Field-Induced Transitions in a Kagomé Antiferromagnet

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The thermal order by disorder effect in magnetic field is studied for a classical Heisenberg antiferromagnet on the Kagomé lattice. Using analytical arguments we predict a unique H-T phase diagram for this strongly frustrated magnet: states with a coplanar and a uniaxial triatic order parameter, respectively, at low and high magnetic fields and an incompressible collinear spin-liquid state at one-third of the saturation field. We also present the Monte Carlo data which confirm the existence of these phases.

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Geometrical frustration in lattice spin models is responsible for complete suppression of conventional magnetic order and appearance of nonmagnetic spin liquids or states with exotic order parameters [1]. Applied magnetic field can further enhance frustration. A well known example is a weakly frustrated triangular lattice antiferromagnet, which acquires an additional continuous degeneracy in external field [2]. Investigation of high-field effects in strongly frustrated magnets poses a new challenge for experimental and theoretical studies [3,4].

The Heisenberg antiferromagnet on the Kagomé lattice (see Fig. 1) is a strongly frustrated spin model, which is approximately realized in a number of insulating layered magnets: $SrCr_8Ga_4O_{19}$ [5] and $Ba_2Sn_2Ga_3ZnCr_7O_{22}$ [6] (both with $S = 3/2$), $KFe_3(OH)_6(SO_4)_2$ ($S = 5/2$) [7]. $Gd_3Ga_5O_{12}$ ($S = 7/2$) is another frustrated magnet on a related three-dimensional garnet lattice of corner-sharing triangles, which is often called a hyper-Kagomé lattice. This magnet has a weak exchange constant $J \sim 1$ K and a peculiar unexplained phase diagram in the magnetic field [3]. Motivated by the above materials with large values of spin we investigate in this Letter the finite-temperature magnetization process of a classical antiferromagnet on the Kagomé lattice. We predict three distinctive field regimes below the saturation field $H_{\rm sat}$, where exotic spin phases are stabilized.

The Hamiltonian of a nearest-neighbor Heisenberg antiferromagnet on the Kagomé lattice with classical unit spins can be written up to a constant term as

$$\hat{\mathcal{H}} = \frac{1}{2}\sum_{\langle\triangle\rangle}^{N_\triangle}(J\mathbf{S}_\triangle^2 - \mathbf{H}\cdot\mathbf{S}_\triangle), \qquad (1)$$

where the sum runs over all triangles, $\mathbf{S}_\triangle$ is the total spin of a triangular plaquette, and $N_\triangle = \frac{2}{3}N$ is the number of plaquettes on an N-site lattice. The zero-field classical constraint $\mathbf{S}_\triangle \equiv 0$ fails to define a unique ground state. The ground state coplanar configurations are constructed by fixing spins on a first plaquette to a 120° structure with left or right chirality for the triad $\hat{\mathbf{e}}_a$, $\hat{\mathbf{e}}_b$, and $\hat{\mathbf{e}}_c$ and, then, tiling this triad over the whole lattice in a way that three spins on every plaquette are different [8–10]. The number of all such states for the Kagomé lattice is known exactly: U^N with $U = 1.1347\ldots$ [11]. Nonplanar ground states are constructed from planar configurations by identifying so-called weather vane defects [9].

The thermal order by disorder effect can appear because of a different entropy of short-wavelength fluctuations above degenerate configurations [12]. All coplanar states for a Kagomé antiferromagnet show identical harmonic spectra. They have one flat zero-energy branch with $N_4 = \frac{1}{3}N$ modes, which corresponds to anharmonic quartic excitations [8–10]. A nonzero harmonic mode described by a classical coordinate y has an energy increase $\Delta E_2 \simeq Jy^2$ and contributes $\frac{1}{2}T\ln(J/T)$ to the thermodynamic potential, whereas a soft quartic mode with $\Delta E_4 \simeq Jy^4$ makes a reduced contribution of $\frac{1}{4}T\ln(J/T)$. Coplanar configurations have the largest number of soft modes and are, therefore, selected by thermal fluctuations [8]. Soft modes in coplanar states correspond to alternate tilting of spins out of the ground state spin plane around elementary hexagons. There are $\frac{1}{3}N$ hexagons on the Kagomé lattice. The counting of soft modes from such a geometrical point of view, thus, agrees with the spin-wave analysis.

