Magnetic phase diagrams of classical triangular and kagome antiferromagnets

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Abstract
We investigate the effect of geometrical frustration on the $H$–$T$ phase diagrams of the classical Heisenberg antiferromagnets on triangular and kagome lattices. The phase diagrams for the two models are obtained from large-scale Monte Carlo simulations. For the kagome antiferromagnet, thermal fluctuations are unable to lift degeneracy completely and stabilize translationally disordered multipolar phases. We find a substantial difference in the temperature scales of the order by disorder effect related to different degeneracy of the low- and the high-field classical ground states in the kagome antiferromagnet. In the low-field regime, the Kosterlitz–Thouless transition into a spin-nematic phase is produced by unbinding of half-quantum vortices.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

One of the important issues in the field of highly frustrated magnets is the universality of the order by disorder phenomenon \cite{1, 2}. If the ‘accidental’ degeneracy of a frustrated spin system is described by a few continuous degrees of freedom \cite{2–7} or by an infinite but non-extensive number of such parameters \cite{8, 9} the order by disorder mechanism is known to stabilize a magnetically ordered state at low temperatures. In the case of a macroscopic number of zero-energy modes the outcome is less universal and only a partial lifting of the degeneracy, if any at all, may take place \cite{10–12}.

In this work we study numerically the entropic order by disorder produced by thermal fluctuations in two classical antiferromagnets on a triangular and a kagome lattice in an external magnetic field. The external field changes continuously the classical ground-states manifold of a frustrated magnet, providing an experimental tool to control and to modify the effect of fluctuations \cite{13, 14}. The magnetic phase diagrams for the two models have been discussed qualitatively in the earlier works \cite{15, 16}. Here we present results of large-scale Monte Carlo simulations, which allow precise mapping of the transition boundaries and clarify the nature of the low-temperature phases. In addition to providing new insights into the order by disorder effect in classical frustrated magnets, our results open the possibility of quantitative comparison with the experimental phase diagrams of recently discovered triangular antiferromagnets RbFe(MoO$_4$)$_2$ \cite{17, 18} and Rb$_3$Mn(MoO$_4$)$_3$ \cite{19}.

The nearest-neighbour exchange Hamiltonian with a Zeeman term is given by

$$\mathcal{H} = J \sum_{\left\langle ij \right\rangle} \mathbf{S}_i \cdot \mathbf{S}_j - H \sum_i S^z_i,$$

where $\mathbf{S}_i$ are three-component classical unit vectors. The model (1) with an antiferromagnetic exchange $J > 0$ is studied for a triangular and a kagome lattice, see figures 1(a) and (b). The common building block of the two lattices, a triangular plaquette, determines frustration at the shortest length scale. In zero field the antiferromagnetic bonds on a single spin-triangle cannot be simultaneously satisfied and magnetic moments form a 120° spin-structure. For the triangular antiferromagnet this leads to an ordered magnetic state with the ordering wavevector $\mathbf{Q} = (4\pi/3, 0)$ and a trivial six-fold degeneracy related to permutations of the spin triad. In contrast, the kagome-lattice model remains infinitely degenerate. The
grows exponentially with the number of sites $N$ as

$$W \approx 1.1347^N.$$  \hfill (2)

In a finite magnetic field the classical ground states may be derived by rewriting the energy (1) as a sum over triangles $[13, 22]$. The classical energy reaches the minimal value for the following magnetization of every plaquette:

$$\sum_{i \in \triangle} S_i = 3 \frac{H}{H_s} \hat{z},$$  \hfill (3)

where $H_s$ stands for the saturation field with $H_s = 9J$ and $6J$ for the triangular and the kagome antiferromagnet, respectively. The constraint (3) leaves three parameters, instead of one, for the choice of the spin triad $S_i$ at $H < H_s$. Physically, extra degrees of freedom correspond to an isotropic susceptibility of a spin-triangle. Thus, an external magnetic field produces new degeneracy in the case of the triangular antiferromagnet and enhances the existing degeneracy for the kagome model.

