$ simplices contribute nonzero terms in (4.5); the right-hand side follows because every spin in the interior
of $D$ appears in two terms of (4.3) with canceling coefficients. (Thus, half-orphan spins are the point charges in our “Gauss’s law”, while $(-1)^{q(i)} s_i$ plays the role of the normal component of the electric field at the surface.) This is a generalization of the sum rules of Ref. (b), Sec. III B.4 By drawing a succession of nested boundaries $\nabla D$ around a single half-orphan spin $s_i$, one shows that neighboring spins $s_j$ have correlations with $s_i$ that alternate in sign (as speculated by Mendels [27]) and decay with distance as $1/|r_i - r_j|^{d-1}$.

If we let $D$ include the entire system, there is no boundary term and the total “charge” (left-hand side of (4.4)) must be zero. This implies that, if there is just one half-orphan spin, it is impossible to exactly satisfy every simplex; if there are just two half-orphan spins, they must be exactly parallel or antiparallel (depending on the relative parity of their respective $q = 1$ simplices). But this law has a very large loophole: the violation of $L_\alpha = 0$ may be spread out uniformly over the simplices, such that the total energy cost is $O(1/N)$, which is negligible in a large system. There is a more physical argument why nearby “charges” ought, nevertheless, to cancel, as the electrostatic analogy would suggest. If they don’t cancel, spins in shells surrounding these “charges” are constrained by (4.3) to have a nonzero mean “charge”, which would reduce the number of possible states, thus reduce the entropy, thus increase the free energy at $T > 0$. I conjecture that, in the diluted quantum system, there is an analogous effective interaction between nearby half-orphan spins, mediated by the harmonic zero-point energy.

E. NMR experiments

An NMR experiment measures the distribution of the local fields $h$ felt by each NMR nucleus (Ga, in SCGO). This is an average of the magnetizations of its neighbor (Cr) spins, $m_i = \langle s_i \rangle$, where the average is taken over all ground states (for a fixed realization). The variance of $h$ would scale as $x_{\text{def}}$, so the NMR linewidth should scale as $\sqrt{x_{\text{def}}} \sim 1 - p$, as is seen experimentally.

The experiments can separate the NMR signal from the Ga(4f) site, which sees 12 Cr neighbors, including three from the linking layer. In the pure system, Eq. (4.4) implies the mean susceptibility is exactly $\mu^2/7J$ per spin, but the mean susceptibility seen by Ga(4f) is $\mu^2(1/4J - 1/8J')$.

V. WHY ARE THE SIMPLICES SATISFIED?

I now present three different – not exclusive – viewpoints for understanding simplex satisfaction.

A. Single-impurity explanation

The original explanation [23] just considered a single nonmagnetic impurity in a pure background, as appropriate to the weak dilution regime. Ref. [25] exhibited a rearrangement of a few spins (as few as 10) around this defect, as in Fig. 2, which completely satisfies every simplex. Farther away, the spin deviations are strictly zero, in contrast to an ordinary frustrated magnet (Sec. III B). The general method – first described for the pyrochlore case [30] – uses $q - 1$ rotatable loops, each connecting a spin on one of the simplices containing the impurity, to a spin on the other affected simplex. (“Rotatable” means that, in the pure lattice, the spins on the loop can be rigidly rotated together to produce other ground states.) The deviations around the impurity remind me of the screening around a test charge in a metal, by the high density of excitations at zero energy – in the present problem, those excitations are the rotatable loops.

Clearly this picture works for well-separated impurities – but that requires $1 - p$ to be quite small. In Fig. 4, all 19 spins must point in the pattern shown, modulo rotations; this conflicts with the pattern forced by a second impurity that sits anywhere on the two hexagons in Fig. 3 or the eight hexagons surrounding a single impurity that excludes other impurities on 40 other sites. (The above counts and Fig. 3 assume a background consisting of the $\sqrt{3} \times \sqrt{3}$ state; in other coplanar backgrounds, more spins are affected.)

The single-impurity picture, then, breaks down at $p \approx 0.97$, where the defect configurations start to overlap. To explain the simulations from this viewpoint, one is forced to postulate that the defect spin deviations obey a nonlinear superposition principle, as magical as that of solitons in certain one-dimensional systems.

B. Constraint propagation explanation

This picture is most appropriate to the strong-dilution limit near $p_c$, where the connected cluster is tenuous.

4Note also that, in the pure system (no “charges”), a vector potential for the “electric field” can be constructed, which is uniquely valued if $d = 2$; in the kagomé case, this is just the “spin origami” embedding of Ref. [23].
Along a simple chain of sites, spins alternate, propagating order as if there were a direct bond between the endpoints. Now allow a few spins neighboring this path; these decorate the path with triangles which I will call “ears” (see Fig. 3). Each “ear” removes constraints on the spins, since the two spins on the path are now constrained merely to differ by a 120° angle. When a path includes two or more “ears”, there is no constraint at all between the endpoint spins.

