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LETTERS

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 $NiGa_2S_4$ is discussed.

KEYWORDS: triangular lattice, frustration, vortex, topological transition, $NiGa_2S_4$
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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 $NiGa_2S_4$[1,2] and $NaCrO_2$,[3] renewed interest has now arisen in this model. The triangular-lattice Heisenberg antiferromagnet 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 noncollinear 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 $NiGa_2S_4$ are of particular interest: While no magnetic LRO is observed to 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, Tusnetsugu 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 anlyses were performed on the basis of the AF $S = 1$ Heisenberg model with the blinear-biquadratic exchange. Experimentally, a weak but clear anomaly, possibly originated from some kind of phase transition, is observed in the susceptibility at $T \simeq 8.5$ K.[1] In the present letter, we address the issue of the nature of the experimentally observed transition-like anomaly of $NiGa_2S_4$.

The model considered is the $S = \infty$ version of the $S = 1$ Hamiltonian used in refs. 5–7, i.e., a classical Heisenberg AF on the 2D triangular lattice with the bilinear-biquadratic exchange described by

$$\mathcal{H} = -J \sum_{\langle ij \rangle} \boldsymbol{S}_i \cdot \boldsymbol{S}_j - K \sum_{\langle ij \rangle} (\boldsymbol{S}_i \cdot \boldsymbol{S}_j)^2, \tag{1}$$

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. 5–7, 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 $(3-6) \times 10^5$ Monte Carlo steps per spin (MCS) at each temperature. Averages are then made over 5–10 independent runs.

Pure bilinear case $K = 0$: In the case of the bilinear interaction only ($K = 0$), the ordering property of the model was studied extensively.[4,8,9] The Z_2 vortex in this case corresponds to a 2π rotation of the 120° spin structure around a vortex core: See Figs. 2–4 of ref. 4. Numerical studies suggested that the model exhibited a Z_2 vortex-induced topological transition at $T = T_V \simeq 0.3$ (in units of $|J|$), at which the spin correlation length remains finite. The

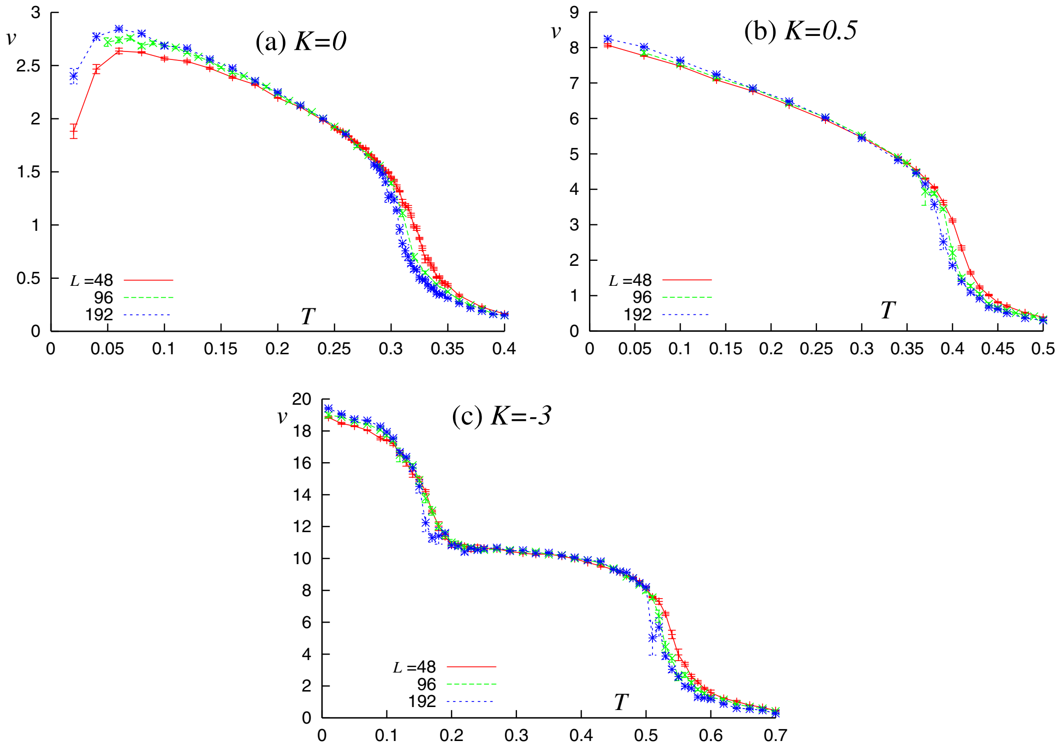

Fig. 1. (Color online) The temperature and size dependence of the vorticity modulus for (a) $K = 0$, (b) $K = 0.5$, and (c) $K = -3$.

