# Classical Spin Liquid on the Maximally Frustrated Honeycomb Lattice

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We show that the honeycomb Heisenberg antiferromagnet with $J_1/2 = J_2 = J_3$, where $J_1$, $J_2$, and $J_3$ are first-, second-, and third-neighbor couplings, respectively, forms a classical spin liquid with pinch-point singularities in the structure factor at the Brillouin zone corners. Upon dilution with nonmagnetic ions, fractionalized degrees of freedom carrying $1/3$ of the free moment emerge. Their effective description in the limit of low temperature is that of spins randomly located on a triangular lattice, with a frustrated sublattice-sensitive interaction of long-ranged logarithmic form. The $XY$ version of this magnet exhibits nematic thermal order by disorder. This comes with a clear experimental diagnostic in neutron scattering, which turns out to apply also to the case of the celebrated planar order by disorder of the kagome Heisenberg antiferromagnet.

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*Motivation.*—The honeycomb lattice has—somewhat belatedly—become one of the prime hunting grounds for spin liquids (SL) in $d = 2$ [1], in addition to the kagome and the $J_1 - J_2$ square lattice Heisenberg models, which have been the focus of much attention over decades, continuing until today. In both these latter cases [2–11], confidence in the existence of a quantum SL state for $S = 1/2$ magnets has ebbed and flowed, while the classical (large-spin) versions evade liquidity by exhibiting rather interesting forms of order by disorder [12–20].

The richness of magnetic models on the honeycomb lattice—bipartite, like the square lattice—has therefore come as somewhat of a surprise. Initially emulating its brethren by appearing to support a quantum SL in a Hubbard model [21], it has been attracting attention in the context of the fractionalized excitations of the Kitaev honeycomb model [22], exhibiting highly unusual exactly soluble quantum SL phases. Particular impetus arose from the suggestion that the Kitaev Hamiltonian may describe the materials $\{\mathrm{Na,Li}\}_2\mathrm{IrO}_3$, provided a Heisenberg term is added [23–25]. In fact, detailed studies of these materials suggest that further nearest neighbor terms play an important role in explaining spiral ordering at low temperatures [26], and one of the models studied in some detail is the $J_1$-$J_2$-$J_3$ Heisenberg model, which had already been subject to considerable earlier attention [27–30]. In determining the Hamiltonian appropriate to these materials, it has turned out to be instructive to consider their response to disorder [31].

*Synopsis.*—Here, we identify and study in detail an unusual, hitherto overlooked, classical SL state on the honeycomb lattice, associated with the (known) degeneracy point $J_1/2 = J_2 = J_3$ of the Heisenberg model on the honeycomb lattice. This represents the first realization of a SL in $d > 1$ of edge-sharing simplices, which here take the form of octahedra. This state exhibits novel disorder effects whereby, upon dilution, fractionalized moments carrying one third of the microscopic spin moment appear. These fractionalized moments interact via a frustrated, sublattice-dependent, long-range interaction in the limit of low temperature, $T$. The structure factor of the pure model exhibits pinch points, rather unexpectedly for a lattice whose dual is not bipartite. These reside at the zone corner three-sublattice wave vector $\mathbf{Q}$ (which distinguishes between the three sublattices of the underlying triangular Bravais lattice).

We show that these phenomena all derive from the fact that the low-temperature behavior of the spins is controlled by the spatial fluctuations of the three-sublattice order parameter of a dual surface with triangular symmetry. This SL thus represents a new class of low-temperature behavior, quite distinct from classical spin liquid states on networks of corner-sharing simplices in which the low-temperature correlations are controlled by fluctuations of a dipolar (divergence-free) vector field defined on the links of the corresponding bipartite dual lattice. Additionally, the $XY$ version of this system is interesting. It exhibits nematic order by disorder: As $T \to 0$, the spins fluctuate predominantly around a nematic axis, making them effectively Ising-like, and causing an algebraic decay of spin correlations at certain wave vectors. This leads to peaks in the structure factor that turn out to be straightforwardly detectable in neutron scattering, providing an unusually direct signature of nematic order.

