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The anisotropic kagomé antiferromagnet: a topological spin glass?

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Abstract. — The anisotropic kagomé antiferromagnet is discussed as a possible candidate for glassiness in the absence of disorder. Numerical studies support the conjecture that this system is not a conventional magnetic system, and we include a discussion of the relevant experimental system $\text{SrCr}_{8-x}\text{Ga}_{4+x}\text{O}_{19}$.

1. Introduction.

The ubiquitous phenomenon of frustration [1], with its accompanying degeneracy and metastability, fascinated Rammal; he studied its manifestation in a rich variety of areas including spin glasses [2], superconducting and random networks [3], and optimization problems [4]. However, as he noted in a Review just a few years ago [5], frustration and disorder usually occur simultaneously and it is often difficult to separate their individual effects. This was one of his key motivations for the study of superconducting networks, where a fine tuning of the frustration can be achieved in the *absence* of disorder. The anisotropic Heisenberg antiferromagnet on a two-dimensional lattice is another system where the effects of strong geometrical frustration can be studied independent of any randomness, and this will be the topic of our discussion here.

Rammal was very generous with his ideas, particularly with young people at the beginning of their careers. When one of us (Chandra) told him of the mysterious experiments on $\text{SrCr}_8\text{Ga}_4\text{O}_{19}$ described below, he eagerly asked for more details and listened carefully to our nascent (and heuristic) ideas on the subject. He suggested that non-Abelian defects might be important in this system, and pointed us to a number of key references on this topic. At the time his remarks were difficult to understand, though they were duly noted. Now, a few years later, the presence of non-Abelian defects play a key role in our proposed topological transition of the kagomé antiferromagnet, and there is even some preliminary numerical support for this conjecture.

The kagomé antiferromagnet is a clear example of a system where strong frustration in the absence of disorder has pronounced effects; though its associated theory is still incomplete, here we describe several of its elusive features. This discussion is not simply abstract; a real experimental system ($\text{SrCr}_{8-x}\text{Ga}_{4+x}\text{O}_{19}$) exists, and we begin with a discussion of its measured properties. Next we turn to the theoretical model, asking whether this “disorder-free” Hamiltonian has a chance of displaying glassy behavior. Preliminary numerical diagnostics are presented that support our hypotheses, and we end with some conclusions, many questions, and lots of plans for further study on this problem.

2. $\text{SrCr}_8\text{Ga}_4\text{O}_{19}$: An atypical spin glass.

Motivated by the possibility of exotic behavior in highly degenerate spin systems [6-8], Obradors and coworkers [9] initiated the experimental study of the magnetoplumbite $\text{SrCr}_{8-x}\text{Ga}_{4+x}\text{O}_{19}$ (SCGO(x)). They attributed its magnetic properties to planes of antiferromagnetically-coupled chromium atoms on a geometrically frustrated kagomé lattice, where each Cr^{3+} ion is associated with an isotropic spin- $\frac{3}{2}$ moment. Obradors et al. [9] performed linear DC susceptibility and neutron diffraction measurements on SCGO(x); they observed the marked absence of conventional antiferromagnetic ordering down to temperatures $T \sim 4$ K, despite a large exchange coupling ($J \sim 100$ K) implied by the significant Curie-Weiss temperature ($\theta_{\text{CW}} = \frac{4}{3}JS(S+1)$).

The underlying magnetoplumbite lattice associated with SCGO(x) is composed of linked two-dimensional spinel structures [10], where crystal-field effects confine the chromium ions to the octahedral sites [9]. Anomalous behavior in this type of magnetic system was first recognized by Anderson, who noted that a [111] projection of the spinel lattice corresponds to a planar kagomé net [6]. Anderson suggested that spins with nearest-neighbor antiferromagnetic coupling residing on this structure could only maintain good *short-range* magnetic order due to a large ground-state degeneracy associated with a finite zero-temperature entropy [6]; at the time there were no known experimental realizations of this model. An exact solution of the Ising kagomé antiferromagnet is now available [11-13] and, by contrast with all other known periodically frustrated (nearest-neighbor) 2d Ising lattices (e.g. the triangular and the fully-frustrated square systems) [14-16] it displays *exponential* rather than power-law spin correlations at $T = 0$ indicating its true lack of zero-temperature long-range spin order. More recently the nearest-neighbor Heisenberg model on a spinel crystal lattice has been called a “cooperative paramagnet” by Villain [8] due to its large ground-state degeneracy; he has suggested that addition of atomic disorder could transform it into a novel spin glass. In SCGO(x), steric hindrance effects present during sample growth [17] lead to unavoidable random substitution of non-magnetic gallium for the (magnetic) chromium ions, and Obradors and coworkers [9] thus proposed SCGO as an experimental realization of Villain’s ideas [8].

Further low-temperature measurements on SCGO(x) were then performed by Ramirez and coworkers [17]; they observed *distinct* zero-field and field-cooled susceptibilities for $T < 4$ K, a feature typical of a spin glass [18]. The presence of a “spin freezing” transition was confirmed [17] by the measured nonlinear susceptibility (χ_3), a direct probe of the Edwards-Anderson spin glass order parameter [19]. In the paramagnetic state, this nonlinear susceptibility is defined in terms of the magnetization

$$M = \chi_1 H + \frac{1}{3!} \chi_3 H^3 + \dots = \sum_{n=1} \chi_{2n-1} H^n \quad (1)$$

in the direction of the applied field. We recall that in a spin glass ensemble and “valley” averages must be performed separately (see Fig. 1); close to the glass transition the observed

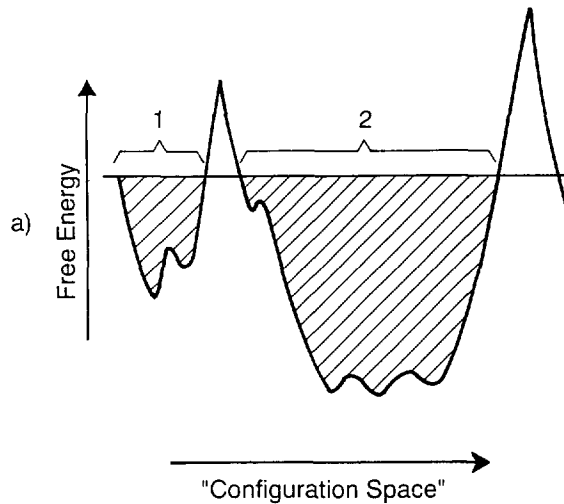
nonlinear susceptibility diverges

$$\overline{\langle \chi_3 \rangle} \sim \frac{\overline{\langle M^4 \rangle}}{T^3} - \frac{3\{(\overline{\chi})^2 + \overline{\delta \chi^2}\}}{T} \rightarrow -\infty \quad (T = T_g) \quad (2)$$

due to large variation in the moment fluctuations

$$\overline{\delta \chi^2} = \overline{\chi^2} - (\overline{\chi})^2 \rightarrow \infty \quad (T = T_g) \quad (3)$$

between different wells (here N is the total number of spins). Though χ_3 could diverge *positively* in an equilibrium system [20,21], we note that a negative divergence can only occur in the presence of free energy "pockets". Thus the observed behavior of χ_3 in SCGO(x) firmly establishes its spin glass character; the associated "spin freezing" was confirmed with quasielastic neutron scattering [22], where a marked increase in intensity at low temperatures was observed in conjunction with a short spin correlation length ($\xi \sim 6 \text{ \AA}$) that remained constant to temperatures well above T_g .



$$b) \quad \overline{\langle A \rangle} = \frac{1}{N} \sum_{\text{Wells}_i} \langle A \rangle_i^{\text{Thermal}}$$

Fig. 1. — (a) A schematic picture of free energy "pockets" in a spin glass and (b) the determination of an observable quantity.

