Hidden glassy behaviors in an ideal Heisenberg Kagomé antiferromagnet

J.D. Lee

W.M. Keck Laboratories, California Institute of Technology, Pasadena, CA 91125

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Dynamics of classical Heisenberg spins, $S_i = (s_i, S_{iz})$, on the Kagomé lattice has been studied. An ideal Heisenberg Kagomé antiferromagnet is known to remain disordered down to $T = 0$ due to the macroscopic degeneracy of the ground state. Through the study, however, we find that $S_i$ and their planar components $s_i$ behave in a qualitatively different way and especially planar spins ($s_i$) show the exotic glass-like transition in the very low temperature $T \sim 0.003J$ ($J$: spin exchange).

The glassy behaviors of $s_i$ would be found to be driven by the spin-nematic fluctuations, different from ordinary spin glasses by disorders or anisotropies.

Magnetic systems in which the exchange energy is not minimized by a simple regular arrangement of the spins are called the frustrated magnetic systems. Frustrated antiferromagnets induced by the geometry of the lattice have been a recent subject attracting much experimental and theoretical interests [1]. One example of such systems is the antiferromagnet on the Kagomé lattice. It consists of triangles. The Kagomé lattice is, however, more frustrated than the triangular lattice because triangles on the former lattice share only one vertex, but on the latter share one side. Indeed, classical Heisenberg spins on the Kagomé lattice have an infinitely degenerate ground state manifold at low temperature $T \sim \frac{1}{T_g}$, which has been a main reason drawing considerable attention to the frustrated magnetic systems [2]. Their experimental realizations are, for instance, the insulating layered compound SrCr$_8$Ga$_{4+x}$O$_{19}$ ($Cr^{3+}$ carries $S = 3/2$) [3, 4, 5] and the jarosites AF$_3$(OH)$_6$(SO$_4$)$_2$ ($A=K,\text{H}_2\text{O},\text{Na},\text{NH}_4$, and so forth; Fe$^{3+}$ carries $S = 5/2$) [6, 7, 8].

Extremely high degeneracy of the ground state inhibits the long-range order at any $T$. But it has been known that thermal fluctuations in the system select a subset of ground state manifold at low $T$ (known as ordering by disorder [4, 6, 7]), even if this subset is not clear enough to lead to long-range Néel order. Chalker et al. [4] have argued by low temperature expansions that thermal fluctuations resolve degeneracy of ground states and cause the system to select a coplanar nematic ground states. There are at least two coplanar ground states possessing long-range Néel order; one is the so-called $q = 0$ state and the other is $\sqrt{3} \times \sqrt{3}$ state (See the inset in Fig. 1a): in the $q = 0$ state, spins along lines are placed in an alternating sequence compared to the $\sqrt{3} \times \sqrt{3}$ state. Reimers and Berlinsky [6] have shown, through the Monte Carlo simulation, that an increase in the order parameter of the $\sqrt{3} \times \sqrt{3}$ state is dominant over the $q = 0$ state. In addition, Sachdev [12] has also argued that the $\sqrt{3} \times \sqrt{3}$ state is further accepted and favored in the low temperature limit by quantum fluctuations.

Magnetic frustration used to be found with the site disorder, even if these two features may appear independently. When both, disorder and frustration, are strong, its interplay gives place to the spin glass phase [13]. On the other hand, the antiferromagnetic Heisenberg model on the Kagomé lattice is an ideal geometrically frustrated magnet without disorders. Nevertheless, in some of prototype systems of the Kagomé lattice, the spin glass ordering at a finite temperature is actually observed because of deviations from an ideal one. In SrCr$_8$Ga$_{4+x}$O$_{19}$, $T_g = 3.3$ K is found in a case of $x = 0$ corresponding to 89% occupation of Cr sites [7, 8]. In the system, Cr$^{3+}$ just enters three different octahedral sites (namely, 2a, 12k, 4f) with an almost random distribution and SrCr$_8$Ga$_4$O$_{19}$ has then $\sim 14\%$ site disorder (nonmagnetic impurities) on the Kagomé layer (12k sites). The glass order is enhanced by the additional dilution of Cr sites [9]. The glassy phase in nondisordered frustrated magnetic systems has been attracting much interests. Most two-dimensional magnetic materials exhibit some kinds of spin anisotropy. A critical slowing down of magnetic fluctuations is found in the hydronium-jarosite (H$_2$O)Fe$_3$(OH)$_6$(SO$_4$)$_2$ with much less disorders [10]. The glass phase in the system may be a bit different from in SrCr$_8$Ga$_4$O$_{19}$, due to the $xy$ anisotropy (“easy-plane”), which could drive the Kagomé antiferromagnet to experience the topological Kosterlitz-Thouless-type transition to a glassy state [11]. The glassy behavior in the Kagomé antiferromagnet with the easy-axis type anisotropy has been also explored [11].

