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Fictitious excitations in the classical Heisenberg antiferromagnet on the kagome lattice

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Using an advanced Monte Carlo algorithm and high precision spin dynamics simulation, we investigated the dynamical behavior of the Heisenberg antiferromagnet on a kagome lattice at extremely low temperatures. We demonstrate that the detection and correct identification of propagating excitations depends crucially on the choice of coordinates and we show how modes are displaced in Fourier space if a single, global Cartesian coordinate system is chosen.

I. INTRODUCTION

Although frustrated magnetic systems have enjoyed the attention of the scientific community for many years, they remain fascinating research subjects and their rich behavior is far from being completely understood. If only classical interactions are considered, it is usually an easy task to find the ground state which is often degenerate. But once the systems are thermally excited, intriguing effects like vortices¹ or magnetic monopoles² can be observed.

Computational physics provides important tools for probing the behavior of these magnets. Furthermore, it bridges the gap between theory and experiment by testing theoretical prediction which are usually stated for idealized systems that do not exist in their pure form in nature and are, therefore, not accessible by experiments. On the other hand more realistic models are often too complex for rigorous analysis but are to some extent treatable in computer simulations which are becoming increasingly powerful due to improved algorithms and faster machines. Spin dynamics methods are valuable in this respect since they allow the determination of the dynamic structure factor which is closely related to results of inelastic neutron scattering experiments.

One system of great interest is the antiferromagnet on the kagome lattice (KHA FM). **Studies of quantum antiferromagnets helped pique interest in corresponding classical systems, see Ref. 3 and 4, and both experiments and numerical studies of quantum systems continue to this day^{5,6}. Twenty years ago an extensive theoretical description^{3,4,7} was developed for the classical Heisenberg model, and a number of numerical studies⁸⁻¹⁰ have been performed since. Predictions for dynamical behavior were, in part, confirmed by recently published results of spin dynamic simulations¹¹. However, a branch of unpredicted modes was found and its origin has not yet been clarified. This makes a closer look at the subject worthwhile.**

The paper is organized as follows: After this introduction we will introduce the model and review some established facts about its behavior. In section III, the methods used in our work for simulation and analysis are then presented. Section IV is devoted to the presentation and discussion of the results the Monte Carlo and spin dynamic simulations and followed by concluding remarks

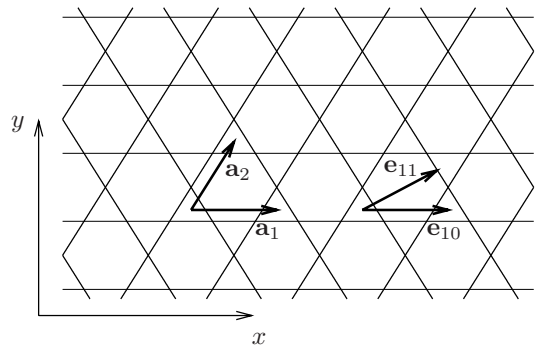


FIG. 1. The kagome lattice and the two lattice vectors \mathbf{a}_1 and \mathbf{a}_2 . Spins are placed at the line intersections. The $[10]$ and $[11]$ lattice directions are shown by (unit) vectors \mathbf{e}_{10} and \mathbf{e}_{11} .

in Section V. The paper concludes with two appendices in which we discuss some calculations in greater detail and introduce a novel modification of the Wang-Landau algorithm¹²⁻¹⁴.

II. BACKGROUND

In the classical Heisenberg model three dimensional vectors \mathbf{s} of unit length interact via the Hamiltonian

$$\mathcal{H} = -J \sum_{\langle ij \rangle} \mathbf{s}_i \cdot \mathbf{s}_j, \quad (1)$$

with a negative interaction constant J favoring antiferromagnetic spin pairs. The kagome lattice is drawn in Fig. 1. We apply periodic boundary conditions and assume in the following that the lattice constant $|\mathbf{a}_i| = 1$. If N is the number of sites, the system contains $2N/3$ triangles. The energy is minimal ($E_{\min} = -N|J|$) if the sum of the three spins belonging to each triangle is zero. This leaves the system underdetermined (frustrated) and the ground state is highly degenerate.

At low enough, but non-zero, temperatures a common spin plane is established, at least in finite systems, in a process called ‘order by disorder’⁹. This ‘coplanar’ state is stabilized by the entropy of local modes which can be constructed on the hexagons of the kagome lattice. Only three distinct spin directions occur separated by angles of

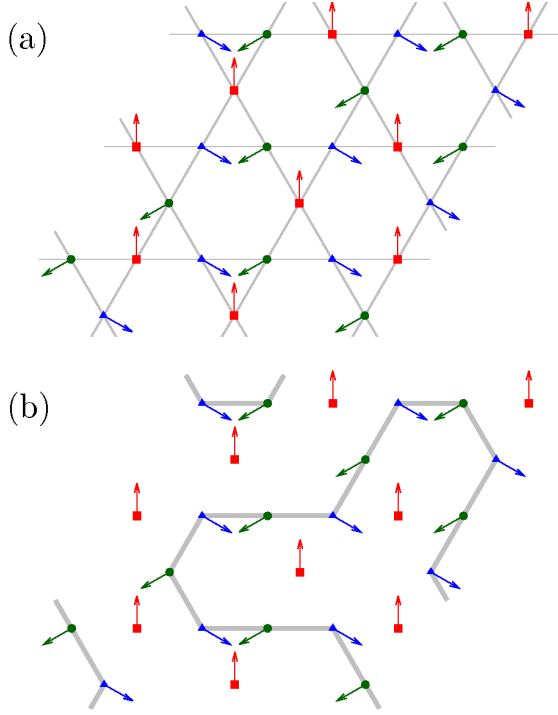


FIG. 2. (a) Typical configuration at low temperatures. Spins fluctuate around three distinct directions which are indicated by different symbols and colors. (b) Solid lines show *weathervane* loops which are constructed if spins of two 'types' are connected.

$2\pi/3$ which allows the assignment of labels $\sigma_i \in \{1, 2, 3\}$ to each spin \mathbf{s}_i according to the direction towards which it is oriented.

It is then possible to classify configurations $\{\mathbf{s}\}$ according to their labels $\{\sigma\}$. The ground state condition requires that each triangle has spins with three different labels; hence, valid $\{\sigma\}$ are also ground state configurations of a three state Potts model. However, while the probability distribution over all $\{\sigma\}$ is uniform in the case of the Potts model, there is a selection in the KHAFM due to entropic effects. This can be understood if the concept of *weathervane* loops is employed. These loops form when two label values are chosen and spins possessing these values are connected along the bonds of the lattice (Fig. 2[b]). Since there are three ways of choosing two out of the three possible values, three overlapping loop structures coexist and each spin belongs to two loops. However, within a single structure loops never touch but are separated by spins pointing in the third direction. Therefore, spins of a single loop can rotate collectively around this direction *while causing only small changes to spin-spin angles and, thus, to energy*. However, unlike in the case of a true weathervane complete rotations rarely occur. Instead, relatively large fluctuations in these collective degrees of freedom occur and account for the difference between the ground state of the Potts model and the distribution over different $\{\sigma\}$