As in Sec. II C, the key question is whether two paths which rejoin (forming a loop) might propagate mutually exclusive constraints. In contrast to the triangular case of Fig. 1(c)), every simple loop has even length. (Derived for the kagomé case in Ref. 3, Appendix; for a general bisimplicial lattice, it follows from the bipartiteness of the underlying network.)

All loops with the same number of “ears” are equivalent, since – by the evenness lemma just stated – they have the same number (modulo 2!) of ear-less links, at which the spins simply invert. Therefore it suffices to study loops of length 6 as in Fig. 3. The one-earred loop shown is frustrated: the two spins in the ear should differ by 120° due to the triangle, but by 180° due to the five simple links connecting them. On the other hand, the two-earred loop forces the same 180° angle as a simple chain (between the endpoint spins where it connects to the rest of the world), and the three-earred loop forces the same 120° angles as a giant triangle would.

The simulations described in Sec. IV A found 38 one-ear loops in 500 realizations of 100 cells each at $p = 0.55$, in full agreement with the predicted frequency $p^7(18(1-p)^3+30(1-p)^5)$, per cell. Three other frustrated clusters were found, each one a one-earred 8-ring, and (in two variations) a hexagon pair sharing two ears, with one added ear. Each of these objects has (at most) one connection to the outside world forming the “ear”, and thus has does not propagate frustration on larger scales.

C. Constraint counting

The preceding discussions detected no frustration in either the strong or weak dilution regime, but are insufficiently general. The constraint-counting (“Maxwellian”) approach of Moessner and Chalker 3 is, I believe, the convincing explanation of simplex satisfaction. 5 The basic aim is to compute the dimensionality $D$ of the manifold of states in which all simplices are satisfied; as long as this set is non-null (i.e. $D > 0$), it is obviously the ground state manifold $X$.

Now let $X_g$ be the manifold of “generic” simplex-satisfied states (having no linear relationships among the spins, apart from the $K$ constraints required to satisfy all the simplices). The dimensionality of $X_g$ is

$$D_g = F - K.$$ (5.1)

Here $F$ is the number of degrees of freedom. Two per spin, so $F/N = 2p\nu$ per unit cell, where $\nu_{site}$ is the number of sites per unit cell, as in Table II. Table II gives the number of constraints per simplex, $K_{sim}(q')$, where $q'$ is the number of magnetic sites remaining after dilution. Naively $K_{sim}(q') = 3$, for the three components of $L_{\alpha} = 0$ (or its generalization to nonzero magnetic field). However, in zero field when the simplex is a single bond ($q' = 2$), the constraint is simply “$s_2 = -s_1$”: attaching $s_2$ does not change $D$, so the added constraints must be two to cancel the added degrees of freedom. Also, $K_{sim}(0) = K_{sim}(1) = 0$. The total number of constraints is

$$K/N = \sum_q \nu(q)C_{q'}^g p^q (1-p)^{q'-q} K_{sim}(q')$$ (5.2)

per unit cell, where $C_{q'}^g$ is the combinations of $q$ things taken $q'$ at a time, and $\nu(q)$ is the number of $q$-corner simplices per cell in the undiluted structure.

Most relevant to propagating order (or frustration) is the generic dimensionality $D_{gc}$ of the extended connected cluster; I approximate this by subtracting from $D_g$ the contribution to $F$ by isolated sites. The results are

$$D_{gc}^{kag}/N = 6p[1 - (1-p)^3] - 6p^2(2-p),$$
$$D_{gc}^{sand}/N = p[14 - 12(1-p)^5 - 2(1-p)^6] - 12p^2(p^3 - 3p + 3),$$
$$D_{gc}^{pyr}/N = 8p[1 - (1-p)^6] - 12p^2(1-p)^2(2-p),$$ (5.3)

for the three lattices. Counterintuitively, removing a single site from the kagomé lattice leaves $D_g$ unchanged, but removing an adjacent pair of sites increases $D_g$ by 1. In a general bisimplicial lattice, after dilution to the point $p_{tri}$) at which the average simplex is a triangle, further dilution ought to (slightly) increase $D_{gc}$. (Note $p_{tri} \approx 1, 0.9$, and 0.75 for the kagomé, sandwich, and pyrochlore lattices respectively.) Indeed, $D_{gc}(p)$ at first decreases rapidly with dilution and and tends to level off below $p_{tri}$, at roughly $D_{gc}(p) \sim 0.2$ on the kagomé, $\sim 1.6$ on the sandwich (SCGO) lattice, and $\sim 1$ on the pyrochlore, per unit cell. Since $D_{gc}(p)$ remains positive, we expect simplex-satisfied ground states at any $p > p_c$. 5

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5 A caution, however, is that “Maxwellian” counting is a mean-field theory. A threshold at which $D_g(p)$ vanishes is not generally the exact threshold, since portions of the structure may be under-constrained while others are over-constrained. For related issues in elastic percolation, see Ref. 3.
1. Generic/non-generic state crossover?