In this paper, through the study of dynamical properties, we report the hidden glassy behaviors of Heisenberg spins on the ideal Kagomé lattice without any disorder or anisotropy. Keren [15] has investigated the stability of the spin order against excitation and the dynamical responses of spins on the Kagomé lattice. Actually, in his study, there is found no transition down to $T = 0$ as there should not be. Nevertheless, we find that behaviors of $S_i$ and its planar component $s_i$ are qualitatively different from each other and $s_i$ undergoes a glassy transition around $T \sim 0.003J$ unlike $S_i$. Taking the classical Heisenberg spins as $S_i = (s_i, S_{iz})$, we have the Hamiltonian

$$H = \sum_{ij} J \mathbf{s}_i \cdot \mathbf{s}_j + \sum_{ij} S_{iz} S_{jz},$$  

where $s_i^2 + S_{iz}^2 = 1$. Reimers and Berlinsky [6] have demonstrated that the tendency toward spin-nematic order starts at $T \sim 0.01J$, which motivates us to explore...
the respective dynamics of $\mathbf{S}_i$ and $s_i$ at the relevant temperature range, i.e. $T \lesssim 0.01J$. Particularly, we have interests in the local spin-spin correlation functions as

$$J(\omega) = \frac{1}{N} \int d\tau e^{-i\omega \tau} \sum_i (\mathbf{S}_i \cdot \mathbf{S}_i(\tau)), \quad j(\omega) = \frac{1}{N} \int d\tau e^{-i\omega \tau} \sum_i (s_i \cdot s_i(\tau)). \quad (2)$$

The correlation functions such as $J(\omega)$ and $j(\omega)$ can be most properly evaluated by the Monte Carlo simulation. The time evolution of spins are governed by the equation of motion $\partial \mathbf{S}_i / \partial \tau = -J \mathbf{S}_i \times \sum_{j \neq i} \mathbf{S}_j$. A practical integration of the equation is done using the Suzuki-Trotter decomposition on the three sublattices (say A, B, and C) of the Kagomé lattice, $\{\mathbf{S}_i(\tau + \delta \tau)\} = e^{(A+B+C)\delta \tau} \{\mathbf{S}_i(\tau)\}$, where $e^{(A+B+C)\delta \tau}$ is decomposed using $e^{(X+Y)\delta \tau} = \prod_{p=1}^3 e^{p_1 X \delta \tau / 2 e^{p_2 Y \delta \tau} e^{p_3 X \delta \tau / 2 + O(\delta \tau^5)}}$ and $p_1 = p_2 = p_4 = p_5 = p = 1/(4 - 4^{1/3})$ and $p_3 = 1 - 4p$. We use the hybrid Monte Carlo procedure which combines the Metropolis update and the overrelaxation update. One hybrid Monte Carlo step in our calculation consists of two Metropolis steps and four overrelaxation steps. Using the hybrid algorithm, $O(10^4)$ hybrid steps are typically used to reach the equilibrium configuration. A system size is defined by the linear dimension $L$ of the lattice and the total number of spins $N$ is $3L^2$. In an actual simulation for dynamical correlation functions, the time integration has been performed up to $\tau_{max} = 26214.4J^{-1}$ with a step of $\delta \tau = 0.2J^{-1}$. The thermal average of time-dependent variables is computed over the observables obtained by evolving all independent initial equilibrium configurations. Typically, $10^3$ configurations were generated at a given $T$. An adopted system size for dynamical calculations is $L = 24$, that is, 1728 spins.