However there are several experimental features of SCGO(x) that do not fit a standard description of a spin glass. For example, quasielastic neutron studies [23] confirm the two-dimensional nature of the spin correlations in its glassy state ($T < T_g$) as first suggested by Obradors and coworkers [9]. This is a somewhat surprising result since the conventional

insulating spin glass, associated with the formation of low-energy domain walls in a *randomly* frustrated antiferromagnet, is unstable in $d = 2$; for the Heisenberg case with infinitesimal anisotropy [24] $d_1 = 3$. Furthermore, in contrast to all known spin glasses, the specific heat of SCGO(x) is *quadratic* at low temperatures [17] and is remarkably robust to dilution [25]; this behavior thus appears to be intrinsic to the ground-state, and is reminiscent of a two-dimensional antiferromagnet with broken spin-rotational symmetry.

Neutron studies are also consistent with a Goldstone mode in the spin channel [22,23]; the inelastic cross-section is *frequency-independent* down to the lowest measured energies. Such behavior is expected for a two-dimensional Heisenberg antiferromagnet with a generalized susceptibility

$$\chi(q, \omega) \approx \frac{\left(\frac{c}{a}\right)}{\omega^2 - \omega_q^2} \quad (4)$$

associated with the Goldstone modes in the vicinity of the zone center Q with the dispersion relation $\omega_q \sim c|\mathbf{q} - \mathbf{Q}|$; the resulting spin scattering has the form

$$\chi''(q, \omega) \approx \frac{\left(\frac{c}{a}\right)}{\omega} \delta(\omega - \omega_q) \quad (5)$$

yielding a frequency-independent integrated scattering intensity

$$\int \frac{d^2q}{(2\pi)^2} \chi''(q, \omega) \approx \frac{1}{4\pi a} \quad (6)$$

For a conventional Heisenberg antiferromagnet with finite-temperature fluctuations, (6) is only valid down to an energy scale defined by $\omega_c = \frac{c}{\xi}$; in SCGO(x) a *flat* inelastic cross-section (6) is measured at energies almost an order of magnitude lower than ω_c , suggesting the presence of long-range spin order inaccessible by standard neutron techniques [22,23].

The combined susceptibility and neutron measurements indicate strong antiferromagnetic correlations ($\Theta_{CW} \sim -500$ K) on length-scales comparable with the planar chromium lattice spacing, suggesting that *only* nearest-neighbor antiferromagnetic couplings are present; such glassy behavior resulting from the interplay between static dilution and purely antiferromagnetic interactions has only been previously observed numerically [26], with the possible exception [27] of La(Sr, Ba)₂CuO₄. The low-temperature (i.e. $T < T_g$) "spin-glass" phase of SCGO(x) is certainly unusual; though here frozen moments are observed with neutrons, they account for only a small fraction of the observed antiferromagnetic correlations. More specifically, Broholm, Aeppli and coworkers [22,23] use the fluctuation-dissipation theorem to relate their measured response function $S(q, \omega)$ to the mean-square of the frozen moment

$$\langle S^2 \rangle_{\Delta-} = \int_{|\omega| < \Delta} \frac{d\omega}{2\pi} \frac{d^2q}{(2\pi)^2} S(q, \omega) \quad (7)$$

in the limit $\Delta \rightarrow 0^+$. Similarly, the mean-square of the fluctuating moment is expressed as

$$\langle S^2 \rangle_{\Delta+} = \int_{|\omega| > \Delta} \frac{d\omega}{2\pi} \frac{d^2q}{(2\pi)^2} S(q, \omega) \quad (8)$$

where the fluctuations are observed on a time-scale $t = \frac{2\pi}{\Delta}$. At $T = 1.5$ K ($T < T_g$) they observe no gap in the excitation spectrum down to energy scales $\Delta \sim 0.2$ MeV ($\Delta < \frac{c}{\xi}$);

furthermore their combined elastic and inelastic data are consistent with a lower bound for the ratio

$$\mathcal{M} = \frac{\langle S^2 \rangle_{\Delta^+}}{\langle S^2 \rangle_{\Delta^-}} > 4 \quad (9)$$

where $\Delta = 0.2$ MeV, thus suggesting large quantum fluctuations in the ground-state of SCGO(x). Since both series expansion [28] and exact diagonalization [29] studies indicate that the spin- $\frac{1}{2}$ Heisenberg antiferromagnet on a kagomé lattice is disordered, strong quantum effects in the $S = \frac{3}{2}$ case are not unexpected.

To summarize, the observed ground-state behavior of SCGO(x) has features mysteriously reminiscent of *both* a spin glass and a quantum antiferromagnet. Simultaneously there is two-dimensional glassiness, a short spin correlation length, large moment fluctuations, and strong indications of a Goldstone mode in the spin channel. Several measured properties of SCGO(x) are surprisingly robust to dilution, suggesting an *intrinsic* glassiness [25] consistent with the fact that disorder-induced spin glasses are *unstable* in two dimensions. Motivated by these experiments, we explore the possibility of a new mechanism for quasi-2D glassy behavior to explain this exotic behavior.

3. Glassiness without disorder: why this system has a chance.

Broken ergodicity, the development of free energy “pockets” in configuration space, is a key feature of spin glasses [30,31]; heuristically the system gets “stuck” in a particular free energy minimum, and thus does not explore all of phase space in the infinite-time limit. It is widely believed that *both* frustration and disorder are needed for the development of this phenomenon; loosely speaking, the standard notion is that the frustration results in multiple ground-states separated by a *macroscopic number of large barriers* stabilized by disorder. Can frustration alone satisfy this behavior? Villain has proposed [15] “fully frustrated models”, where each elementary plaquette is “unsatisfied”, as possible spin glasses with *non-random* interactions, and there has been much work in this area following his suggestion. Because the effects of frustration are known to be more pronounced in lower dimensions most of the effort has been focussed on two-dimensional models with the hope of finding experimental analogues.

Though several two-dimensional periodically frustrated spin systems display large ground-state degeneracies, even accompanied by *extensive* ground-state entropies, none to date definitely possess a finite-temperature glass transition. Usually the ground-state degeneracy is lifted by *anisotropic* thermal fluctuations; they select and stabilize magnetically ordered spin states from the ground-state manifold, thus dashing any hopes for glassy behavior. A possible exception is the two-dimensional triangular Heisenberg antiferromagnet with Ising anisotropy; here the original proposal [32] breaks down for the classical case [33], but may remain intact [34] for $S = \frac{1}{2}$. However, in general, frustration does not seem to be able to produce a sufficiently “rough” free energy landscape necessary for the development of glassiness; though it often results in a multitude of ground-states at $T = 0$, the barriers separating them are not “high” enough to “localize” the system in phase space.

