

Chiral phase of the Heisenberg antiferromagnet with a triangular lattice

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In the presence of next-to-nearest-neighbor exchange interaction the classical ground state of the Heisenberg antiferromagnet with a triangular lattice in a wide domain of parameters has a four-sublattice structure. This ground state is highly degenerate: the only restriction for the spins on four sublattices is that their sum should be equal to zero. The accidental degeneracy is removed by fluctuations or nonbilinear interactions of spins. Fluctuations (quantum or thermodynamical) favor the collinear arrangement of spins, but a four-spin exchange interaction favors a nonplanar configuration that can be characterized by positive or negative chirality. In the presence of such an interaction, the long-range order in chiralities will survive up to some finite temperature although the long-range order in spin variables will be destroyed by fluctuations at an arbitrarily low temperature.

Due to recent developments in high- T_c superconductivity there was a revival of interest in the quantum Heisenberg antiferromagnet with a triangular lattice (HAFMTL), which as early as in 1973 was conjectured to possess a nontrivial spin-liquid ground state.¹ In particular the idea was put forward that if the quantum fluctuations destroy the long-range order in spin variables the long-range ordering of chiralities may still persist.^{2,3} Chirality is a pseudoscalar variable defined on each elementary triangular plaquette:

$$\chi = \mathbf{S}_i[\mathbf{S}_j \times \mathbf{S}_k] \quad (1)$$

that can be associated with some kind of short-range order corresponding to the nonplanar arrangement of spins. Here i , j , and k are the sites at the corners of the plaquette.

In this paper we show that the classical ground state of HAFMTL with nearest-neighbor (NN), next-to-nearest-neighbor (NNN), and also four-particle exchange interaction in a wide domain of parameters has a four-sublattice structure with a nonplanar configuration of spins. That makes possible the existence at finite temperatures of the chiral phase in which there is no long-range order in spin variables but the long-range order in chirality does exist. Baskaran³ had looked for such a phase in the quantum HAFMTL with $S = \frac{1}{2}$ and in the framework of the mean-field approach has obtained the result that it can be stabilized just by NNN exchange interaction. Our analysis shows that fluctuations (both quantum and thermal) do not favor the nonplanar short-range order, so to overcome them the four-spin interaction should be also included into the Hamiltonian.

The ground state of the classical HAFMTL with only NN exchange interactions is known to consist of three sublattices [Fig. 1(a)], the spins belonging to the same sublattice being parallel to each other. The spins on different sublattices should be lying in the same plane and form angles 120° with each other.⁴ The degeneracy space

for such a state is given by the group of three-dimensional rotations $SO(3)$. As in the case of the nonlinear σ model,⁵ thermal fluctuations destroy the ordering in such a system, making the correlation radius finite for any nonzero temperature.⁶

Recently Jolicoeur *et al.*⁷ have studied the HAFMTL with both NN and NNN exchange interaction, that is the model described by the Hamiltonian

$$H = J_1 \sum_{NN} \mathbf{S}_i \cdot \mathbf{S}_j + J_2 \sum_{NNN} \mathbf{S}_i \cdot \mathbf{S}_j \quad (2)$$

and have found that antiferromagnetic ($J_2 > 0$) NNN exchange favors a four-sublattice state [Fig. 1(b)], which in the classical limit becomes the ground state for

$$J_1/8 < J_2 < J_1. \quad (3)$$

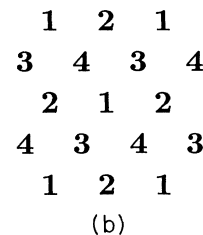
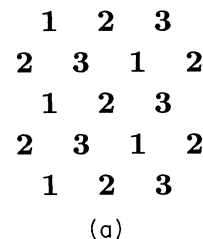


FIG. 1. Partition of the triangular lattice into three (a) or four (b) sublattices.

When speaking about the classical limit (or classical model) we always have in mind the case of $S \rightarrow \infty$ when spin \mathbf{S} can be treated not as an operator but as a vector with constant length. In that case one can always make a rescaling and consider \mathbf{S} as a unit vector.

The authors of Ref. 7 assumed that in the ground state all the spins should be lying in the same plane and obtained that the four sublattices should be forming two

pairs so that in two sublattices belonging to the same pair the spins would be antiparallel to each other. But the angle between the spins belonging to different pairs of sublattices was found to be a free variable with respect to which the ground state is degenerate.