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 ΔF 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. 1(a). The data indicate the occurrence of a vortex-induced topological transition at $T \simeq 0.28$, consistently with the previous results.[4,8,9)]

Ferroquadrapolar 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. 2(a), 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. 2(a). 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

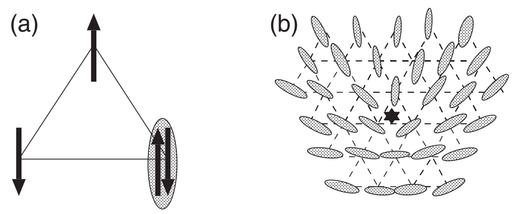

Fig. 2. (a) In the FQ state, frustrated spins on a triangle give rise to a director. (b) Z_2 vortex in the FQ state, where the director makes a π rotation around a vortex core indicated by the asterisk.

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 quadrapole variable, $q_{i\mu\nu} = S_{i\mu}S_{i\nu} - (1/3)\delta_{\mu\nu}$, the FQ order parameter Q_F might be defined by,

$$(Q_F)^2 = \frac{3}{2} \sum_{\mu,\nu=x,y,z} \left\langle \left(\frac{1}{N} \sum_i q_{i\mu\nu} \right)^2 \right\rangle, \tag{2}$$

where $\langle \cdots \rangle$ represents a thermal average.

In Fig. 3(a), 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

$$(m_f)^2 = 2\langle |\boldsymbol{m}(\boldsymbol{q})|^2 \rangle, \quad \boldsymbol{m}(\boldsymbol{q}) = \frac{1}{N} \sum_i \boldsymbol{S}_i e^{i\boldsymbol{q}\cdot\boldsymbol{r}_i}, \tag{3}$$

where $\boldsymbol{q} = (4\pi/3, 0)$. 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

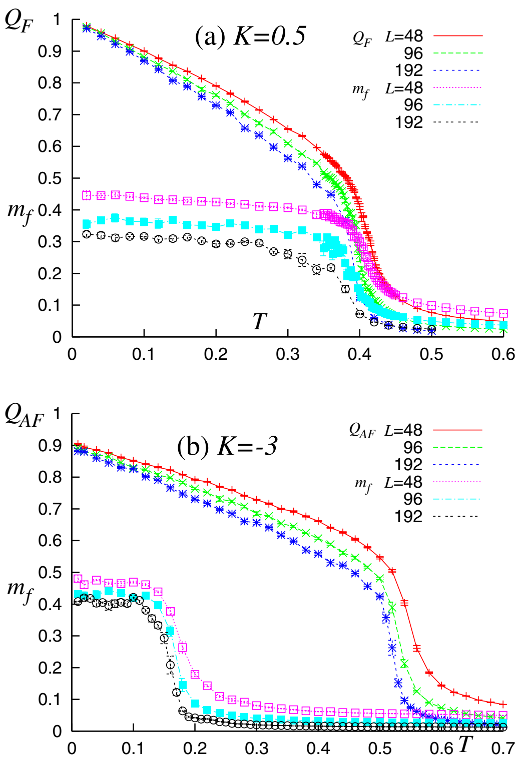

Fig. 3. (Color online) The temperature and size dependence of the FQ and AFQ order parameters, Q_{F} and Q_{AF}, and the Fourier magnetization m_{f} for the cases of (a) $K = 0.5$ and (b) $K = -3$.

Fig. 3(a), 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 topological quantum number.[13] A typical example of such Z_2 vortex is illustrated in Fig. 2(b): It corresponds to a π turn (π disclination) of the director around a vortex core.

Figure 1(b) exhibits the vorticity modulus for $K = 0.5$. As can be seen from the figure, a vortex-induced topological transition occurs at $T_{\mathrm{V}} \simeq 0.37$ 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 $T \lesssim T_{\mathrm{V}}$, 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.

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° spin structure for $K > -1$, whereas for $K < -1$ it takes a non-coplanar structure with an angle between two spins θ 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 $\boldsymbol{S}_a \cdot \boldsymbol{S}_b \times \boldsymbol{S}_c$. 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

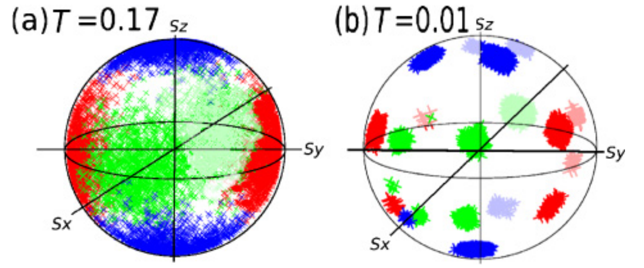

Fig. 4. (Color online) Snapshots of spin directions mapped onto a unit sphere in spin space for $K = -3$, at temperatures (a) $T = 0.17$ and (b) $T = 0.01$. Each color represents each sublattice. Thick color represents the point on the front side of a sphere, while thin color represents the back side.

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. 4(a), 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. 4(b), 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-sate condition.