In Fig. 1 the temperature-dependences of $J(0)$ and $j(0)$ are provided both when relaxing and annealing the system. $J(0)$ (or $j(0)$) is a quasi-elastic response of local spin-spin correlation function and a quantity directly related to the spin-lattice relaxation rate $T_1$ of a local probe like the nuclear magnetic resonance (NMR). For a case of relaxing, the nominal state at $T = 0$ is taken as the $\sqrt{3} \times \sqrt{3}$ state. For a case of annealing, on the other hand, after heating the $\sqrt{3} \times \sqrt{3}$ state to $T = 0.05J$, we obtain the equilibrium state by gradual cooling. Fig. 1 shows that $J(0)$ is history-independent, whereas $j(0)$ is history-dependent roughly below $T \sim 0.003J$. History-dependent magnetic responses can be one of indications of the glassy phase because of the slow dynamics of spins in the phase. Hereafter the aimed configuration is obtained by warming the system from the $\sqrt{3} \times \sqrt{3}$ state, if not stated otherwise. In the inset of the figure, $J(0) - j(0)$, i.e. $J_0(0)$, are given. A much sharper comparison between $J(0)$ and $j(0)$ is done by taking their inverses. In Fig. 2 $1/J(0)$ and $1/j(0)$ are shown more or less linear with respect to $T$ and $1/j(0)$ suffers an abrupt and appreciable change of the proportionality constant near $T \sim 0.003J$, which is not found in $1/J(0)$.
and is, \( \nu \) Fig. 3, the low energy responses of the spin-spin correlation

\( T \) differences, we pay attention to the low energy sector qualitatively different way. In order to understand the changes to \( j(\omega) / j(0) \), we note that \( \nu(T) \) and especially \( \nu(T) \sim 0.09 \) to \( \nu(T \gtrsim T_g) \sim 0.4 \) around the freezing temperature \( T_g \).

Broholm et al. [3] have also reported the similar power law behavior like \( \omega^\nu(T) \) of the local response function in SrCr\(_{0.995}\)Ge\(_{0.005}\) and changes of \( \nu(T) \) around \( T_g \), that is, \( \nu(T) \sim 0 \) below \( T_g \) and \( \sim 1 \) in high temperatures. In Fig 3, the low energy responses of the spin-spin correlation functions are given. Behaviors of \( j(\omega) \) in the lower panel of Fig 3 is appealing in that \( j(\omega) \sim \omega^\nu \) (\( j(\omega) \) is expected to follow \( \sim 1/\omega^2 + \Gamma(T^2) \)) below \( T \sim 0.003J \) changes to \( j(\omega) \sim \omega^\nu \) (\( \nu > 0 \) above \( T \sim 0.003J \). No similar changes are found in \( J(\omega) \) as shown in the upper panel of the figure. All previous results of Figs 1, 3 on \( j(\omega) \) are subordinate to the glass-like phase transition at \( T \sim 0.003J \) and quite consistent with each other. On the contrary, any indication of spin phase transition down to \( T \rightarrow 0 \) is not found in magnetic responses of \( S_1 \) (i.e. \( J(0) \) and \( j(0) \)), which is consistent with Kerens' [12].

We now importantly note that \( S_1 \) and \( s_i \) behave in a qualitatively different way. In order to understand the differences, we pay attention to the low energy sector of the Hamiltonian. Following Chalker et al. [4], \( H_0 \) in

\[
H = H_0 + H_h(\epsilon_\alpha^2, \epsilon_\beta^2) + H_{anh}(\epsilon_\alpha^2 \epsilon_\beta^2, \epsilon_\beta^4, ...),
\]

where \( \epsilon_\alpha \) (\( \epsilon_\beta \)) is the fluctuation perpendicular to \( S_1 \) in the particular planar ground state and in (out of) the spin plane and \( H_h \) and \( H_{anh} \) includes harmonic and anharmonic fluctuations, respectively. In harmonic fluctuations (within the linear spin waves), \( \epsilon_\alpha^2 \) gives three transverse modes and \( \epsilon_\beta^2 \) two transverse modes and one zero mode per unit cell comprising three spins. The role of quartic potential for the zero mode is then unique and is to stabilize the planar states by inducing the spin-nematic fluctuation. Now, let us think of the planar counterpart of \( H \), i.e. \( H_a \),

\[
H_a = H_0 + H_h(\epsilon_\alpha^2) + H_{anh}(\epsilon_\alpha^2 \epsilon_\beta^2, \epsilon_\beta^4, ...).
\]