So what is so different about the kagomé antiferromagnet, and why do we believe it has a chance at being glassy without any disorder? In order to respond to this question, we must examine the effects of geometrical frustration and the nature of the resulting degeneracies in this particular spin system. Our starting point, the simplest magnetic model for SCGO, is the two-dimensional nearest-neighbor classical Heisenberg antiferromagnet (afm)

$$H = J \sum_{ij} \mathbf{S}_i \cdot \mathbf{S}_j \quad (10)$$

on a kagomé lattice (see Fig. 2); we shall discuss our neglect of quantum effects later, once we have determined the important energy scales of the problem. Energetically, the vector sum of the classical spins on each triangular plaquette must vanish in the ground-state (i.e. $\sum_i \mathbf{S}_i = 0$); performing a leading order spinwave calculation within a standard Holstein-Primakoff spin representation, we can reexpress the Hamiltonian (10) as

$$H = \frac{JS}{2} \sum_{ij} \left\{ (b_i^\dagger b_j + b_j^\dagger b_i) \cos^2 \frac{\theta_{ij}}{2} - (b_i^\dagger b_j^\dagger + b_j b_i) \sin^2 \frac{\theta_{ij}}{2} - b_i^\dagger b_i \cos \theta_{ij} \right\} \quad (11)$$

where $\theta_{ij} = \frac{2\pi}{3}$, reflecting the 120° orientation of the spins. In momentum space this Hamiltonian (11) on a kagomé lattice becomes

$$\underline{H} = \frac{1}{2} \sum_{\mathbf{q}} B_{\mathbf{q}}^\dagger \underline{H}_{\mathbf{q}} B_{\mathbf{q}} \quad (12)$$

with

$$B_{\mathbf{q}}^\dagger = (b_{q1}^\dagger, b_{q2}^\dagger, b_{q3}^\dagger, b_{-q1}, b_{-q2}, b_{-q3}) \quad (13)$$

where the sum on \mathbf{q} runs over the Brillouin zone, and

$$\underline{H}_{\mathbf{q}} = \frac{JS}{2} \begin{pmatrix} 3 + \underline{h}_{\mathbf{q}} & 3(\underline{h}_{\mathbf{q}} - 1) \\ 3(\underline{h}_{\mathbf{q}} - 1) & 3 + \underline{h}_{\mathbf{q}} \end{pmatrix} \quad (14)$$

where

$$\underline{h}_{\mathbf{q}} = \begin{pmatrix} 1 & c_x & c_y \\ c_x & 1 & c_z \\ c_y & c_z & 1 \end{pmatrix} \quad (15)$$

and $c_x \equiv \cos q_x$, $c_y \equiv \cos q_y$, and $c_z \equiv \cos(q_y - q_x)$. Diagonalization of (12) yields the spinwave spectrum

$$\omega_{\mathbf{q}}^2 = \frac{JS}{2} \left\{ \begin{array}{c} 0 \\ 4\sqrt{1 - c_x c_y c_z} \end{array} \right\} \quad (16)$$

We see that this simple calculation yields a flat band of zero-modes in the spinwave spectrum ($\omega_{\mathbf{q}} = 0$), the consequence of a continuous local degeneracy in the Néel state on this lattice. More specifically, like its triangular counterpart, the kagomé antiferromagnet admits a coplanar ground-state; however its novel geometry permits continuous *spin folding* zero-energy modes that retain the 120° spin orientation. Figure 2 shows an example, a “weathervane mode”; here the six central spins of an elementary plaquette can rotate freely about the axis of the external spins independently of the rest of the lattice. We note, however, that there are strong interactions between these modes; the rotation of one star prevents the rotation of its neighboring ones, since their external spins will no longer be parallel. Thus, classically, we see that for a lattice of N spins there are at minimum $N/9$ independent local zero modes, corresponding to the number of stars that are not adjoining one another; the zero-point entropy per spin then has the lower-bound of one-nineth that associated with each local “weathervane” mode.

The presence of a large ground-state manifold enhances the role of high-frequency short-wavelength fluctuations in the kagomé antiferromagnet; their associated fluctuation free-energy often selects spin configurations that break the underlying lattice symmetry [35-37]. In the quantum case, these states minimize their zero-point energy, thereby maximizing their number

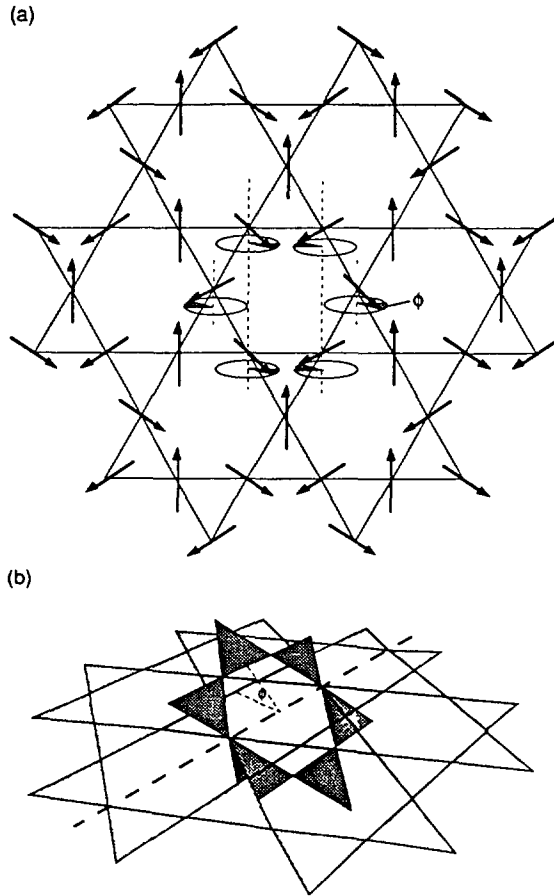


Fig. 2. — (a) A “weathervane” mode; the six central spins can rotate freely about the axis defined by the parallel external spins, here indicated with a dashed line. (b) A schematic view of the orientation of the spin plane during the rotation.

of zero modes. This phenomenon, first discussed by Villain [35], of “order from disorder” is somewhat counterintuitive: normally stability is associated with rigidity. In frustrated magnetic systems, Villain has turned this standard argument on its head: he has observed that the “most flexible” spin configurations are least affected by the presence of fluctuations, and thus outlast their more “rigid” counterparts in the degenerate ground-state manifold [35-37].

Because “order from disorder” plays an important role in the highly degenerate kagomé antiferromagnet, let us take a brief moment to look at this fluctuation-induced ordering effect in a simple illustrative example (Fig. 3). We turn to the two-dimensional frustrated square Heisenberg Hamiltonian

$$H = J_1 \sum_{ij} \mathbf{S}_i \cdot \mathbf{S}_j + J_2 \sum_{ik} \mathbf{S}_i \cdot \mathbf{S}_k \tag{17}$$

with nearest and next-nearest neighbor couplings J_1 and J_2 respectively; in the limit $\eta = \frac{J_1}{2J_2} \ll 1$ this simple model has a ground-state with a continuous global degeneracy [38]. For

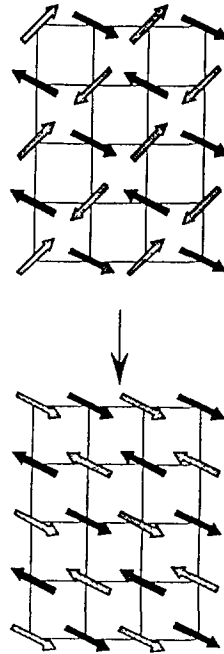


Fig. 3. — Fluctuation-selection of ground states in the 2D frustrated square Heisenberg antiferromagnet. Note that though fluctuations lift the continuous degeneracy of the classical manifold, a discrete Z_2 degeneracy remains.

