Surprise(s) in magnets without net moments

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(Dated: January 12, 2009)

We are all familiar with ferromagnetic and antiferromagnetic materials, in which the localized
ionic moments (in case of ionic insulators) or the electronic spins (in case of metals) go into a long-
range ordered state with a net macroscopic moment (in case of ferromagnets) or a net macroscopic
sublattice magnetization (in case of antiferromagnets). However this behaviour is far from ubiquitous
even in ionic insulators with well-developed local moments. Indeed, there are many ionic insulators
in which the dominant interactions between the local moments compete with each other, leading to
a cooperative paramagnetic state with no ordering of the moments down to the lowest temperatures
accessible to experiments. The physics of such magnets without net moments has some interesting
and surprising aspects, which are touched upon in this brief review.

PACS numbers: 75.10.Jm 05.30.Jp 71.27.+a

Overview: In many ionic insulators, some of the ions
have non-zero ground-state angular momentum, and an
associated magnetic moment. These localized magnetic
moments usually interact with each other in two ways:
On the one hand, by virtue of being magnetic dipoles,
each moment reacts to the dipole field of all other mag-
netic dipoles, giving rise to a long-range anisotropic mag-
netic dipolar interaction. On the other hand, the virtual
hopping of charge carriers between neighbouring mag-
netic ions gives rise to a short-ranged (typically nearest
neighbour) interaction generally known as the exchange
interaction $\tilde{J}$.

This exchange energy can be written as $E = J \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j ; J > 0$, where $J$ is the exchange constant
and the subscripts refer to pairs of nearest-neighbour
moments. In many commonly occuring cases, $J$ is positive
and therefore antiferromagnetic in nature, in that it
encourages antiparallel alignment of pairs of neighbouring
moments. In many situations, this exchange constant
is much larger than the long-range dipolar coupling,
which can then be left out of the analysis to a very good
approximation, while in other cases, the dipolar cou-
pling can dominate over the nearest-neighbour exchange.
[Here, the spins $\mathbf{S}$ are of course quantum-mechanical op-
erators; however, for many purposes at not too low tem-
peratures, they can be usefully approximated by classical
vectors of fixed length, particularly if the spin quantum
number $S$ is 3/2 or higher.]

When the magnetic ions form a bipartite lattice (in
which the lattice can be broken up into two sublattices
in such a way that the nearest neighbour exchange coupling
connects only pairs of spins belonging to different
sublattices), this antiferromagnetic exchange energy is
minimized by the so-called Neel state in which all spins
lie along a spontaneously chosen axis $\mathbf{n}$ and every spin
points anti-parallel to its nearest neighbours [In two and
higher dimensions, this picture also gives an essentially
correct caricature of the ground state of the full quantum
problem on a square or hypercubic lattice.] This is the
case, for instance, in the compound MnO which has a
Neel ordering temperature of approximately 116 K.

However, there are many other examples in which the
magnetic ions are coupled by exchange couplings that do
not obey this bipartite constraint—in other words, there
are triangles in the nearest neighbour connectivity of the
lattice. Such magnetic lattices with triangular motifs in
them are good examples of geometric frustration. To see
the significance of such triangular motifs, it is enough
to note that the Neel (antiferromagnetic) state along any
axis $\mathbf{n}$ is frustrated in the presence of such triangles, since
there is no unique way of satisfying all the exchange in-
teractions (Fig 1) fully.