The entropic degeneracy lifting in the classical triangular antiferromagnet was first studied by Kawamura, who calculated the free-energy contribution of the harmonic excitations $[3]$. Thermal fluctuations lift the degeneracy and stabilize in the interval $0 < H < H_s$ one collinear and two coplanar states. These are shown in figures 1(c)–(e) and will be referred to in the following as the coplanar Y-state, $0 < H < \frac{4}{3}H_s$, the coplanar V-state, $\frac{4}{3}H_s < H < H_s$, and the collinear up–up–down (uud) state, $H \sim \frac{1}{3}H_s$.

The harmonic fluctuations in the kagome antiferromagnet are dominated by zero-energy modes, which contribute most to the free-energy decrease at low temperatures $[20]$. Their number for different classical ground states (3) can be counted by using purely geometrical arguments. As a result, one ends up with the same sequence of three-sublattice configurations in a magnetic field, figures 1(c)–(e) $[16]$. In addition, the zero-energy modes favour the $\sqrt{3} \times \sqrt{3}$ periodic pattern for the V-state, whereas no such selection takes place for the Y- and for the uud-state. Note, also that two sublattices become identical for uud and V configurations. Consequently, the total number of these states is smaller than (2) and is given by the number of dimer coverings of the dual hexagonal lattice $[23, 16]$:

$$W \approx 1.1137^N.$$  \hfill (4)

Lower degeneracy in the high-field region $\frac{1}{3}H_s < H < H_s$ may significantly affect the fate of the order by disorder selection.

In the following we present the Monte Carlo results for the phase diagrams of the triangular (section 2) and of the kagome antiferromagnet (section 3.2). In section 3.1 we also give a brief symmetry analysis of various multipolar states in zero and finite magnetic fields. The obtained results are summarized in section 4.

2. Triangular antiferromagnet

Monte Carlo simulations have been performed using the standard Metropolis algorithm in combination with the microcanonical over-relaxation steps, see $[24]$ for further details. Periodic boundary conditions were implemented for $N = L \times L$ site clusters with the linear size $L$ up to 192. At every temperature/magnetic field we discarded $5 \times 10^4$ Monte Carlo steps (MCS) for initial relaxation and data were collected during subsequent $10^6$ MCS. The error bars were estimated from 10 to 20 independent runs initialized by different random numbers. Instantaneous values of the antiferromagnetic order parameter

$$m_Q = \frac{1}{N} \sum_i S_i e^{-iQr_i}, \quad Q = (n \pi, 0)$$  \hfill (5)

have been used to measure longitudinal and transverse components of the staggered spin susceptibility and the
corresponding Binder cumulants:

\[
\chi_\parallel^\alpha = \frac{N}{T} \langle (m_\parallel^\alpha)^2 \rangle, \quad U_\parallel^\alpha = \frac{\langle (m_\parallel^\alpha)^4 \rangle}{\langle (m_\parallel^\alpha)^2 \rangle^2},
\]

(6)

where \(\alpha = z, \perp\) and \(m_\parallel^\alpha = (m_\parallel^z + m_\parallel^\perp)^{1/2}\).

Following the standard procedure, the second-order transition may be located by the crossing point of the Binder cumulant \(U_\parallel\) measured for different clusters. We illustrate this method in the top panel of figure 2 for the transition between the paramagnetic state and the uud-state at \(H/J = 1\). The alternative approach is to use the normalized transverse susceptibility, provided the critical exponent \(\eta\) is known precisely. In the critical region the susceptibility scales as

\[
\chi = L^{2-\eta} f \left( \frac{\tau}{L^{1/\nu}} \right), \quad \tau = 1 - T/T_c.
\]

(7)

Hence, the normalized susceptibility \(\chi/L^{2-\eta}\) becomes size independent at \(\tau = 0\) and curves for different \(L\) plotted as functions of \(T\) exhibit a crossing point, similar to the behaviour of the Binder cumulant. The collinear uud-state of the triangular antiferromagnet breaks the \(Z_3\) lattice symmetry. The associated phase transition belongs to the universality class of the two-dimensional three-state Potts model, for which the critical exponents are known exactly including \(\eta = 4/15\) [25].