In Fig. 3(b), we show m_{f} and the AFQ order parameter Q_{AF} defined by

$$(Q_{\mathrm{AF}})^2 = 3 \sum_{\mu,\nu=x,y,z} \left\langle \left(\frac{1}{N} \sum_i q_{i\mu\nu} \mathrm{e}^{\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{r}_i} \right)^2 \right\rangle, \tag{4}$$

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. 4(a) is realized in the intermediate temperature range $0.55 \gtrsim T \gtrsim 0.15$, whereas the non-orthogonal AFQ state illustrated in Fig. 4(b) is realized in the lower temperature range $T \lesssim 0.15$.

In the AFQ state, the order-parameter space is isomorphic 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 quarternion group, or more precisely, its five conjugacy classes. In addition to the vortex-free state, there are four topologically distinct vortices. Three of these four correspond to π rotations of the biaxial director with respect to three distinct rotation-axes in spin space, while one of them corresponds to a 2π rotation of the biaxial director: See Figs. 35 and 36 of ref. 13. Interestingly, the quarternion group is non-Abelian, which might lead to a glassy dynamics via a peculiar combination rule of vortices. Even in such an exotic case, the vortex binding-unbinding mechanism is expected to operate, i.e., one expects a vortex-induced topological transition.

Figure 1(c) exhibits the vorticity modulus for $K = -3$. As can be seen from the figure, a vortex-induced topological transition takes place at $T_{\mathrm{V}} \simeq 0.5$ in the temperature region where the AFQ SRO order is developed but the magnetic

SRO is kept suppressed. In contrast to the $K > 0$ case, the vorticity modulus exhibits a second anomaly around a temperature $T_2 \simeq 0.15$ considerably lower than the vortex transition temperature. Details of this second transition (or crossover) remains to be elucidated.

Implications to $NiGa_2S_4$: Based on our finding that the bilinear-biquadratic triangular Heisenberg AF exhibits a vortex-induced topological transition, we wish to discuss its possible implications to $NiGa_2S_4$. We argue that the experimentally observed "transition" of $NiGa_2S_4$ might be originated from a vortex-induced topological transition. The relevant vortices could be (i) Z_2 vortices based on the noncollinear AF order for smaller $|K|$, (ii) Z_2 vortices based on the FQ order for largely positive K, and (iii) quarternion vortices based on the AFQ order for largely negative K. Whichever situation (i)–(iii) applies, the scenario immediately explains the experimental observation that the spin correlation length remains finite even at and below 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 effect have revealed that, as the impurity concentration is reduced 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 transition 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 $NiGa_2S_4$. Very recent NQR and μSR measurements indicate that static internal fields set in below T_f accompanied with a divergent increase of the correlation time toward T_f, at least within experimental time window.[14] 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 correlation 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 excitations of the noncollinear AF order of the $S = 1$ quantum 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 critical behavior realized below T_V seems consistent with the NQR observation.[14] One may suspect that a weak interplane coupling J', which should exist in real $NiGa_2S_4$, inevitably induces the 3D AF LRO immediately below T_V. However, this is not necessarily the case: If J' is sufficiently small satisfying $J'\xi(T_V)^2 \lesssim k_B T_V$, $\xi(T_V)$ being the spin correlation length at T_V, the 3D AF LRO needs not set in even below T_V. Finiteness of ξ and smallness of J' are essential in preventing the vortex ordered state from forming the 3D AF LRO. At still lower temperatures, ξ diverges exponentially toward $T = 0$, eventually leading to the onset of the magnetic LRO at a certain temperature $T' < T_V$.

In $NiGa_2S_4$, distant neighbor interactions neglected in the present analysis, particularly the third-neighbor interaction, compete with the nearest-neighbor one leading to an incommensurate spin structure at low temperature.[1] We note that the vortex transition discussed here is not specific to the 120° spin structure realized in the nearest-neighbor model, but generically expected for the noncollinear spin order including the incommensurate one, although details of the transition needs to be clarified further. The vortex scenario might also apply to the $S = 3/2$ triangular AF $NiCrO_2$.[3]

Finally, the noncollinear AF order might explain another noticeable feature of experiments that the T^2 specific heat is quite robust against applied magnetic fields.[1] This experimental observation is rather surprising since applied fields reduce the Hamiltonian symmetry from O(3) to O(2), leading to smaller number of Goldstone modes, i.e., from three to one. As discussed in ref. 16, an interesting observation here is that the noncollinear AF ground state in magnetic fields is capable of keeping an "accidental" degeneracy not related to the Hamiltonian symmetry O(2), essentially of the same amount as in the zero-field case. In fact, even in applied fileds, the ground-state manifold still retains *three* continuous parameters which can be set freely, just as in the zero-field case. One of these three is of symmetry origin, i.e., a true Goldstone mode, while other two are not of symmetry origin (accidental), i.e., *pseudo*-Goldstone modes. Thus, at the classical level, such pseudo-Goldstone modes may account for the robustness of the low-temperature specific heat in applied fields, 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 transition. The relevant vortices could be of three different types, depending on the value of the biquadratic coupling. It was then suggested that the peculiar phase transition recently observed in $NiGa_2S_4$ might have its origin in such a vortex-induced topological transition.

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