Magnets with strong geometric frustration

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A non-technical introduction to the theory of magnets with strong geometric frustration is given, concentrating on magnets on corner-sharing (kagome, pyrochlore, SCGO and GGG) lattices. Their rich behaviour is traced back to a large ground-state degeneracy in model systems, which renders them highly unstable towards perturbations. A systematic classification according to properties of their ground states is discussed. Other topics addressed in this overview article include a general theoretical framework for thermal order by disorder; the dynamics of how the vast regions of phase space accessible at low temperature are explored; the origin of the featureless magnetic susceptibility fingerprint of geometric frustration; the role of perturbations; and spin ice. The rich field of quantum frustrated magnets is also touched on.

The concept of geometric frustration dates back to 1950, when it was noticed that Ising antiferromagnets on the triangular lattice have properties very different from those of ferromagnets or bipartite antiferromagnets. Geometric frustration has been a topic of constant interest over the half century between then and now. Bursts of activity have originated from developments such as the discovery of high-temperature superconductors and the subsequent search for unconventional magnets, or, more recently, a still ongoing systematic study of frustrated magnetic compounds on the highly frustrated SCGO, GGG, kagome and pyrochlore lattices (see Fig. 1).

Geometric frustration arises when the arrangement of spins on a lattice precludes satisfying all interactions at the same time. The simplest case is provided by a group of three antiferromagnetically coupled spins: once two spins point in opposite directions, the third one cannot be antiparallel to both of them. Geometrically frustrated magnets are considered to be in a separate class both from unfrustrated and from disordered magnets (spin glasses and the like). This article concentrates on continuous, classical, disorder-free geometrically frustrated magnetism, although discrete, quantum and disordered models are also briefly discussed.

The popularity of geometrically frustrated magnets stems from the very rich behaviour they present. For example, magnetic analogues of solid, glassy, liquid and even ice phases have been identified in this class of magnets, which is increasingly seen as providing a stage for studying generic questions in many-body physics in a set of well-characterised compounds described by simple model Hamiltonians. A wide range of experimental probes are available for their study – including neutron and X-ray scattering, muon spin rotation (µSR), nuclear magnetic resonance (NMR), susceptibility and heat capacity measurements – which yield complementary information. For instance, recently begun NMR measurements on SCGO are providing information about the local physics at the different inequivalent sites of the magnetic Cr ions, complementing our knowledge obtained from the probes from which such local information is harder to extract.

In the following, however, only cur-

FIG. 1. Corner-sharing lattices, clockwise from top left: The pyrochlore lattice. A projection of the lattice of the Gadolinium Gallium Garnet (GGG), which consists of two separate, interpenetrating sublattices of corner-sharing triangles. The kagome lattice. A side-on view of the trilayer lattice of SCGO, consisting of triangles and tetrahedra. It can be thought of as two kagome layers coupled by an intermediate triangular layer (circles).
main weak although the temperature is below the scale 
set by the interactions.

This observation suggests a two step strategy for un-
derstanding magnets in this class. For the cooperative 
paramagnet, it should be sufficient to study a fairly sim-
ple model system to capture the generic behaviour char-
acterising this regime. Building on this, perturbations 
to the simple model Hamiltonian, appropriately chosen 
for each compound, are introduced to describe the non-
generic regime.

The remainder of this article adheres to this struc-
ture in that we first identify and discuss an appropriate 
class of model cooperative paramagnets and then con-
sider the effect of perturbations. In the process, we shall 
see that classical models of highly frustrated magnets 
have in common that, once the leading, frustrated ex-
change interaction has been optimised energetically, a 
large ground-state degeneracy remains. The collection 
of degenerate ground states (the ground-state manifold) 
provides no energy scale of its own and hence any per-
turbation has to be considered strong. Frustrated mag-
nets are thus model strongly interacting systems. The 
richness of their behaviour in the non-generic regime can 
be understood as a consequence of the non-perturbative 
nature of any term added to the leading, frustrated ex-
change Hamiltonian.