The bottom panel in figure 2 demonstrates that the normalized susceptibility curves for the above value of \(\eta\) cross precisely at the same point as the Binder cumulants. On one side this proves that the critical behaviour for the transition into the uud-state belongs to the three-state Potts universality class, on the other side it allows one to employ stochastically less noisy \(\chi_\parallel^\alpha\) to determine the full transition boundary in the \(H–T\) plane.

Figure 2. Transition between the paramagnetic phase and the uud-state in the triangular antiferromagnet at \(H/J = 1\); the Binder cumulant \(U_\parallel^\alpha\) (upper panel) and the staggered susceptibility \(\chi_\parallel^\alpha\) (lower panel) versus temperature for different lattice sizes \(L\).

At low temperatures the transverse spin components exhibit a quasi-long-range order. The Binder cumulant method is not very convenient for a precise location of the corresponding Kosterlitz–Thouless (KT) transition. However, one can still use the normalized transverse susceptibility \(\chi_\perp^\alpha/L^{2-\eta}\) with the exact KT exponent \(\eta = 1/4\) [25]. We also measured the spin stiffness with respect to twists about the \(\hat{z}\)-axis, given for the triangular antiferromagnet by [22]

\[
\rho_s = -\frac{J}{N\sqrt{2}} \sum_{\langle ij \rangle} \left( \langle S_x^i S_y^j + S_y^i S_x^j \rangle \right) - \frac{2J^2}{NT\sqrt{3}} \left( \left( \sum_{\langle ij \rangle} (S_x^i S_y^j - S_y^i S_x^j) (\hat{e} \cdot \delta_{ij}) \right)^2 \right).
\]

(8)

Here, \(\delta_{ij} = r_j - r_i\) and \(\hat{e}\) stands for an arbitrary unit vector in the lattice plane. The spin stiffness shows a universal jump from zero to \(\rho_s = 2\pi/\xi\) at the KT transition. Measurements of \(\rho_s(T)\) provide, therefore, an independent check for the susceptibility method.

Results of various temperature and magnetic field scans are summarized in the \(H–T\) phase diagram presented in figure 3. The high-temperature paramagnetic phase remains invariant under the \(O(2) \times Z_3\) symmetry group, where the discrete symmetry \(Z_3\) is related to the permutation of three antiferromagnetic sublattices imposed by lattice translations: \(\hat{T}_m \mathbf{m}_Q \to e^{i2\pi m} \mathbf{m}_Q\). Three ordered phases in figure 3 break in different ways the full symmetry group. The boundary between the paramagnetic and the uud-state is formed by the second-order transition points, all other boundaries correspond to the KT transitions. The notable feature of the phase diagram is a wide region with the collinear uud magnetic order [15, 22]. At \(T = 0\) this collinear spin configuration appears only for one specific value of the applied magnetic field \(H = \frac{1}{2} H_s\). Thermal
fluctuations expand its stability range, such that the uud-state (up up down) has the highest transition temperature among the three ordered spin structures. The collinear ordering produces a clear 1/3-magnetization plateau, which is most pronounced in the plots \(dM/dH\) versus \(H\).

Overall, the phase diagram shown in figure 3 is similar to the diagram proposed by Kawamura and Miyashita in [15]. The only qualitative difference concerns the low-field part. For temperature scans in a constant field \(H < 4H\), we always find two successive transitions: one from the paramagnetic state into the uud phase with the \(\mathbb{Z}_3\)-order of \(z\)-components and, then, into the Y-state with an algebraic order of the transverse spin components. There is no direct transition between the paramagnetic state and the Y-phase suggested in [15]. The double transition is completely natural from the symmetry point of view, because ordering processes for longitudinal and transverse spin components are generally decoupled from each other. Note also that the Y-state and the uud-state should be separated from the disordered state in zero field by additional line(s) of phase transitions at \(H/J \lesssim 0.1\). The present study remains uncertain about the precise location of these transition boundaries. Numerical investigation of the low-field regime requires Monte Carlo simulations of substantially bigger clusters with \(L \sim 10^3\) because of the rapidly growing correlation length \(\xi\) below \(T^* \approx 0.3J\) [26].