Harmonic fluctuations do not select between various planar configurations, though the highest statistical weight corresponds to a so-called $\sqrt{3}\times\sqrt{3}$ structure [10]. The low-temperature phase has nematic correlations of the

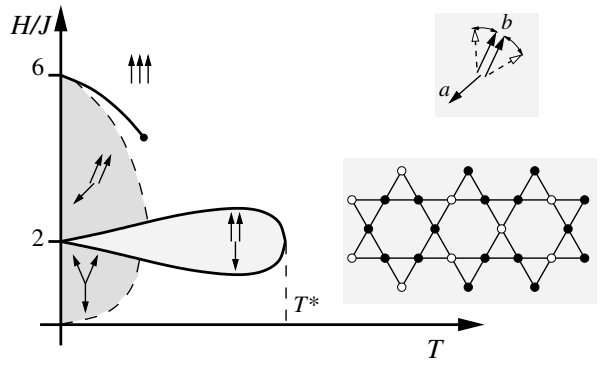

FIG. 1. Schematic phase diagram of the classical Kagomé antiferromagnet. The top inset shows the clapping mode for quasicollinear states. The lower inset shows a unit cell of the $\sqrt{3}\times\sqrt{3}$ quasicollinear state: filled and empty circles denote the b- and a-type spins, respectively.

chirality vectors defined on every plaquette [8]. Besides, the broken symmetries include selection of the spin triad. The residual symmetry group is determined by the two elements: $C_6\hat{\mathcal{T}}$ and U_2, where C_6 is a spin rotation by $\pi/3$ about normal to the spin plane, $\hat{\mathcal{T}}$ is the time reversal, and U_2 is a rotation by π about one of the three in-plane axes. The order parameter for low-temperature phases of the Kagomé antiferromagnet is a three-point correlation function taken on one plaquette:

$$\langle S^{\alpha}(\mathbf{r}_1)S^{\beta}(\mathbf{r}_2)S^{\gamma}(\mathbf{r}_3)\rangle = S^{\alpha\beta\gamma}, \tag{2}$$

which has only a spin part described by a fully symmetric traceless tensor $S^{\alpha\beta\gamma}$ [9,13]. For coplanar states at $H = 0$ this tensor is parametrized as

$$S^{\alpha\beta\gamma} = \sum_{a\leftrightarrow b\leftrightarrow c} \hat{e}_a^{\alpha}\hat{e}_b^{\beta}\hat{e}_c^{\gamma}\,. \tag{3}$$

Note that the three-spin (triatic) order parameter breaks explicitly the time-reversal symmetry, which determines its nontrivial coupling to an applied field.

In a finite magnetic field the classical energy Eq. (1) reaches the minimum provided that

$$\mathbf{S}_{\triangle} = \mathbf{H}/(2J) \tag{4}$$

for every plaquette. Since $|\mathbf{S}_{\triangle}| \leq 3$, above the saturation field $H_{\rm sat} = 6J$ all spins are aligned parallel to $\mathbf{H}$. We are interested in the field range $0 < H < H_{\rm sat}$, where the classical ground state possesses an infinite degeneracy. The degeneracy is further enhanced by a field because the minimum energy constraint does not require spins on one plaquette to lie in the same plane. Shender and Holdsworth made the only attempt to understand the finite field behavior of the Kagomé antiferromagnet [14]. They noticed that spin coplanarity responsible for the increased number of soft modes is preserved if the spin plane is parallel to an external field. Hence, thermal fluctuations create magnetic anisotropy, which orients at low temperatures the spin plane.