*Model.*—The Hamiltonian for classical $O(n)$ spins $\vec{S}_i$ of unit length on sites $i$ of the honeycomb lattice reads

$$H = J_1 \sum_{\langle \vec{r}, \vec{r}_1 \rangle} \vec{S}_{\vec{r}} \vec{S}_{\vec{r}_1} + J_2 \sum_{\langle\langle \vec{r}, \vec{r}_2 \rangle\rangle} \vec{S}_{\vec{r}} \vec{S}_{\vec{r}_2} + J_3 \sum_{\langle\langle\langle \vec{r}, \vec{r}_3 \rangle\rangle\rangle} \vec{S}_{\vec{r}} \vec{S}_{\vec{r}_3}$$
$$= \frac{J}{2} \sum_{\vec{R}} [\vec{S}_{\bigcirc}(\vec{R})]^2 + \text{const}, \quad (1)$$

 

where $\langle\vec{r},\vec{r}_1\rangle$, $\langle\langle\vec{r},\vec{r}_2\rangle\rangle$, and $\langle\langle\langle\vec{r},\vec{r}_3\rangle\rangle\rangle$ refer respectively to first, second, and third nearest neighbor pairs. In the second line, which follows from fixing $J_1/2 = J_2 = J_3 = J$, $\vec{S}_{\bigcirc}(\vec{R})$ is the total spin of the hexagon labeled by the dual triangular lattice site $\vec{R}$ at its center.

This form shows that any, and each, configuration where every hexagon $\vec{R}$ has vanishing total spin, $\vec{S}_{\bigcirc}(\vec{R}) = 0$, is a ground state. Such a rewriting is helpful for geometrically frustrated lattices with a corner-sharing structure of elementary simplices [19,20], examples being pyrochlore (corner-sharing tetrahedra) or kagome (corner-sharing triangles) lattices. It immediately allows us to estimate the dimensionality of the ground state manifold, $F$. This proceeds by subtracting the number of constraints, $K$, imposed by Eq. (1), from the total number of degrees of freedom, $D$, of the spin system.

*Constraint counting.*—For a system of $n$-component spins with $N$ such simplices, and each spin part of $b$ simplices, $D = q(n-1)/b$ per simplex, where the number of spins in a simplex $q = 3, 4, 6$ for a triangle, tetrahedron, and octahedron, respectively. Each simplex imposes $K = n$ constraints, as each component of its total spin must vanish. Hence,

$$F = \frac{q(n-1)}{b} - n. \tag{2}$$

To maximize $F$, and hence enhance the chance of finding a SL [19,20], one should minimize $b$, or maximize $n$ and $q$. Indeed, $b$ is minimal for corner-sharing arrangements, and $q = 4$, $n = 3$ result in the well-established classical SL on the pyrochlore lattice. Triangle-based lattices (kagome has $q = 3$) need higher, $n \geq 4$, component spins for a similar SL to arise [32].

The $J_1$-$J_2$ model on the square lattice with $J_2 = J_1/2$ can be thought of as edge-sharing tetrahedra, with a large $q = 4$; it does not support $F > 0$ for any $n$. Indeed, no such Heisenberg model with $F > 0$ has been identified for edge-sharing simplices at all so far. However, from Eq. (2), $F = 1$ for $q = 6$ and $b = 3$, which corresponds to the frustration point of the honeycomb lattice, Eq. (1). It can be thought of as edge-sharing octahedra (Fig. 1), and thus presents the first instance of a possible SL on an edge-sharing lattice. It is also the first with $b > 2$.

*Effective theory and numerics.*—To explore the consequences of this unusual geometry for the low-temperature behavior of $n$ component spins, we now develop a low-energy effective description. Consider an $A$-sublattice ($B$-sublattice) site $\vec{r}_A$ ($\vec{r}_B$) of the honeycomb lattice, which sits at the center of an "up-pointing" ("down-pointing") triangle comprising dual lattice points $\vec{R}_a$, $\vec{R}_b$, and $\vec{R}_c$ belonging to the three sublattices of the tripartite dual triangular lattice. One writes the corresponding $O(n)$ spins $\vec{S}_{\vec{r}}$ in terms of $\vec{\zeta}_{\vec{R}}$ and $\vec{\tau}_{\vec{R}}$, two $O(n)$ vector fields on the dual triangular lattice.