Actually the degeneracy is even higher. The energy of the four-sublattice state (per site) is equal to

$$E_4 = \frac{1}{2}(J_1 + J_2)(\mathbf{S}_1 \cdot \mathbf{S}_2 + \mathbf{S}_1 \cdot \mathbf{S}_3 + \mathbf{S}_1 \cdot \mathbf{S}_4 + \mathbf{S}_2 \cdot \mathbf{S}_3 + \mathbf{S}_2 \cdot \mathbf{S}_4 + \mathbf{S}_3 \cdot \mathbf{S}_4) \\ \equiv \frac{1}{4}(J_1 + J_2)[(\mathbf{S}_1 + \mathbf{S}_2 + \mathbf{S}_3 + \mathbf{S}_4)^2 - (\mathbf{S}_1^2 + \mathbf{S}_2^2 + \mathbf{S}_3^2 + \mathbf{S}_4^2)], \quad (4)$$

and therefore the only restriction for the ground state is that the sum of four spins should be equal to zero, but there is no need for them to be coplanar. Thus in addition to global rotations of all spins there are *two* additional continuous degrees of freedom. Essentially different ground states can be parametrized, for example, by the angle Θ between \mathbf{S}_1 and \mathbf{S}_2 (which equals the angle between \mathbf{S}_3 and \mathbf{S}_4) and the angle Φ between the planes in which the former and the latter pairs of spins are lying.

This degeneracy is accidental but still survives if bilinear exchange interaction of further neighbors is taken into account. It seems worthwhile to note that in the case of the square lattice analogous accidental degeneracy of the four-sublattice state appears only on the critical line $J_1 = 2J_2$,⁸ whereas for the triangular lattice it is present in a wide domain of parameters.

It is well known that accidental degeneracy in systems with continuous degrees of freedom is usually removed by thermal or quantum fluctuations.⁹⁻¹² Jolicœur *et al.*⁷ have calculated the energy of quantum fluctuations for the Hamiltonian (2) (the first term in $1/S$ expansion) and have discovered that among the coplanar states it is always minimal for collinear configurations of spins when the four-sublattice state reduces to a two-sublattice state (Fig. 2). Our extension of the calculations of Ref. 6 to nonplanar states has shown that for them the energy of quantum fluctuations (or the free energy of thermal fluctuations) is also larger than for the collinear configuration.

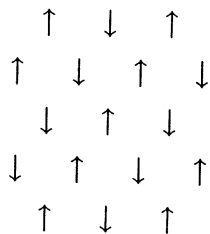


FIG. 2. Two-sublattice state. There are three equivalent possibilities to divide a triangular lattice into two sublattices. For each of these states the short-range correlations are ferromagnetic in one direction and antiferromagnetic in the perpendicular direction.

We considered the states in which two pair of spins are lying in the planes that are perpendicular to each other ($\sin \Phi = \pm 1$). In the harmonic approximation (the leading term in the $1/S$ expansion) the spin-wave spectrum for such states has the form

$$\omega^2 = S\{ K_1[K_1 \cos \Theta + (K_2 + K_3) \sin^2(\Theta/2)] \\ - (K_2 - K_3)^2 \cos^2(\Theta/2) \}, \quad (5)$$

where

$$K_1 = J_1(1 - \cos q_1) + J_2[1 - \cos(q_2 - q_3)], \\ K_2 = J_1(1 - \cos q_2) + J_2[1 - \cos(q_3 - q_1)], \\ K_3 = J_1(1 - \cos q_3) + J_2[1 - \cos(q_1 - q_2)],$$

and $q_\alpha \equiv \mathbf{q} \cdot \mathbf{a}_\alpha$ ($\alpha = 1, 2, 3$) are the products of the momentum \mathbf{q} on the three smallest periods \mathbf{a}_α of the triangular lattice ($q_1 + q_2 + q_3 \equiv 0$). This spectrum incorporates four gapless modes: three with linear dispersion and one with quadratic. Summation over the Brillouin zone shows that the maximum of the fluctuations energy (the sum of $\omega/2$) or free energy (the sum of $T \ln[2 \sinh(\omega/2T)]$) is always achieved at the tetrahedral configuration in which all four spins are forming equal angles $\Theta = \arccos(-\frac{1}{3})$ with each other and the minimum at the collinear configuration ($\cos \Theta = 1$). This property still survives if the next-to-next-to-nearest-neighbor interaction is included into the analysis.