$\eta > 1$, the classical ground state has conventional Néel order, but when $\eta < 1$ the two sublattices become decoupled and can have *arbitrary* angular orientation with respect to one another. The spinwave spectrum, which is sensitive to short-wavelength fluctuations, suggests that “order from disorder” effects could occur; in the limit $\eta \ll 1$ it is given by [37]

$$\omega(\mathbf{q}, \theta)^2 = (4SJ_2)^2 \{ [1 + \eta(\alpha \cos q_x + \beta \cos q_y)]^2 - [\cos q_x \cos q_y + \eta(\alpha \cos q_y + \beta \cos q_x)]^2 \} \quad (18)$$

where $(\alpha, \beta) = (\cos^2 \frac{\theta}{2}, \sin^2 \frac{\theta}{2})$ and θ is the angle between the two-sublattices. The angle-dependent part of the classical fluctuation free energy, $\delta F(\theta) = F(\theta) - F(0)$ can be estimated to leading-order by incorporating the zero-temperature spinwave dispersion(18) into

$$F(\theta) = T \sum_{\mathbf{q}} \ln \frac{\omega(\mathbf{q}, \theta)}{2T} \quad (19)$$

which, upon integration, yields

$$\delta F(T, \theta) \sim -E(T)(1 + \cos^2 \theta) \quad (20)$$

for $\eta \ll 1$ where $E(T) = 0.636\eta^2 T$.

The fluctuation free energy (20) clearly favors spin configurations where $\cos \theta = \pm 1$ (Fig. 3), thereby retaining a *discrete* two-fold degeneracy in the ground-state manifold. It selects states that break Z_4 lattice symmetry; the spins are ferromagnetically aligned ($\omega \sim k^2$)

in one lattice direction, thereby *maximizing* the coupling between the two sublattices and minimizing the overall dispersion. From a more technical standpoint, the coupling between two antiferromagnetic sublattices enters as off-diagonal matrix elements in a standard spinwave calculation; thus maximum coupling lead to a minimization of the dispersion, and thus the free energy.

Let us now return to the kagomé spin system, and discuss the effects of “order from disorder” here. We have already mentioned that the special geometry of this structure yields continuous *spin folding* zero modes that preserve the ground-state 120° spin orientation: spin folds can be “open”, traversing the entire lattice (Fig. 4a) or they can be “closed” weathervane modes (Fig. 4b). In particular, we note that spins along fold lines share a common orientation; thus two folds with noncollinear spin axes can only intersect in a *coplanar* region. Specifically, the presence of a spin fold “stiffens” all intersecting folds, increasing their frequency; the kagomé magnet thus acquires a geometric spin rigidity similar to that of a paper Origami structure. In a coplanar spin state overlapping zero modes are energetically decoupled to Gaussian order, maximizing their number. These configurations therefore minimize zero-point energy and thus are fluctuation-selected from the ground-state manifold.

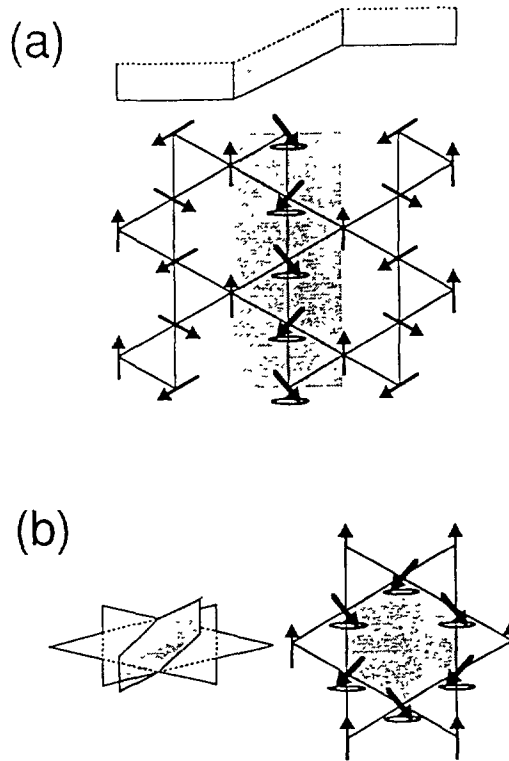


Fig. 4. — (a) Open and (b) Closed (“weathervane”) spin folds in the states of uniform and staggered spin chirality respectively. Spin orientations in diagram denote orientations in the internal *spin* space. A weak *xy* anisotropy aligns the *z* axis of the internal *spin* space with the physical *z* axis.

Quantifying these remarks, we have calculated the energy barrier associated with the cre-

ation of a spin fold semiclassically. Gaussian order spin wave theory for all fully coplanar classical ground states is formally identical; intersecting spin folds are independent, consistent with a zero-energy spinwave band. If the plane is uniformly bent, the spin configuration becomes stiff in the direction of zero curvature, replacing the flat band by a line of gapless modes in momentum space. The zero-point energy is

$$V = E - E_{cl} = \frac{1}{2} \sum_n \omega_n + \mathcal{O}(J) \quad (21)$$

where E_{cl} is $\mathcal{P}(JS^2)$ and the ω_n ($\mathcal{O}(JS)$) are the Bogoliubov quasiparticle energies in the standard Holstein-Primakoff representation. A spin fold through an angle ϕ introduces a phase factor $\Delta \rightarrow e^{2i\phi}\Delta$ to the magnon pairing field on one side of the fold; ϕ -folds are thus invisible to spinwaves. Intermediate fold angles ($0 < \phi < \pi$) constitute a *degenerate* perturbation within this manifold; for small ϕ all intersecting folds have an increased frequency $\delta\omega_n \sim JS|\delta\phi|$. The resulting energy barrier appears in the simple illustrative example of applied uniform curvature; for the planar configuration with *only* open folds (Fig. 4a), a uniform phase gradient $\nabla\phi(\mathbf{x}) = \Phi$ is introduced to the magnon pairing field along a crystal axis. A projection of the Hamiltonian into the low-energy subspace yields $H = \sum_{\mathbf{q}} \mathcal{A}_{\mathbf{q}}^\dagger \mathcal{H}_{\mathbf{q}} \mathcal{A}_{\mathbf{q}}$, with

$$\begin{aligned} \mathcal{H}_{\mathbf{q}} &= \gamma [\mathbb{1} + \underline{\sigma}_{\mathbf{x}}(\xi_{\mathbf{q}^+} \cdot \xi_{\mathbf{q}^-})] \\ \mathcal{A}_{\mathbf{q}}^\dagger &= (a_{\mathbf{q}^+}^\dagger, a_{-\mathbf{q}^-}) \end{aligned} \quad (22)$$