Do not be misled by the term “generic”! The typical (physical) ground state is “non-generic” if such states have a higher dimensionality than the “generic” ones. In particular, in the pure kagomé lattice, starting with the $\sqrt{3} \times \sqrt{3}$ coplanar state, one can rotate one of every three six-loops (“weathervanes”) by an independent angle. This ground-state manifold has dimensionality $D_n = 1/3$ per cell which dominates $D_g = 0$. Such states are non-generic because some spins (e.g., every second one in a rotatable loop) have exactly the same directions. (Indeed, in ground states relaxed from a random spin configuration, second-neighbor spins often point in nearly the same direction.) For small dilution, the non-generic states still dominate; these are precisely the coplanar states with rare impurities discussed in Sec. IV A. But the spin rearrangement at each impurity (Fig. 2) touches, and thus immobilizes, 10 six-loops, and the frequency of impurities is $3(1-p)$ per cell. So, I estimate

$$D_n(p)/N \approx \frac{1}{3}(1 - 30[1-p])$$

(5.4)

as the dimension per unit cell of the non-generic manifold. The non-generic manifold loses out to the generic one at $p \approx 0.97$, in the kagomé lattice. By contrast, the pure pyrochlore lattice has $D_n(1) = 2$, while it appears $D_n(1) = 1$ (and presumably $D_n(p)$ also plummets upon dilution). Thus, only the generic manifold is relevant in the pyrochlore. (essentially this paraphrases the absence of (local) “order-by-disorder”)

2. Back to frustration

Up to now, I considered the isotropic classical Heisenberg antiferromagnet, with possible dilution. The degeneracies retained by the classical ground state manifold, even under dilution, may be broken by various realistic perturbations. These can be handled by the constraint-counting framework, as in Table I (with extensions to accommodate constraints that involve two simplices).

Bond disorder has been identified in pyrochlore antiferromagnets and was modeled theoretically. It can produce “canted local states” of deviated spins in ordinary Heisenberg antiferromagnets (interacting as a spin glass). In a bisimplex lattice, it has a different effect: the constraints $K_{\text{sim}}(q)$ are greatly increased. Assuming a fraction $x_F$ of ferromagnetic bonds, inserting $K_{\text{sim}}(q)$ from Table I into Eq. (5.1) gives

$$D_{\text{pyr}}^g = 2(1 - 18x_F),$$

$$D_{\text{pyr}}^g = 2(1 - 21x_F)$$

(5.5)

in place of (5.3). Eq. (5.5) predicts that $x_F \approx 0.05$ is a threshold, beyond which the system becomes overconstrained and (presumably) spin-glassy.

Quantum fluctuations also impose collinearity in a tetrahedron. For easy-axis exchange anisotropy, rather surprisingly the $g = 3$ ground state manifold’s dimensionality is unchanged (3). (The constraint count for easy-plane exchange anisotropy is the same as for XY spins, see 8). Magnetic field is a special case (already covered in Sec. V B). The spins along a chain when $B \neq 0$ alternate between two possible directions, indicating that each extra link contributes only two constraints, as accounted in footnote c of Table I. Substitution into (5.2) and (5.3) shows that most of these perturbations make the generic states overconstrained, and presumably glassy, though (in the undiluted cases) one needs to rule out possible non-generic ground states.

VI. CONCLUSIONS

Large-$S$, non-random antiferromagnet should have periodic long-range order at $T = 0$, even on a highly frustrated lattice. A purely classical picture is invalid, even for $S \gg 1$, when temperature is far below the spin-wave energy $JS$, but this may be fixed up by using the effective Hamiltonians in the simulation (Sec. IV B).

Dilution does not engender a spin-glass in classical ground states, as it does in ordinary frustrated magnets, since simplices remain satisfied, as confirmed by simulations (Sec. IV A). This was understood most generally from constraint-counting arguments (Sec. IV A). The observed spin-glass state in SCGO might be attributed to a competition of dilution with the coplanarity tendency due to quantum fluctuations. Effective Hamiltonians (Sec. IV A) serve as a first (perhaps only) way to model the zero-point energy contribution in a disordered lattice – even if the functional form is not quite right – since a more exact treatment would be intractable, in the presence of the multiple perturbations of Sec. IV A.

Some physical corollaries are deduced about the structure factor, paramagnetic susceptibility, and NMR response, including a “Gauss’s Law” rule (Sec. IV D), which manifests the long-range effects of the paramagnetic (“half-orphan”) spins. Experiments sensitive to percolation on SCGO ought to be performed with occupations below the threshold $p_c \approx 0.5$, which is estimated here for the first time (Appendix A).

