Vortex-induced topological transition of the bilinear-biquadratic Heisenberg antiferromagnet on the triangular lattice

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The ordering of the classical Heisenberg antiferromagnet on the triangular lattice with the bilinear-biquadratic interaction is studied by Monte Carlo simulations. It is shown that the model exhibits a topological phase transition at a finite-temperature driven by topologically stable vortices, while the spin correlation length remains finite even at and below the transition point. The relevant vortices could be of three different types, depending on the value of the biquadratic coupling. Implications to recent experiments on the triangular antiferromagnet NiGa$_2$S$_4$ is discussed.

Antiferromagnetic (AF) Heisenberg model on the two-dimensional (2D) triangular lattice has been studied extensively as a typical example of geometrically frustrated magnets. Inspired by recent experiments on a variety of triangular magnets, including NiGa$_2$S$_4$[1, 2] and NaCrO$_2$[3], renewed interest has now arisen in this model. The triangular-lattice Heisenberg antiferromagnet (AF) with the nearest-neighbor bilinear exchange is known to exhibit a magnetic long-range order (LRO) at $T=0$, the so-called 120° structure, in either case of quantum $S=1/2$ or classical $S=\infty$ spin. Because of the two-dimensionality of the lattice, the AF LRO is established only at $T=0$, while the associated spin correlation length diverges exponentially toward $T=0$.

Some time ago, it was demonstrated by Kawamura and Miyashita (KM) that the triangular Heisenberg AF bears a topologically stable point defect characterized by a two-valued topological quantum number, $Z_2$ vortex, in contrast to its unfrustrated counterpart. [4]. Existence of such a vortex has become possible owing to the non-collinear nature of the spin order induced by frustration. KM suggested that the triangular Heisenberg AF might exhibit a genuine thermodynamic transition at a finite temperature associated with the condensation (binding-unbinding) of $Z_2$ vortices. This topological transition is of different character from the standard Kosterlitz-Thouless (KT) transition in that the spin correlation length does not diverge even at and below the transition point and the spin correlation in the low-temperature phase decays exponentially. The topological transition occurs between the two spin paramagnetic states.

On experimental side, recent data by Nakatsuji et al on the $S=1$ triangular Heisenberg AF NiGa$_2$S$_4$ are of particular interest: While no magnetic LRO is observed at low temperature, the low-temperature specific heat exhibits a $T^2$ behavior, suggesting the existence of Goldstone modes associated with a broken continuous symmetry. Meanwhile, neutron scattering measurements suggested that the spin correlation length stayed short even at low temperature [1]. To account for such peculiar experimental results, Tusnetsugi and Arikawa [5], Läuchli et al [6] and Bhattacharjee et al [7] proposed a scenario where the spin nematic order, either ferroquadratic (FQ) or antiferroquadratic (AFQ), play a dominant role. Their theoretical analyses were performed on the basis of the AF $S=1$ Heisenberg model with the bilinear-biquadratic exchange. Experimentally, a weak but clear anomaly, possibly originated from some kind of phase transition, is observed in the susceptibility at $T\approx 8.5K$ [1]. In the present letter, we address the issue of the nature of the experimentally observed transition-like anomaly of NiGa$_2$S$_4$.

The model considered is the $S=\infty$ version of the $S=1$ Hamiltonian used in Refs. [3, 2, 7], i.e., a classical Heisenberg AF on the 2D triangular lattice with the bilinear-biquadratic exchange described by

$$\mathcal{H} = -J \sum_{<ij>} \vec{S}_i \cdot \vec{S}_j - K \sum_{<ij>} (\vec{S}_i \cdot \vec{S}_j)^2,$$

where $J<0$ is the antiferromagnetic bilinear exchange, $K$ the biquadratic exchange which is either FQ ($K>0$) or AFQ ($K<0$), and the sum is taken over all nearest-neighbor pairs on the lattice. While the biquadratic term is essential in stabilizing a hypothetical spin nematic order, its significance in real systems has not been established yet. The biquadratic term is usually small, while it has been argued that it could be large near the Mott transition or due to the effect of orbitals [6]. In the present letter, following Refs. [5, 6, 7], we assume (1), and investigate its finite-temperature ordering properties by means of Monte Carlo (MC) simulations.

MC simulations are performed based on the standard heat-bath method. The system studied is of size $L \times L$, $L$ being in the range from 48 to 192, with periodic boundary conditions. The system is gradually cooled from the high temperature, each run containing $(3 \sim 6) \times 10^5$ Monte Carlo steps per spin (MCS) at each temperature. Averages are then made over $5 \sim 10$ independent runs.