Thus the fluctuations reduce the degeneracy space of the system from the five-dimensional manifold to $O(3) \times Z_3$, where $O(3)$ is the group of rotations of the three-dimensional vector and Z_3 corresponds to three possibilities to form the collinear state by dividing the triangular lattice into two sublattices (that is, to form two pairs from four sublattices). The reduction of the degeneracy is exactly the same as in three-dimensional antiferromagnet with an fcc lattice.¹¹ But in contrast to the three-dimensional case the continuous symmetry related to the non-Abelian group in accordance with the general result of Polyakov⁵ is restored at arbitrarily low temperature, whereas the phase transition related to the Z_3 group takes place at a finite temperature. That means that at low temperatures there is no long-range order in the orientation of spins, but the direction (in the real

space) for which the short-range correlations are ferromagnetic is the same for the whole system. In the high-temperature phase this triple degeneracy is removed.

Let us now discuss what happens if the multiparticle exchange processes are also included into consideration. For the most popular case of $S = \frac{1}{2}$ the form of the spin interaction in the presence of multiparticle exchange was thoroughly analyzed in Ref. 13. In that case the term related to three-particle cyclic exchange reduces to renormalization of two-particle interaction, but the four-particle cyclic exchange: $i \rightarrow j \rightarrow k \rightarrow l \rightarrow i$ leads to four-spin interaction of the form

$$V_4 = J_4[(\mathbf{S}_i \cdot \mathbf{S}_j)(\mathbf{S}_k \cdot \mathbf{S}_l) + (\mathbf{S}_i \cdot \mathbf{S}_l)(\mathbf{S}_k \cdot \mathbf{S}_j) - (\mathbf{S}_i \cdot \mathbf{S}_k)(\mathbf{S}_j \cdot \mathbf{S}_l)], \quad J_4 > 0. \quad (6)$$

An analogous four-spin interaction appears also in the next-to-leading term of the t/U expansion if the Hamiltonian of HAFMTL is deduced from the Hamiltonian of the Hubbard model.¹⁴

After summation over all possible rhombic exchange paths (formed by two elementary triangles) the correction to classical energy of the four-sublattice state will be given by

$$\Delta E_4 = J_4[(\mathbf{S}_1 \cdot \mathbf{S}_2)(\mathbf{S}_3 \cdot \mathbf{S}_4) + (\mathbf{S}_1 \cdot \mathbf{S}_3)(\mathbf{S}_2 \cdot \mathbf{S}_4) + (\mathbf{S}_1 \cdot \mathbf{S}_4)(\mathbf{S}_2 \cdot \mathbf{S}_3)]. \quad (7)$$

In terms of the angles Θ and Φ that we introduced earlier ΔE_4 can be expressed as

$$\Delta E_4 = J_4 S^2 [1 + 2 \cos^2 \Theta - \frac{1}{2}(1 - \cos \Theta)^2 \sin^2 \Phi] \quad (8)$$

and for $J_4 > 0$ is minimal when all four spins are forming equal angles with each other ($\cos \Theta = -\frac{1}{3}$, $\sin \Phi = \pm 1$), that is, for the nonplanar tetrahedral configuration of spins which from the point of view of fluctuations is the worst. On the other hand, the maximum of ΔE_4 is achieved for the two-sublattice state (Fig. 2) with a collinear arrangement of spins ($\cos \Theta = -1$, $\sin \Phi = 0$, or $\cos \Theta = 1$) for which the free energy of the fluctuations is the lowest.

For the nonplanar configuration of spins minimizing ΔE_4 the value of chirality χ is equal either to $(\frac{16}{27})^{1/2}$ or $-(\frac{16}{27})^{1/2}$, but is the same for all elementary plaquettes of the lattice. That means that arbitrarily small positive J_4 reduces the degeneracy of the classical ground state from the five-dimensional to the three-dimensional manifold consisting of two disconnected parts: $\text{SO}(3) \times Z_2$. Here $\text{SO}(3)$ corresponds to global rotation of all spins and Z_2 is related to twofold degeneracy in the sign of χ . As in the previous case arbitrarily low temperature will restore the continuous symmetry related to the non-Abelian group but the ferromagnetic ordering of chiralities will persist up to some finite temperature where the Ising-type transition will take place.