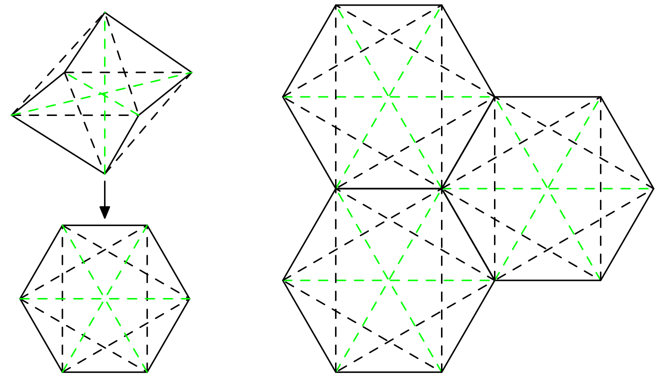

FIG. 1. Projection of the octahedron into the hexagon and the $J_1$-$J_2$-$J_3$ model on the honeycomb lattice. The $J_3$ interactions are differentiated with colors.

$$\vec{S}_{\vec{r}_A} = \sum_{\alpha=a,b,c} (\vec{\tau}_{\vec{R}_\alpha} + \vec{\zeta}_{\vec{R}_\alpha}), \qquad \vec{S}_{\vec{r}_B} = \sum_{\alpha=a,b,c} (\vec{\tau}_{\vec{R}_\alpha} - \vec{\zeta}_{\vec{R}_\alpha}).$$

Next, we note that $\{\vec{\zeta}\}$ drop out of the classical Hamiltonian Eq. (1) when rewritten in these variables. We have

$$H(\{\vec{S}\}) = F_2(\{\vec{\tau}\}) + \text{const}, \tag{3}$$

where

$$F_2(\{\vec{\tau}\}) = \frac{\beta J}{2} \sum_{\vec{R}} \Big(6\vec{\tau}_{\vec{R}} + 2 \sum_{\vec{R}_n \in \partial\vec{R}} \vec{\tau}_{\vec{R}_n}\Big)^2. \tag{4}$$

Here, $\vec{R}_n \in \partial\vec{R}$ denotes the six dual triangular lattice sites $\vec{R}_n$ that are nearest neighbors of the dual triangular lattice site $\vec{R}$. Thus $\vec{\zeta}$ encodes the $T = 0$ fluctuations of the classical SL, while $\vec{\tau}$ captures thermal fluctuations.

To obtain the form of the entropic contribution to the phenomenological low-temperature free-energy density, we take guidance from the self-consistent Gaussian approximation (equivalently, the large-$n$ limit) [33]. As is well known, this predicts an entropic contribution that takes the form $\frac{\rho}{2}\sum_{\vec{r}}\vec{S}_{\vec{r}}^2$ at low temperature. Incorporating this into our description, we see that the partition function can be written as a product of $\vec{\zeta}$ and $\vec{\tau}$ partition functions, with actions

$$\mathcal{S}_\zeta = F_1(\{\vec{\zeta}\}), \qquad \mathcal{S}_\tau = F_1(\{\vec{\tau}\}) + F_2(\{\vec{\tau}\}), \tag{5}$$

where

$$F_1(\{\vec{v}\}) = \frac{\rho}{2} \sum_{\vec{r}} \Big[\vec{v}_{\vec{R}_a(\vec{r})} + \vec{v}_{\vec{R}_b(\vec{r})} + \vec{v}_{\vec{R}_c(\vec{r})}\Big]^2 \tag{6}$$

for $\{\vec{v}\} = \{\vec{\zeta}\}$ or $\{\vec{\tau}\}$. Here, the phenomenological spin stiffness $\rho$ is chosen to ensure $\langle\vec{S}_{\vec{r}}^2\rangle = 1$.