Pure bilinear case $K=0$

In the case of the bilinear interaction only ($K=0$), the ordering property of the model were studied extensively [5, 6, 7]. Numerical studies suggested that the model exhibited a $Z_2$ vortex-induced topological transition at $T = T_v \approx 0.3$ (in units of $|J|$), at which the spin correlation length remains finite. The specific peak exhibits
a rounded peak above $T_V$, while no appreciable anomaly is observed at $T_V$. The transition manifests itself as a dynamical anomaly [4, 10].

A convenient quantity characterizing such a vortex transition might be the vorticity modulus, which measures the stiffness of the system against spin deformation corresponding to vortex formation [11]. The vorticity modulus is defined by $v = \Delta F / \ln L$ where $\Delta F$ is the free-energy cost due to the introduction of an isolated vortex into the system. In MC simulations, $v$ can be calculated from appropriately defined fluctuations [8]. In the vortex-unbounded phase, the system does not exhibit macroscopic stiffness against vortex formation with $v = 0$, while, in the vortex-bounded phase, the system becomes stiff against vortex formation and $v > 0$.

Our MC result of the vorticity modulus is shown in Fig.1a. The data indicate the occurrence of a vortex-induced topological transition at $T \simeq 0.28$, consistently with the previous results [4, 8, 9].

Ferroquadrupolar case $K > 0$

Next, we analyze the FQ case with $K > 0$. The ground-state of three spins on a triangle is the 120° structure for $K < 2/9$ (measured in units of $|J|$), whereas at $K = 2/9$ it exhibits a discontinuous change into the collinear state with up-up-down (down-down-up) state as illustrated in Fig.2a, which remains to be the ground state up to $K = \infty$. Such a collinear ground state resembles the one of the triangular Ising AF, although in the present Heisenberg case the axis of spin collinearity can be arbitrary. In the collinear ground state, whether each spin points either up or down is not uniquely determined due to the frustration-induced local degeneracy: See Fig.2a. Such a local degeneracy leads to a macroscopic degeneracy in an infinite triangular lattice. Indeed, one sees from exact information about the corresponding Ising model that the collinear ground state does not possess a true AF LRO, but only a quasi-LRO with power-law-decaying spin correlations [12]. Meanwhile, since spins are aligned all parallel or antiparallel selecting a unique axis in spin space, the collinear ground state is characterized by the FQ LRO. The order parameter of the FQ state is a director, rather than the spin itself. In terms of a local quadrupole variable, $q_{\mu\nu} = S_{\mu i} S_{\nu i} - (1/3)\delta_{\mu\nu}$, the FQ order parameter $Q_F$ might be defined by

$$ (Q_F)^2 = \frac{3}{2} \sum_{\mu,\nu=x,y,z} < \frac{1}{N} \sum_i q_{\mu\nu}^2 >, $$(2)

where $< \cdots >$ represents a thermal average.

In Fig.3a, we show for the case of $K = 0.5$ the temperature dependence of $Q_F$ together with that of the Fourier magnetization $m_f$ defined by

$$ (m_f)^2 = 2 < |\vec{m}(q)|^2 >, \quad \vec{m}(q) = \frac{1}{N} \sum_i \vec{s}_i e^{i\vec{q} \cdot \vec{r}_i}, $$(3)

where $\vec{q} = (4\pi/3, 0)$. Although both $Q_F$ and $m_f$ vanish in the thermodynamic limit at any $T > 0$, one can still get useful information about the short-range order (SRO) from the corresponding finite-size quantities. As can be seen from Fig.3a, the FQ SRO develops rather sharply at $T \simeq 0.4$, while the standard AF SRO is kept smaller.

The $Z_2$ vortex based on the noncollinear spin order is expected to survive at least up to $K = 2/9$. Different situation, however, is expected for $K > 2/9$ since the ground state changes from the 120° structure to the FQ state. Interestingly, one sees that the FQ state also sustains a topologically stable $Z_2$ vortex with a parity-like
Antiferroquadrapolar case $K < 0$

In the case of the AFQ coupling $K < 0$, the ground state of three spins on a triangle remains to be a $120^\circ$ spin structure for $K > -1$, whereas for $K < -1$ it takes a non-coplanar structure with an angle between two spins $\theta$ equal to $\cos \theta = 1/(2K)$. The change in the spin configuration at $K = -1$ is continuous. For $K < -1$, due to the non-coplanarity of the spin structure, the ground state possesses two distinct “chiral” states with mutually opposite signs of the scalar chirality $\vec{S}_a \cdot \vec{S}_b \times \vec{S}_c$. This local chiral degeneracy has important consequence on the property of an infinite lattice, as the sign of the local chirality tends to take random spatial pattern in the ground state, destroying the three-sublattice AF LRO. As we shall see below, such a ground state still can sustain the AFQ order with the three-sublattice periodicity.

