Dynamic spin-glass behavior in a disorder-free, two-component model of quantum frustrated magnets

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Motivated by the observation of a spin-glass transition in almost disorder-free Kagome antiferromagnets, and by the specific form of the effective low-energy model of the S=1/2, trimerized Kagome antiferromagnet, we investigate the possibility to obtain a spin-glass behavior in two-component, disorder-free models. We concentrate on a toy-model, a modified Ashkin-Teller model in a magnetic field that couples only to one species of spins, for which we prove that a dynamic spin-glass behavior occurs. The dynamics of the magnetization is closely related to that of the underlying Ising model in zero field in which spins and pseudo-spins are intimately coupled. The spin-glass like history dependence of the magnetization is a consequence of the ageing of the underlying Ising model.

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I. INTRODUCTION

The experimental investigation of quantum magnets is undergoing rapid progress with the synthesis of new and better controlled samples. Of special interest are frustrated magnets, for which very unusual behavior has been reported\(^{(1)}\). In particular, a behavior reminiscent of spin glasses has been reported in a number of Kagome antiferromagnets (AF)\(^{(2,3)}\). While the role of disorder in these phenomena cannot be excluded yet, the persistence of this behavior in progressively cleaner samples calls for explanations in terms of disorder-free models.

The possibility of a spin-glass behavior in the spin 1/2 Heisenberg model on the Kagome lattice without any disorder was first suggested in the pioneering work of Chandra et al.\(^{(4)}\) in 1993. Since the classical Heisenberg model on the Kagome lattice does not exhibit a spin-glass behavior, the main problem one is facing is how to include the quantum aspects of the problem into a description in terms of classical variables for which one can use standard techniques to study the dynamics. In Ref.\(^{(5)}\), the authors assumed that quantum fluctuations select coplanar configurations, which led them to concentrate on the anisotropic XY version of the model. For this model, they suggested the presence at finite temperature of 3-spin order, and of a glassy transition related to the binding of non-abelian defects that are expected to be the natural point defects associated with this type of order. For details, the reader should consult Ref.\(^{(5)}\). To prove or disprove this scenario turned out to be difficult, and whether this indeed provides an explanation of the spin-glass behavior of Kagome antiferromagnets without introducing any disorder remains unsettled. Since then, clear evidence of glassiness has been reported by Chandra et al.\(^{(6)}\) for another class of disorder-free models describing periodic Josephson arrays in a transverse magnetic field, but these models are not directly related to the Kagome AF.

In parallel, a lot of progress has been made in the understanding of the spectrum of the S=1/2 AF Heisenberg model on various frustrated lattices, in particular the Kagome and the pyrochlore ones\(^{(7)}\). So far, it is well established that frustration can have two effects: It can open a gap to triplet excitations\(^{(7)}\), like for the non-frustrated spin 1 chain\(^{(8)}\), but it can also lead to a proliferation of low-lying singlets inside this gap, like for the S=1/2 Kagome antiferromagnet\(^{(9)}\). These singlets can be interpreted as RVB (Resonating Valence Bond) states\(^{(10)}\), and they could lead to a power-law behavior of the low-temperature specific heat\(^{(11–13)}\). Experimental systems known so far have a larger spin however\(^{(3/2, 5/2,...)}\), and there is room for new physics in these cases since the presence of a singlet-triplet gap is unlikely given the rather small value already reported for S=1/2. Possible implications of this strange spectrum regarding in particular a possible spin-glass behavior have not been discussed yet, mainly due to the lack of methods to attack this problem.