In many situations [2], this results in a macroscopic
degeneracy of classical minimum energy configurations.
At intermediate temperatures $T$ that are less than the
exchange $J$, but are not small enough for the quantum
mechanical nature of spins to matter, the spin correla-
tions (measured, say, by neutron scattering experiments)
in the system simply reflect this macroscopic degeneracy,
and can be modeled in a universal way in terms of aver-
ages over an ensemble that gives a certain weight to each
of these minimum energy configurations [3]. To a first
approximation, this weight is of course uniform and the
same for each minimum energy microstate, and the sub-
leading effects of canonical fluctuations (that increase the
energy from this minimal value) can also be included in
a more sophisticated treatment. Many examples of such
frustrated magnets are known. On the pyrochlore lattice,
these include the $Cu^{2+}$ based $S = 1/2$ magnet paramela-
conite [4] and the $Cr^{3+}$ based $S = 3/2$ magnets CdCr$_2$O$_4$
and HgCr$_2$O$_4$ [5]. Several interesting examples have also
been studied on the kagome lattice—these include $Cu^{2+}$
based $S = 1/2$ volborthite and other systems [6], $Ni^{2+}$
based $S = 1$ magnets Ni$_2$V$_2$O$_4$ [7], $Cr^{3+}$ based $S = 3/2$
states [8], and $Fe^{3+}$ based $S = 5/2$ magnets Fe jarosite
[9].

Much of the interest in frustrated magnetism arises
from the non-trivial nature of the resulting state. In par-
FIG. 1: Three spins interacting antiferromagnetically with each other cannot satisfy the demands of all the exchange interactions.

particular, as we will see explicitly in examples below, the resulting intermediate state is not a simple paramagnet with each moment fluctuating more or less independently of the others. Instead, it is typically a non-trivial cooperative paramagnet, with a complicated pattern of correlations between far away moments. Such a cooperative paramagnetic state can have some very unusual properties, and that is the really interesting thing about these systems. Finally, it is also useful to keep in mind that the ultimate fate of such magnets at very low temperatures is less universal, and depends sensitively on the effects of quantum fluctuations and other (subdominant) interactions acting in this subspace.

Toy model—The triangular lattice antiferromagnet: Fortunately for us, the triangular lattice Ising antiferromagnet provides a simple example where a great deal of the foregoing can be illustrated quite explicitly. Here ‘Ising’ refers to the fact that we consider a simplified situation in which ‘spins’ that are not unit vectors, but instead discrete variables that can take two values ±1. Such models are generally referred to as Ising models, after Ising, who studied them first in his Ph.D thesis. Although this model looks very over-simplified at first sight, it does have the potential to describe real magnets at least in some cases. This is because real magnets have local moments that often correspond to ionic ground state multiplets with a non-zero value of orbital angular-momentum. In such cases, spin-orbit coupling in the presence of strong crystal field effects can induce a single-ion anisotropy term \(-D \sum_i (S_i^z)^2\) in the magnetic Hamiltonian. If \(D\) is large (compared to \(J\)) and positive, then the spins predominantly prefer to be in one of two states \(S^z = \pm S\) (for spin \(S\) moments), which can be thought of as the two Ising states in our foregoing description [One example of this is the Kagome lattice antiferromagnet Nd-Langasite, where a description in terms of a Ising magnet on the Kagome lattice apparently works quite well].

With that background, we now ask: What configurations minimize the nearest neighbour Ising exchange energy \(E = J \sum_{(ij)} \sigma_i \sigma_j\)? Clearly, the answer is all configurations in which each triangle has either two ‘up’ (+1) spins and one ‘down’ (−1) spin, or vice versa. These configurations minimize the ‘frustration’ induced by the competing antiferromagnetic interactions by ensuring that each triangle has exactly one frustrated bond (pair of parallel Ising spins).

Furthermore, these minimally frustrated configurations have a relatively ‘clean’ characterization in terms of dimer covers of the dual honeycomb lattice. More explicitly, consider the honeycomb net formed by forming links between the centers of triangles across the shared side of the triangle. If we place hard-rods on each honeycomb link that crosses a frustrated bond in a minimally frustrated configuration, then each honeycomb lattice site will have exactly one hard-rod covering it. Such configurations of hard-rods define so-called ‘dimer covers’ of the honeycomb lattice, and clearly, there is a one-to-one mapping between dimer covers of the honeycomb lattice and minimum frustration states of the classical Ising model on the triangular lattice.