In Fig.4a, we show a typical snapshot of spin directions observed at a temperature $T = 0.17$, where a typical three-sublattice AFQ pattern is realized, with the A-, B- and C-sublattice spins pointing to, say, $\pm S_x$, $\pm S_y$ and $\pm S_z$ directions with equal probability. Since such a locally orthogonal spin structure is not a ground state for $|K| < \infty$, its stabilization should be an entropic effect. In Fig.4b, we show a typical snapshot of spin directions at a lower temperature $T = 0.01$, where a non-orthogonal AFQ state is realized in which spins on each triangle locally satisfy the above-mentioned ground-state condition.

In Fig.3b, we show $m_f$ and the AFQ order parameter $Q_{AF}$ defined by

$$ (Q_{AF})^2 = 3 \sum_{\mu,\nu=x,y,z} \frac{1}{N} \sum_i q_{i\mu\nu} e^{i\vec{q} \cdot \vec{r}_i} < i >, $$

for the case of $K = -3$. The AFQ SRO turns out to develop rather sharply at $T \simeq 0.55$, where the standard AF SRO is still kept small. The AF SRO grows at a lower temperature $T \simeq 0.15$. The orthogonal AFQ spin structure illustrated in Fig.4a is realized in the intermediate temperature range $0.55 \simeq T \simeq 0.15$, whereas the non-orthogonal AFQ state illustrated in Fig.4b is realized in the lower temperature range $T \lesssim 0.15$.

topological quantum number \[13\]. A typical example of such $Z_2$ vortex is illustrated in Fig.2b. It corresponds to a $\pi$ turn (π disclination) of the director vector.

Fig.1b exhibits the vorticity modulus for $K = 0.5$. As can be seen from the figure, a vortex-induced topological transition occurs at $T_V \simeq 0.37$ in the temperature region where the FQ SRO has been developed. Here note the difference in the size dependences of $v$ and of $Q_F$ (or $m_f$) at low temperatures: With increasing $L$ at $T \lesssim T_V$, while $v$ tends to increase slightly tending to a nonzero value, $Q_F$ or $m_f$ tends to decrease. Each size dependence corresponds to the LRO and the SRO, respectively.

![FIG. 2: (a) Frustrated spins on a triangle in the FQ state. (b) $Z_2$ vortex formed by directors in the FQ state.](image)

![FIG. 3: The temperature and size dependence of the FQ and AFQ order parameters, $Q_F$ and $Q_{AF}$, and the Fourier magnetization $m_f$ for the cases of (a) $K = 0.5$ and (b) $K = -3$.](image)

![FIG. 4: Snapshots of spin directions mapped onto a unit sphere in spin space for $K = -3$, at temperatures (a) $T = 0.17$ and (b) $T = 0.01$. Each color represents each sublattice.](image)
In the AFQ state, the order-parameter space is isomorpho
to that of biaxial nematics. The topological defect
structure of biaxial nematics has been analyzed [13]: It
sustains a vortex whose topological quantum number is
given by the quaternion group, or more precisely, its
five conjugacy classes. In addition to the vortex-free
state, there are four topologically distinct vortices. In-
terestingly, the quaternion group is non-Abelian, which
might lead to a glassy dynamics via a peculiar combina-
tion rule of vortices. Even in such an exotic case, the vor-
tex binding-unbinding mechanism is expected to operate,
i.e., one expects a vortex-induced topological transition.

Fig.1c exhibits the vorticity modulus for \( K = -3 \). As
can be seen from the figure, a vortex-induced topological
transition takes place at \( T_V \approx 0.5 \) in the temperature
region where the AFQ SRO order is developed but the mag-
etic SRO is kept suppressed. In contrast to the \( K > 0 \) case, the vorticity modulus exhibits a second anomaly
around a temperature \( T_2 \approx 0.15 \) considerably lower than the vortex transition temperature. Details of this second
transition (or crossover) remains to be elucidated.