In this paper, we continue the quest initiated in Ref.\(^{(5)}\) for spin-glass behavior in the disorder-free, quantum Heisenberg model on the Kagome lattice. However, building on the recent results obtained on the low-energy spectrum of the model, we propose another approximate way of including quantum fluctuations into a classical description. The starting point is the effective model obtained in Ref.\(^{(14)}\) for the spin 1/2 Heisenberg model on the trimerized Kagome lattice (see Fig.\(^{(1)}\)). This is a modified version of the spin 1/2 Heisenberg model on the Kagome lattice in which the exchange integrals take two different values \(J\) and \(J'\) according to the pattern of...
Fig. 1. This is actually the relevant description of the Kagome layers in SrCr$_9$pGa$_{12-9}$O$_{19}$ since the presence of a triangular layer between pairs of Kagome layers lead to two types of bonds with precisely the pattern of Fig. 1. Then, since the ground state of a triangle is fourfold degenerate and can be described by two spin 1/2 degrees of freedom, the total spin $\vec{\sigma}$ and the chirality pseudospin $\vec{\tau}$, it was shown in Ref. [14] that the low-energy effective Hamiltonian in the limit $J' \ll J$ can be written

$$\hat{\mathcal{H}} = \left( \frac{J'}{9} \right) \sum_{<i,j>} \hat{H}_{ij}^\sigma \hat{H}_{ij}^\tau, \quad \hat{H}_{ij}^\sigma = \sum_{<i,j>} \hat{\sigma}_i \cdot \hat{\sigma}_j,$$

$$\hat{H}_{ij}^\tau = (1 - 2(\alpha_{ij} \tau_i^- + \alpha_{ij} \tau_j^+))(1 - 2(\beta_{ij} \tau_j^- + \beta_{ij} \tau_j^+)) \quad (1)$$

where $<i,j>$ denotes pairs of nearest neighbors. In $\hat{H}_{ij}^\sigma$, $\alpha_{ij}$ and $\beta_{ij}$ are complex parameters that take the values $1$, $\exp(2\pi i/3)$ or $\exp(4\pi i/3)$ depending on the bond (for details, see Ref. [14]).

The analysis of the model of Eq. (1) with the spin and the pseudo-spin treated as classical Heisenberg variables is a considerable task, and before starting such an endeavor, one would like to know whether the scenario outlined in the previous paragraph can indeed lead to a spin glass behavior. In fact, other models, like the fully frustrated XY model [16] or some vector models [17], can be described in terms of spin and chirality variables, and no spin-glass behavior was ever reported for these models. Consequently the rest of the paper is devoted to a detailed analysis of a toy model to test whether the presence of two local degrees of freedom can indeed lead to a spin-glass behavior. The simplest model of this kind is a modified Ashkin-Teller model defined by the Hamiltonian:

$$\mathcal{H} = J \sum_{<i,j>} S_i S_j T_i T_j - h \sum_i S_i \quad (2)$$

In this Hamiltonian, $S_i$ and $T_i$ are Ising variables that describe the spin and the pseudo-spin respectively. Note that the magnetic field $h$ is coupled only to the spin degree of freedom. Since frustration has already been included in the model as an extra degree of freedom, there is no need to work on a frustrated lattice any more, and for simplicity we study this model on a square lattice. This model is of course a very dramatic simplification since the Heisenberg spins and pseudo-spins are replaced by Ising variables. Still, as we shall see, the physics of this model is very rich, and to a large extent confirms the simple picture of the previous paragraph. Shortcomings that might be overcome by going to the more physical description in terms of Heisenberg spins will be discussed in the last section.

II. MONTE-CARLO RESULTS

Let us first discuss the equilibrium properties of this model. If $h = 0$, this model is equivalent to the antiferromagnetic Ising model after the local gauge transformation $\sigma_i \equiv S_i/T_i$, and all states have an additional degeneracy of $2^{N_{sites}}$, since $\sigma_i = 1$ (resp. $-1$) can be achieved with $(S_i, T_i) = (1, 1)$ and $(-1, -1)$ (resp. $(1, -1)$ and $(-1, 1)$). In fact, for any value of $h$, the partition function can be factorized as $Z = Z_{\text{Ising}} Z_S$ where $Z_{\text{Ising}}$ is the...
partition function of the Ising model on a square lattice, and \( Z_S \) is the partition function of paramagnetic spins in a magnetic field. Accordingly, the free energy per site \( f \) is given by:

\[
f = f_{\text{Ising}} - k_B T \ln(2 \cosh(\beta h))
\]

with \( \beta = 1/k_B T \). The magnetization per site is defined as usually by \( m = -\partial f/\partial h \), and since \( f_{\text{Ising}} \) does not depend on \( h \), we obtain \( m = \tanh(\beta h) \). So if the system has reached its equilibrium state, the magnetization smoothly saturates as the temperature goes to zero.