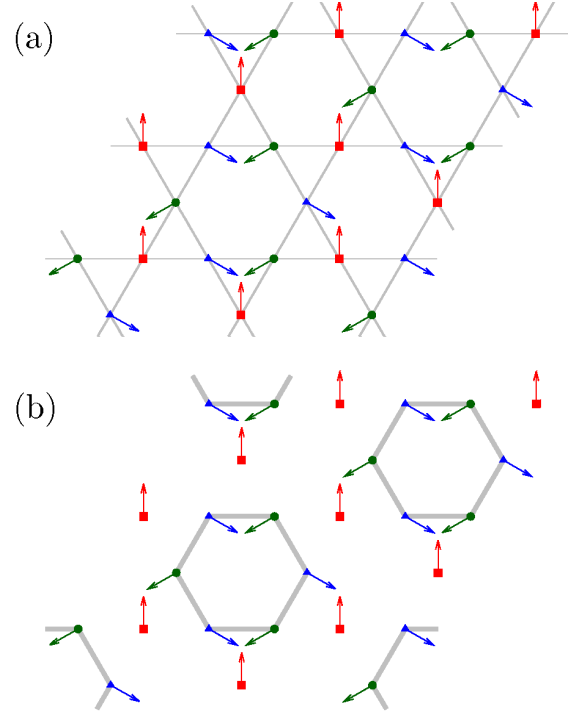


FIG. 3. (a) The $\sqrt{3} \times \sqrt{3}$ structure; (b) *weathervane* loops for the $\sqrt{3} \times \sqrt{3}$ structure are shown by solid lines.

in the KHAFM. Since each loop provides one degree of freedom for fluctuations, a structure with many loops has a higher entropy than a structure with few loops. In a counter effect, however, entropy decreases if the loop number becomes very large; and the maximum loop number does not occur in large enough systems, since it is realized in only six distinct configurations $\{\sigma\}$ possessing the so-called $\sqrt{3} \times \sqrt{3}$ structure (Fig. 3). In the $\sqrt{3} \times \sqrt{3}$ state each hexagon is a *weathervane* loop and the loop structure is periodic in three directions with *the six* wave vectors $\mathbf{k}_{\sqrt{3} \times \sqrt{3}} = \pm \frac{8}{3}\pi \mathbf{a}_1, \pm \frac{8}{3}\pi \mathbf{a}_2, \pm \frac{8}{3}\pi(\mathbf{a}_1 - \mathbf{a}_2)$.

In an analytical study⁴ Harris et al. examined the KHAFM with and without second and third nearest neighbor interactions and predicted a branch of soft modes with frequencies approaching zero as the temperature decreases as well as a two-fold degenerate acoustic branch. In the case of only nearest neighbor interactions the frequencies of the acoustic branch were given by

$$\omega(\mathbf{k}) = |J| \sqrt{2 (\sin^2 q_1 + \sin^2 q_2 + \sin^2(q_1 - q_2))}, \quad (2)$$

where $q_1 = k_x$, $q_2 = (k_x - \sqrt{3}k_y)/2$, and \mathbf{k} is the wave vector.

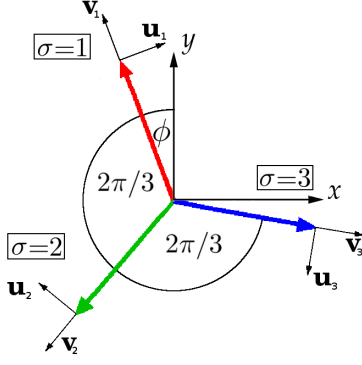


FIG. 4. With decreasing temperature spins tend towards three basic directions and labels, σ , can be assigned to each spin accordingly. Fluctuations around these directions are described in the alternative coordinates u, v given in Eqn. 4.

III. METHODS

A. Alternative Coordinates

Since no anisotropy and no external field are considered, the Hamiltonian has spherical symmetry in spin space and the system can be freely rotated. In the following we will choose the orientation in such a way that the spin plane coincides with the xy -plane, and we name the angle between the average direction of the $\sigma = 1$ spins and the y -axis ϕ . Alternative in-plane coordinates s^u, s^v, s^z as used by Harris et al.⁴ are now given by

$$\begin{pmatrix} s_i^u \\ s_i^v \\ s_i^z \end{pmatrix} = \begin{pmatrix} \cos \psi_i & \sin \psi_i & 0 \\ -\sin \psi_i & \cos \psi_i & 0 \\ 0 & 0 & 1 \end{pmatrix} \begin{pmatrix} s_i^x \\ s_i^y \\ s_i^z \end{pmatrix}, \quad (3)$$

where $\psi_i = \phi + 2\pi/3(\sigma_i - 1)$ is the angle enclosed by the y -axis and the basic spin direction is denoted by σ_i . The new u -axis is perpendicular and the v -axis is parallel to this direction (Fig. 4).

B. Monte Carlo simulations

We used the simulated tempering¹⁵ technique which allowed us to cover a large temperature range. It provides a generalized ensemble that is a composition of N_T canonical ensembles with a priori defined temperatures T_i . The probability to find the system in a configuration $\{\mathbf{s}\}$ at temperature T_i is

$$p_i(\{\mathbf{s}\}) = \frac{W_i}{Z_{\text{ST}}} \exp\left(-\frac{\mathcal{H}(\{\mathbf{s}\})}{k_B T_i}\right), \quad (4)$$

where W_i is a weight factor and the normalization constant Z_{ST} is the partition sum of the entire ensemble

$$Z_{\text{ST}} = \sum_{i=1}^{N_T} W_i Z(T_i) \quad (5)$$

which is here written as a weighted sum of canonical partition sums Z . The acceptance probability for a Monte Carlo step from configuration $\{\mathbf{s}\}$ to configuration $\{\mathbf{s}'\}$ and temperature T_i to temperature T_j is given by

$$P_{ij}^{\text{acc}}(\{\mathbf{s}\}, \{\mathbf{s}'\}) = \min\left(1, \frac{W_j}{W_i} \exp\left(\frac{\mathcal{H}(\{\mathbf{s}\})}{k_B T_i} - \frac{\mathcal{H}(\{\mathbf{s}'\})}{k_B T_j}\right)\right) \quad (6)$$

It follows from Eqn. (6) that conformational updates can be performed just like in a Metropolis simulation as long as the temperature remains unchanged. Usually, in order to move in temperature space, separate Monte Carlo steps which do not alter the configuration of the system are employed. To ensure that all temperatures are sampled with equal probability the weights have to be chosen such that $W_i \propto Z(T_i)^{-1}$. Since the partition function is not known at the beginning, we performed preliminary simulations to determine W_i using a modification of the Wang-Landau algorithm that is described in Appendix A. For our simulation we used $N_T = 10000$ different temperatures distributed equidistantly on a logarithmic temperature scale: $-6 \leq \log_{10}(k_B T_i / |J|) \leq 3$. In order to update configurations at constant temperature, we mainly used the heatbath¹⁶ method; and, in doing so, we avoided the problem of too low or too high acceptance rates as result of improperly chosen step length. Details of the heatbath technique for a Heisenberg spin system have been published by Miyatake et al.¹⁷.

Obtaining thermodynamic quantities from simulated tempering simulations is relatively simple. Although the reweighted histogram technique¹⁸ could be applied, it is not required here. It follows from Eqn. (4) that the distribution $P(E)$ can be written in terms of a density of states $g(E)$:

$$P(E) \propto g(E) \sum_{i=1}^{N_T} W_i \exp\left(-\frac{E}{k_B T_i}\right). \quad (7)$$

Once the weight factors are chosen it is easy to calculate the sum on the right hand side, and the density of states can be estimated via a histogram over E .