where $\mathbf{q}^\pm = \mathbf{q} \pm \Phi$, $a_{\mathbf{q}} = \xi_l b_{\mathbf{q}}^l$ defines the zero mode at each site $l = (1, 2, 3)$ in the unit cell and $\gamma = \frac{3JS}{4}$. Using the convention $-q_3 = q_1 + q_2$, and denoting $s_l = \sin q_l$, the eigenvector $\xi_l = s_l / \sqrt{\sum_\lambda s_\lambda^2}$. The zero point energy of this band is $V(\phi) = \gamma L \int_{\mathbf{q}} \sin \theta_{\mathbf{q}}$ per fold of length L , where $\cos \theta_{\mathbf{q}} = \xi_{\mathbf{q}^+} \cdot \xi_{\mathbf{q}^-}$. For a curvature along the x axis, $\Phi = (\phi, 0)$; this function is well approximated by $V(\phi) = \eta L |\sin \phi|$, where $\eta = 2\gamma \int_{\mathbf{q}} |\nabla_x \xi_{\mathbf{q}}| = 0.14JS$. At finite temperatures,

$$V(\phi) \sim TL \ln[2 \sinh |\beta \eta \sin \phi|] \quad (23)$$

indicating an analogous entropic selection of coplanarity, recently confirmed by numerical studies [39,40]. xy anisotropy $\delta J_z = -\epsilon J$ could also yield a coplanar state ($V(\phi) \sim L\epsilon JS^2 \sin^2 \phi$); the latter will be fluctuation-selected only if $\epsilon < \epsilon^* \equiv V_o/JS^2$. ϵ^* is a fluctuation-induced anisotropy; as for $\epsilon \neq 0$, out-of-plane spin fluctuations acquire a frequency $\Delta \sim \sqrt{\epsilon^*} JS$, that sets the scale ($\gamma \sim \Delta$) for the planar spin stiffness.

“Order from disorder” at the Gaussian level cannot distinguish between coplanar states and a residual local *discrete* degeneracy remains; is there further fluctuation-selection within this manifold? Each coplanar configuration can be identified with a ground-state of the Potts model on the kagomé lattice; there are W^N states where $W = 1.1833..$ and N is the number of sites. [41] Loops of alternating spin orientations (*abab..*), π -folds, are observed in all coplanar configurations (see Fig. 5); loop-flipping (i.e. *abab* \rightarrow *baba*) at zero-energy cost thus allows the system to explore its many ground-states. Numerical studies indicate that the *typical* coplanar state has loops on all length scales; more specifically the probability distribution for the loop length passing through a given site is a power-law distribution $P(L) \sim L^{-\zeta}$ with $\zeta \sim 1.34(\pm 0.02)$ in agreement with a recent SU(3) symmetry analysis by Read [42]. The number of loops greater than a lower cutoff length L_0 through a given site is then

$$n(L_0) = \sum_{L > L_0} P(L) dL \sim L_0^{-\frac{1}{\zeta}} \quad (24)$$

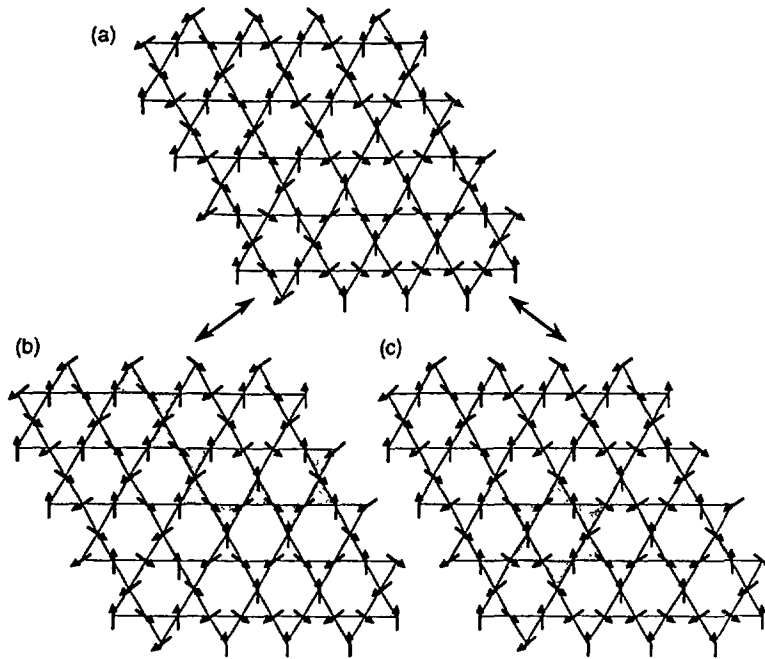


Fig. 5. — A typical coplanar state (a) and two states (b) and (c) related to it by π -folds (shown shaded) on different length-scales.

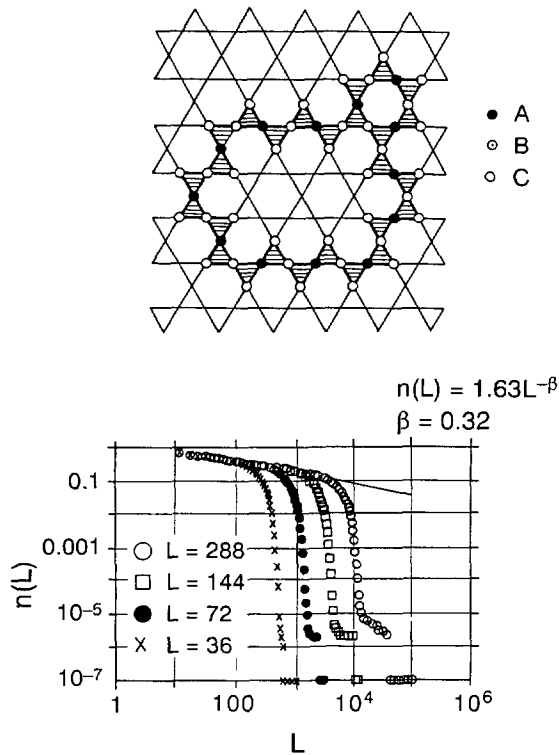


Fig. 6. — (upper) A typical loop in a coplanar state where the thick line indicates the loop perimeter (lower) The total number of loops $n(L)$ greater than length L as a function of lattice size.

as shown in figure 6. The probability distribution associated with a loop length on the lattice is

$$\tilde{P}(L) = \frac{\frac{P(L)}{L}}{\sum \frac{P(L)}{L}} \quad (25)$$

leading to

$$\langle L \rangle_{\text{lat}} = \frac{\sum L \tilde{P}(L)}{\sum \tilde{P}(L)} = \frac{\sum P(L)}{\sum \frac{P(L)}{L}} = \left\langle \frac{1}{L} \right\rangle_{\text{site}} \quad (26)$$

so that though the average loop length through any given lattice site is finite, the average loop length in the lattice is short; this is another way of saying that the typical state has a finite but small number of loops of length the lattice size. The presence of these “infinite” loops (in the thermodynamic limit) will play a crucial role in our discussion of possible glassiness; we argue that the associated energy barriers scale as $E \sim L^\alpha$ and are *macroscopic*, thus providing a necessary means for “system localization” in phase space.