The $T \to 0$ limit is thus characterized by a particularly simple action in which the $\vec{\tau}$ fields do not contribute. This action, as well as the expressions for the physical spins $\vec{S}$, are both invariant under $\vec{\zeta}(\vec{R}) \to \vec{\zeta}(\vec{R}) + \mathrm{Re}[\vec{\chi}\exp(2\pi i \mathbf{Q}\cdot\vec{R})]$ for any constant $\vec{\chi}$, where $\mathbf{Q}$ is the three-sublattice ordering wave vector. Thus, this limit of our effective theory describes the spatial fluctuations (parametrized by $\vec{\zeta}$) of the three-sublattice order parameter of a dual surface with triangular symmetry. At nonzero temperature, the partition function also receives contributions from fluctuations of $\vec{\tau}$, which exhibit a similar symmetry.

Using this effective description, we have analytically computed spin correlations in the $T \to 0$ limit. The corresponding spin structure factor features pinch points at $\mathbf{Q}$. While such pinch points are the defining characteristic of algebraic SLs [34], their present location at the three-sublattice wave vector reflects the unusual underlying tripartite structure of the low-temperature correlations implied by our theory. Indeed, the very existence of pinch points in the Heisenberg case comes as somewhat of a surprise given the nonbipartite nature of the dual triangular lattice. In the corresponding corner-sharing models, the bipartiteness of the dual lattice (square, honeycomb, or diamond lattice) is a crucial ingredient for such pinch points [34]. For instance, in work close in spirit to the present one, on bosons on a honeycomb and the dual triangular lattice [35], one finds an emergent Ising gauge field implying the absence of pinch points.

In order to test these predictions, we have performed Monte Carlo (MC) simulations that employ a combination of heat-bath and microcanonical moves as well as parallel tempering moves. The structure factor from MC simulation of Heisenberg spins (Figs. 2) agrees with the analytical prediction of our effective theory. In sharp contrast, for $n = 2$, the corresponding $XY$ model, low-temperature peaks develop in addition to the pinch points. This is an instance of nematic (collinear) order by disorder (see Supplemental Material [36] for more details), as is readily verified by constraint counting [19,20]. The appearance of these peaks is a consequence of, and provides an unusually direct signature of, nematic ordering. This interpretation is confirmed (Fig. 3) by a low-$T$ specific heat of $c = 0.375k_B$ per spin, reduced from the value of $c = bnk_B/2q$ expected from equipartition in the absence of order by disorder [15–19], as is found in the Heisenberg magnet with $c = 0.75k_B$ (see Supplemental Material [36]).

We note that the unexpected appearance of these peaks is a diagnostic for nematic ordering more generally—indeed, they appear even for the coplanar order by disorder [15,16] of the kagome Heisenberg antiferromagnet. The reason for their appearance is beautifully indirect: nematic ordering goes along with the emergence of discrete (Ising for our model, Potts for the kagome case [15,16]) effective degrees of freedom from the continuous Heisenberg ones. Now, in two dimensions, such a discreteness leads to the appearance of additional operators in the effective low-energy theories, as described in the pedagogical introduction by Zeng and Henley on height models [37]. They, in turn, lead to the peaks in the structure factor generally reflecting algebraic (rather than long-range) spin correlations in addition to the pinch points as shown in Fig. 2 for our case; or, for kagome, in the comparison between large-$N$ [33] and low-temperature $N = 3$ [38] structure factors. The nematic diagnostic is therefore fundamentally a diagnostic of emergent discreteness.

*Dilution effects.*—The ground states of SLs often are less revealing of their topological nature than their excitations.

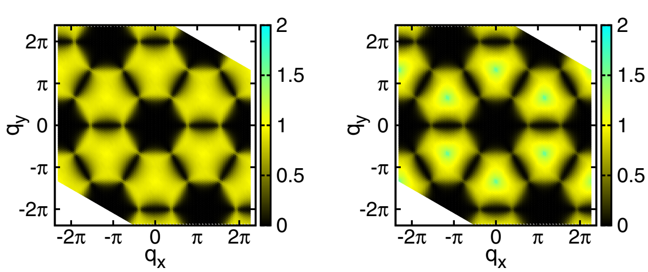


FIG. 2. Structure factor as obtained in Monte Carlo (MC) simulations of the pristine Heisenberg (left) and $XY$ (right) systems. Both results correspond to $N = 1800$ spins at $T/J = 0.01$.

