Spin waves in the classical Heisenberg antiferromagnet on the kagome lattice

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Abstract. We investigate spin waves in the classical Heisenberg antiferromagnet on the kagome lattice by means of Monte Carlo and spin dynamics simulations. Using sophisticated sampling algorithms we were able to establish thermal equilibrium at temperatures as low as $T = 10^{-6} |J|/k_B$, and from the dynamic structure factor we could precisely determine frequencies of spin waves. Results agree well with theoretical predictions for acoustic modes. Measurements in different coordinate systems as well as theoretical considerations showed that optical modes, which were apparently found in a recent study [1], are in fact artifacts and do not indicate the existence of additional spin wave modes.

1. Introduction

Although geometrically frustrated magnets are not easily investigated, they are subject of strong interest since their behavior is often both surprising and counter-intuitive. In these systems the spatial arrangement of spins prevents the simultaneous minimization of all interactions which usually leads to a degenerate ground state and/or glass-like behavior at low temperature. One example is the Heisenberg kagome antiferromagnet which has been investigated in a number of studies [2–5]. The classical Hamiltonian for Heisenberg model is given by

$$\mathcal{H} = -J \sum_{\langle ij \rangle} \mathbf{s}_i \cdot \mathbf{s}_j,$$

where the sum goes over nearest neighbor pairs $\langle ij \rangle$. The spins $\mathbf{s}_i$ are three-dimensional vectors of length unity and the interaction constant is negative ($J < 0$), i.e., two interacting spins have minimal energy if they are pointing in opposite directions. The lattice (Fig. 1) includes hexagons and triangles, which causes frustration, since antiparallel alignment of all spin pairs of a triangle is impossible. The ground state is highly degenerate without long-range order. However, it has been found that at low, but finite, temperatures ($T < 5 \times 10^{-3} |J|/k_B$) in a process called 'order by disorder' [6], coplanar configurations are selected due to their higher entropy. It has been shown analytically that this state gives rise to a degenerate acoustic branch and a dispersionless branch of soft modes. A recent numerical study [1] also seemed to suggest that a branch of nearly dispersionless optical modes exists as well.

The paper is organized as follows: In the next section we will give a short overview of the low temperature state of the system, which is followed in section 2 by a description of the methods we used. In section 3 and 4 we present results of Monte Carlo and spin dynamics simulations.
and in section 4 we will discuss how the apparent conflict between theory and simulation is resolved.

2. Coplanar state

It is easy to see that the energy of one spin triangle is minimal if the sum of the three spins is zero, i.e. if the three spins lie within one plane and pairwise include angles of $2\pi/3$. Applying this condition to every triangle in the lattice still leaves the system under-determined, hence, the ground state is highly degenerate. However, it turns out that the harmonic constraints for certain degrees of freedom vanish if all spins lie within the same plane. These degrees of freedom then provide additional 'soft' modes which strongly increase the system entropy, and such spin configurations are, therefore, favored at low finite temperatures. We will call these configurations 'coplanar' and the macrostate comprising them the 'coplanar state'. Since the Hamiltonian is symmetric under arbitrary rotations we are free to orient the system as necessary, and in the following we assume that the spin plane coincides with the $xy$-plane. In the coplanar state each spin fluctuates around one of only three possible basic directions [Fig.2(a)] and labels $\{\sigma\}$ can be assigned to each spin such that $\sigma_i = 1, 2, 3$ if $s_i$ points in the first, second or third direction. For neighboring spins $s_i, s_j$ which always point in different directions $\sigma_i \neq \sigma_j$. This means that $\{\sigma\}$ is also a ground state configuration of the three-state Potts model, although, in contrast to that model, due to the entropic effects the probability distribution over all $\{\sigma\}$ that fulfill this condition is non-uniform [2].

In order to understand these effects it is helpful to consider the so-called Weathervane loops. These are created by connecting adjacent spins which belong to two of the three different types [Fig.2(b)], i.e. neighboring spins $s_i, s_j$ are connected if $\sigma_i \neq \nu$ and $\sigma_j \neq \nu$, with $\nu = 1, 2$ or 3. Each loop is surrounded only by spins pointing in the third direction $\nu$; and its own spins can, therefore, rotate collectively around this direction with low energy cost. In consequence, fluctuations in these rotational degrees of freedom are large increasing the entropy of the coplanar state. It also explains the deviation from the ground state of the three state Potts model: Since additional loops provide additional entropy there is a bias towards configurations $\{\sigma\}$ with many loops. On the other hand, there are few configurations with high loop numbers. For example the so-called $\sqrt{3} \times \sqrt{3}$ state (Fig. 3) providing the maximum number of loops - one on every hexagon - is only six-fold degenerate. This means that there is a counter effect reducing entropy with
increasing loop number and in consequence, the system neither shows the uniform probability distribution of the Potts ground-state nor does it solely occupy the $\sqrt{3} \times \sqrt{3}$ configurations.