There is absolutely no reason for two transitions to

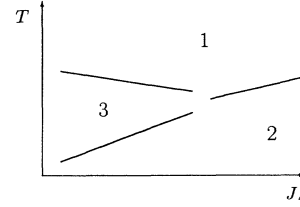


FIG. 3. General structure of the phase diagram of classical HAFMTL induced by the four-particle exchange interaction. Two-particle interactions are assumed to correspond to the domain of stability of the four-sublattice ground state. 1—disordered phase, 2—doubly degenerate phase with long-range order in chirality (nonplanar short-range order), 3—triply degenerate phase with collinear short-range order.

occur simultaneously. The disordering related to continuous degeneracy takes place due to gapless spin-wave fluctuations whereas the destruction of twofold Ising-type symmetry calls for the proliferation of the domain walls. As the domain wall energy per unit length E_{DW} is finite this can happen only at finite temperature which is proportional to E_{DW} . If the low-temperature corrections to different quantities are calculated the correction to \mathbf{S}_i or $\mathbf{S}_i \times \mathbf{S}_j$ will be logarithmically divergent whereas the correction to χ will be finite. The gapless modes cannot make a divergent contribution to $\Delta\chi$ since a strictly uniform rotation of all spins does not change the chirality and any correction from the gapless modes appears only due to the nonuniformity of the rotation. This provides an additional small factor which makes the integral convergent.

For $J_4 S^2 \ll J_1, J_2$ the doubly degenerate nonplanar phase is stable only at low temperatures and with increase in temperature a first-order transition takes place into the triply degenerate collinear phase for which the free energy of fluctuations is lower. The general structure of the phase diagram is depicted in Fig. 3. An analogous phase diagram would be obtained if instead of four-particle exchange interaction (6) we add to Hamiltonian (2) biquadratic terms for two-particle NN exchange: $J_{12}(\mathbf{S}_i \cdot \mathbf{S}_j)^2$ with $J_{12} > 0$.

Thus we have shown that in the classical HAFMTL a reasonable choice of the interactions leads to the stabilization of the phase with nonplanar short-range order in the configuration of spins and therefore with the long-range order in chiral variable χ which survives at finite temperatures. Quantum fluctuations reduce (not increase) the domain of stability of this nontrivial phase, shifting it to higher J_4 . Nonetheless for high enough J_4 it can be expected to survive even for the case of $S = \frac{1}{2}$. On the other hand, for small values of S quantum fluctuations may destroy long-range order in spin variables even at zero temperature.

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¹P.W. Anderson, *Mater. Res. Bull.* **8**, 153 (1973).

²X.-G. Wen, F. Wilczek, and A. Zee, *Phys. Rev. B* **39**, 11 413 (1989).

³G. Baskaran, *Phys. Rev. Lett.* **63**, 2524 (1989).

⁴H. Kawamura and S. Miyashita, *J. Phys. Soc. Jpn.* **53**, 4138 (1984).

⁵A.M. Polyakov, *Phys. Lett.* **59B**, 79 (1975).

⁶P. Azaria, B. Delamotte, and Th. Jolicoeur, *Phys. Rev. Lett.* **64**, 3175 (1990).

⁷Th. Jolicoeur, E. Dagotto, E. Gagliano, and S. Bacci, *Phys. Rev. B* **42**, 4800 (1990).

⁸P. Chandra and B. Douçout, *Phys. Rev. B* **38**, 9335 (1988).

⁹J. Villain, R. Bidaux, J.P. Carton, and R. Conte, *J. Phys. (Paris)* **41**, 1263 (1980).

¹⁰S.E. Korshunov, *J. Phys. C* **19**, 5927 (1986).

¹¹C.L. Henley, *J. Appl. Phys.* **61**, 3962 (1987).

¹²A.V. Chubukov and D.I. Golosov, *Pis'ma Zh. Eksp. Teor. Fiz.* **50**, 416 (1989) [*JETP Lett.* **50**, 451 (1989)]; *J. Phys. Condens. Matter* **3**, 69 (1991).

¹³M.C. Cross and D.S. Fisher, *Rev. Mod. Phys.* **57**, 881 (1985).

¹⁴A.H. MacDonald, S.M. Girvin, and D. Yoshioka, *Phys. Rev. B* **37**, 9753 (1988).