**Implications to NiGa\textsubscript{2}S\textsubscript{4}**

Based on our finding that the bilinear-biquadratic tri-
angular Heisenberg AF exhibits a vortex-induced topolo-
gical transition, we wish to discuss its possible impli-
cations to NiGa\textsubscript{2}S\textsubscript{4}. We argue that the experimentally
observed “transition” of NiGa\textsubscript{2}S\textsubscript{4} might be originated
from a vortex-induced topological transition. The rele-
vant vortices could be (i) \( Z_2 \)-vortices based on the non-
collinear AF order for smaller \( |K| \), (ii) \( Z_2 \)-vortices based
on the FQ order for largely positive \( K \), and (iii) quar-
ternion vortices based on the AFQ order for largely nega-
tive \( K \). Whichever situation (i)-(iii) applies, the scenario
immediately explains the experimental observation that the
spin correlation length remains finite even at and be-
low the transition. The specific heat is expected to show
no appreciable anomaly at the transition, only a rounded
peak above it, which seems consistent with experiments.
Recent experiments on the nonmagnetic impurity effec-
t have revealed that, as the impurity concentration is re-
duced toward the pure limit, the extent of the spin-glass-
like hysteretic behavior is suppressed, while the freezing
temperature \( T_f \) itself increases [2]. This observation is
also consistent with our view that the topological transi-
tion intrinsic to the pure system induces a spin-glass-like
freezing in the corresponding impure system.

The next question is obviously which type of vortex is
relevant in NiGa\textsubscript{2}S\textsubscript{4}. Very recent NQR and \( \mu \)SR mea-
surements indicate that static internal fields set in below
\( T_f \) accompanied with a divergent increase of the corre-
lation time toward \( T_f \), at least within experimental time
window [13]. This observation of internal fields appears
compatible only with the case (i) above. In the case (i),
the low-temperature phase should be dominated by spin-
wave excitations: It is a near critical phase characterized
by large but still finite spin correlation length and corre-
lation time. Then, spin waves would be responsible for
the \( T^2 \) specific heat. Indeed, Fujimoto recently accounted
for the \( T^2 \) specific heat based on the spin-wave excita-
tions of the noncollinear AF order of the \( S = 1 \) quan-
tum magnets, neglecting the vortex degrees of freedom
[15]. Vortex-free assumption of Ref. [15] is well justified
at \( T < T_V \), if there occurs a topological transition. Note
that, in this vortex scenario, the correlation time does
not truly diverge at \( T_V (= T_f) \), but only grows sharply at
\( T_V \) exceeding the experimental time scale, and stays long
in a wide temperature range below \( T_V \). Such a near criti-
cal behavior realized below \( T_V \) seems consistent with the
NQR observation [14]. One may suspect that a weak in-
terplane coupling \( J^' \), which should exist in real NiGa\textsubscript{2}S\textsubscript{4},
inherently induces the 3D AF LRO immediately below
\( T_V \). However, this is not necessarily the case: If \( J^' \) is suf-
ficiently small satisfying \( J^' (T_f)^2 \lesssim k_B T_V \), \( \xi(T_V) \) being
the spin correlation length at \( T_V \), the 3D AF LRO needs
not set in even below \( T_V \). Finiteness of \( \xi \) and smallness of \( J^' \) are essential in preventing the vortex ordered state
from forming the 3D AF LRO. At still lower tempera-
tures, \( \xi \) diverges exponentially toward \( T = 0 \), eventually
leading to the onset of the magnetic LRO at a certain
temperature \( T < T_V \).

In NiGa\textsubscript{2}S\textsubscript{4}, distant neighbor interactions neglected in
the present analysis, particularly the third-neighbor in-
teraction, compete with the nearest-neighbor one lead-
ing to an incommensurate spin structure at low temper-
ature [1]. We note that the vortex transition discussed
here is not specific to the 120\degree spin structure realized
in the nearest-neighbor model, but generically expected for
the noncollinear spin order including the incommensu-
rate one, although details of the transition needs to be
clarified further. The vortex scenario might also apply to
the S=3/2 triangular AF NiCrO\textsubscript{2} [3].

Finally, the noncollinear AF order might explain an-
other noticeable feature of experiments that the \( T^2 \) spe-
cific heat is quite robust against applied magnetic fields
[1]. It is because the noncollinear AF ground state
in magnetic fields is capable of keeping an accidental de-
genzer not related to the Hamiltonian symmetry, es-
entially of the same amount as in the zero-field case
[10]. Hence, at the classical level, this accidental de-
genzer gives rise to pseudo-Goldstone modes even in
applied fields, which may account for the robustness of
the low-temperature specific heat, while this degeneracy
would become approximate in quantum systems.

In summary, we studied the ordering properties of the
AF Heisenberg model on the triangular lattice with the
the bilinear-biquadratic coupling, and have shown that
the model exhibits a vortex-induced topological transi-
tion. The relevant vortices could be of three different
types, depending on the value of the biquadratic cou-
pling. It was then suggested that the peculiar phase
transition recently observed in NiGa\textsubscript{2}S\textsubscript{4} might have its
origin in such a vortex-induced topological transition.

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