I speculated that – in the plain kagomé lattice, at least – there is a threshold $p_c \approx 0.98$ separating two regimes of dilution: nongeneric coplanar states, peppered with defects, at $p > p_c$, but at $p < p_c$ generic states, in which every trace is lost of the coplanar background (Secs. IV A and IV C). This would imply, of course, that it
is invalid to extrapolate experimental measurements for $p \in (0.9, 0.95)$ up to $p = 1$. 6

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APPENDIX A: PERCOLATION THRESHOLD OF KAGOMÉ SANDWICH LATTICE

A very simple program (no spins are involved) generated random configurations with a fixed number of diluted sites. Having found the number of sites $m_j$ in the $j$-th connected cluster, and letting $m_1$ be the largest of them, I computed two percolation quantities: the “percolation susceptibility” $\chi_p = N^{-1} \sum_{j \neq 1} m_j^2$, and $P_p = m_1/pN$, the fraction in the extensive (“infinite”) cluster. In the limit $N \to \infty$, one expects $\chi_p \sim |p - p_c|^{-\gamma}$ on either side of $p_c$, while $P_p \sim |p - p_c|^\beta$ on the high side, and $P_p \equiv 0$ on the low side.

From the behavior of these two quantities, I estimated $p_c \approx 0.50(2)$. To probe the systematic error due to size-dependence, several sizes were tried (up to $16 \times 16$, i.e. 1792 sites before dilution); however, a genuine scaling fit was not carried out.

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TABLE I. Bisimplex lattices. Number of sites \( \nu_{\text{site}} \) and of simplices \( \nu(q) \) are per Bravais unit cell.

| Name              | Derived from   | Bravais lattice | \( d \) | \( (q, q') \) | \( \nu_{\text{site}} \) | \( \nu(3) \) | \( \nu(4) \) | \( p_c \) |
|-------------------|----------------|-----------------|--------|---------------|--------------------------|-------------|-------------|--------|
| kagomé            | honeycomb      | triangular      | 2      | 3,3           | 3                        | 2           | 0           | 0.65   |
| garnet            | “gyroid” graph | bcc             | 3      | 3,3           | 6                        | 4           | 0           | > 0.5 ?|
| crossed-square    | square         | square          | 2      | 4,4           | 2                        | 0           | 1           | 0.50   |
| pyrochlore        | diamond        | fcc             | 3      | 4,4           | 4                        | 0           | 2           | 0.39   |
| kag. sandwich     | –              | triangular      | 2      | 3,4           | 7                        | 2           | 2           | 0.50(2)|

TABLE II. Constraint count \( K_{\text{sim}}(q) \) in \( q \)-corner simplices

| Case                          | \( q = 2 \) | \( q = 3 \) | \( q = 4 \) |
|------------------------------|-------------|-------------|-------------|
| Isotropic Heisenberg spins\(^a\) | 2           | 3           | 3           |
| The same, with spin waves    | 2           | 3\(^b\)    | 5           |
| One ferromagnetic bond       | 2           | 4           | 6           |
| Easy-axis exchange anisotropy| 4           | 4           | 5           |
| Magnetic field\(^c\)         | 3\(^d\)    | 3           | 3           |

\(^a\) Also the case \( J' \neq J \)
\(^b\) For the coplanarity constraint, add +1 for every site with \( q = 3 \) on both sides,
\(^c\) Also \( K_{\text{sim}}(1) = 2 \), since \( \mathbf{s}_i \parallel \mathbf{B} \) in that case.
\(^d\) But subtract 1 for every site with \( q = 2 \) on both sides of it.

FIG. 1. Diluting ordinary antiferromagnets; diluted sites and their bonds are always shown dashed. (a). Unfrustrated antiferromagnet: the total moment here is one down spin, the excess of odd sites over even sites (b). Removal of one spin in the triangular Heisenberg antiferromagnet, causing neighboring spins to deviate in the direction of the missing spin. Spins on the second ring outwards are given the directions they would have in the pure lattice, though in reality a small distortion is found at any radius. (c). A loop of sites with an odd number of steps introduces random frustration and spin relationships absent in the pure lattice. The upper (lower) path favors the endpoint spins to be parallel (antiparallel).

FIG. 2. One removed spin in the kagomé antiferromagnet, and the 10-site defect loop induced around it. Circles with dots or crosses are spins pointing directly out of or into the paper; “+” and “−” signs indicate spins with an outward or inward component. The surroundings are part of a coplanar \( \sqrt{3} \times \sqrt{3} \) ground state.
FIG. 3. (a,b,c). “One-ear”, “two-ear”, and “three-ear” loops with their spin configurations (unique, modulo rotations in spin space). “A”, “B”, and “C” mark three spin directions which differ by $120^\circ$. 