3. Method

We investigated systems of different sizes, although most of the data presented in the following were obtained from simulations of a system containing $N = 3888$ spins with periodic boundary conditions.

In order to perform spin dynamic simulations at low temperatures we first have to perform a Monte Carlo simulation that produces equilibrium configurations at that temperature. Since ergodicity is surely broken for Metropolis [7] simulations at these temperatures, the application of a generalized ensemble method is obligatory. For the classical Heisenberg antiferromagnet on the kagome lattice there is no phase transition involving latent heat [8] and it is not necessary to overcome barriers in energy. This gives us the freedom to rely on methods like simulated tempering [9] or parallel tempering [10] which are based on canonical ensembles. Canonical
Figure 4. (Color online) At low temperature larger changes to the configuration can still be achieved when using the loop-flip update which flips all spins in one Weathervane loop.

ensembles on the other hand allow for the application of the Heatbath algorithm [11, 12] for which single spin updates are always accepted. As a consequence, we do not have to worry about cone opening angles or acceptance rates at low \( T \). During the simulated or parallel tempering simulation the system is required to change its temperature frequently. These steps in temperature space can only be performed if there is a large enough overlap between the energy distributions of neighboring temperatures. From the previous knowledge of the specific heat [8] we can estimate the width of these distributions and we find that we need a few thousand canonical ensembles in order to cover many orders of magnitude in temperature. To keep the simulation simple and stable we chose the simulated tempering technique which requires a preliminary iteration for weight determination. The algorithm can, however, be performed entirely on a single processor core. We used 10000 temperatures such that \(-6 \leq \log_{10}(T_i k_B / |J|) \leq 3\) with constant intervals in the logarithmic temperature scale.

Although spin-freezing occurs only at extremely low temperatures (we still observe sporadic spin-flips at \( T < 10^{-5} |J| / k_B \)) the dynamics within the coplanar state is slow, especially changes in \( \{\sigma\} \) are rare. In order to reduce autocorrelation times we exploited the specific properties of the coplanar state to design a novel Monte Carlo move. In this update all spins of a particular Weathervane loop are flipped such that their directions \( \sigma \) are exchanged (Fig. 4). It turned out that simple \( \pi \) rotations around the direction of the neighboring spins result in low acceptance rates. Spins pointing above the spin plane become spins pointing below the plane and vice versa, which distorts the second loop to which each spin belongs. Instead the spins of the loop have to be reflected in a mirror plane that contains the neighboring spins and is simultaneously perpendicular to the spin plane. We find that such an update is accepted with a probability of roughly 10% independently of temperature. Thereby, most of the flipped loops are small and involve less than 30 spins; but sometimes loops of more than 100 spins are flipped as well.

In order to capture the dynamics of the system we integrated the coupled equations of motion for multiple configuration using a 4th order Adams-Bashforth predictor-Adams-Moulton corrector calculations [13] which were initialized with the Runge-Kutta method. The derived trajectories \( \{s(t)\} \) were used to obtain the space-time correlation function:

\[
C(\mathbf{r}, t) = \langle s(\mathbf{r'}, 0) \cdot s(\mathbf{r'} + \mathbf{r}, t) \rangle_{\mathbf{r'}},
\]

where \( \mathbf{r'} \) goes over the three points in one unit cell, i.e. one triangle of the lattice. A Fourier transform provides the dynamic structure factor

\[
S(\mathbf{k}, \omega) = \sum_{\mathbf{r}} \int dt \, C(\mathbf{r}, t) e^{-i\mathbf{r} \cdot \mathbf{k}} e^{-i\omega t},
\]
Figure 5. (Color online) Spins fluctuate around three basic directions (arrows). Angles \( \phi_{\sigma_i} \) are used to define alternative in-plane coordinate systems \( u_{\sigma}, v_{\sigma} \) (4), which allow to measure transversal and longitudinal fluctuations separately.

with the wave vector \( k \) and frequency \( \omega \). In the context of neutron scattering experiments \( S(k, \omega) \) corresponds to the scattering function, \( k \) to the momentum transfer, and \( \omega \) to energy.