Each triangular plaquette in a coplanar configuration can be characterized by its chirality [43]

$$\tau = \frac{1}{3 \sin 120^\circ} \left| \sum_{j,k} \mathbf{S}_j \times \mathbf{S}_k \right| \quad (27)$$

where cross-products are evaluated in a clockwise sense around plaquettes; $\tau_s = \tau_\Delta - \tau_\nabla$ is the chirality difference on opposite sublattices. τ_s is the chiral analogue of the staggered magnetization, and geometrically one can show that its values are constrained to ± 6 . The chirality-chirality overlap between neighboring triangular plaquettes $\tau_1 \cdot \tau_2 = \cos \theta$ gives us a flavor for the “chirality ordering” in this system; here $\theta = 0$ and $\theta = \pi$ correspond to states with staggered and uniform chirality respectively. In the pure Potts model our numerical studies (Fig. 7) yield a ratio $\frac{P(0)}{P(\pi)} = \frac{2}{3} : \frac{1}{3}$, suggesting that the $T = 0$ Potts model on the kagomé is a zero-temperature chiral glass.

Is it possible to preserve this “glassy” behavior at finite temperatures? For the Potts model the answer is no; at finite temperatures the energy cost of a defect (*aba*) on a triangular plaquette is offset by the associated entropy, and the typical state no longer has loops on all length scales. In particular, it does not have loops that span the entire length of its lattice, and thus does not have a finite number of *macroscopic* free-energy barriers crucial for glassiness. Is the situation different for a continuous order parameter where spinwaves might “delocalize” the entropy of such a defect? In general, there will be *two* contributions to the anisotropic fluctuation free energy in the coplanar manifold of the Heisenberg kagomé magnet: one associated with the discrete in-plane degrees of freedom which we have just discussed, and the other with out-of-plane continuous angular fluctuations? What is their combined effect? Numerical results for the “chirality overlap” are shown in figure 7 for the full Heisenberg model at low temperatures; a sole peak at $P(0)$ corresponds to the ground-state of the triangular antiferromagnet and the development of a magnetic moment. We see that the distribution is certainly shifted from the Potts case, but is inconclusive about the nature of the ground-state. Analytically the nature of the ground-state is a difficult question to tackle, since the coplanar degeneracy persists to Gaussian order. Nevertheless, several groups have tried to tackle this question from a variety of perspectives. Using a large N approach, Sachdev [44] finds that the staggered chiral state is selected from the manifold, though the value of the zero-temperature moment μ is a subtle point due to an order of limits issue ($T \rightarrow 0$, $\mu \rightarrow 0$). Series expansion calculations by Berlinsky and coworkers [45] also reached this conclusion; they studied the modified αJ triangular-kagomé model and then took the limit $\alpha \rightarrow 0$, a procedure that may

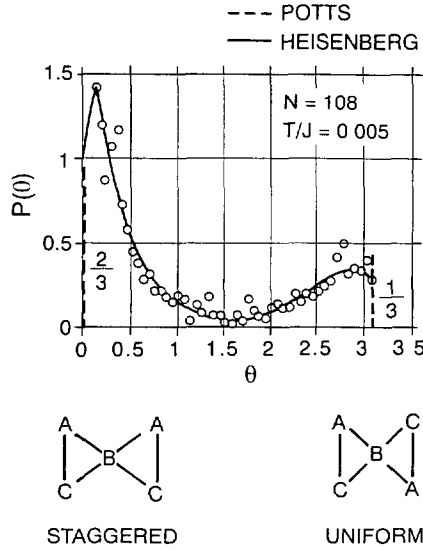


Fig. 7. — The chirality-chirality overlap between neighboring plaquettes in the Potts ($T = 0$) and the Heisenberg ($T/J = 0.005$) models where $P(0)$ and $P(\pi)$ refer to the staggered and uniform chiral states respectively.

not be appropriate if $\alpha = 0$ corresponds to an *unstable* fixed point. Recently two groups have reported that higher-order spinwave fluctuations stabilize the magnetic staggered chiral state in a *local* minimum [46,47]; because the self-energy corrections for arbitrary wavevector are very complicated for arbitrary wavevector the issue of the *global* minimum is still outstanding.

Let us recall our goal: a possible mechanism for finite-temperature glass behavior in the *absence* of disorder. This whole issue of the true thermodynamic ground-state of the Heisenberg kagomé antiferromagnet can be sidestepped by the addition of a small xy anisotropy (ϵ) to the Hamiltonian

$$H = J \sum_{i,j} \{ \mathbf{S}_i \cdot \mathbf{S}_j - \epsilon S_i^z S_j^z \} \tag{28}$$

thus putting an energy cost on out-of-plane fluctuations and favoring the “typical” coplanar spin configuration. In such a state there is *equal* probability for spins of three orientations to reside on a single sublattice; this is seen numerically (Fig. 8) in Monte Carlo simulations of the anisotropic kagomé antiferromagnet (28) at low temperatures, thus suggesting that the order parameter is a planar triad of vectors with a director normal to the plane. The associated tensor order parameters, described by three unit vectors \hat{e}_Λ ($\Lambda = 1, 3$) with $\sum \hat{e}_\Lambda = 0$, have *discrete* hexagonal C_{3h} symmetry

$$\begin{aligned}
 \langle S^\alpha(x) S^\beta(y) S^\gamma(z) \rangle &= t(x, y, z) f_{\alpha\beta\gamma} \hat{e}_1^\alpha \hat{e}_2^\beta \hat{e}_3^\gamma \\
 \langle S^\alpha(x) S^\beta(y) \rangle - \frac{1}{3} \delta^{\alpha\beta} \langle \mathbf{S}(x) \cdot \mathbf{S}(y) \rangle &= q(x - y) [\hat{n}^\alpha \hat{n}^\beta - (1/3) \delta^{\alpha\beta}]
 \end{aligned}
 \tag{29}$$

where \hat{n} is the director normal to the spin plane and $f_{\alpha\beta\gamma} = |\epsilon_{\alpha\beta\gamma}|$ is the fully-symmetric tensor. We note that π rotations of \hat{n} about the \hat{e}_λ , do *not* commute, and the homotopy group associated with the allowed point defects of (29) is *non-Abelian* [48,49,50]. Thus we expect

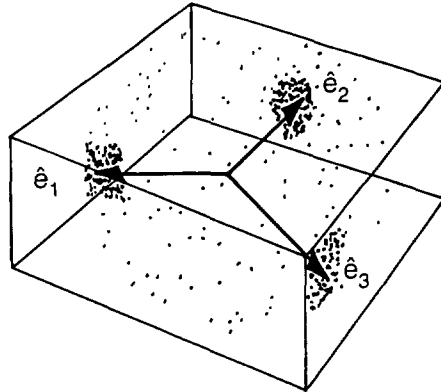


Fig. 8. — A polar plot of the spins on a *single* sublattice of the anisotropic kagomé antiferromagnet with $\epsilon = 0.01$ and $T/J = .005$; the three spin orientations appear with equal probability, thus suggesting three-spin ordering.

that these allowed *non-Abelian* textures, unlike their $U(1)$ counterparts, will affect the *local* spin state, possibly leading to a large ground-state degeneracy and to glassy behavior.