As described above the coplanar state is highly degenerate. Different configurations $\{\sigma\}$ exist in great number and, although requiring no increase in energy, transitions between them are inhibited by entropic bottlenecks. Hence, autocorrelation times are large in the coplanar state and ergodicity might even be broken at very low temperatures. This is partially corrected for by use of a generalized ensemble method, like simulated tempering in our case; but further improvement is possible if jumps between different configurations σ are enabled at low T . To do this we introduce a new Monte Carlo step

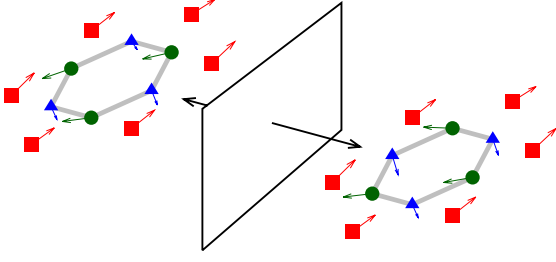


FIG. 5. Spins of a **weathervane** loop can be updated collectively in a reflection. The boundary spins (red) remain unchanged. The mirror plane (rectangle) contains the average direction of the boundary spins and is perpendicular to the (local) spin plane.

that is designed to “flip” all spins along a **weathervane** loop. Here, “flipping” means exchanging spin directions σ that already occur in the loop, i.e., if a **weathervane** loop is composed of spins with numbers $l_i, i \leq N_{\text{loop}}$ and $\sigma_{l_i} = 1$ or $\sigma_{l_i} = 2$ for all spins in the loop then in the modified loop it is still $\sigma'_i = 1$ or $\sigma'_i = 2$ for all i but at exchanged positions $\sigma_i \neq \sigma'_i$ (Fig.5). A local reference frame is established with the use of the loop spins and the neighboring (boundary) spins b_i for which in this example $\sigma_{b_i} = 3$.

The first step is the identification of a **weathervane** loop. While this can be done on a global scale by determination of the configuration $\{\sigma\}$, we used a more flexible procedure that, in principle, can result in accepted updates even if the system is in a disordered state. In the beginning, one spin is randomly chosen as the first spin of the loop s_{l_1} and one of its neighbors is - again randomly - drawn as the second s_{l_2} . Their common neighbor is recognized as a boundary spin s_{b_1} which is not, and cannot be, part of the loop. To continue the construction of the loop the following procedure is repeated until the loop is closed: Assuming that n spins are part of the incomplete loop, then one neighbor of s_{l_n} is part of the loop ($s_{l_{n+1}}$) and one neighbor is the boundary spin $s_{b_{n+1}}$. The next spin ($s_{l_{n+1}}$) must, thus, be chosen from the two remaining neighbors s_{m_1} and s_{m_2} which are neighbors themselves. Given the nature of a **weathervane** loop, this spin has to be oriented in approximately the same direction as $s_{l_{n-1}}$ so $l_{n+1} = m_1$ if $s_{l_{n-1}} \cdot s_{m_1} > s_{l_{n-1}} \cdot s_{m_2}$ and $l_{n+1} = m_2$ otherwise. The spin that is not chosen is consequently the boundary spin s_{b_n} . If during this procedure a spin is included that is already a boundary spin or part of the loop but not s_{l_1} , the procedure has failed and the Monte Carlo move is rejected.

There are two intuitive possibilities to update the loop. The first one is a rotation of the spins of the loop in spin-space by an angle of π around the direction given by the boundary spins. This, however, also alters the orientation of the spins with respect to the spin plane. A spin that is pointing above the plane will afterwards point below it and vice versa. This is not desirable since each spin also belongs to a second loop, and the out-of plane

component of the spin coordinates is mainly determined by the collective excitation of each loop. Hence, a switch in this component distorts the second loop and increases the energy. One has to choose the alternative which can be described as a reflection on a plane spanned by the direction of the boundary spins and the normal vector on the spin plane leaving the out-of plane component untouched.

If a closed loop could be constructed, then two vectors $\mathbf{v}_b, \mathbf{v}_l$ spanning the local spin plane could easily be found. The first one is the sum of all boundary spins

$$\mathbf{v}_b = \sum_{i=1}^{N_{\text{loop}}} \mathbf{s}_{b_i} \quad (8)$$

and a roughly perpendicular second vector is created by alternating the addition and subtraction of the loop spins

$$\mathbf{v}_l = \sum_{i=1}^{N_{\text{loop}}/2} \mathbf{s}_{l_{2i}} - \mathbf{s}_{l_{2i-1}}. \quad (9)$$

A normal vector of the plane on which the spins are reflected is then given by

$$\mathbf{u} = \mathbf{v}_b \times (\mathbf{v}_b \times \mathbf{v}_l), \quad (10)$$

and the updated spins are

$$\mathbf{s}'_{l_i} = \mathbf{s}_{l_i} - 2\mathbf{u} \frac{\mathbf{s}_{l_i} \cdot \mathbf{u}}{|\mathbf{u}|^2}. \quad (11)$$

After each loop flip we must test if the above procedure would also identify the flipped loop. If this is not the case, the inverse move is impossible and the update has to be rejected. The spin plane and the mirror plane will always be identified correctly in the inverse update. Finally, the change in energy has to be determined, and the move is accepted with the probability given in Eqn.(6).

It turns out that in the coplanar state the acceptance rates for this update are independent of temperature and decrease exponentially with increasing loop length. This suggests that the average change in energy is positive and proportional to the loop length.

C. Spin dynamics simulations

Employing the described Monte Carlo algorithm we extracted configuration $\{\mathbf{s}\}$ from canonical ensembles at temperatures T_i . The coupled equations of motion

$$\dot{\mathbf{s}}_i(t) = J\mathbf{H}_i(t) \times \mathbf{s}_i(t) \quad (12)$$

were then integrated over a time interval of $t = 2000|J|^{-1}$ using a 4th order Adams-Bashforth predictor- Adams-Moulton corrector method¹⁹ with a time step of $\Delta t = 10^{-4}|J|^{-1}$. Here, the local magnetic field H_i is the sum of the spins adjacent to \mathbf{s}_i . The resulting trajectory is subsequently used to calculate space displaced, time displaced

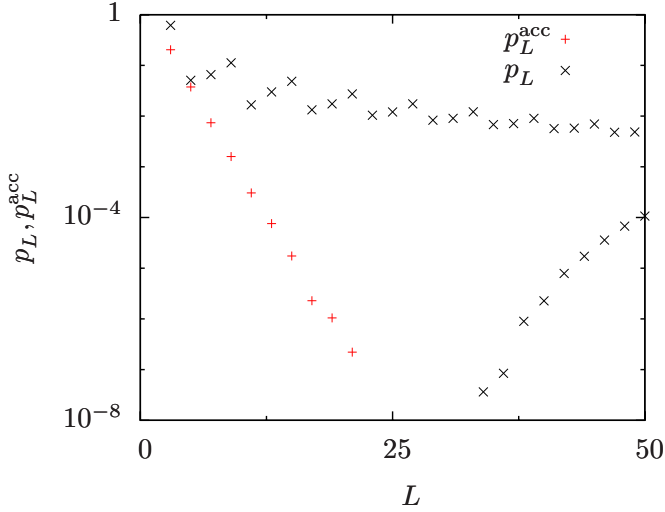


FIG. 6. Probability p_L that a constructed loop is of length L , and probability p_L^{acc} that an attempted flip of a loop of length L is accepted. Data in the lower right represent loops that span the entire system which for this case ($24^2 \times 3$ spins) requires $L \geq 24$. (Note that the number of spins in a loop N_{loop} is twice its length L in units of $|\mathbf{a}_i|$.)

correlation functions, e.g., correlations of the spin's x -component:

$$C_{\mathbf{r}}^x(t) = \langle s_{\mathbf{r}'}^x(0) \cdot s_{\mathbf{r}'+\mathbf{r}}^x(t) \rangle_{\mathbf{r}'}, \quad (13)$$

where \mathbf{r}' runs over all lattice sites. Finally, a space-time Fourier transform results in the dynamic structure factor

$$S^x(\mathbf{k}, \omega) = \sum_{\mathbf{r}} \int dt C_{\mathbf{r}}^x(t) e^{-i\mathbf{k} \cdot \mathbf{r}} e^{-i\omega t} \quad (14)$$

(S^y, S^z similarly) which in turn is closely related to outcomes of inelastic neutron scattering experiments, if global coordinates (x, y, z) are used. For the KHAFM the more genuine description, however, is given by the alternative coordinates (u, v, z) and we also calculated the respective structure factors (S^u, S^v).