I. GROUND-STATE DEGENERACY OF 
FRUSTRATED MAGNETS

The main distinction between frustrated and unfrus-
trated magnets appears to be the presence of a large 
ground-state degeneracy in the former. In the following, 
we first give a description of how the degeneracy arises, 
and then provide a general quantitative determination of 
the size of the ground-state degeneracy based on a simple 
Maxwellian counting argument.

Our starting point is the classical nearest neighbour an-
tiferromagnetic Hamiltonian, $H_J = J \sum_{<i,j>} S_i \cdot S_j$, where 
the sum on $(i,j)$ runs over nearest-neighbour pairs and 
the spins $S$ are represented by classical vectors of unit 
length. $J > 0$ is the strength of the antiferromagnetic 
exchange. Let us first consider the case of a group of 
$q$ mutually interacting spins, for which the Hamiltonian 
can be rewritten, up to a constant, as

$$H_J = J \sum_{<i,j>} S_i \cdot S_j = \frac{1}{2} \sum_{i=1}^{N} \mathbf{L}_i^2,$$

(1)

where $\mathbf{L}_i \equiv \sum_{\alpha=1}^{q} S_{i,\alpha}$ is the total spin of the unit.

From this it can be read off that the ground states 
are those states in which the total spin $L$ vanishes. The 
appropriate configurations for Heisenberg spins are de-
picted in Fig. 2. Note that, up to global rotations, the 
ground states for pairs and triplets of spins ($q = 2, 3$) 
are non-degenerate, whereas there are two degrees of free-
dom, $\alpha$ and $\phi$, in the groundstate for a quartet of spins.

The origin of this difference is the following. For Heisen-
berg spins, the condition $L = 0$ imposes three constraints 
($L_x = L_y = L_z = 0$), independent of $q$. The number of 
degrees of freedom, however, increases with $q$. For $q = 4$, 
the constraints no longer suffice to determine the ground 
state uniquely, and the underconstraint shows up as the 
ground-state degrees of freedom.

For a lattice built up of frustrated units, this argument 
can be generalised: the dimension of the ground state, $D$, 
is given by the difference between the degrees of freedom, 
$F$, and the ground-state constraints, $K$. Note that the 
Hamiltonian can be written as $H_J = \frac{1}{2} \sum_{i=1}^{N} \mathbf{L}_i^2$, where 
$\alpha$ runs over all $N$ units.

For spins with $n$ components ($n = 2, 3$ being XY and 
Heisenberg spins, respectively), there are $n$ ground-state 
constraints per unit: the $n$-component vector $\mathbf{L} = 0$. 
The number of degrees of freedom per unit is largest for 
lattices where the frustrated units share sites, since the 
degrees of freedom of each spin are only shared between 
two units in this case. The kagome lattice is thus made 
up of corner-sharing triangles, whereas the pyrochlore 
lattice consists of corner-sharing tetrahedra. In addition, 
more complicated lattices are possible, for example the 
SCGO lattice consisting of triangles and tetrahedra, or 
the GGG lattice made up of non-planar corner-sharing 
triangles.

For lattices of corner-sharing units of $q$ spins with $n$ components, one thus obtains $D = F - K = N[q(q - 2) - q]/2$. The ground-state dimension $D$ grows with $n$ and $q$. For Heisenberg spins, it becomes extensive ($D = N$) at 
$q = 4$, i.e. the pyrochlore antiferromagnet has an exten-
sive ground-state dimension. Since physically it is hard 
to realise $n > 3$ or $q > 4$. Heisenberg magnets containing 
corner-sharing tetrahedra are the realistic systems where 
the effects of frustration are strongest.