As the temperature \(T\) falls well below the exchange energy scale \(J\), most triangles of the lattice will satisfy the minimum frustration condition and have exactly one frustrated bond. Indeed, one expects that a typically low-temperature configuration will differ from a minimally frustrated \(T = 0\) configuration only by an exponentially small \((O(e^{-J/k_B T}))\) density of triangles with three frustrated bonds. If we ignore the effects of such defects, the \(T \ll J\) properties of this system can thus be modeled by calculating the properties of the ensemble of minimum frustration states, with equal weight to each such state.

This is most conveniently done in dimer language, and one learns two important things upon translating back to the language of Ising spins: The first is that the Ising spins are not ordered even at \(T = 0\), i.e there will be no magnetic Bragg peaks even in a hypothetical neutron scattering experiment performed on our Ising magnet. The second is that the correlations between spins do not decay away to zero on the scale of a few lattice spacings, as would be expected for a simple paramagnet in which the moments fluctuate independently of each other. Instead, the correlations decay to zero very slowly, as the inverse square-root of the separation between the spins.

The \(T = 0\) state is thus a cooperative paramagnet in which the moments are correlated with each other over macroscopic distances although there is no long-range ordering. In this simple toy model, it is also possible to put back the exponentially small density of defect triangles into our description, and it can be shown that these do not change the \(T = 0\) picture in any striking way—all that happens is that the defect triangles disrupt the slow power-law decay of correlations beyond a length-scale \(\xi\) that corresponds to the typical inter-defect distance, and the correlations decay exponentially rapidly to zero for \(r \gg \xi\).

There are thus two different but related questions that one needs to keep in mind when thinking about the low temperature properties of such magnets: The first is the nature of the degenerate minimum exchange energy configurations, and the ensemble they define. In particular,
in the limiting case of zero temperature, physical quantities can be modeled by averages over this restricted ensemble of minimum energy configurations. The second is the nature of the thermally induced defects that allow a system to locally deviate from a minimum energy configuration, and the properties of a dilute, extremely cold gas of these defects—the low temperature properties of the magnet depend both on the nature of the ground state ensemble, and the statistical mechanics of the defect gas.

**Effective field theory for the \( T \to 0 \) limit:** In order to prepare the ground for our later discussion, it is useful to spend a little time understanding these results from the perspective of a coarse-grained effective field theory. The idea is think of the dimer occupation on a link as the value of an electric field \( e \) on the corresponding link of the honeycomb lattice, with the sign convention that the \( e \) on each link always points from the \( A \) sublattice site to the \( B \) sublattice site of that link (see Fig 2). Clearly, the dimer constraint now translates to the statement that there is a static + charge on each \( A \) sublattice site of the honeycomb net, and a static − charge on each \( B \) sublattice site. Now, we solve for this Gauss’s law divergence constraint by writing \( e \) in terms of a height field (which is the two dimensional analog of the vector potential of ordinary electrodynamics).

This height field \( h \) is defined on the original triangular lattice sites, but is quite distinct from the original spins, and in terms of \( h \), we may write the electric field on link \( l \) as

\[
e_l - \frac{1}{3} = h_{L(l)} - h_{R(l)}
\]

where \( R(l) \) and \( L(l) \) are triangular lattice sites to the right and left of this link (as defined when looking down the link from its \( A \) sublattice end (see Fig 2).

We now note that the staggered dimer configuration in which we occupy all links of one orientation and leave all others free corresponds to a height configuration with maximum tilt (gradient) in one of the principal directions of the triangular lattice. By inspection, we also see that such a staggered configuration cannot be changed into any other dimer configuration by any local moves that do not involve a macroscopically large number of links, and thus has very few nearby dimer configurations. Conversely, dimer configurations that can be transformed into other valid dimer configurations in a large number of ways tend to have zero average tilt in height language.

With this motivation, one postulates a coarse-grained ‘free energy’ that captures the entropic weight of different height configurations and writes the \( T = 0 \) partition function as

\[
Z = \int \mathcal{D}h(x) \exp \left( -\frac{K}{2} \int d^2x (\nabla h)^2 \right),
\]

where \( K \) is a phenomenological ‘stiffness’ parameter. Of course, in order to use this effective field theory, one needs a prescription for writing the local spin density \( \sigma(r) \) in terms of the height fields. To understand this correspondence, one may start by fixing one spin, say the spin on the site at the origin, to be up \( \sigma(r = 0) = +1 \), and the corresponding height to be zero \( h(r = 0) = 0 \). Now, we note that \( 3h \) jumps by an odd number whenever one crosses an unfrazed bond (across which the spin flips sign), while the height jump is even across a frustrated bond (across which the spin remains unchanged).