Spin dynamics methods have proven to be valuable for the study of diverse dynamic excitations, e.g., spin-waves²⁰, vortices¹, and solitons²¹.

IV. RESULTS AND DISCUSSION

A. Static Properties

As a first result we obtained the density of states $g(E)$ and calculated the specific heat (Fig. 7). At low T on each of the $N/3$ hexagons a local soft mode can be constructed. These modes experience anharmonic potentials of fourth order with a thermal energy of $\frac{1}{4}k_B T$. All other degrees of freedom contain $\frac{1}{2}k_B T$ leading to a total average energy of $\langle E \rangle / N|J| = \frac{11}{12}k_B T - 1$ at small T . As in previous

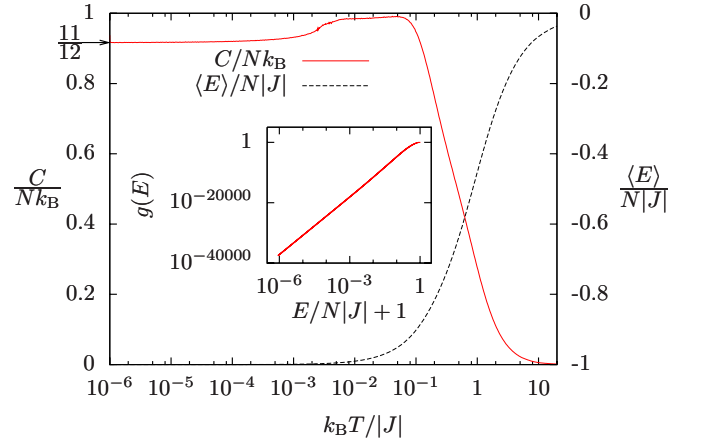


FIG. 7. Specific heat C/N , mean energy $\langle E \rangle / N$, and density of states $g(E)$ for a system containing $N = 6912$ ($= 48^2 \times 3$) spins. Statistical errors do not exceed the line width.

studies^{7,9,10} this theoretical prediction characterizing the coplanar state is fulfilled for $T \lesssim 3 \cdot 10^{-3} |J| / k_B$.

At intermediate temperatures $C/Nk_B \approx 1$ and low E indicate that only short range correlations exist and that spherical symmetry is not yet broken in favor of a common spin plane.

Note the linear shape of the density of states in the double-logarithmic plot in the inset of Fig. 7. Although of minor relevance here, this behavior is a general feature of classical systems corresponding to a finite limit of the specific heat for $T \rightarrow 0$. This can be shown easily using the following Ansatz for the density of states:

$$\log \frac{g(E)}{G} = \gamma \log ((E - E_0) / |J|), \quad (15)$$

where E_0 is the ground-state energy $E_0 = -N|J|$ and G a normalization constant. One finds that the specific heat $C = \gamma k_B$ and the mean energy $\langle E \rangle = E_0 + \gamma k_B T$. A fit of the data in Fig. 7 for $\log_{10} k_B T / |J| < -5.5$ gives $\gamma = 6335.9(2)$ which is close to the value $N \cdot 11/12 = 6336$ which we expect for $T \rightarrow 0$.

From simulation and theory^{4,8,9} it is known that the probability distribution over different configurations $\{\sigma\}$ is non-uniform and biased towards the $\sqrt{3} \times \sqrt{3}$ state. It has been under discussion whether this distribution changes with temperature and if long-range order is established for $T \rightarrow 0$. To investigate this possibility and to test the algorithm, we produced thousands of conformations of a 432 spin system at different temperatures, cooled them below $T = 10^{-5} |J| / k_B$, and performed simulated tempering simulations without loop flip updates at $10^{-6} \leq k_B T / |J| \leq 10^{-5}$. Due to the low temperature in more than half of the simulations no loop flips occurred spontaneously which allowed the comparison of average logarithmic temperatures of single configurations $\{\sigma\}$. If certain configurations are favored at low temperatures, then this would result in values closer to the lower

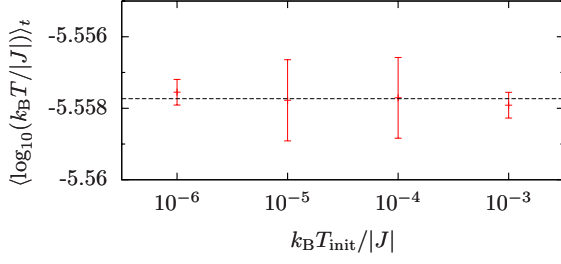


FIG. 8. If the simulation is constrained to $\log_{10}(k_B T / |J|) \leq -5$ the time-averaged logarithmic temperatures are independent of the original temperatures T_{init} . The broken line is a guide for the eye.

bound while configurations taken at higher T would consequently show higher average logarithmic temperatures. The results are shown in Fig. 8, and within the precision achieved no evidence for such a trend can be found. The exact values have no physical meaning; with perfectly tuned weights W_i the distribution over the logarithmic temperature would be flat and the averages would converge to 5.5.

We conclude that for $\log_{10}(k_B T / |J|) \leq -3$ the ensemble of loop structures is practically independent of temperature. Consequently, no further fundamental order is established and changes in energy, sub-lattice magnetization, or other order parameters¹⁰ are based solely on decreasing excitations on a fixed set of ground-state configurations.

It should be noted, however, that Henley²² recently studied effective Hamiltonians and suggested that long range order may exist for $T \rightarrow 0$ at length scales that exceed the size of the systems we could investigate and which could, therefore, be neither confirmed nor excluded by our simulations.

It is instructive to investigate the static correlations between labels σ which we define as:

$$\Gamma_{\mathbf{r}_i - \mathbf{r}_j} = \langle \delta_{\sigma_i, \sigma_j} \rangle - \frac{1}{3}. \quad (16)$$

This function bears no noticeable temperature dependence as long as the system is in the coplanar state. This confirms once more that the distribution over the loop structures does not change with T . Yet, $\Gamma_{\mathbf{r}}$ is best measured at lower temperatures where $\{\sigma\}$ can be determined most easily. As previous studies observed, we find that the systems show $\sqrt{3} \times \sqrt{3}$ correlations which decrease with increasing distance. The data are well described by a power law:

$$\Gamma_{\mathbf{r}} = \Gamma_{\mathbf{r}}^{\sqrt{3} \times \sqrt{3}} |\mathbf{r}|^{-\kappa}, \quad (17)$$

where

$$\Gamma_{\mathbf{r}}^{\sqrt{3} \times \sqrt{3}} = \frac{2}{3} \cos(\mathbf{r} \mathbf{k}_{\sqrt{3} \times \sqrt{3}}). \quad (18)$$