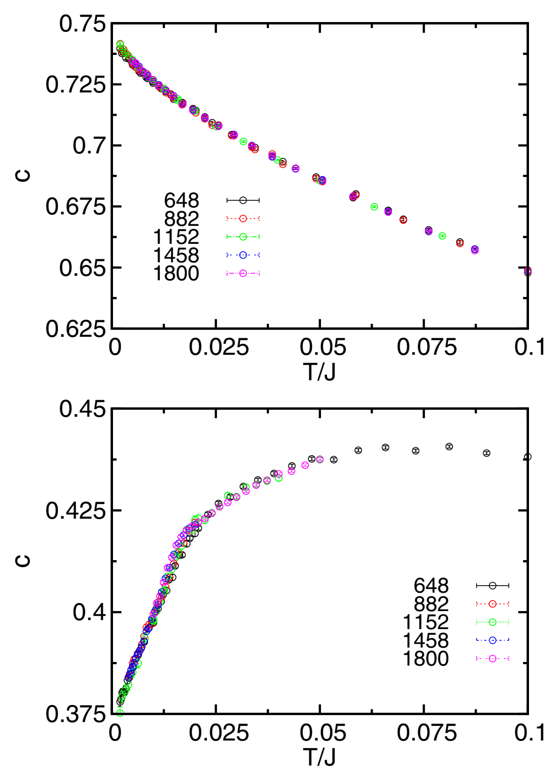


FIG. 3. Specific heat obtained in Monte Carlo simulations of the pristine Heisenberg ($n = 3$, top) and $XY$ ($n = 2$, bottom) systems, with $c = \frac{3}{8}k_B < (bn/2q)k_B = k_B/2$ indicating nematic order by disorder for the $XY$ case.

An elegant way to visualize the latter as effectively a ground state property is to introduce disorder that then nucleates excitations. In SLs, this is perhaps most easily done by replacing some of the magnetic ions with non-magnetic ones. For classical SLs, this dilution problem has been studied in some detail both experimentally [39–42], and theoretically [43–47]. In particular, for the cases of $SrCr_{9p}Ga_{12-9p}O_{19}$, the checkerboard and the pyrochlore lattices, it was found that fractional impurity moments carrying one half of the moment of a free spin arise as a cooperative phenomenon. These so-called orphan spins occur when all but one of the spins of a simplex are replaced—so that the total spin of that simplex [see Eq. (1)] can no longer possibly vanish.

With this motivation, we have analyzed the response of the honeycomb Heisenberg SL to dilution within our effective field theory, incorporating missing sites as a constraint $\vec{S} - \vec{r} = 0$, and retaining the orphan spin degree of freedom as a unit vector in our description [45,46]. We find that all other kinds of defects (i.e., hexagons with more than one spin left) do not lead to fractionalization or a Curie tail contribution to the magnetic susceptibility at low temperature for the same reasons as for the corner-sharing lattices [44]. In contrast, the orphans provide a number of signatures of the new structure of the honeycomb SL. First of all, they directly reflect the fact that we have $b = 3$ edge-sharing octahedra meeting at each site—the fractional impurity moment is not one half but one third of that of a free spin. This is displayed in Fig. 4 (top panel) where our analytical prediction is compared with numerical results for the impurity susceptibility. This is, to our knowledge, the first instance of fractionalization into three items in a classical spin model.

Interactions between these orphans are entropic in nature and take the form of an effective Heisenberg exchange $J_{\mathrm{eff}}$ mediated by the bulk SL, and hence reflect the structure of the latter. In the classical SLs known so far, these effective interactions can be written in a form that is uniformly antiferromagnetic [48]. Here, this is not possible: We now find that these interactions are antiferromagnetic (ferromagnetic) for orphans residing on the same sublattice (different sublattices) of the dual triangular lattice, respectively, with the antiferromagnetic interactions being twice as strong as the ferromagnetic ones. This intricate structure in the effective exchange couplings follows from our field theory, which relates these entropic interactions to correlations between the thermally excited net spins $\vec{S}_{\bigcirc}(\vec{R})$ [Eq. (1)] in the pristine bulk spin liquid,

$$\beta J_{\mathrm{eff}}(\vec{R}_1, \vec{R}_2) \approx \frac{-\langle \vec{S}_{\bigcirc}(\vec{R}_1) \cdot \vec{S}_{\bigcirc}(\vec{R}_2)\rangle}{\langle \vec{S}_{\bigcirc}(\vec{R}) \cdot \vec{S}_{\bigcirc}(\vec{R})\rangle^2}. \tag{7}$$