3. Kagome antiferromagnet

3.1. Multipolar phases in zero and finite magnetic fields

The thermal order by disorder effect in the kagome antiferromagnet at zero field leads to a selection of coplanar states below the crossover temperature \(T^*/J \approx 0.005\) [20, 24]. A true long-range order is not possible in 2D, though one can still discuss an asymptotic breaking of the spin-rotation symmetry at distances smaller than the correlation length \(\xi \sim \exp(J/T)\). The symmetry breaking in the ensemble of coplanar states is described by an unconventional octupolar order parameter [24]. For classical spin models magnetic octupoles are represented by the on-site third-rank tensor
\[
T_{i}^{\alpha\beta\gamma} = S_i^{\alpha} S_i^{\beta} S_i^{\gamma} - \frac{1}{5} S_i^{\alpha} \delta_{\beta\gamma} - \frac{1}{5} S_i^{\beta} \delta_{\alpha\gamma} - \frac{1}{5} S_i^{\gamma} \delta_{\alpha\beta}. \tag{9}
\]

The other common unconventional order parameter is the quadrupolar tensor [27]
\[
Q_{i}^{\alpha\beta} = S_i^{\alpha} S_i^{\beta} - \frac{1}{3} \delta_{\alpha\beta}. \tag{10}
\]

Spin tensors (9) and (10) transform under \(l = 3\) and \(l = 2\) representations of the \(O(3)\) rotation group, respectively. The usual (anti)ferromagnetic order parameter \(m_q\) corresponds to the \(l = 1\) representation. In a typical coplanar ground state of the kagome antiferromagnet, spin on a given site may be oriented parallel to any of the three principal directions of the \(120^\circ\) spin triad. Thus, \(\langle S_i \rangle = 0\) and the lowest order spherical tensor, which captures the \(D_3\) (triatic) symmetry of the ensemble of coplanar states, is the uniform octupole tensor
\[
T^{\alpha\beta\gamma} = \frac{1}{N} \sum_i \langle T_{i}^{\alpha\beta\gamma} \rangle. \tag{11}
\]

The quadrupole tensor
\[
Q^{\alpha\beta} = \frac{1}{N} \sum_i \langle Q_{i}^{\alpha\beta} \rangle \tag{12}
\]
is the second order parameter and is induced via the rotationally invariant coupling term \(Q^{\alpha\beta} T_{\alpha\mu\nu} T_{\beta\mu\nu}\).

In an applied magnetic field, the spin symmetry is reduced to the \(O(2)\) group. Its irreducible representations are labelled by the projection of the angular momentum \(l_z\) on the field direction. Therefore, different components of the same spin tensor may now describe different instabilities of the paramagnetic phase:
\[
l_z = \pm 1, \quad Q^{\alpha\alpha}, T^{\gamma\gamma\alpha}, \tag{13a}
\]
\[
l_z = \pm 2, \quad Q^{\alpha\beta}, T^{\gamma\gamma\beta}, \tag{13b}
\]
\[
l_z = \pm 3, \quad T^{\gamma\gamma\gamma}, \quad \alpha, \beta, \gamma = x, y. \tag{13c}
\]

One can arbitrarily select either \(Q^{\alpha\beta}\) or \(T^{\alpha\beta\gamma}\) to characterize states with \(|l_z| = 1, 2\). In the presence of a uniform magnetization \(m^z\), the two tensors are coupled via a bilinear term \(m^z Q^{\alpha\beta} T^{\gamma\gamma\beta}\) and become completely equivalent from the symmetry point of view, see also [28].

The two coplanar states stabilized in the kagome antiferromagnet at finite fields break the \(O(2)\) rotation symmetry differently. The translationally disordered Y-state has the residual nematic symmetry \(C_2\) and its order parameter is given by a \(2 \times 2\) symmetric traceless matrix (13b). The \(V\)-state may break the translational symmetry, then, its order parameter is the antiferromagnetic vector of the \(\sqrt{3} \times \sqrt{3}\) structure [16]. If fluctuations are not able to select this periodic pattern, the proper order parameter of the \(V\)-state is transverse vector components of the quadrupolar tensor (13a).