In order to estimate the exponent κ we fitted a power-law

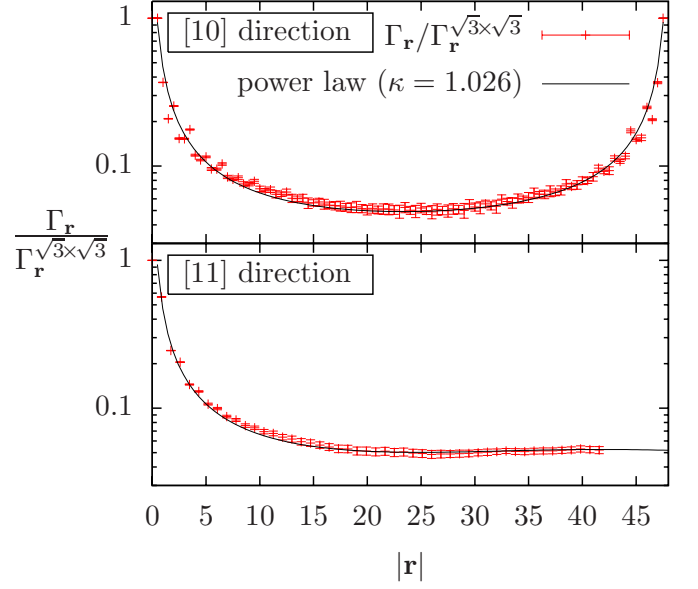


FIG. 9. Static correlations $\Gamma_{\mathbf{r}}$ for a $48^2 \times 3$ lattice follow a decaying $\Gamma_{\mathbf{r}}^{\sqrt{3} \times \sqrt{3}}$ pattern if the system occupies the coplanar state. The solid lines show a power law $\propto |\mathbf{r}|^{-1.026}$ with included periodic boundary conditions. (In [11] direction $\Gamma_{\mathbf{r}}$ has a period of $\sqrt{3} \cdot 48 \approx 83.1$)

function G with included periodic boundary conditions

$$G(\mathbf{r}) = G_0 \sum_i \sum_j |\mathbf{r} - i\mathbf{L}\mathbf{a}_1 - j\mathbf{L}\mathbf{a}_2|^{-\kappa} \quad (19)$$

to $\Gamma_{\mathbf{r}} / \Gamma_{\mathbf{r}}^{\sqrt{3} \times \sqrt{3}}$ and obtained $\kappa \approx 1$. Some of the relevant data and the fit for a system with $L = 48$ is shown in Fig. 9.

B. Dynamic properties

In our investigation we consider solely wave vectors parallel and perpendicular to the bonds of the system which simplifies Eqn. (2) to

$$\omega(\mathbf{l}\mathbf{e}_{10}) = |J| \sqrt{3 - 2 \cos l - \cos 2l}, \quad (20)$$

with the unit vector $\mathbf{e}_{10} = \mathbf{a}_1, \mathbf{a}_2, \mathbf{a}_1 - \mathbf{a}_2$ and

$$\omega(\mathbf{l}\mathbf{e}_{11}) = 2|J| \left| \sin \left(\sqrt{3}l/2 \right) \right|, \quad (21)$$

with $\mathbf{e}_{11} = \frac{\mathbf{a}_1 + \mathbf{a}_2}{|\mathbf{a}_1 + \mathbf{a}_2|}, \frac{2\mathbf{a}_1 - \mathbf{a}_2}{|2\mathbf{a}_1 - \mathbf{a}_2|}, \frac{-\mathbf{a}_1 + 2\mathbf{a}_2}{|-\mathbf{a}_1 + 2\mathbf{a}_2|}$ (Fig. 1).

All data presented in this section are from simulations of a system containing $48^2 \times 3$ spins at $T = 10^{-6}|J|/k_B$. We produced 2457 independent configurations and performed spin dynamics simulations of length $t_{SD} = 2000|J|^{-1}$ with a step length of $\Delta t = 10^{-4}|J|^{-1}$. Cauchy-distributions were fitted to the maxima and the Jackknife method was employed to estimate statistical errors in peak positions. While we will focus on the modes

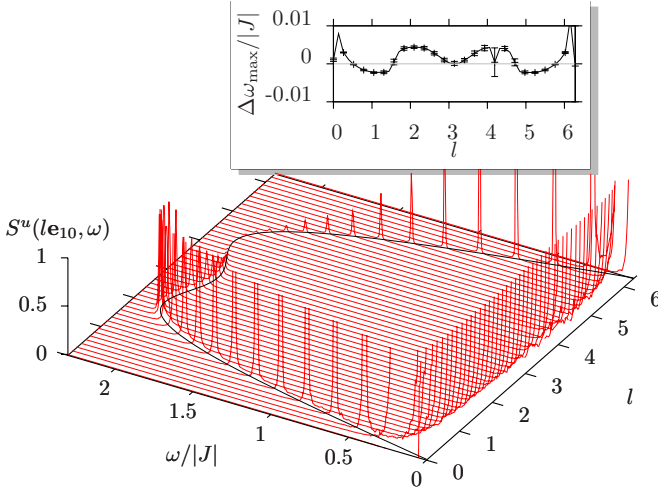


FIG. 10. Dynamic structure factor in the (10) direction based on the alternative in-plane coordinate u . Maxima agree well with the dispersion relation from Eqn. (20) which is plotted as a line in $l\omega$ -plane. Due to the low temperature systematic deviations (inset) are very small. For the sake of convenience, we increased S by a factor of 10^3 here and in Figs. 11, 12, and 13.

of the acoustic branch, we acknowledge that prominent signals at $\omega = 0$ give strong evidence for soft modes.

We also obtained results for smaller lattices and observed the systematic variation in the dynamic structure factor with lattice size. The results for smaller lattices were useful for the eventual interpretation of the large lattice data but do not themselves show different phenomena. For this reason we do not show those data here.

We find that the results for alternative coordinates u, v, z agree very well with the predictions. In both direction the soft modes produce prominent peaks at very low frequencies. While no trace of acoustic modes can be found in S^v prominent maxima exist in S^u (Figs. 10, 11) and S^z at the frequencies given by Eqns. (20, 21).

The in-plane dynamic structure factor in global coordinates $S^{xy} = S^x + S^y$, however, shows a complete different behavior. In the (10) direction (Fig. 12[a]) arcs of maxima intersect, but no excitations of comparable intensity occur at the frequencies of the acoustic branch according to Eqn. (20). In the (11) direction an almost dispersionless branch of maxima with high frequencies resembles an optical mode and was, in fact, interpreted as such by Robert et al.¹¹.

Additionally, numerous ridges of low intensity form washboard-like patterns in S^{xy} in both directions, but these vanish if a pure $\sqrt{3} \times \sqrt{3}$ structure is considered (Fig. 12[b]). This washboard pattern is pronounced for small lattices but washes out as the lattice size increases.