For low $T$ and large distances $|\vec{R}_1 - \vec{R}_2| \gg a$ between the orphan spins, where $a$ is the lattice spacing, this gives a scaling form (Fig. 4),

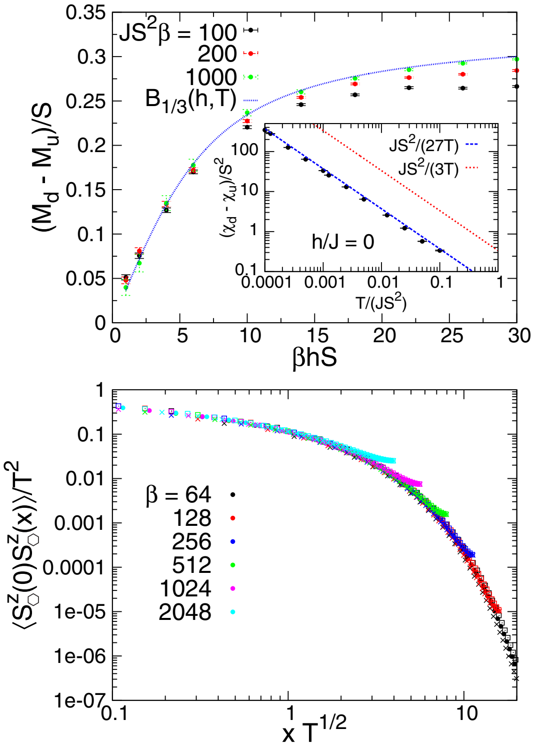


FIG. 4. Top: "Impurity magnetization," defined as the difference of total magnetization in the diluted and undiluted systems, as observed in MC simulations of the model. The solid curve corresponds to the theoretical prediction for a free spin $S/3$ in a field $h$, i.e., the Langevin function $\mathcal{B}_{S/3}(h, T)$. The inset shows the "impurity susceptibility" at zero external field, consistent with a Curie law for fractionalized spins $S/3$. Bottom: The scaling prediction for the charge correlations on a finite lattice of linear size $L = 210$ using the effective field theory expression for correlations. Crucially, correlations between sites on the same sublattice have been multiplied by an extra scaling factor of $-1/2$.

$$\beta J_{\mathrm{eff}} = \eta(\vec{R}_1, \vec{R}_2)\mathcal{F}[(\vec{R}_1 - \vec{R}_2)\sqrt{T}] \tag{8}$$

$$\overset{T\to 0}{=} \frac{1}{2\pi}\eta(\vec{R}_1, \vec{R}_2)\log(|\vec{R}_1 - \vec{R}_2|), \tag{9}$$

where $\eta = +1$ ($\eta = -1/2$) if $\vec{R}_1$ and $\vec{R}_2$ are on the same sublattice (different sublattices) of the dual triangular lattice.

*Outlook.*—Our model, notwithstanding its simplicity, displays a plethora of phenomena of current interest; the unusual emergent $\vec{\tau}$ and $\vec{\zeta}$ fields and the new fractionalized behavior of $1/3$ for the impurity spin moments show that these nontrivial phenomena, usually associated with the quantum realm, can emerge even in a classical setting. Additionally, the resulting pattern of frustrated logarithmic interactions between the impurity moments is as yet unstudied, and will possibly lead to a spin glass, unlike in the bipartite cases [48].

As for realizations, the 2:1:1 ratio of exchange interactions is natural if exchange is via an ion on the hexagon center with no angular dependence, as the nearest neighbor bonds are part of two hexagons. Known experimental values are encouragingly nearby, being close to 2∶1.6∶1.6 [24]. Hence direct observation of these phenomena might be possible, the main obstacle perhaps being the effects of quantum fluctuations for $S = 1/2$. Quite generally, at finite $T$, the classical SL behavior is favored over competing phases on account of its large entropy, and, in particular, fans out from the degeneracy point (see Supplemental Material [36]). We hope that this work stimulates further investigation on appropriate honeycomb materials.

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