An interesting observation is that the symmetry

Figure 4. Magnetic field phase diagram of the Heisenberg kagome-lattice antiferromagnet. Transition points determined by the Monte Carlo simulations are shown by squares. Solid lines are guides for the eye.
of the translationally disordered $\mathcal{V}$-state coincides with the symmetry of the transverse ferromagnetic polarization $\mathbf{m}^\perp$, allowing the interaction term $m^\perp Q^\perp m^\perp$. For the Heisenberg model, the classical constraint (3) ensures that $m^\perp = 0$, so that the corresponding coupling constant exactly vanishes. Nevertheless, the ground-state constraint may be modified, for example, by a single-ion anisotropy $D \sum_i S_i^2$. We have checked that in the easy-axis case $D < 0$ the anisotropy produces a substantial transverse magnetization $m^\perp \propto |D|$ in the $\mathcal{V}$-state of the kagome antiferromagnet.

Partial breaking of the spin-rotation symmetry by multipoles order parameters leads, generally, to fractional topological defects [24]. The $C_2$ symmetry of the Y-state is compatible with elementary half-quantum vortices, whereas the $\mathcal{V}$-multi-polar state supports only the usual integer quantized defects. The universal jump of the spin stiffness at the KT transition is given by [25, 29]

$$\rho_s / T = \frac{2}{\pi \nu^2} \times 3^\nu,$$

where $\nu$ is the vortex winding number. For the half-quantum vortices with $\nu = 1/2$, the jump in the spin-stiffness amounts to $\rho_s / T = 8/\pi$.

3.2. Monte Carlo results

A Monte Carlo investigation of the kagome antiferromagnet in an external magnetic field has been performed using the same simulation protocol as described previously for the triangular antiferromagnet in section 2. To obtain good statistics, one needs, however, to increase the number independent cooling runs to 30–50. We have studied periodic lattices with $N = 3L^2$ sites and $L$ in the range 18–96. The obtained phase diagram of the kagome antiferromagnet is presented in figure 4. The KT transition boundaries for the two coplanar states were determined from the plots of the rescaled quadrupolar susceptibility with the exact KT exponent $\eta = 1/4$ [25]. We also measured the spin stiffness of the kagome antiferromagnet, which is obtained from the previous expression (8) by applying the renormalization factor $3^\nu / \pi$ [24]. The temperature variation of $\rho_s$ for two values of the external field is shown in figure 5. The stiffness jump at the transition into the Y-state ($H / J = 1$) is equal with good accuracy to $8T / \pi$. This large jump demonstrates that the corresponding KT transition is mediated by the unbinding of the half-quantum vortices. The jump of $\rho_s$ at $H / J = 5$ is substantially smaller and agrees with the standard value $2T / \pi$ expected for the XY vector order parameter.

Fractional vortices are incompatible with a conventional antiferromagnetic ordering, therefore, the spin-stiffness data prove the presence of the quasi-long-range nematic order in the low-field Y-state. In order to identify the type of symmetry breaking in the high-field $\mathcal{V}$-state we consider in figure 6 the temperature evolution at $H / J = 3$ of the vector component of the quadrupole tensor $Q_{\perp \perp} = (Q^{\perp x})^2 + (Q^{\perp y})^2$ and the transverse antiferromagnetic amplitude $m_{Q\perp}$ of the $\sqrt{3} \times \sqrt{3}$ configuration. The order parameters take on a finite value in the presence of a long-range order, but scale down as $1/N$, if correlations decay exponentially. The quadrupole tensor $Q_{\perp \perp}$ demonstrates a very weak decrease with the lattice size at low temperatures, which is completely consistent with the power-law decay $\langle Q_{\perp \perp} \rangle \sim r^{-\eta}$ and a $T$-dependent exponent $\eta < 1/4$. The antiferromagnetic order parameter goes down much faster, though still slower than $1/N$. This is related to emergence of the Coulomb-type correlations that are typical to the constrained spin models; see, for example, [30]. Overall,
The two particularly simple spin structures are the $q$ between the two coplanar phases, see figure 4. The corresponding 1/3 selection, and a collinear spin-liquid phase at the 1/3-plateau. Nevertheless, there is no symmetry breaking related to this transition, and a collinear spin-liquid phase at the 1/3-plateau is connected to the paramagnetic phase by a simple crossover.