This immediately implies that a spin at some other site \( r \) will be up if and only if \( 3h(r) \) is even [10]. This provides one piece of the correspondence between the height field and the spin field. The second, and in many contexts more crucial, piece of the correspondence is slightly trickier to understand, and is best appreciated by trying to predict the spin value at site \( r = (x, y) \) by going across \( x \) links of the dual honeycomb lattice in direction \( T_0 \), followed by \( y \) links of the dual lattice in direction \( T_1 \) [11]. Now, each dimer crossed in the process guarantees that the spin state has not changed, while each empty link corresponds to a flip in the spin state. We may therefore write \( \sigma(r) = \exp(i\pi \sum_i (1 - n_i)) \sigma(0) \), where \( n_i \) is the dimer number on all the links \( l \) thus encountered. Rewriting this in terms of the height field allows us to argue that \( \sigma(r) \sim \exp(2\pi i (x+y)/3 + i\pi h(x,y)) + h.c. \)

Thus the zero momentum and momentum \( \pm Q = \pm (2\pi/3, 2\pi/3) \) components of the spin field have a simple local representation in terms of exponentials of the height field, and the long-distance properties of the spin correlations may be captured by the correspondence: \( \sigma(r) = c_Q \cos(\pi h(r) + 2\pi(x+y)/3) + c_0 \cos(3\pi h(r)) \), where \( c_Q \) and \( c_0 \) are non-universal scale factors. Using this and the known value of \( K \) [10], we can calculate the \( T = 0 \) correlators of the Ising spins, and find that the leading term at large separation \( r \) goes as \( \cos(2\pi r/3)/\sqrt{r} \). Thus, the triangular Ising magnet is anything but a simple uncorrelated paramagnet, although it does not order at all.
even at \(T = 0\) — instead, it forms a non-trivial correlated state with very slowly decaying correlations.

This simple example illustrates the non-trivial nature of the cooperative paramagnetic state of frustrated magnets. Such non-trivial correlated states can have interesting properties, including unusual low-lying excitations. Below, we will describe one example of this in some detail, and briefly allude to another.

**Magnetic monopoles in spin-ice materials:** With this background, we now come to the first surprise we advertise in our title and abstract, namely, the existence of genuine magnetic monopoles in the low-energy spectrum of the easy-axis pyrochlore lattice antiferromagnets Dy\(_2\)TiO\(_3\) and Ho\(_2\)TiO\(_3\), that are generally referred to as spin-ice compounds [The interested reader should also refer to the original articles Refs [12] [13] for a more detailed discussion of the various technical aspects and subtleties involved.]

In these materials, Dy\(^{3+}\) and Ho\(^{3+}\) occupy the vertices of the pyrochlore lattice tetrahedra (shown in Fig 3) and carry a ground state magnetic dipole moment \(\mu = 10\mu_B\) (where \(\mu_B\) is the Bohr magneton) that has its origins in the spin-orbit coupled ground state multiplet for this valence state. Crystal field effects result in a strong ‘easy-axis’ energy that forces each moment to lie along the tetrahedral body diagonal that passes through the corresponding pyrochlore lattice site. Thus, if one considers a single tetrahedron, each moment has two choices: It can either point inward towards the center of the tetrahedron, or outward away from the center of the tetrahedron — in both cases, it must lie precisely along the corresponding body-diagonal.

This degree of freedom can be thought of as an Ising spin \(\sigma\), and the magnetic properties of these materials can again be modeled as some sort of Ising model. Since each dipole is shared by one up-pointing tetrahedron and one down-pointing tetrahedron, a convenient sign-convention for this mapping is that \(\sigma = +1\) if the corresponding dipole points outwards when viewed from the up-pointing tetrahedron to which it belongs. Conversely, \(\sigma = -1\) if the corresponding dipole points outwards when viewed from the down-pointing tetrahedron to which it belongs.