To understand this behavior we need to know the relationships between the in-plane correlation functions in global and alternative coordinates. In Appendix B we

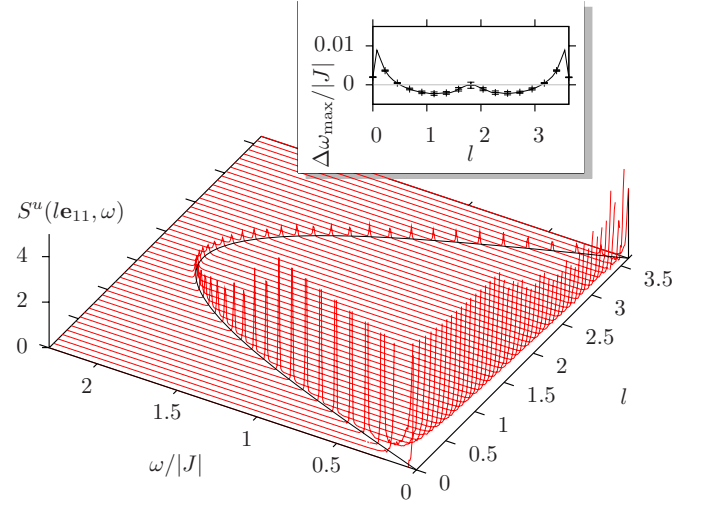


FIG. 11. Dynamic structure factor for the alternative in-plane coordinate u in the 11-direction. The behavior is well described by the dispersion relation in Eqn. (21) (solid curve). Deviations of peak positions are small (inset).

show that

$$C_{\mathbf{r}}^{xy}(t) \propto \Gamma_{\mathbf{r}} C_{\mathbf{r}}^u(t). \quad (22)$$

It follows that $C_{\mathbf{r}}^{xy}(t)$ and $S^{xy}(\mathbf{k}, \omega)$ depend on the underlying loop structure $\{\sigma\}$ even if $C_{\mathbf{r}}^u(t)$ does not. For the $\sqrt{3} \times \sqrt{3}$ structure the Fourier transform of $\Gamma_{\mathbf{r}}^{\sqrt{3} \times \sqrt{3}}$ is

$$\hat{\Gamma}^{\sqrt{3} \times \sqrt{3}}(\mathbf{k}) \propto \sum_{i=1}^6 \delta(\mathbf{k} - \mathbf{k}_{\sqrt{3} \times \sqrt{3}}^i), \quad (23)$$

where δ is Dirac's delta. Using the convolution theorem we find

$$S^{xy}(\mathbf{k}, \omega) \propto \sum_{i=1}^6 S^u(\mathbf{k} - \mathbf{k}_{\sqrt{3} \times \sqrt{3}}^i, \omega). \quad (24)$$

This means that modes seen in S^u contribute **six-fold** to S^{xy} as fictitious modes displaced by $\mathbf{k}_{\sqrt{3} \times \sqrt{3}}^i$ as shown in Figs. 14 and 15. The particular choice of $\mathbf{k}_{\sqrt{3} \times \sqrt{3}}^i$ is irrelevant **unless intensities are considered**. Due to the periodicity of ω each of the three directions will cause the same apparent branches by shifting the center of the Brillouin zone onto a corner and the opposite corner into the center. In the (10) directions this results in acoustic branches which follow the same dispersion relation as Eqn. (20) but shifted by $\pm 2\pi/3$.

In the (11) direction the apparent optical mode turns out to be a **part of the acoustic branch** and the dispersion relation (for the shifted positions) reads

$$\begin{aligned} \tilde{\omega}(l e_{11}) &= \omega(l e_{11} \pm \mathbf{k}_{\sqrt{3} \times \sqrt{3}}), \\ &= \sqrt{\frac{7}{2} + \cos(\sqrt{3}l)}. \end{aligned} \quad (25)$$

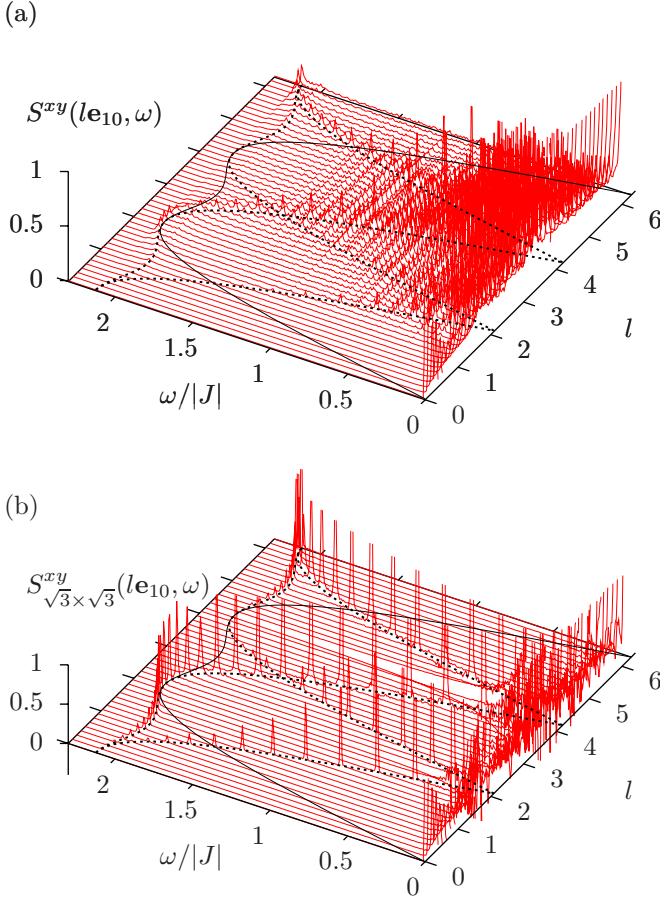


FIG. 12. Dynamic structure factor in the (10) direction based on global in-plane coordinates. A shift in Fourier space occurs (see text) and no signals matching the analytical dispersion relation (continuous lines) are observed. In the canonical ensemble (a) a washboard-like pattern is caused by the power law-like decay of the $\sqrt{3} \times \sqrt{3}$ correlations; (b) In case of long-range correlations, e.g. for the pure $\sqrt{3} \times \sqrt{3}$ structure, this pattern vanishes.

The described behavior is induced by the $\sqrt{3} \times \sqrt{3}$ correlations which in the general case of free $\{\sigma\}$ are present but appear to decay in a power law (Fig. 9). This means that we have to apply the convolution theorem a second time to describe the shifts k -space if an ensemble of typical loop structures (not pure $\sqrt{3} \times \sqrt{3}$) is considered. Since the Fourier transform of the power law is again a power law it follows that in principle each mode can be measured at each point in Fourier space with maximal intensity at the position given by the initial shifts $\pm \mathbf{k}_{\sqrt{3} \times \sqrt{3}}$. This effect causes the washboard pattern by smearing the peaks in k -space; however, as mentioned earlier, the shifted peaks can only be observed for finite systems. We expect no signs of acoustic modes in $S^{xy}(\mathbf{k}, \omega)$ in the thermodynamic limit. If, on the other hand, $\sqrt{3} \times \sqrt{3}$ correlations are stabilized by second and third nearest neighbor interactions⁴ the shifted acoustic branch without the washboard **would be detectable in experiments**

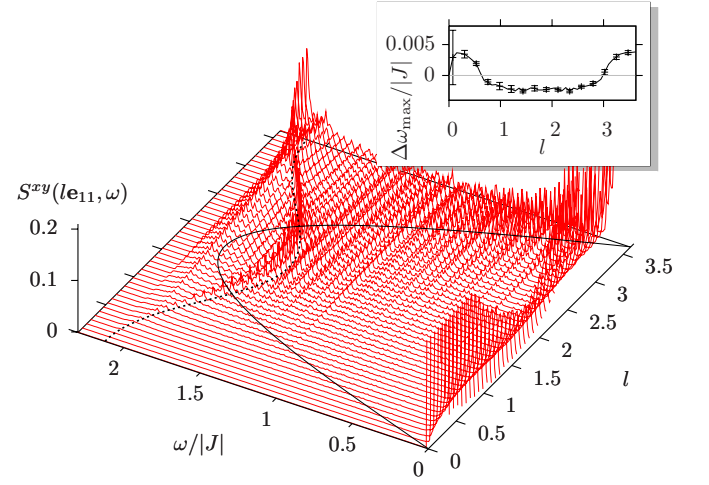


FIG. 13. As in Fig. 12 a shift in Fourier space can also be observed in the (11) direction, and modes from a different part of the Brillouin zone manifest at frequencies given by Eqn. (25) drawn as a broken line. Peak positions used for calculating the deviations in the inset were determined from simulations of the $\sqrt{3} \times \sqrt{3}$ structure allowing a higher precision.