This argument has relied on the constraints being in-
dependent and mutually compatible. For example, the 
ground state in a very strong magnetic field is always non-
degenerate (all spins aligned), although the Hamiltonian 
can still be written as a sum of squares of $n$-component

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{diagram.png}
\caption{Left: ‘Susceptibility fingerprint’ of strongly frus-
trated magnets. Top right: Units of $q$ spins with total spin $L = 0$. The shaded pair of spins is rotated out of the plane 
by an angle $\phi$. Bottom right: The easy axes of a pyrochlore 
magnet.}
\end{figure}
of thermal fluctuations around them, which give a different entropic weighting to each ground state. The softer the fluctuations around a particular ground state, the larger the region of phase space accessible near it, and the more time the system spends fluctuating around this state. It can now happen that the fluctuations around a special state (or set of states) are so soft that the systems at low temperature effectively spends all its time nearby. For this to occur, the entropy of this special set must dominate the total entropy of all the other states taken together. In Fig. 3, this is represented as the area of the shaded region near a special point becoming much larger at low temperature than the total shaded area elsewhere taken together.

In practice, the states with the softest fluctuations tend to incorporate some degree of long-range order (see Fig. 3), so that their selection implies an ordering transition. This phenomenon is thus known as order by disorder, since order is induced by thermal fluctuations, which are normally associated with a disordering tendency (and indeed, upon raising the temperature, the increasingly violent fluctuations do destroy the order they initially stabilised).

The concept of order by disorder was initially proposed by Villain and Shender, and it has received a great deal of attention in connection with the selection of coplanar order in kagome Heisenberg magnets. Order by disorder, although counterintuitive, has since been found to be almost ubiquitous – after a hard search, the first magnet shown to avoid ordering to my knowledge was one on a Bethe lattice.

In Ref. 8, a theory based on the Maxwellian mode counting described above was worked out to determine the presence of order by disorder for the general case of n-component spins arranged in corner-sharing units of q. The result is that ordered states, provided they exist, are selected for q and n both small, as depicted in Fig. 3. This region includes all the realistic cases (q ≤ 4, n ≤ 3), with the exception of the case q = 4, n = 3 which is marginal. This system, the Heisenberg pyrochlore antiferromagnet, is now universally agreed to remain disordered at all T, a result first suggested by Villain.

This concludes the first part of our program, namely the quest for a robust cooperative paramagnet. We can choose a classical magnet with sufficiently large q and n and expect it to reproduce the qualitative features of that regime faithfully, as has been done for the Heisenberg pyrochlore antiferromagnet and subsequently for a large-n kagome magnet. The former concentrated on the low-temperature statistical mechanics and dynamics of a cooperative paramagnet, whereas the latter contains a detailed treatment of the thermodynamics of this regime.
III. DYNAMICS

Having established that the cooperative paramagnet can explore a vast region in phase space even down to the lowest temperatures, the question naturally arises how it in fact does so. This question about its dynamics is one of the most intriguing aspects of cooperative paramagnetism, but one which has not received its fair share of attention over the years, despite the fact that experimental studies of this problem are not at all uncommon.

The semiclassical equations of motion for a spin precessing in the exchange field set up by its neighbours can be written as $dS_{\alpha \beta}/dt = -J S_{\alpha \beta} \times (L_{\alpha} + L_{\beta})$, with $\hbar = 1$, and $S_{\alpha \beta}$ being the spin shared by tetrahedra $\alpha$ and $\beta$. This leads to a simple equation of motion for the $L$: $dL_{\alpha}/dt = -J \sum_{\beta} S_{\alpha \beta} \times L_{\beta}$.

A pioneering numerical study of this dynamics was undertaken by Keren, who contrasted the behaviour of Heisenberg spins on the square and kagome lattices and who found that kagome lattice correlations decayed qualitatively more rapidly. This is because, in ordered magnets, the dynamics can usually be described satisfactorily by considering excitations around the ordered structure only, since overall changes in the ordered structure (rotation of the ordering direction) occur at exponentially longer timescales. In cooperative paramagnets, however, the motion from one ground state to another (parametrised by the $x$-coordinates), typically not related by symmetry, can occur on relatively short timescales.