In the transition to the coplanar state rotational symmetry is broken, hence, we cannot expect the in-plane components \( \{s^x(t)\}, \{s^y(t)\} \) and the out-of-plane components \( \{s^z(t)\} \) to show the same collective excitations. We, therefore, also calculate correlations \( C^x, C^y, C^z \) and dynamic structure factors \( S^x, S^y, S^z \) for single components only.

Considering the unique geometry of the coplanar state it is apparent that the system is poorly described within a single Cartesian coordinate system. While the out-of-plane excitation can well be measured in the \( z \)-direction, the in-plane behavior is inadequately reflected in the \( x \) and \( y \)-components (Fig. 5). If, for instance, an average is taken over spins which rotate collectively around the \( z \)-axis, the different contributions will add up to zero if an equal number of spins belongs to each of the three basic spin directions. It is, therefore, helpful to introduce in-plane coordinates that are parallel \( (s^u) \) and perpendicular \( (s^v) \) to the respective basic directions similarly to the coordinates used in [5]:

\[
\begin{pmatrix}
  s^u_i \\ s^v_i \\ s^z_i 
\end{pmatrix} =
\begin{pmatrix}
  \cos \phi_{\sigma_i} & \sin \phi_{\sigma_i} & 0 \\
  -\sin \phi_{\sigma_i} & \cos \phi_{\sigma_i} & 0 \\
  0 & 0 & 1 
\end{pmatrix}
\begin{pmatrix}
  s^x_i \\ s^y_i \\ 1 
\end{pmatrix} -
\begin{pmatrix}
  0 \\ 1 \\ 0 
\end{pmatrix},
\]

(4)

where \( \phi_{\sigma_i} \) denotes the angle between the \( y \)-axis and the basic direction of the \( i \)th spin, i.e. its equilibrium direction. These coordinates have two advantages. Once the spin-plane is aligned with the \( xy \)-plane (i.e. \( uv \)-planes) further orientation is not necessary, which means that \( \phi_{\sigma_i} \) don’t have to be adjusted, and the system is at minimal energy \( E_{\text{min}} = -N|J| \) if all \( (s^u, s^v, s^z)^T = 0 \). We also calculated correlations and dynamic structure factors \( S^u, S^v \) for these coordinates, although there is no direct link to experimentally accessible quantities.

4. Monte Carlo results

During the simulated tempering simulations we recorded the energy distribution and derived the density of states. Since the simulation covers nine orders of magnitude in temperature and since the investigated systems contain thousands of degrees of freedom, the density of states extends over ten-thousands of orders of magnitude. In return we use the density of states to
calculate the specific heat (Fig. 6) and obtain a relatively precise result considering the size of the system or the large temperature interval. In the high temperature region the system is driven by entropy, changes in temperature have little influence on the internal energy and the specific heat is, therefore, small. In the intermediate region spins on each triangle are correlated and energy is minimized, but the overall symmetry remains unbroken. At very low temperatures we observe a good agreement with the theoretical prediction: \( \lim_{T \to 0} C(T)/N = 11/12 \) [4] which indicates the existence of soft modes and proves that the system has settled in the coplanar state. It should be mentioned here that methods like simulated tempering allow a more direct access to thermodynamic quantities at the temperatures used in the simulation. The approach based on the density of states, however, provides data for all intermediate temperatures as well, while at the same time using all existing data for each data point. It is, therefore, more flexible and more precise.

To observe the formation of the coplanar state we also determined averages of absolute in-plane \( \langle |s^u| \rangle \) and out-of-plane \( \langle |s^z| \rangle \) components as functions of temperature (Fig. 6). It should be noted here that both are defined only in the oriented system, i.e. within a coordinate system with the \( xy \)-plane identical to the spin plane. Of course the system can only be oriented when in the coplanar state, since there is no spin plane otherwise, i.e., although there are finite values of \( \langle |s^u| \rangle \) and \( \langle |s^z| \rangle \) for \( T > 10^{-2}|J|/k_B \) these have no physical meaning, because the coplanar state does not exist at these temperatures. We find that in the coplanar state \( \langle |s^u| \rangle \) and \( \langle |s^z| \rangle \) show a power law behavior and, as expected, the in-plane excitations are much smaller and decrease faster than the out-of-plane excitations. This is not surprising, since the latter include the rotations of the Weathervane loops which are comparatively weakly constrained.