What is the Hamiltonian or energy functional that describes the energetics of these Ising spins? The answer is a little complicated: It turns out that the nearest neighbour exchange coupling in these systems is weak (of order \(1K\) in temperature units), and the long range magnetic dipole interactions between the magnetic moments is actually more important.

The Hamiltonian is thus the well-known classical expression for the interaction energy of a bunch of magnetic dipoles, oriented along body-diagonals of the tetrahedra. Rather than write this big expression down explicitly, it is useful to use a pedagogical device and think in terms of a ‘dumbbell-model’ [14] for the interaction energy. The idea is quite simple: We know that the interaction energy between two spatially separated groups of electric charges, each of which is overall charge-neutral, can be approximated by a multipole expansion, of which the dipole-dipole interaction energy is the leading term at large distances, with corrections that fall off as a faster power of the distance between the two groups of charges. We can turn this standard fact around, and view each magnetic dipole as being made up of a dumbbell with a ‘blue’ and ‘red’ end located at the body-centers of the two tetrahedra that share the pyrochlore lattice site on which the magnetic dipole is located (as shown in Fig 1). These blue and red ends thus lie on sites of the dual diamond lattice whose sites are the body-centers of the pyrochlore tetrahedra, and whose links pass through the sites of the pyrochlore lattice.

The blue end represents a fictitious positive magnetic
charge \( +q_m/2 \), and the red end represents a fictitious negative magnetic charge \( -q_m/2 \) (the reason for the factor of two in this definition will be clear below). The magnitudes of these charges are adjusted to give the correct magnetic dipole strength of \( \mu = 10\mu_B \) by requiring that \( q_m/2 = \mu/a_d \), where \( a_d \) is twice the distance from the vertex of a tetrahedron to its body-center along the body diagonal (equivalently, \( a_d \) is the nearest-neighbour distance of the dual diamond lattice). The Ising degree of freedom \( \sigma \) at each pyrochlore site now corresponds to the orientation of this dumbbell, and the Ising spin \( \sigma \) at the vertex of an up-pointing tetrahedron is +1 if the red end of the dumbbell is located at its body-center, and −1 if the blue end of the dumbbell is located at its body-center.

The original energy functional can now be simply (but approximately) reproduced by postulating a fictitious Coulomb interaction between these fictitious magnetic charges:

\[
V_m(r_{\alpha\beta}) = \frac{\mu_0}{4\pi} \frac{Q_{\alpha}Q_{\beta}}{r_{\alpha\beta}}; \quad \alpha \neq \beta
\]

\[
= \frac{1}{2} v_0 Q_{\alpha}^2; \quad \alpha = \beta
\]

Here \( \mu_0 \) is the vacuum permeability, \( Q_{\alpha} \) is the total magnetic charge on diamond lattice site \( \alpha \) (corresponding to the body-center of tetrahedron \( \alpha \)) and the ‘self-energy’ constant \( v_0 \) is adjusted to correctly reproduce the interaction energy between nearest neighbour dipoles.

Naturally, this statement is only approximate, but the approximation involved is such that the difference between the real interaction energy and the approximate form obtained by our device of introducing fictitious magnetic charges falls off rapidly with distance \( r \) between the magnetic dipoles, and is very small everywhere.

This way of thinking immediately yields dividends when we ask for the nature of the minimum energy configurations of the Ising spins \( \sigma \). Using the electrostatic analogy, it becomes clear that the minimum energy configurations are precisely those configurations for which the total (fictitious) magnetic charge on the body-center of each tetrahedron is zero \( Q_{\alpha} = 0 \) for all \( \alpha \). Translated to Ising variables, this is the ‘two-in two-out’ ‘ice rule’ that says that two vertices of each tetrahedron must have Ising spins pointing inwards towards its body-center, while two must have Ising spins pointing outwards away from its body-center. [Here ‘ice-rule’ refers to the analogy to Pauling’s ideas about the entropy of ice, and Nagle’s unit model for ice [15].]