These ground-state modes have zero frequency in the harmonic approximation (see Fig. 3). On top of the ‘spin-waves’ (locally parametrised by the $y$-coordinates), one therefore has to consider anharmonic effects (which are of course also of relevance for many other properties). This was done in Ref. 8, noticing that at low temperatures a separation of timescales occurs. The three timescales of was done in Ref. 8, noticing that at low temperatures a separation of timescales occurs. This leads to a simple equation of motion for the $L$: $dL_{\alpha}/dt = -J \sum_{\beta} S_{\alpha \beta} \times L_{\beta}$.

The salient feature of this result is that the decay time $\tau$ grows only algebraically (and not, for example, according to an Arrhenius law as the temperature is lowered. Note that this leads to a width, $\Gamma \propto 1/T$, in inelastic neutron scattering linear in $T$, which is close to what is observed in SCGO to which this theory should also apply. There is no sign of a phase transition, the cooperative paramagnetic phase extends all the way down to zero temperature. Note also that the excitation lifetime is much shorter than in an ordered magnet because the dynamics along the ground-state manifold induces a powerful scattering mechanism.

IV. THE SINGLE-UNIT APPROXIMATION AND THE SUSCEPTIBILITY FINGERPRINT

We have seen that cooperative paramagnets have a short-range correlations both in space and time, even in a regime where the temperature is below the energy scale set by interactions. One can thus hope that there might be a description in terms of variables which behave approximately as if they were decoupled. A good candidate set of variables are the total magnetisation vectors $L_{\alpha}$ of the basic units: these are the variables appearing in the Hamiltonian, and the equations of motion (see above) can also be cast in a simple form with their aid. In the low-temperature limit, they follow simple equipartition, while at high temperatures, they provide a Curie-Weiss law. These properties are shared in quantitative detail, upon inclusion of factors of two to account for the decomposition, by the magnet on full lattice.

The partition function for isolated units of $q$ spins were obtained in Ref. 24, and the pyrochlore magnet was approximated by a set of isolated tetrahedra. In Fig. 3, the resulting expressions for energy and susceptibility are compared against Monte Carlo simulations. The agreement is very satisfactory, the error being below 5% everywhere for the susceptibility and much less than that for the energy.

Note that the shape of the susceptibility is that of the susceptibility fingerprint pictured in Fig. 3 in that it follows the Curie-Weiss law down to temperatures well below $\Theta_{CW}$, before bending upwards (in this case, towards the exact $T = 0$ result for the susceptibility). The single-unit approximation thus provides a simple model resulting in a closed-form expression for the susceptibility of the cooperative paramagnet at all temperatures. This model has been extended to describe quantum spins on
frustrated lattices by Garcia-Adeva and Huber.

V. PERTURBATIONS

In real systems, the validity of the nearest-neighbour classical Heisenberg exchange Hamiltonian $H_J$ can at best be approximate. The real Hamiltonian will symbolically have the shape $H(P) = H_J + H_P$ where $H_P$ denotes one of many possible ‘perturbation’ such as anisotropies, quenched disorder, further-neighbour exchange, dipolar interactions, coupling to lattice degrees of freedom... If $P$ is the energy scale (perturbation strength) attached to $H_P$, we expect the theory of the cooperative paramagnet to be useful in the temperature regime $P < T < \Theta_{CW}$.

For $T \ll P$, the perturbation $H_P$ is singular, in the following sense. Consider, as the simplest case, a perturbation with a non-degenerate ground state, $\Lambda$, which is at the same time a ground state of the exchange Hamiltonian $H(0) = H_J$. This case is for example realised, as explained in Sect. VI in the case of a pyrochlore magnet with easy axis anisotropy.

For $P = 0$, the ground-state properties are obtained from the entropy-weighted average over the entire ground-state manifold in the limit $T \to 0$. For an infinitesimal $P$, the ground-state manifold (and along with it the correlation functions) discontinuously collapses onto the state $\Lambda$. Since $\Lambda$ can be any one of the multitude of states in the ground-state manifold of $H_J$, different perturbations can result in entirely different correlations. This in a nutshell is the origin of the richness of the behaviour encountered in geometrically frustrated magnets.