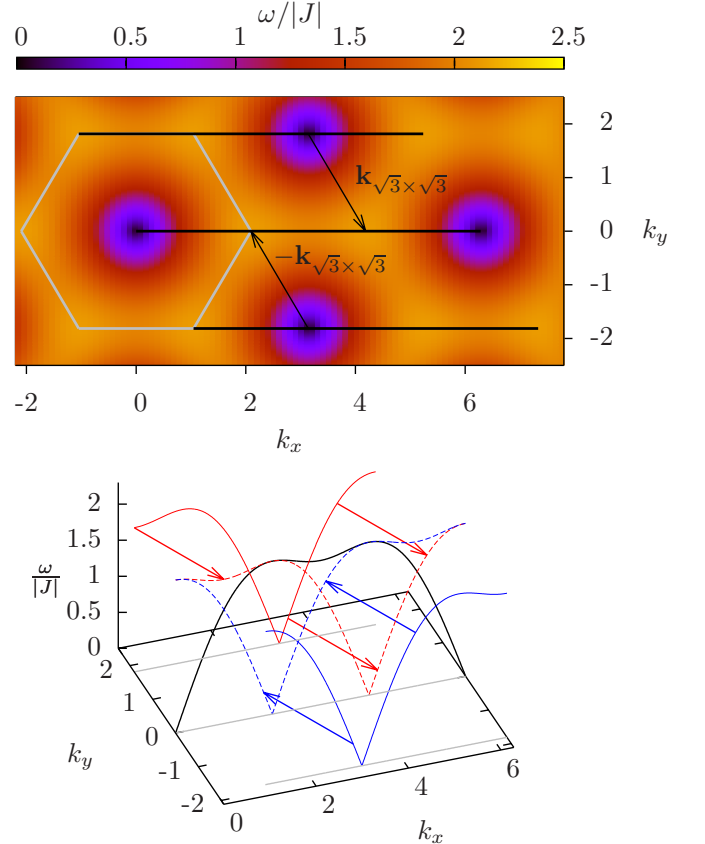


FIG. 14. Analytical frequencies of the acoustic branch as function of wave vector \mathbf{k} . In the (10) direction the shifts by $\mathbf{k}_{\sqrt{3} \times \sqrt{3}}$ and by $-\mathbf{k}_{\sqrt{3} \times \sqrt{3}}$ contribute differently, resulting in two apparent branches. The gray hexagon marks the edge of the first Brillouin zone.

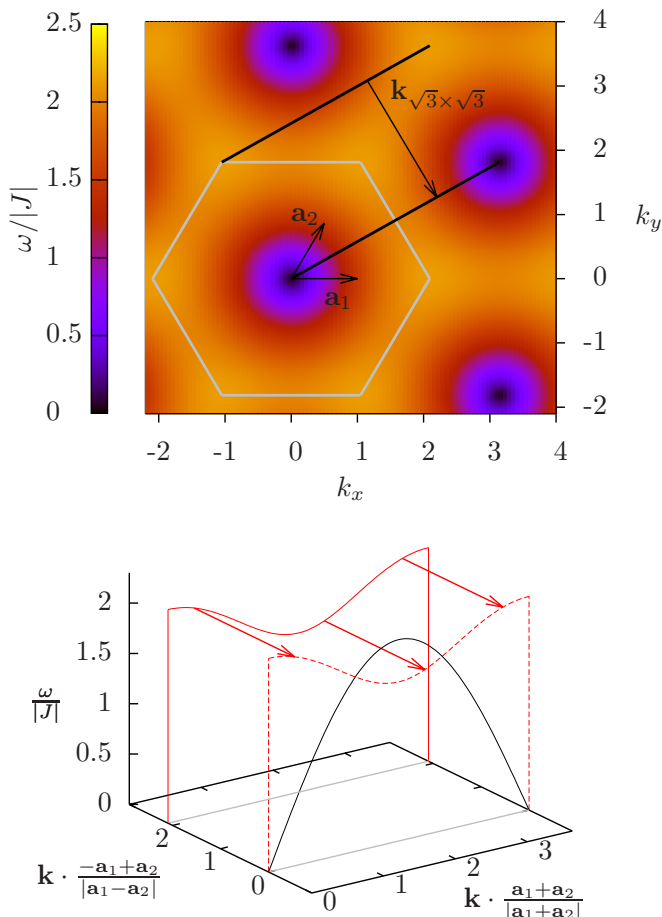


FIG. 15. Analytical frequencies of the acoustic branch as function of wave vector \mathbf{k} . In the (11) direction the shift by $\mathbf{k}_{\sqrt{3} \times \sqrt{3}}$ creates a fictitious optical branch. The shift by $-\mathbf{k}_{\sqrt{3} \times \sqrt{3}}$ (not shown) has an identical effect.

and might actually have been observed already⁵.

Considering the described displacements, we find that all observations become consistent. Not only do the positions of maxima in $S^u(\mathbf{k}, \omega)$ and $S^{xy}(\mathbf{k}, \omega)$ agree nicely with theory, the deviations caused by non-zero temperature also match. From Fig. 10 and Fig. 13 it follows that at the corners of the Brillouin zone acoustic modes have frequencies above the analytically calculated values while no deviation is noticeable at the midpoints of the zone edges according to Fig. 10 and Fig. 11. These two plots also show that in the proximity of the center frequencies are slightly increased, while they are below theoretical predictions at intermediate distance from the center (for the latter see also Fig. 13).

V. CONCLUSIONS

For this work we devised a novel Monte Carlo algorithm able to equilibrate the Heisenberg antiferromagnet on the kagome lattice at temperatures which are

more than two orders of magnitude lower than previously reached. To do so we designed a new trial move that allows the “flipping” of **weathervane** loops and combined the simulated tempering and heatbath techniques with a novel modification of the Wang-Landau algorithm. We could show that, in agreement with theory, minimal temperature dependent selection of loop structures occurs over three orders of magnitude in temperature.

Extensive spin dynamics simulations at $k_B T_i / |J| = 10^{-6}$ provided results that agree very well with analytical calculations: Using suitable coordinates we observed harmonic modes with the predicted frequencies directly within the spin plane and transverse to it. Considering global Cartesian coordinates we showed that signals in the dynamic structure factor are shifted due to $\sqrt{3} \times \sqrt{3}$ correlations. Apparent in-plane excitation at the center of the Brillouin zone with non-zero frequency actually belong to acoustic modes at the zone edge. Hence, the classification of these signals as a branch of optical modes by Robert et al.¹¹ was premature.

We argue that due to the decay of the correlations in-plane excitations will not be measurable for large systems unless next-nearest neighbor interactions stabilize the $\sqrt{3} \times \sqrt{3}$ correlations.