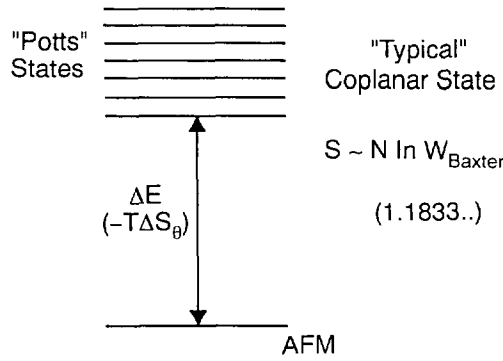
It is well-known that exponentially small amounts of xy anisotropy are sufficient to drive the spin correlation length of a two-dimensional Heisenberg antiferromagnet to infinity [51], thereby leading to a true Kosterlitz-Thouless phase transition at $T = T_{KT}$. The development of a low-temperature “typical” coplanar state demands the entropic selection of the coplanar manifold (Fig. 9) that must occur at a temperature where its free energy is lower than that of its competing ordered states. In the worst case scenario, this coplanar degeneracy may be lifted by higher-order fluctuations that favor an ordered configuration with an energy $\Delta E \sim J$ (per site) relative to that of the “typical” state [52] with an entropy $\ln W$. However, if

$$T_{KT} > T_0 = \frac{\Delta E}{\ln W} \quad (30)$$

the system will cool into a “typical” configuration with a macroscopic number of infinite energy barriers; as in conventional window glass, stability becomes a kinetic not a thermodynamic issue and the “typical” state is the effective ground-state (Fig. 9).

We would like to associate the Kosterlitz-Thouless transition in this system with the binding of non-Abelian defects, and a proposed phase diagram is displayed in figure 10. The finite xy anisotropy (ϵ) provides a new length-scale $l_0 \sim a \sim \sqrt{\epsilon}$ beyond which renormalization of the spin stiffness is suppressed; in other words, there is an energy cost for rotating a spin cluster of length l_0 out of the plane. The phase diagram is generated by comparing l_0 with the other length scale of the problem ξ , the spin correlation length. If ξ is less than l_0 ($T > T_\pi$, the defects are free. However if $l_0 > \xi$ ($T_\pi > T > T_{KT}$) they bind (similar to mesons!) to avoid anisotropic energy costs; since a small number of “quarks” still exist due to “meson-meson” scattering, T_π is only a crossover temperature. Eventually at $T = T_{KT}$ the spin correlation length diverges and all defects are bound.

This proposed binding has several distinctive features that are different from those observed in a conventional Kosterlitz-Thouless transition [53]. First of all, it is a true lattice symmetry-breaking (second-order) transition, and thus is accompanied by a specific heat *divergence*. Next the gradient field associated with the 120° defect is a third that of a standard 360° one,



Entropy Selection of "Typical" State

if

$$T_{KT} > \frac{\Delta E}{\ln W}$$

Fig. 9. — A schematic representation of the kinetic stability of the typical coplanar configuration.

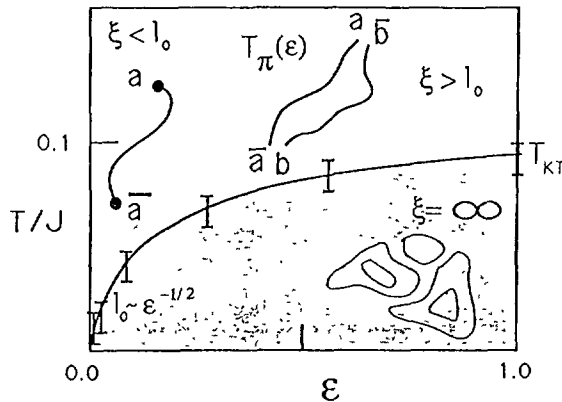


Fig. 10. — The proposed phase diagram for the anisotropic kagomé antiferromagnet, with superimposed numerical dates points for the Kosterlitz-Thouless temperature T_{KT} as a function of anisotropy (ϵ).

leading to a reduction of the transition temperature by a factor $\frac{1}{9}$. Finally we believe that the low-temperature phase will be determined by the detailed braiding of the defect paths; unlike the conventional case, the ground-state configurations cannot be characterized by two winding numbers due to the non-commuting nature of the allowed defect structures.

4. Preliminary numerical diagonalistics.

In the last section we put forth many ideas about the anisotropic kagomé antiferromagnet; let us now try to test them. Unfortunately the proposed three-spin ordering is inaccessible from existing mean-field treatments of magnetism and may require a new type of gauge theory, but further characterization with “numerical diagonalistics” is possible before plunging into new analytic technology. Following this approach, we are currently performing Monte Carlo simulations on the classical Heisenberg kagomé antiferromagnet with a weak xy anisotropy $\delta J_z = -\epsilon J$. Typically, arrays of 108, 432 and 864 spins are sequentially cooled from a random configuration with 1.25×10^6 spin flips per site per temperature, and thermal averages are also performed over periods of 1.25×10^3 updates [52]. Monitoring local spin current fluctuations, we can measure the spin stiffness γ [54]; we identify a jump in this stiffness (Fig. 11) with the Kosterlitz-Thouless transition, and note that $\gamma \rightarrow 0$ for $\epsilon \rightarrow 0$. For finite ϵ we observe a divergence in the specific heat at $T = T_{KT}$, in contrast to the conventional Kosterlitz-Thouless case [53].

Could this specific heat behavior be due to an “order from disorder” Ising transition? Perhaps, but figure 11 (lower) makes it clear that chirality is certainly *not* the relevant order parameter. The generalized Edwards-Anderson order parameter, $\chi_{ch} = \frac{1}{N}(\sum_i \tau_s)^2$ measures the overlap between of a given state with one of uniform staggered spin chirality ($\tau_s = \tau_\Delta - \tau_\nabla$); at low temperatures χ_{ch} remains finite indicating the *absence* of magnetic ordering. By contrast, $\overline{\chi_{ch}^2}$ fluctuates rapidly at a temperature $T \leq T_{KT}$, suggesting the possibility of glassy behavior. Figure 11 (lower) also displays the distribution of χ_{ch} at low temperatures that is similar to that of the typical Potts state at $T = 0$ on the same lattice ($P(\chi_{ch}) \sim e^{-\chi_{ch}/\chi_0}$ with $\chi_0 \sim 3.29$). χ_{ch} can also be monitored as a function of time (Fig. 12); preliminary results suggest a hierarchy of time-scales in the problem, probably associated with the freezing of loops on different length-scales. The loop distribution can be determined from the power spectrum of this data; early results suggest that it is very similar to that of the $T = 0$ Potts model, and that somehow its continuous degrees of freedom permit it to have a “spin roughening” transition at finite temperatures.

5. Discussion.

At this point, the numerical results clearly indicate that the anisotropic kagomé spin system is *not* a conventional antiferromagnet. Numerical noise spectra and multi-spin overlap studies are currently in progress to further characterize the low-temperature phase of this system. We believe that this system is one-spin disordered, two-spin glassy and three-spin ordered, but require further “diagnostics” before a detailed description of the ground-state(s) is possible. In many ways, the challenge of this model is to maintain the spin Goldstone mode at low temperatures despite the presence of a discrete transition with many relaxation time-scales; we are still far from this goal.