However, as we shall see below, this equilibrium state might be very difficult to reach depending on the history of the system. To be specific, let us consider the following protocol: The quench takes place at \( t = 0 \), the field is switched on after a waiting time \( t_w \), and the magnetization is measured after a measuring time \( t_m \) elapsed after the field has been switched on. The time elapsed between the quench and the measurement is denoted by \( t \). In a FC experiment, \( t_w = 0 \) and \( t = t_m \), while in a ZFC one, \( t_w > 0 \) and \( t = t_w + t_m \).

To mimic such experiments, we have performed Monte Carlo simulations of the model of Eq. (2). The elementary step consists in flipping either a spin or a pseudo-spin according to Glauber prescription. The site and the variable (spin or pseudo spin) are chosen randomly, and the time unit corresponds to a number of steps equal to the number of degrees of freedom, i.e. twice the number of sites. In all numerical experiments reported below, the starting configuration is completely random, corresponding to an infinite temperature, and when it is not switched off, the magnetic field \( h \) is equal to 0.2 in units of \( J \). Typical results for the magnetization as a function of temperature are shown in Fig. (3) for a system of size 400 \( \times \) 400. Below a temperature \( T_g \) which is smaller than the Curie temperature \( T_c \approx 2.27J \) of the underlying Ising model, there is a clear difference between the ZFC and FC measurements: The ZFC magnetization drops quite abruptly, as in typical spin-glasses. Note that these curves depend on \( t_m \) and \( t_w \) (this dependence should of course vanish if \( t_m \) was infinite) but only weakly in a large parameter range, as we shall explain below.

To gain some insight into the origin of this behavior, it is very useful to study the time evolution of the magnetization at fixed temperature for different values of \( t_w \). Typical results are given in Fig. (4). The most salient features are: i) A much faster increase at short times for the FC experiment (\( t_w = 0 \)) than for the ZFC ones; ii) A similarity of the shape of the ZFC curves. In fact, a very good scaling can be obtained if we plot the magnetization as a function of \( t_m/t_w \) (see inset of Fig. (3)). Such a scaling is typical of ageing phenomena [17].

![FIG. 2. Temperature dependence of the magnetization for different measurement conditions.](image)

![FIG. 3. Time dependence of the magnetization at given temperature for various measurement conditions (FC and ZFC with \( t_w = 500, 1000, 2000, 3000 \)). Inset: Plot of the ZFC data as a function of \( t_m/t_w \).](image)

### III. ANALYTICAL APPROACH: AGEING AND PERSISTENCE

To understand the behavior of the ZFC and FC experiments let us first consider the dynamics of the underlying Ising model in the absence of a magnetic field. After a quench below the Curie temperature \( T_c \), the model has a spontaneous staggered magnetization in the variables \( \sigma_i \) in terms of statics. However, in the absence of a field that breaks the symmetry between the two ground states, no global magnetization develops for an infinite system and the system stays out of equilibrium. Domains of the two phases form and coarsen with a characteristic length scale \( \sqrt{T/\xi} \) as the evolution is via diffusion of the domain walls and coalescence of domains. The ergodic time \( t_{\text{erg}} \) for such a system scales with the linear size \( L \) of the system like \( L^2 \), and thus as long as \( t \ll t_{\text{erg}} \), the system is out of equilibrium and has no global magnetization. We have checked that for \( T = 0.6J \) and \( L = 400 \), the ergodic time is of the order of 4000. Since this regime is the only one accessible for very large samples, we will limit our discussion to that regime.
Another point worth noticing about the dynamics of the Ising model is that, after the short initial stage where spins with 3 or 4 parallel neighbors have disappeared, flips take place only at sites with two neighbors up and two neighbors down. For such sites, the effective field coming from the coupling to the neighbors is equal to zero, and we call them 0-sites. Since there is no energy cost to flip the spin $s_i$ of such a site, the probability to flip according to Glauber dynamics is $1/2$. So the flipping rate should be half the concentration of 0-sites. We have checked numerically that this is indeed the case after a few sweeps. For $T = 0.6J$, this is true as soon as $t > 40$.