The relationship between the ground states of $H_J$ and of $H_P$ can of course be different from the simple case described above. Two other generic scenarios are illustrated by the case of quenched disorder. Consider first site dilution, where some ions on the lattice are replaced by vacancies or non-magnetic ions. The corresponding perturbation consists of shortening the spin on the site to be diluted (until it vanishes). In the case of pyrochlore Ising antiferromagnets, at small dilution, the size of the ground-state manifold changes but its dimension remains extensive. In the latter case, non-coplanar spin clusters are generated by the dilution. In the former, spins with no neighbours in one of the tetrahedron act as uncorrelated effective (classical) spin-$1/2$ impurities. The presence of such a population of ‘orphan’ spins was first pointed out by Schiffer and Daruka in a phenomenological model for the susceptibility of SCGO.

The other scenario is provided by bond disorder, where the strength of the bonds has a distribution of non-zero width. Here none of the ground states of $H_P$ and $H_J$ coincide. For small but nonzero (‘finite’) $P$, the ground states thus do not lie on the ground-state manifold of $H_J$, and many of the ways of finding compromises between $H_P$ and $H_J$ lead to a rugged energy landscape with barrier heights of order $P$. The existence of barriers is necessary to account for the glassiness seen in many compounds. Recent XAFS experiments on lattice disorder in one such pyrochlore compound $(Y_2Mo_2O_7)$ do suggest a distribution of bond lengths wide enough to give rise to a substantial degree of bond disorder.

VI. QUANTUM FRUSTRATION

Of all perturbations, the introduction of quantum fluctuations is probably the most interesting as the ground-state wavefunction can turn out to be any linear combination of the classical ground states. Unusual correlated or disordered (‘spin liquid’) magnetic states, with unconventional excitations and quantum phase transitions, can thus arise.

A gentle approach to quantum magnetism lies in a semi-classical (large-$S$) treatment of our model Hamiltonian $H_J$. The leading effect in $1/S$ is the generation of a zero-point contribution to the effective energy of a classical ground state, which looks like a classical perturbation. This energy may be represented by a bilinear term favouring collinearity in an effective energy functional. However, the harmonic analysis at $O(1/S)$ may still preserve a massive degeneracy, as happens in the case of the kagome magnet, where all the coplanar states have the same zero-point energy. Selection of a single state in that case is believed to be caused by anharmonic interactions, which select a $\sqrt{3} \times \sqrt{3}$ configuration with a tripled unit cell. Similarly, a biquadratic term favouring collinear configurations retains the extensive zero-point entropy of a pyrochlore Ising antiferromagnet.

As the spin length is decreased further towards $S = 1/2$, the strength of the quantum fluctuations increases. Sufficiently violent quantum fluctuations might destabilise any ordered structure present at large $S$, the same way that strong thermal fluctuations destroy thermal order by disorder. There is strong evidence from numerics that the $S = 1/2$ kagome Heisenberg antiferromagnet is indeed quantum disordered, as are the kagome Ising antiferromagnet with quantum fluctuations introduced via a transverse field or the triangular lattice Heisenberg magnet either with a multiple-spin exchange term added to increase the strength of quantum fluctuation in a “large-$S$” treatment or in a valence bond dominated phase. Similarly, a perturbative analysis by Canals and Lacroix of the $S = 1/2$ pyrochlore antiferromagnet finds a quantum-disordered phase. For this case, however, Harris et al. have suggested that long-range order may nonetheless be present in higher-order spin correlation functions and Isoda and Moriya have proposed the existence of a valence-bond crystal.

As indicated above, there is much more to quantum frustration than the discovery of such quantum spin liquids. However, a proper discussion of quantum frustra-
tion lies beyond the scope of this article, and we now move on to quite a different subject, the magnetic version of ice.

VII. SPIN ICE

Probably the most remarkable recent experimental development has been the discovery of spin ice, in experiments on the compound \( \text{Ho}_2\text{Ti}_2\text{O}_7 \) \(^{43}\), which was supplemented by another titanate compound, \( \text{Dy}_2\text{Ti}_2\text{O}_7 \) \(^{44}\).