There is an additional effect missed in the above consideration and related to the triatic nature of the spin order parameter. Once selection of the spin plane takes place, an extra *macroscopic* degree of freedom appears: orientation of the spin triad inside the plane relative to $\mathbf{H}$. At $T = 0$ all orientations have the same classical energy, though the spin triad distorts differently, when it forms different angles φ with the magnetic field. This distortion modifies harmonic fluctuations inside the spin plane. In such cases with a one-parameter continuous degeneracy thermal fluctuations always produce the order by disorder effect and select a homogeneous state for one particular value of the parameter [12]. The orientational field effect for the triatic order parameter is described by an invariant $E_{\rm an} \simeq H^{\alpha}H^{\beta}H^{\gamma}S^{\alpha\beta\gamma} \propto H^3\cos 3\varphi$, which is present in the Landau functional because of a broken time-reversal symmetry in the triatic phase. The minimum energy is reached for $\varphi = 0$ or π depending on a sign of the prefactor, i.e., one of the triad vectors has to be either parallel or antiparallel to the applied field.

To find the equilibrium orientation of the spin triad in external field, we performed spin-wave calculations. The harmonic spectrum of a coplanar state at $H \neq 0$ depends not only on the triad orientation but also on the way the triad is tiled over the lattice. We used, therefore, the $\sqrt{3} \times \sqrt{3}$ and the $q = 0$ structures [10] as two example cases. For both structures $\varphi = \pi$ is selected by fluctuations with $E_{\rm an} = 5.0T(H/H_{\rm sat})^3N\cos 3\varphi$ and $1.3T(H/H_{\rm sat})^3N\cos 3\varphi$, respectively. Thus, thermal fluctuations in a weak magnetic field create anisotropy for the spin plane as well as inside the plane. The Kagomé antiferromagnet at low fields is described by the coplanar triatic order parameter (3) with one of the spin triad vectors being antiparallel to the field direction (Fig. 1).

Generally, classical ground states at an arbitrary field $0 < H < H_{\rm sat}$ are all noncollinear. Collinear configurations appear among the ground state manifold only at special rational values of the applied field. The up-up-down (*uud*) structure has $|\mathbf{S}_{\triangle}| = 1$ and, hence, is stable only at $H_c = 2J = \frac{1}{3}H_{\rm sat}$. The problem of calculating the total number of the *uud* states for the Kagomé lattice at $H = H_c$ can be mapped onto the problem of dimer coverings of a dual hexagonal lattice, which is solved exactly [11]. The number of *uud* states scales as V^N, with $V = 1.1137\ldots$. A special role of such collinear spin configurations in magnetic field for various frustrated models has been recently emphasized in [4]. Standard harmonic analysis for the Kagomé antiferromagnet at $H = \frac{1}{3}H_{\rm sat}$ yields three twofold degenerate excitation branches for an arbitrary *uud* state:

$$\omega_{\mathbf{k}}^{1} = 0, \qquad \omega_{\mathbf{k}}^{2,3} = 3 \pm \sqrt{3(1 + 2\gamma_{\mathbf{k}})}\,, \tag{5}$$

with $\gamma_{\mathbf{k}} = \frac{1}{3}(\cos k_x + 2\cos\frac{1}{2}k_x\cos\frac{\sqrt{3}}{2}k_y)$. Therefore, the collinear states have $N_4 = \frac{2}{3}N$ soft quartic modes, twice more than any coplanar state. This fact has a simple geometrical origin. Local soft modes correspond to alternate tilting of spins around elementary hexagons. For collinear states such distortions have two polarizations in two directions perpendicular to the field, while coplanar states have soft modes only in the polarization transverse to the spin plane.