VI. ACKNOWLEDGEMENTS

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VII. APPENDIX A

The Wang-Landau algorithm^{12–14} is a method that is designed to adjust a weight function $w_i(Q)$ over time i such that a flat histogram $H(Q)$ can be produced. The parameter Q is often the system energy which allows the determination of thermodynamic quantities like specific heat and mean energy via the estimation of the density of states as function of energy. Since we are interested in producing a flat histogram over a logarithmic temperature range, we will treat Q as a general variable in these considerations. In the simplest case the probability of a configuration \mathbf{X} is desired to be proportional to $w(Q(\mathbf{X}))$ which means that proposed Monte Carlo moves $\mathbf{X} \rightarrow \mathbf{X}'$ are accepted with probability $P_{\text{acc}}(\mathbf{X}, \mathbf{X}') = \min(1, w(Q(\mathbf{X}')) / w(Q(\mathbf{X})))$. In that way a series of configurations \mathbf{X}_i and values $q_i = Q(\mathbf{X}_i)$ is produced. In order to achieve flatness we modify the weight function after each step:

$$w_{i+1}(Q) = \begin{cases} \frac{w_i(Q)}{f} & \text{if } Q = q_i \\ w_i(Q) & \text{otherwise} \end{cases}, \quad (26)$$

where f is a modification factor which is initially set to $f = e$ (Euler’s constant) and gradually reduced to unity,

thus causing decreasing changes to w . We propose a modification of the original Wang-Landau algorithm which is based on the assumption that especially early in the iteration the deviations from the detailed balance criterion can be reduced by delaying the modification of the weight function. It is self-evident that a change in w affects the simulation's balance most if it is performed in the proximity of the current position q_i of the system. We, therefore, introduce a delay for the modification of d time steps which allows the simulation to proceed undisturbed, decorrelate, and be less influenced by alterations of w . When a modification at q_{i-d} is eventually made, it is necessary to take into account the changes of w at that position between time $i-d$ and i . The corrected modification formula reads:

$$w_{i+1}(Q) = \begin{cases} \frac{w_i(Q)}{f^{w_{i-d}(q_{i-d})}} & \text{if } Q = q_{i-d} \\ w_i(Q) & \text{otherwise} \end{cases}. \quad (27)$$

Note that these equations become equal to Eqn. 26 if $d = 0$, i.e., in case of no delay. In order to calculate this formula it is necessary to know q_{i-d} and $w_{i-d}(q_{i-d})$, i.e. at each time i the values q_i and $w_i(q_i)$ have to be stored for d time steps giving a total of $2d$ numbers to be held

If $\sigma_i = \sigma_j$:

$$\begin{aligned} \langle s_{\mathbf{r}_i}^x s_{\mathbf{r}_j}^x \rangle &= \frac{1}{3} s_{\mathbf{r}_i}^u s_{\mathbf{r}_j}^u \left(\cos^2 \phi + \cos^2(\phi + \frac{2}{3}\pi) + \cos^2(\phi - \frac{2}{3}\pi) \right), \\ &= \frac{1}{6} s_{\mathbf{r}_i}^u s_{\mathbf{r}_j}^u \left(3 + \cos 2\phi + \cos(2\phi + \frac{4}{3}\pi) + \cos(2\phi - \frac{4}{3}\pi) \right), \\ &= \frac{1}{2} s_{\mathbf{r}_i}^u s_{\mathbf{r}_j}^u. \end{aligned} \quad (28)$$

If $\sigma_i \neq \sigma_j$:

$$\begin{aligned} \langle s_{\mathbf{r}_i}^x s_{\mathbf{r}_j}^x \rangle &= \frac{1}{3} s_{\mathbf{r}_i}^u s_{\mathbf{r}_j}^u \left(\cos \phi \cos(\phi + \frac{2}{3}\pi) + \cos \phi \cos(\phi - \frac{2}{3}\pi) + \cos(\phi + \frac{2}{3}\pi) \cos(\phi - \frac{2}{3}\pi) \right), \\ &= \frac{1}{6} s_{\mathbf{r}_i}^u s_{\mathbf{r}_j}^u \left(\cos(2\phi + \frac{2}{3}\pi) + \cos(-\frac{2}{3}\pi) + \cos(2\phi - \frac{2}{3}\pi) + \cos(\frac{2}{3}\pi) + \cos(2\phi) + \cos(\frac{4}{3}\pi) \right), \\ &= \frac{1}{6} s_{\mathbf{r}_i}^u s_{\mathbf{r}_j}^u \left(\cos(-\frac{2}{3}\pi) + \cos(\frac{2}{3}\pi) + \cos(\frac{4}{3}\pi) \right), \\ &= -\frac{1}{4} s_{\mathbf{r}_i}^u s_{\mathbf{r}_j}^u. \end{aligned} \quad (29)$$

In both cases the contributions of ϕ vanish which means that the average is equivalent to an average over ϕ . Furthermore, $\langle s_{\mathbf{r}_i}^x s_{\mathbf{r}_j}^x \rangle_{\{\sigma\}} = \langle s_{\mathbf{r}_i}^y s_{\mathbf{r}_j}^y \rangle_{\{\sigma\}}$ even if ϕ is constant since an average over all loop structures $\{\sigma\}$ involves the average over permutations. Following Harris et al.⁴ we exploit the fact that collective harmonic exci-

in memory. On modern computers this can be done for large values of d , however, we found that a good compromise between increased balance and fast convergence is achieved at $d \approx 10^3, 10^4$.

It turns out that systematic errors are much smaller in the beginning of the simulations and that the overall convergence is often faster, but in our experience never slower, than in the original Wang-Landau method. In the past this method was successfully used to calibrate one and two dimensional weight functions for polymer simulations^{23,24}.

In the present study the weight function w corresponds to the weight factors W_i and the quantity Q is $\log_{10} k_B T / |J|$. Modifications of W were performed after each temperature update.

VIII. APPENDIX B

Consider the product of the x -components of two spins as a function of local coordinates s^u . The three main spin directions lie in the xy -plane, while one of which and the y -axis include an angle ϕ (Fig. 4). Instead of taking the average over all possible angles ϕ , we average only over circular permutations of spin directions σ , which corresponds to rotations of all spins by angles $\pm \frac{2}{3}\pi$ around the z -axis.

tations s^u are independent of the spin configuration $\{\sigma\}$ and average over all $\{\sigma\}$:

$$\langle s_{\mathbf{r}_i}^x s_{\mathbf{r}_j}^x \rangle_{\{\sigma\}} = s_{\mathbf{r}_i}^u s_{\mathbf{r}_j}^u \left(\frac{\tilde{\Gamma}_{\mathbf{r}_i - \mathbf{r}_j}}{2} - \frac{1 - \tilde{\Gamma}_{\mathbf{r}_i - \mathbf{r}_j}}{4} \right),$$

$$= s_{\mathbf{r}_i}^u s_{\mathbf{r}_j}^u \left(\frac{3}{4} \tilde{\Gamma}_{\mathbf{r}_i - \mathbf{r}_j} - \frac{1}{4} \right), \quad (30)$$

where $\tilde{\Gamma}_{\mathbf{r}_i - \mathbf{r}_j}$ denotes the probability for $\sigma_i = \sigma_j$:

$$\tilde{\Gamma}_{\mathbf{r}_i - \mathbf{r}_j} = \langle \delta_{\sigma_i, \sigma_j} \rangle, \quad (31)$$

with δ being the Kronecker delta. Inserting the aforementioned correlation function Γ , we obtain

$$\langle s_{\mathbf{r}_i}^x s_{\mathbf{r}_j}^x \rangle_{\{\sigma\}} = \frac{3}{4} s_{\mathbf{r}_i}^u s_{\mathbf{r}_j}^u \Gamma_{\mathbf{r}_i - \mathbf{r}_j}. \quad (32)$$

It follows that

$$C_{\mathbf{r}}^x(t) = \frac{3}{4} \Gamma_{\mathbf{r}} C_{\mathbf{r}}^u(t) \quad (33)$$

and

$$C_{\mathbf{r}}^{xy}(t) = C_{\mathbf{r}}^x(t) + C_{\mathbf{r}}^y(t) = \frac{3}{2} \Gamma_{\mathbf{r}} C_{\mathbf{r}}^u(t). \quad (34)$$

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