Indeed, the collinear configurations preserve O(2) rotations about the field direction, and the only type of symmetry breaking at the plateau may be related to selection of a specific periodic pattern. The harmonic spectra of classical fluctuations are identical for different ud structures, though the anharmonic corrections are, generally, not [16]. The two particularly simple spin structures are the $q = 0$ configuration and the $\sqrt{3} \times \sqrt{3}$-state. In the dimer language, the two states correspond to the columnar array of dimers and the staggered arrangement, which maximizes the number of three-dimer hexagons. In our Monte Carlo simulations we did not find any significant tendency for the $q = 0$ ordering in the classical model, though this configuration is favoured by the zero-point fluctuations in the large-$S$ quantum kagome antiferromagnet [31]. The numerical data for the antiferromagnetic order parameter of the longitudinal $\sqrt{3} \times \sqrt{3}$ spin-structure are presented in figure 7. One finds again a fast, but slower than $1/N$ decrease of $m^2_O$ with increasing lattice size. There is no evidence of the long-range ordering down to $T/J \sim 5 \times 10^{-4}$.

The overall behaviour of the antiferromagnetic order parameter is consistent with the pseudo-dipolar correlations in the 2D Coulomb phase [30]. At finite temperatures the Coulomb gas description of the ensemble of the collinear ground states at $H = \frac{1}{3} H_c$ is extended by allowing a nonzero density of monomers, which correspond to triangles with broken constraint condition (3). The monomer doping destroys the power-law correlations at large distances, producing a finite correlation length [32]. A detailed investigation of the crossover from short-distance power-law decay to exponential decrease at large distances requires systematic investigation of much bigger lattices.

### 4. Summary and discussion

In this paper, we have considered the effect of thermal fluctuations in an external magnetic field for two classical frustrated antiferromagnets on 2D triangular and kagome lattices. The common property of the two models is selection of two coplanar and one collinear spin configurations by short-range fluctuations. For the triangular antiferromagnet, such an order by disorder mechanism produces three ordered magnetic structures, see figure 3. The most dramatic effect concerns the collinear up–up–down configuration, which changes from being marginally stable at the mean field level to having the highest transition temperature among the three states.

For the kagome antiferromagnet, in addition to an arbitrary choice of the spin triad subject to the constraint (3), there is also a massive degeneracy related to different arrangements of the triad over the whole lattice. No periodic magnetic structure is selected down to at least $T/J \sim 4 \times 10^{-4}$. We find instead unconventional magnetic ordering described by multipolar (tensor) order parameters, see the phase diagram of the kagome antiferromagnet in figure 4. The most significant change again concerns the ud-state, which does not break any symmetry and corresponds to a collinear spin-liquid phase. Despite producing a pronounced 1/3-magnetization plateau, this state is connected to a high-temperature paramagnetic phase by a crossover rather than a transition.

The surprising feature of the phase diagram of the kagome antiferromagnet, figure 4, is an order of magnitude difference between transition temperatures of the high-field and the low-field multipolar state. Such a remarkable dissimilarity of the two temperature scales is produced by the combined effect of (i) 20% difference in the ground-state entropy, see (2) and (4), and (ii) different harmonic spectra with additional zero-energy modes present for the Y-state [16]. One can speculate, therefore, that if other terms are added to the spin Hamiltonian (1), the system would more easily find an optimal periodic structure in the V-state rather than in the more degenerate Y-state. Let us also note that there is an interesting similarity between the phase diagram in figure 4 and the experimental phase diagram of the 3D hyperkagome antiferromagnet Gd$_3$Ga$_5$O$_{12}$, which orders only in finite magnetic fields [33]. Thus, a Monte Carlo investigation of the magnetic diagram of the hyperkagome model is a pressing extension of the present study. One should be also interested in the effect of the single-ion anisotropy on the phase diagram of the triangular Heisenberg antiferromagnet in relation to the experimental studies of RbFe(MoO$_4$)$_2$ [17, 18] and Rb$_3$Mn(MoO$_4$)$_3$ [19].
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