### 5. Spin dynamics results

In this section we will present data for structure factors \( S(k, \omega) \), with \( k_l = l k_{11} \), where the vector \( k_{11} \) is perpendicular to bonds of the lattice, i.e. \( k_{11} \parallel (a_1 + a_2) \) or \( k_{11} \parallel (2a_1 - a_2) \) or \( k_{11} \parallel (-a_1 + 2a_2) \). The length of \( k_{11} \) is chosen such that \( l = 1 \) at the edge of the Brillouin zone. From the analytical work [5] the dispersion relation for the acoustic branch is known, and we expect maxima in \( S(k_l, \omega) \) for

\[
\omega = 2 \sin(\pi l).
\]

All measurements were taken at \( T = 10^{-6}|J|/k_B \).

The results agree well with the aforementioned numerical study [1]. We, too, observe the apparent, novel optical mode (Fig. 7) and the acoustic branch. But, additionally we find that the acoustic branch only appears in the out-of-plane component \( S^z \), while the ‘optical branch’ manifests only in the in-plane component of the dynamic structure factor \( S^{xy} = S^x + S^y \) together with a number of dispersionless signals which form a washboard-like pattern. Furthermore, if we use the alternative coordinates \( u, v \) (Fig. 8), we observe the acoustic branch in \( S^u \) while no maxima are obtained in \( S^v \). This is consistent with the picture of spins precessing collectively around their equilibrium positions - the respective basic direction - while maintaining a constant angle with respect to it. We conclude that the features of \( S^{xy} \) are either artifacts that arise in the transition from \( s^u, s^v \) to \( s^x, s^y \) or, less likely, different unpredicted modes that leave no sign in \( S^{uv} \) while at the same time the acoustic modes are invisible in \( S^{xy} \).

### 6. Optical illusion

In this section we briefly present results of considerations that explain the features of \( S^{xy} \). Detailed calculations will soon be published elsewhere.

To understand which signals the acoustic modes cause in the in-plane dynamic structure factor \( S^{xy} \) we calculated how these modes shape the correlation function. We could show that for the \( \sqrt{3} \times \sqrt{3} \) state the correlations in \( \{ \sigma \} \) cause a shift in the Brillouin zone such that
Fig. 6. (Color online) Specific heat and averaged absolute deviations from ground state.

signals found in $S^{xy}(k, \omega)$ belong to modes with wave vectors $k \pm \pi a_1 / 3|a_1|^2$. Since we know the dispersion relation for the acoustic branch for the entire zone from analytical studies [5] we could calculate the resulting positions of maxima in $S^{xy}(k, \omega)$:

$$\omega = \sqrt{\frac{7}{2} + \cos(2\pi l)},$$

which is in good agreement with the 'optical' branch in Fig. 7.

The coplanar state, however, differs from the $\sqrt{3} \times \sqrt{3}$ state. The correlations are basically the $\sqrt{3} \times \sqrt{3}$ - correlations enveloped in a power law, which causes additional signals. The Fourier transform of the central peak leads to contributions of each mode in $S^u(k, \omega)$ to $S^{xy}(k', \omega)$ at each point $k'$. In consequence, regardless of wave vectors the frequency of every mode of the system can in principle be measured everywhere leading to the dispersionless wash-board-like pattern in $S^{xy}(k, \omega)$. In the thermodynamic limit the short-range $\sqrt{3} \times \sqrt{3}$ correlations will have no important influence and no maxima related to acoustic modes will be observed in $S^{xy}$. Next-nearest neighbor interactions on the other hand can stabilize the $\sqrt{3} \times \sqrt{3}$ structure [5] and in that case we expect the described effects to be detectable in experiments.

7. Conclusions

Combining various advanced simulation techniques we could sample the kagome Heisenberg antiferromagnet at temperatures much lower than previously reached. Thermodynamic and structural quantities were in good agreement with previous analytical and numerical studies.
Figure 7. (Color online) Rescaled in-plane component of the dynamic structure factor. The wave vector $\mathbf{k}_l = l\mathbf{k}_1$ with $l = 1$ at the end of the Brillouin zone. The dispersion relation, Eqn.(6), is drawn with a dotted line.

Spin dynamic simulations were used to obtain dynamic structure factors with a very good signal-noise ratio due to the low temperature. We found that appropriate coordinates must be chosen to directly observe the predicted acoustic modes within the spin plane. Analytical calculations showed that signals found when using global coordinates do not belong to a novel optical mode, but origin from a different part of the Brillouin zone and are displaced due to the special structure of the coplanar state.

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Figure 8. (Color online) Rescaled in-plane component of the local coordinate structure factor. Signals are one hundred times larger compared to Fig. 7. The dispersion relation is given by Eqn.(5).

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