Let us now turn to an analysis of the problem in the presence of a magnetic field. Let us forget the first few steps and concentrate on times where flips take place at 0-sites. If the magnetic field is small compared to $J$, a condition usually fulfilled in experiments, the dynamics of the Ising spins $s_i$ will be essentially unaffected, and the magnetic field will just favor configurations where the spins $S_i$ are parallel to the field $\mathbf{h}$.

Now let us suppose that a 0-site remains so for a while. The equation that governs the appearance of a magnetization for the $S$ spins can be easily deduced. If we denote by $p_+$ (resp. $p_-$) the probability for $S$ to be up (resp. down), Glauber dynamics implies that $p_+$ satisfies the equation

$$\frac{dp_+}{dt} = \frac{e^{\beta h}p_+ - e^{-\beta h}p_-}{e^{\beta h} + e^{-\beta h}}$$

(4)

The magnetization, which is related to $p_+$ and $p_-$ by $p_+ = (1 + m)/2$ and $p_- = (1 - m)/2$, is thus given by

$$m = \tanh(\beta h)(1 - \exp(-t/\tau))$$

(5)

where the relaxation time $\tau$ is equal to 1 in the chosen time units. This is a very short time, especially considering the very small flipping rate which is achieved after a few steps (already below $10^{-2}$ after 100 sweeps for $T = 0.6J$). Under these circumstances, the sites that have been 0-sites in the presence of the magnetic field should have enough time to reach equilibrium, and on average their magnetization should be equal to $\tanh(\beta h)$. So if we call $c_0(t_m, t_w)$ the proportion of sites that have been 0-sites before $t_w$ and $t_w + t_m$ this simple argument leads to the prediction that

$$m(t_m, t_w) = \tanh(\beta h)c_0(t_m, t_w)$$

(6)

Note that $c_0(t_m, t_w)$ depends only on the dynamics of the underlying Ising model and not on the magnetic field. To check this prediction, we have calculated $c_0(t_m, t_w)$ for different $t_w$ corresponding to our ZFC numerical experiments. The agreement is very good - the curves are indistinguishable from the ZFC calculations of $m$ on the scale of Fig. 4 - and we have checked that it remains so as long as $t_w$ is not too small so that the magnetization is indeed controlled by 0-sites. This analysis shows that the ageing behavior of the model of Eq. (1) is indeed closely related to the dynamics of the underlying Ising model.

To understand the magnetization process we therefore only have to consider $c_0(t_m, t_w)$ for the Ising model. After the first few steps, domains can be identified, and 0-sites are on the boundary of the domains. On a square lattice, they correspond to the diagonal portions of the boundary. Now, after a time $t_w$ the characteristic size of the domains is of order $\sqrt{t_w}$. Consequently the total number of domains is of order $1/\sqrt{t_w}$, and the total length of domain walls is proportional to $1/\sqrt{t_w}$. When $t_m \ll t_w$ we remark two important points: (i) the domain walls appear locally flat, the effects of surface tension and hence curvature are negligible and hence the domain walls diffuse (as zero modes are the only ones generating dynamics by this stage), a given point on a domain wall will therefore diffuse a length $\sqrt{t_m}$; (ii) one can neglect the interaction between domain walls, that is to say the coalescence of domains. The total number of spins which flip between $t_m$ and $t_m + t_w$ is therefore proportional to $\sqrt{t_m} \times$ total length of domain walls. We therefore find that at short times $t_m$ (compared to $t_w$) one has

$$c_0(t_m, t_w) = \text{Const.} \sqrt{t_m / t_w}$$

where the constant should be independent of $t_w$. This provides an excellent fit of the data at short times (see Fig. 4). We have checked that $\text{Const.} \approx 0.05$ at $T = 0.6J$ is indeed independent of $t_w$. This argument provides a very simple explanation of the fact that the larger $t_w$, the slower the initial increase in the magnetization.