How many such minimum energy configurations are there? The answer is that the set of minimum energy configurations has macroscopic entropy, i.e. it scales as the exponential of the number of system sites. This is closely analogous to the triangular lattice example we discussed earlier, and indeed, the spin-ice minimum energy configurations can also be characterized in terms of dimer configurations as before: One simply considers all up-pointing tetrahedra, and agrees to put a dimer through each outward pointing magnetic moment. To ensure consistency, the rule is reversed for all down-pointing tetrahedra, that is, each inward pointing spin of a down-pointing tetrahedron corresponds to a dimer on the diamond lattice link passing through that spin. This gives a dimer model on the diamond lattice, with two dimers touching each diamond lattice vertex, and one can then use efficient loop algorithms to sample all the minimum energy configurations in this dimer representation [16, 17].

What about excited states? Naively, one might imagine that the lowest lying excited states may be constructed by starting with an arbitrary minimum energy configuration, and flipping one Ising spin. Such a flipped spin would give rise to two nearest neighbour tetrahedra that violate the ice rule—one of them will have three out-pointing spins, and one of them will have three in-pointing spins.

However, the language of fictitious magnetic charges allows us to think a little bit more deeply. For consider flipping a single spin \( \sigma_{\alpha\beta} \) from +1 to −1. This creates two equal and opposite fictitious magnetic charges at nearest neighbour sites \( \alpha \) and \( \beta \): \( Q_{\alpha} = +q_m, \quad Q_{\beta} = -q_m \). Thought of in this way, there is nothing special about having these two charges \( \pm q_m \) at nearest neighbour locations. By flipping a string of Ising spins, we can pull these charges further and further apart from each other until they are separated by distance \( r \). Now, these two charges interact with an attractive Coulombic potential that falls off as \( 1/r \). As we know, a \( 1/r \) attraction is not confining, in the sense that it only takes a finite amount of work to separate the two charges to infinity. Thus, the dominant excitations will not correspond to two charges \( \pm q_m \) bound tightly at nearest-neighbour distance, but rather two independent and ‘free’ charges that can be at arbitrary separations from each other.

We may now translate back to the language of the original magnetic moments and Ising spins: The dominant low energy excitations consist of arbitrarily long strings of flipped Ising spins. Some more analysis reveals that the end points of these strings are genuine magnetic monopoles of strength \( \pm q_m \), in the sense that the associated magnetic field configuration would induce a current in a superconducting ring if one end of the string passed through the ring. This current would be identified as a monopole signal in a standard monopole search experiment such as the Stanford experiment to detect fundamental magnetic monopoles in cosmic radiation!

So far, all of this is strictly a \( T = 0 \) argument about ground states and low-lying excited states. One may worry that entropic effects associated with thermal fluctuations at non-zero temperature would somehow spoil all this. Perhaps fortuitously, the answer is no! It turns out that the most important consequence of entropic effects is a modification of the prefactor of the Coulombic
attraction between our emergent magnetic monopoles, by an amount proportional to $T$ that can be calculated precisely by computer simulations \cite{17, 18, 20}. This is a fairly innocuous effect, and is not expected to change the basic picture outlined above.

Impurity induced half-orphan $S = 3/4$ spins in SCGO: The second of our promised surprises is Henley’s \cite{22} identification of the unusual defect induced local moments that may exist in the pyrochlore slab magnet SCGO. SCGO is an abbreviation for the oxide SrCr$_9$Ga$_3$O$_{19}$, where the listed formula is only notional since this ideal stoichiometric composition can never be in the laboratory. What is instead commonly prepared is SrCr$_9$Ga$_{12-p}$O$_{19}$, with $p$ ranging all the way from roughly 0.5 to 0.98 \cite{21}.