Harris et al. noticed that a strongly anisotropic ferromagnet on the pyrochlore lattice is frustrated, whereas an antiferromagnet is not. The reason for this lies in the orientation of the easy axes, depicted in Fig. 2, which the spins are constrained to point along by the anisotropy. The exchange Hamiltonian (Eq. 1) seeks to minimise (maximise) the total spin of the tetrahedron in case of antiferromagnetic (ferromagnetic) exchange. The two configurations with the spins pointing all in and all out on alternating tetrahedra have \( L_a = 0 \) everywhere. The antiferromagnetic ground state is thus only trivially degenerate and appears unfrustrated. By contrast, there are exponentially many configurations maximising the total spin on each tetrahedron, namely all those with two spins pointing in and two pointing out on each tetrahedron. This is equivalent to the ice model, as the ice rules state that each oxygen has two protons sitting near and two far from it in the ice structure. In this model, a spin pointing out/in is taken to represent a proton sitting near/far from an imaginary oxygen placed at the centre of the tetrahedron. These ice states are in fact the ground states of an Ising model on the pyrochlore lattice, which curiously had already been noticed by Anderson \(^{45}\) in the first discussion of frustration on the pyrochlore lattice in 1956. Thus we now have a magnetic compound which cannot only be used to study ice physics but which also turns out to have a large number of other interesting aspects. \(^{46,47}\)

Theoretical work has so far thrust in two directions. Firstly, unusual properties of spin ice as a model many-body system are being explored. For example, the spin ice ground states are massively (discretely) degenerate, giving the system an extensive zero-point entropy. Harris et al. \(^{48}\) noted that a magnetic field, \( B \), applied in the \([100] \) direction can lift this degeneracy completely as a result of the orientations of the easy axes. The contributions of both the field and the entropy to the free energy are extensive, the latter being weighted by the temperature. At \( T = 0 \), the magnetic field energy thus dominates, and the ground state (termed an entropy-poor, “liquid” state) has the maximal magnetisation compatible with the easy axis constraints. However, as \( T \) is increased, the entropic contribution eventually dominates, and a first order transition to the entropy-rich (“gas”) state ensues, with a discontinuous drop in the magnetisation.

The curious feature of this transition is that it takes place between two states not differing in symmetry. In particular, the line of first order transitions in the \( B - T \) plane terminates in a critical point, very much the same way as happens in a conventional liquid-gas phase diagram, allowing a continuous path from the liquid to the gas without encountering any transition (Fig. 3). Secondly, two groups are studying the microscopic details and the resulting behaviour of the two titanate compounds mentioned above. Both are incorporating long-range dipolar interactions in addition to the nearest neighbour exchange. Siddharthan et al. find \( \text{Ho}_2\text{Ti}_2\text{O}_7 \) to be in a partially ordered state, whereas den Hertog et al. conclude it to be a bona fide spin ice compound. The origins of this disagreement are not entirely clear and may be due to problems involved in approximating the long-range nature of the dipolar interactions.

VIII. WHAT’S NOT HERE

In this article, I have tried to give a non-technical introduction to and a short overview of the theory of strongly frustrated magnets. I hope to have conveyed to the reader the idea that this field is a rich one, and, if nothing else, that a review article not constrained by size limits is by now overdue. I had to skip many exciting topics; these include the unconventional heavy fermion behaviour in \( \text{LiV}_2\text{O}_4 \) \(^{50,51}\) interactions of orbital and spin degrees of freedom \(^{52,53}\) rigorous results on ground states of frustrated quantum magnets, and the many facets of quantum itinerant magnetism to name just a few. I have completely omitted any mention of one-dimensional systems. Other overview articles can be found in Refs. 1, 19, 43, 44, 56.

For magnets on the triangular lattice, there exists a very thorough review by Collins and Petrenko. Classical frustrated Ising magnets on a wide range of lattices are reviewed in Ref. 37.

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