FIG. 4. Fits of the FC and ZFC data at $T/J = 0.6$ with the help of Eqs. (7-9).

In the case where $t_w = 0$, the above argument cannot be applied as it is because of the initial steps, where flips occur at sites with 3 and 4 parallel neighbors as well, and because domains are not present right away. Still the same kind of reasoning suggest that every spin which has flipped has an average magnetization $\tanh(\beta h)$. Therefore one has $m(t, 0) = \tanh(\beta h)(1 - p(t, 0))$ where $p(t, 0)$ is the probability that a given spin has not flipped before time $t$. The quantity $p(t, 0)$ has received much attention in the literature and is called the probability of
Extensive numerical simulations have shown that for large times \( p(t) \sim 1/t^\theta \), where \( \theta \) is the persistence exponent which has been measured as \( \theta \approx 0.2 \) for the two dimensional Ising model. One therefore deduces that

\[
m(t,0) = \tanh(\beta h)(1 - (\tau/t)^\theta)
\]  

(8)

Here \( \tau \) is a microscopic time scale related to the flipping rate of the spins. This provides and excellent fit of the autocorrelation function up into units of size \( \sqrt{\tau} \). These blocks may be regarded as effective spins of the sign of the average block persistence introduced in \cite{21}. Moreover, the characteristic time scale \( \tau(t_w) \) for these effective spins is proportional to the time it takes to reverse the staggered magnetization of a given block, since this is done by diffusion one has \( \tau(t_w) \approx t_w \). One may now apply the reasoning of the case \( t_w = 0 \) to obtain

\[
m(t,t_w) = \tanh(\beta h)(1 - (\tau(t_w)/t)^\theta)
\]  

(9)

for \( t \gg t_w \). This again provides a reasonable fit of the data (see Fig. \ref{fig:figure1}), and we have checked that \( \tau(t_w) \) is proportional to \( t_w \).

To summarize, the very slow onset of magnetization in ZFC experiments follows scaling laws typical of ageing, and this is the origin of the spin-glass behavior. This ageing is a consequence of the very slow march towards equilibrium of the underlying model in which spins and pseudo-spins are coupled into an effective Ising spins since the physical field, which couples only to the spin and not to the pseudo-spin, does not act as a symmetry breaking field for that model.

\section{IV. DISCUSSION}

While the difference between FC and ZFC measurements of the magnetization is very typical of spin-glasses, a \textit{bona fide} spin-glass has many other characteristics which are not shared by the present model.

First of all, for an infinite system the freezing transition of a standard spin glass is believed to be a thermodynamic transition, and the freezing temperature is expected to be independent of the protocol. In the present case, in order to flip, a spin inside a domain must be able to cross an energy barrier of \( 8J \). The Arrhenius law gives therefore a characteristic flipping time of \( \tau_a \sim \exp(8J/T) \). Such activated flips are only observed after a time of measurement of order \( \tau_a \). One may therefore define a temperature depending on the time scale of the measurement \( T_g(t_m) \), such that if \( \tau_a \gg t_m \) such activated spin flips do not occur and hence the equilibration of the magnetization within domains is not possible. The crossover between the two regimes is therefore given by \( T_g(t_m)/J \sim 8/\ln(t_m) \). So, unlike the standard thermodynamic spin glass transition, the freezing temperature depends on \( t_m \). This estimate of \( T_g \) agrees with our Monte Carlo results. If one takes a measurement time of \( t_m = 500 \), as for the curves shown in Fig. \ref{fig:figure2}, one finds that \( T_g/J \sim 1.287 \), close to the value of \( T_g \) shown in Fig. \ref{fig:figure1}.