The ideal stoichiometry corresponds to $S = 3/2$ Cr$^{3+}$ ions occupying the sites of a ‘pyrochlore slab’, in addition to forming a layer of decoupled pairs (Fig.\ref{fig:5}). The spins in the pyrochlore slab interact with each other through an isotropic nearest neighbour antiferromagnetic exchange coupling $J \approx 100K$ (in temperature units), forming two Kagome layers coupled through apical sites in the middle (Fig.\ref{fig:5}). The spins in the dimer layer only interact within a pair via an isotropic antiferromagnetic exchange $J' \approx 200K$ (in temperature units), thus forming a system of decoupled spin-dimers. The excess Ga introduced by having $p < 1$, substitute for the Cr$^{3+}$ ions and introduce non-magnetic impurities with $S = 0$. From detailed studies \cite{21}, it is known that the Ga have a slight preference for going into the Kagome and isolated dimer layers, rather than substitute for the apical Cr$^{3+}$.

With this background, let us now think classically ($S = 3/2$ is large enough that we expect a classical analysis to be accurate except at extremely low temperatures at which quantum fluctuations start to play an important role) and ask for the classical minimum energy configurations of the exchange Hamiltonian

$$H = J \frac{1}{2} \sum_{i \in \bigotimes} (\sum_{i \in \bigotimes} \vec{S}_i - \frac{\hbar}{2J})^2 + J' \frac{1}{2} \sum_{\triangle} (\sum_{i \in \triangle} \vec{S}_i - \frac{\hbar}{2J})^2$$

where $\bigotimes$ refers to the tetrahedra that have as one of their faces the up-pointing (down-pointing) triangles in the upper (lower) Kagome layer, and $\triangle$ refers to the down-pointing (up-pointing) triangles in the upper (lower) Kagome layer, and $\hbar$ is the external magnetic field, which we now proceed to set to zero.

When written in this form, it is clear that this energy functional has enormously many ground states, which correspond to all possible configurations in which each simplex (a tetrahedron or a triangle) has zero net spin. Let us now dope the system with non-magnetic impurities. If a simplex has a single non-magnetic site, it can still arrange the spins on the other sites to add up to zero, and thus a single vacancy on a simplex has no significant effect on the properties of the system.

What about a correlated defect consisting of two vacancies on a single simplex? If this simplex is a tetrahedron, again, nothing much happens. However, if this simplex is a triangle, then it becomes impossible to satisfy the zero spin constraint on this simplex. However, all neighbouring simplices can still satisfy the zero net spin constraint. One therefore expects an infinitesimal magnetic field, say $\hbar = \epsilon \vec{z}$ to immediately polarize the net spin of the triangle with two defects, while having no effect on any other simplex.

What is the total spin of the resulting state? Since each physical spin is shared by two simplices, and only one simplex has non-zero net spin, we may write $S^z_{\text{tot}} = \frac{1}{2} \left( \sum_{\bigotimes} S_\bigotimes^z + \sum_{\triangle} S_\triangle^z \right) = 3/4$! Thus, such a correlated defect gives rise to a spin of $S = 3/4$—these have been referred to in the literature as ‘half-orphans’ \cite{22}, and constitute the second of our promised surprises.

To complete our discussion, we must also note that these half-orphans come with a statutory warning: As
in our earlier example, this is again a purely $T = 0$
statement relying on minimizing the interaction energy.
Again, it is not at all obvious that any of this survives
the entropic effects of thermal fluctuations at non-zero
temperature. Indeed, unlike in the previous example in
which entropic effects have been analyzed and are now
well-understood, not much is known about the effects
of thermal fluctuations on these half-orphans, although
diluted SCGO has been studied using computer simula-
tions and phenomenological approaches [23, 24]. It thus
remains an open question whether these $S = 3/4$ spins
survive the effects of non-zero temperature and can be
‘seen’ in experiments, and we are working on providing
some definite answers soon [25].

Acknowledgements: The author acknowledges use-
ful discussions with D. Dhar, and with his collaborators
A. Banerjee, D. Heidarian, R. Moessner, and A. Sen.
Funding from DST SR/S2/RJN-25/2006, and the hospi-
tality of School of Physical Sciences, JNU and Physics
Department, IIT Bombay during the completion of this
contribution is also gratefully acknowledged.

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