In \( H_a \), terms of \( \epsilon_\alpha \epsilon_\beta^2 \) and \( \epsilon_\beta^4 \) disturb the system like disorders (but not static) in a complicated way. For a weakly (or non-) disordered case, it is known that the ground state configuration is such that the energy of each separate triangle is minimized ("rule of satisfied triangles"), implying the glass phase will not occur [22]. In \( H_a \), however, in Fig 4 the energy minimization of each triangle depends on the local spin distortions produced by neighboring triangles, in other words, by correlations between fluctuating degrees of freedom (e.g. \( \epsilon_\alpha^4 \epsilon_\beta^2 \) or \( \epsilon_\beta^2 \epsilon_\beta^4 \)). Alternatively, terms of \( \epsilon_\alpha^2 \epsilon_\beta^2 \) and \( \epsilon_\beta^4 \) can also be understood to lead to planar spins of dynamically varied magnitudes, which act as strong disorders such that glassy behavior may occur. To integrate out \{\( \epsilon_\alpha \)\} in the partition function or correlation functions leaves terms of \( \epsilon_\beta^4 \). If we note its role in \( H \) and the ground state, the glass-like order of planar spins can be attributed to the spin-nematic fluctuations. The glass-like order in the system is different from conventional spin glasses in that it is originated by the dynamical fluctuations of inherent degrees of freedom, not by the static physical disorders or anisotropies in the systems.
The spin susceptibilities are defined for at the transition temperature in the spin glass [13, 24]. It is also a good indicator of the spin ordering and shows a cusp to examine the static spin susceptibility. It is also a XY spins (planar spins $\chi_{\text{planar}}$ and its planar component $\chi_s$) without such fluctuations [23], for which the easiest way is probably to examine the static spin susceptibility. It is also a good indicator of the spin ordering and shows a cusp at the transition temperature in the spin glass [13, 24]. The spin susceptibilities are defined for $\mathbf{S}_i$ and $\mathbf{s}_i$ as $\chi_{\mathbf{S}}(T) \propto 1/T \sum_{i \neq j} [\mathbf{S}_i \cdot \mathbf{S}_j - \langle \mathbf{S}_i \rangle \cdot \langle \mathbf{S}_j \rangle]$ and $\chi_s(T) \propto 1/T \sum_{i \neq j} [\mathbf{s}_i \cdot \mathbf{s}_j - \langle \mathbf{s}_i \rangle \cdot \langle \mathbf{s}_j \rangle]$, respectively. In Fig. 5 it is found that only $\chi_s(T)$ shows a cusp at $T \sim 0.003J$ and other $\chi_{\mathbf{S}}(T)$ and $\chi_{\text{XY}}(T)$ increases smoothly without any singularities with respect to $T$, even if all the three cases are started from the seemingly same spin configuration (i.e. $\sqrt{3} \times \sqrt{3}$ state). Spin susceptibilities in Fig. 5 ascertain the glassy behavior of the planar components of Heisenberg spins on the Kagomé lattice and further support that it would be induced by the spin-nematic fluctuation. As mentioned, one of the most outstanding features of the frustrated magnet is the high degeneracy of the ground state, whose peculiarity could result in the spin-nematic ordering in the limit of very low temperatures ($T \ll J$). The nematic glassy state should then be crucial and eminently important in understanding the novel ground state of the frustrated magnet.

In summary, we have found and discussed qualitative differences between $\mathbf{S}_i$ and its planar component $\mathbf{s}_i$ in an ideal Heisenberg Kagomé antiferromagnet. It is remarkable to find the hidden glass-like order of $\mathbf{s}_i$ even in the Heisenberg model on the Kagomé lattice without any disorder or anisotropy. The glassy behavior has been shown through dynamical spin responses and static susceptibilities. We find that it would be driven by the dynamical fluctuation of spin-nematic order, different from ordinary spin glasses by static disorders or anisotropies.

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![FIG. 5: Static spin susceptibilities for $\mathbf{S}_i$ and $\mathbf{s}_i$, $\chi_{\mathbf{S}}$ and $\chi_s$. The inset gives the spin susceptibility for XY spins, $\chi_{\text{XY}}$. Calculations are done for 2700 spins on the Kagomé lattice ($L = 30$).](image_url)

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