Another original aspect of our data with respect to standard spin glasses is the minimum of the ZFC magnetization at a temperature far below \( T_g \). To understand this, one may make an approximate decomposition of the magnetization in terms of the activated component \( m_a \) within the domains and the component generated by the domain walls \( m_d \). We find \( m_a(t_m,t_w) = \tanh(\beta h)(1 - \exp(-\alpha(h)t_m/\tau_a)) \) (solving the Glauber dynamics explicitly for a single spin with its four neighbors fixed parallel shows that \( \alpha = 1 + \cosh(2\beta h) + O(1/\tau_a^2) \)).

As \( m_a \) is generated within domains and hence in the bulk of the system (that is to say not at domain interfaces), it is very weakly dependent on \( t_w \). Another type of activated process may occur at domain interfaces where a spin with three neighbors antiparallel and one parallel flips. The energy barrier for this is \( 4J \). This gives another characteristic time \( \tau_a^* \) with a corresponding time dependent temperature \( T^*(t_m)/J \sim 4/\ln(t_m) \). For \( t_m = 500 \) as in Fig. \ref{fig:figure1} one finds \( T^*/J = 0.6436 \) which corresponds to the minimum seen on the ZFC curve. For \( T < T^* \), the system behaves as if \( J \) is infinite on the experimental time scale and only domain wall diffusion occurs, the only energy involved being the external field energy \( h \). Here the dynamics is well described by Eq. \ref{eq:5}, hence the measured magnetization \( m(t_m,t_w) \) increases as \( T \) is lowered below \( T^* \).

Both these effects are dynamic in nature and show that the behavior we have observed is typical of the out of equilibrium dynamics observed in spin-glasses. This conclusion is in fact consistent with another aspect of our data, namely the sensitivity of the results to the dynamics used in the simulation. To perform Monte Carlo simulations, we have made the assumption that spins and pseudo-spins flip independently of each other. However if one allows simultaneous flips as well, the behavior will be very different: These moves will allow the magnetization to develop inside the Ising domains, and the system will polarize after only a few sweeps. In other words, the breaking of ergodicity is related to the dynamics.

Another way to check how different the present model is from standard spin glasses is to study the temperature dependence of the non-linear susceptibility, which is expected to diverge at the transition in a spin-glass. Preliminary results \cite{22} indicate that the non-linear susceptibility is not singular in the present case. The absence of another thermodynamic singularity below the Curie temperature \( T_c \) is also clear from Eq. \ref{eq:5}.

However, other characteristic aspects of spin-glasses are also shared by the present model. For instance, the thermo-remanent magnetization \( m_{TMR} \) shows a very strong dependence on the time \( t_w \) elapsed between the
quench and the switching off of the magnetic field: The longer $t_w$, the slower the decay. A detailed analysis of these results along the same line is in progress.

Coming back to our original purpose, namely the explanation of the low-temperature behavior of Kagome antiferromagnets, the present model has both merits and drawbacks. The very clear difference between FC and ZFC magnetizations is the most interesting aspect of the results. It shows that the presence of an extra degree of freedom which is not coupled to the external field can indeed lead to a glassy behavior at low temperatures. However most of the differences with standard spin-glasses are problematic in that respect: No experimental indication of a dependence of $T_g$ on the protocol was reported, and the non-linear susceptibility is indeed enhanced close to $T_g$. This is not a final blow however. In fact, these differences with standard spin glasses all depend on the fact that flipping simultaneously a spin and a pseudo-spin leaves the Ising spin unchanged. This symmetry will not be present in more realistic models where spins and pseudo-spins are treated as Heisenberg variables, while the underlying mechanism for the difficulty that the system will have to magnetize after being cooled in zero-field is still expected to apply. More realistic models with two degrees of freedom treated as Heisenberg spins are therefore good candidates for effective models to get a spin-glass behavior in disorder free magnets. Work is in progress along these lines.

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