Thermal fluctuations reduce the free energy of the *uud* states compared to other noncollinear classical ground states at $H = \frac{1}{3}H_{\rm sat}$. Further lifting of degeneracy within the discrete subset of the *uud* states does not occur due to the same mechanism as for coplanar states at $H = 0$ [8,9,14]. Every collinear state has special lines of weather vane defects, which contain alternating sequences of *u-d-u-d* spins. Flipping all spins along such a line by 180° produces another collinear state at the free energy cost $\Delta F \simeq T$. A macroscopic temperature independent number of such defects is generated at low T's leaving

all *uud* states accessible for the magnet and creating a collinear spin liquid. An enhanced number of soft modes is also reflected in the specific heat C: out of $2N$ degrees of freedom for N classical spins, every quadratic mode contributes $\frac{1}{2}k_B$ to the specific heat, while a quartic mode does only $\frac{1}{4}k_B$ [8]. Hence, $C = [1 - N_4/(4N)]k_B$ per one spin and we expect $C = \frac{5}{6}k_B$ for the collinear spin liquid compared to $C = \frac{11}{12}k_B$ for coplanar states. Specific heat measurement in numerical simulations provides a useful way to distinguish different spin states.

The free energy for the collinear spin liquid is lower by $\sim NT \ln(J/T)$ compared to any noncollinear state. But a small deviation of field away from $H = \frac{1}{3}H_{\rm sat}$ produces only a little change in the classical energy. The collinear spin liquid is, therefore, stabilized by anharmonic interactions to a finite field range in a way similar to stabilization of the incompressible quantum fluid states in the fractional quantum Hall effect. The collinear spin liquid phase yields a *weak* magnetization plateau at $1/3$ of the saturated magnetization: spins in the *uud* states cannot tilt towards the field and a weak variation of the magnetization for the collinear spin liquid is produced by thermal excitations alone. The above arguments also suggest the first-order transition from the collinear spin liquid state to the low- and the high-field phases at low temperatures.

In the field range $\frac{1}{3}H_{\rm sat} < H < H_{\rm sat}$ the ground state configurations are again all noncollinear. There appear, however, new quasicollinear states with two coinciding spins of the basis triad $\hat{\mathbf{e}}_b = \hat{\mathbf{e}}_c$, which have zero chirality on every plaquette (Fig. 1). These configurations belong to the subset of coplanar states. Hence, a quasicollinear state has all the soft excitations of coplanar states: $\frac{1}{3}N$ quartic modes in the polarization transverse to the spin plane. In addition, they acquire extra in-plane soft modes. For a single plaquette, such a clapping-type mode corresponds to simultaneous tilting of the two parallel spins in opposite directions inside the spin plane (Fig. 1). To preserve a weak anharmonic energy change the clapping deformation has to be extended on the lattice along a loop which contains only the b-type spins. The shortest such loop is the perimeter of a hexagon, while the number of appropriate hexagons is maximized if a quasicollinear state is tiled in the $\sqrt{3} \times \sqrt{3}$ structure (Fig. 1). Thus, the $\sqrt{3} \times \sqrt{3}$ quasicollinear state has the largest number of soft modes $N_4 = \frac{4}{9}N$ at $\frac{1}{3}H_{\rm sat} < H < H_{\rm sat}$.

Nevertheless, the true breaking of the translational symmetry does not take place. The disordering mechanism is again related to the presence of loops of alternating spin orientations or weather vane defects [9]. Loop flipping *a-b-a-b* $\rightarrow$ *b-a-b-a* at zero-energy cost contributes $\sim T \ln(J/T)$ to the thermodynamic potential because of the destruction of a few soft modes. Such losses are outweighed at small concentrations of defects by the entropy gain. At finite temperatures there will be a macroscopic number of defects, which destroy the long-range $\sqrt{3} \times \sqrt{3}$ translational symmetry and create instead a phase with a uniaxial triatic order parameter:

$$S^{\alpha\beta\gamma} = n^\alpha n^\beta n^\gamma - \tfrac{1}{4}(n^\alpha \delta_{\beta\gamma} + n^\beta \delta_{\gamma\alpha} + n^\gamma \delta_{\alpha\beta}), \tag{6}$$

where indices run over x, y and $\mathbf{n} = \hat{\mathbf{e}}_a - (\hat{\mathbf{e}}_a \cdot \hat{\mathbf{z}})$. Higher free energy costs make the number of defects in the uniaxial triatic state to be temperature dependent: $N_{\rm def} \sim TN$ with diverging magnetic correlation length $\xi \sim 1/T$. Lacking a conventional magnetic order the high-field triatic phase asymptotically approaches the ordered $\sqrt{3} \times \sqrt{3}$ quasicollinear structure. This behavior is reflected in the temperature dependence of the specific heat which should tend to $C = \frac{8}{9}k_B$ at low T.

At the saturation field $H_{\rm sat} = 6J$ the ground state becomes nondegenerate and corresponds to parallel alignment of spins. The harmonic excitations in this collinear *uuu* phase are given by the same expressions (5) as for the *uud* states. There are $N_4 = \frac{2}{3}N$ soft modes at this magnetic field and the specific heat again reaches the value $C = \frac{5}{6}k_B$. Soft modes also produce a universal magnetic behavior of the frustrated spin system at the saturation. For $H > H_{\rm sat}$ all three branches (5) acquire an additional field dependent shift $\delta\omega = (H - H_{\rm sat})$. Taking into account only the most singular contribution of the first flat branch, we derive that at $H \geq H_{\rm sat}$ the classical partition function scales as

$$Z(H,T) = T^{1/4} f(u), \qquad u = (H - H_{\rm sat})/\sqrt{T}. \tag{7}$$

The susceptibility $\chi = dM/dH$ determined from (7) is $\chi = [f''(u)/f(u)] - [f'(u)/f(u)]^2$. At $u = 0$ ($H = H_{\rm sat}$) the susceptibility becomes temperature independent and all the curves $\chi(H,T)$ cross at one point; see Fig. 2.

So far we considered only the short-wavelength degrees of freedom and completely disregarded long-wavelength excitations, which determine the nature of phase transitions in two dimensions. The non-Abelian topological defects discussed previously for the triatic order parameter [9] are suppressed by the field. The two triatic phases break only U(1) symmetry for rotations about the field direction. Berezinskii-Kosterlitz-Thouless–type transitions separate them from the high-temperature paramagnetic phase; see Fig. 1. In contrast, the first-order transition to the collinear spin-liquid state survives and corresponds to a gas-liquid–type transition on the boundary with the paramagnetic phase. The collinear *uuu* phase also has an enhanced entropy contribution and is stabilized below $H_{\rm sat}$. The corresponding line of first-order transitions ends at a critical point as sketched in Fig. 1.

We have also performed Monte Carlo (MC) simulations for the model (1). The standard Metropolis algorithm was used with up to 10^7 MC steps per every point. Simulations were done for periodic $3L^2$-site lattices with $L = 6$, 12, 18; the data presented in Fig. 2 are for a 972-spin cluster. The magnetization curve at $T = 0$ is a straight line with a slope $\chi = dM/dH = \frac{1}{6}$. At low temperatures $\chi(H)$