Here, following the ideas of Villain [55] and De Sze [56], we have once again raised the controversial issue of glassiness in the absence of disorder, bringing together several preexisting themes in the literature. The binding of non-Abelian defects has been already proposed several times as a mechanism for a glass transition [57-61], and the anisotropic kagomé spin system may provide a natural setting for the realization of this scenario. The importance of chirality in vector spin glasses was originally discussed by Villain [43] with much subsequent work by Kawamura [62], who has also been interested in the connection between multi-spin ordering and glassiness in vector spin systems. To our knowledge the discussion of defect topology in

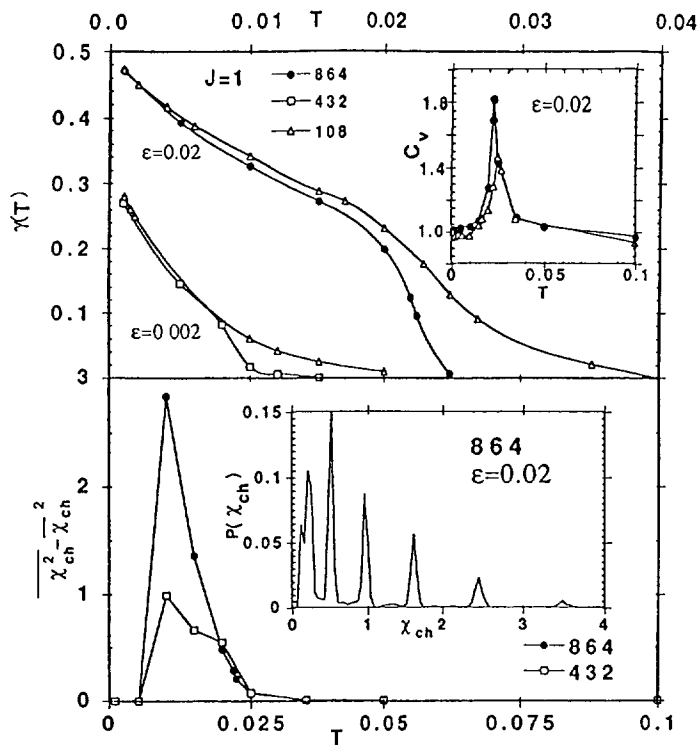


Fig. 11. — (upper) Spin stiffness about z axis for 108, 432 and 864 spins at $\epsilon = 0.002$ and $\epsilon = 0.02$; γ is determined by monitoring local spin current fluctuations. (lower) Divergence in $\overline{\chi_{ch}^2}$ for $\epsilon = 0.02$ with 432 and 864 spins; Inset: Distribution of χ_{ch} for $\epsilon = 0.02$ and $\overline{T} = 0.010$ for 432 and 864 spins.

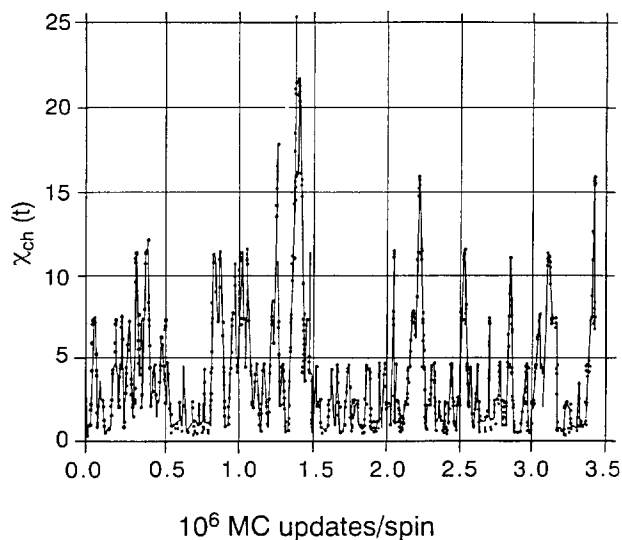


Fig. 12. — χ_{ch} as a function of time for $\epsilon = 0.02$ and $\overline{T} = 0.02$; note that χ_{ch} changes by values ± 6 , as required geometrically, and that there appear to exist a *distribution* of relaxation times (confirmed by the noise spectrum of this data).

spin glasses was initiated by Toulouse [63], followed by Henley's extensive computer search [64] for defect structures in the $d = 3$ Heisenberg spin glass. We do not know of any similar studies for two-dimensional periodically frustrated systems, and are currently beginning such a project for the kagomé case. The hope is that once we can classify its allowed defect structures, we will be closer to understanding its low-temperature behavior; perhaps we could then even develop a analytic description of this phenomenon.

There are already several aspects of our quasi-two dimensional picture of the kagomé system that pertain directly to the SCGO(x) measurements. Of course, we have not included dilution in our discussion that is bound to have consequences. In particular, a defect centered on a vacancy will have a reduced core energy leading to a finite density of unbound pinned vortices; in short, we do expect the character of the problem to be affected by disorder. However, the fully-occupied anisotropic kagomé antiferromagnet is a good start as a first step, and even here there are plenty of unsolved questions. We do argue that our semiclassical approach to the "simplified" problem is justified for the experimental parameters, because the classical energy scale is set by the *reduced* Kosterlitz-Thouless temperature $T_{KT} \approx \frac{\alpha^2 \pi \gamma}{2} = \frac{\pi \gamma}{18}$ where γ is the spin stiffness. The upper limit for the stiffness is the pure xy case where $\gamma = \frac{JS^2}{2}$; the resulting energy scale $T_{KT} = \frac{JS^2 \pi}{2} \approx \frac{T_{CW}}{48}$ is within the mysterious low-temperature regime of SCGO(x). Heuristically, a finite ϵ could easily originate from site (dilution?) or bond anisotropy, and provides the small energy scale associated with the ratio of the glass and the Curie-Weiss temperatures. In our scenario the "freezing" of the typical state results in a negative divergence, as observed, of the nonlinear susceptibility: here higher-order fluctuations will generate a chirality coupling $E = \alpha_0 \sum_{\Delta, \nabla} \langle \tau_{\Delta} \cdot \tau_{\nabla} \rangle \propto \chi_{ch}$ between neighboring plaquettes, so that the energy per site has an exponential distribution $P(E) \sim P(\alpha_0 \chi_{ch})$. Within any given coplanar configuration, the kagomé system has a frozen moment and thus a Golstone mode in the spin channel, consistent with the T^2 specific heat and the flat inelastic neutron scattering cross section. The field-dependence of the glass transition T_g is a definite test of our ideas; in a conventional spin glass an external applied field suppresses T_g , whereas in the picture of topological freezing such a field *increases* the anisotropy, thereby enhancing the transition temperature.

Clearly at this time there are many more questions than answers, though there are lots of ideas. We would like to believe that the strong frustration present in the kagomé antiferromagnet stabilizes an exotic three-spin order parameter that permits new types of defect and possibly a new class of phase transition, but there is much work to be done before this conjecture can be confirmed. As Rammal would tell us, "Allons-y!"

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