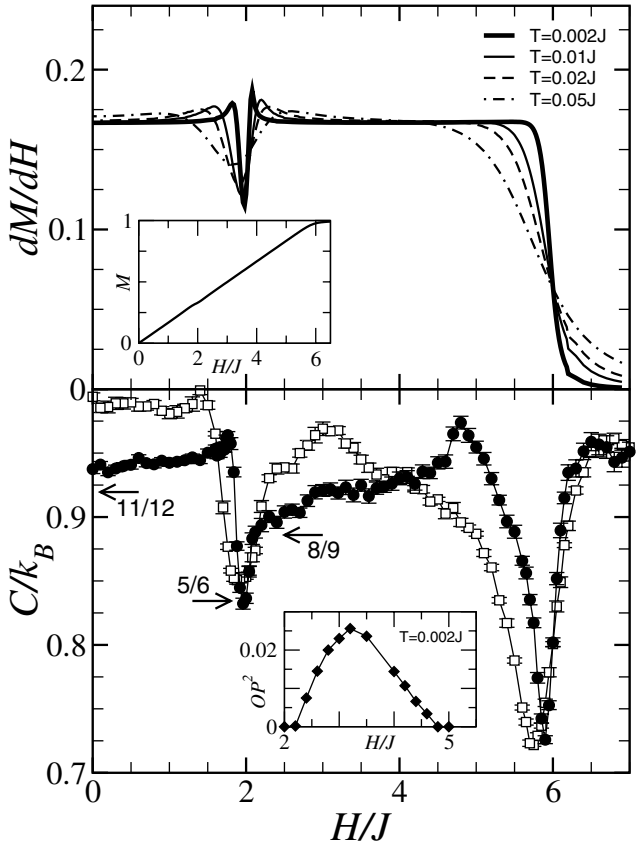

FIG. 2. MC results for the classical Kagomé antiferromagnet. Top panel: Susceptibility vs magnetic field for several temperatures. The inset shows the magnetization curve at $T = 0.01J$. Bottom panel: Specific heat vs magnetic field for $T = 0.002J$ (full circles) and $T = 0.01J$ (open squares). The inset shows the finite-size extrapolation for the squared triatic order parameter (6).

develops a dip near $H = \frac{1}{3}H_{\rm sat}$ corresponding to a weak plateau at $\frac{1}{3}M_{\rm sat}$. Two peaks, which surround the dip, indicate first-order transitions to the plateau phase. The peaks become rounded and then completely disappear at higher temperatures. We estimated $T^* \simeq 0.025J$ as the highest temperature, where the collinear spin liquid still exists (Fig. 1). Above this temperature the dip corresponds to a smooth crossover. The field dependence of the specific heat also follows the above predictions. At $T = 0.002J$, the specific heat starts near $\frac{11}{12}k_B$, corresponding to the coplanar triatic state, at low fields. As the field approaches $\frac{1}{3}H_{\rm sat}$, $C(H)$ goes down to $\frac{5}{6}k_B$, corresponding to the collinear spin liquid, and then recovers back to $\frac{8}{9}k_B$, corresponding to the uniaxial triatic state. The specific heat drops again near the saturation with $C|_{H_{\rm sat}} = \frac{5}{6}k_B$, while the minimum value is reached at somewhat lower fields. Such a behavior signals stabilization of the *uuu* phase beyond its classical boundary: bare negative modes below $H_{\rm sat}$ are renormalized into soft modes, which further decrease $C(H)$. The specific heat also exhibits a peak at $H \approx 4.8J$ ($T = 0.002J$) and $H \approx 3J$ ($T = 0.01J$) with a significant finite-size dependence. We attribute this peak to a phase transition from the collinear *uuu* state into a noncollinear uniaxial triatic state (6). To check this we have calculated field dependence of the squared triatic order parameter (2) at $T = 0.002J$ for three cluster sizes and extrapolated it to the thermodynamic limit. Results are shown in the inset in Fig. 2. The uniaxial triatic phase disappears at about the same field as the position of the peak in the specific heat. The present data do not resolve clearly the order of the phase transition, which must be determined in a detailed numerical study.

We have shown that the geometrical approach based on soft mode counting is a powerful tool for the investigation of the field behavior of a classical Kagomé lattice antiferromagnet. Our preliminary analysis shows that a Heisenberg antiferromagnet on the related garnet lattice has the same type of phase diagram (Fig. 1). In particular, the field range of an asymptotically ordered quasicollinear state coincides roughly with a region for a field-induced ordering observed in $Gd_3Ga_5O_{12}$ [3].

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