Incommensurate Single-Angle Spiral Orderings of Classical Heisenberg Spins on Zigzag Ladder Lattices

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Exact and rigorous solutions of the ground-state problem for the classical Heisenberg model with nearest-neighbor interactions on two- and three-dimensional lattices composed of zigzag (triangular) ladders are obtained in a very simple way, with the use of a cluster method. It is shown how the geometrical frustration due to the presence of triangles as structural units leads to the emergence of incommensurate spiral orderings and their collinear limits. Interestingly, these orderings are determined by a single angle (along with the signs of the interactions between neighboring spins); therefore, the term “single-angle spiral orderings” is proposed.

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In geometrically frustrated systems, the energies of all interactions cannot be simultaneously minimized because of geometric constraints [1]. There is no general algorithm for determining ground states even for classical frustrated systems. For this reason, the ground-state problem for many classical Heisenberg spin models still remains a challenging task for theorists [2, 3]. The most known and used method for these purposes is that of Luttinger and Tisza [4] and its generalizations [5]. However, it appears not to be useful for complicated Hamiltonians (for instance, with an external field or a biquadratic exchange) and difficult to apply to non-Bravais lattices.

There is also a cluster method proposed by Lyons and Kaplan half a century ago [6], but it is “rather unknown,” although simple and intuitively clear [7, 8]. We developed this method in our studies of the ground states for some Ising-type models [9–11]. Here, we consider the classical Heisenberg model on a set of lattices for which, despite the presence of frustration and a non-Bravais character of the lattices, a solution of the ground-state problem can be obtained in a remarkably simple and clear way, with the use of the cluster method. These are two- and three-dimensional lattices composed of zigzag (triangular) ladders. There are quite a few compounds where magnetic atoms are arranged in lattices of this type [12–22]. Making use of the cluster method, we clearly and rigorously show how the presence of structural triangles and, hence, the geometric frustration, leads to the emergence of incommensurate spiral orderings. An important point is that, despite the presence of several interaction parameters, these orderings are determined by a single angle (together with the signs of the interactions between neighboring spins along the ladder rungs). Therefore, we introduce the term “single-angle spiral orderings.”

Since triangular ladders are composed of identical isosceles triangles, let us start with a single isosceles triangle and classical spins (unit 3 vectors) at its vertexes (Fig. 1). \( L \) and \( K \) are the strengths of couplings, and \( \alpha \), \( \beta \), and \( \gamma \) are the angles between the spins.

The energy of the system reads

\[
E = K \cos \alpha + L(\cos \beta + \cos \gamma).
\]  

(1)

What are the values of \( \alpha \), \( \beta \), and \( \gamma \) that minimize the energy? The \( K < 0 \) case is trivial: the spins are collinear, because there is no frustration, that is the interaction energy can be minimized by pairs. If \( K > 0 \) (the case of frustration), then, at a fixed angle \( \alpha = 2\phi \), the minimum value of the energy is attained when \( \cos \beta + \cos \gamma \) is maximum or minimum, depending on the sign of \( L \). Since \( \alpha + \beta + \gamma \leq 2\pi \), it is easy to see that, in the \( L < 0 \) case, the sum is maximum, if the spins are coplanar and \( \beta = \gamma = \alpha/2 = \phi \) [Fig. 1(b)], and in the \( L > 0 \) case, this sum is minimum, if the spins are coplanar and \( \beta = \gamma = \pi - \alpha/2 = \pi - \phi \) [Fig. 1(c)].

The ground-state energy of the triangle equals to

\[
E_{gs} = K \cos 2\phi - 2|L| \cos \phi.
\]  

(2)

The energy is minimum if \( \cos \phi = \frac{|L|}{2K} \). Thus, at \( |L| \leq 2K \), the ground state is determined by the angle \( \phi \) and

\[
\phi = \frac{1}{2} \arccos \left( \frac{|L|}{2K} \right).
\]
the spins are coplanar (the polarization plane, the orientation of one of the three spins, and the chirality are arbitrary), otherwise the spins are collinear.

Consider now a triangular ladder (Fig. 2). It is composed of identical triangle clusters. Each $J$-bond belongs to two such clusters; therefore, one should minimize the energy of a triangle cluster with the interactions $J_0$ and $J/2$. In view of the previous result, we can assert that the ground state is spiral and determined by the angle $\phi = \arccos \frac{J}{J_0}$ if $|J| \leq 4J_0$, otherwise all the spins are collinear. Two examples of the ground-state ordering for a triangular ladder are shown in Fig. 2. It should be emphasized that all the spins are coplanar but the polarization plane is arbitrary. Since the angle $\phi$ depends on the strengths of couplings only, the spin ordering is, in general, incommensurate along legs.

It is of interest to consider a 2D-lattice composed of two types of triangular ladders with nearest-neighbor interaction $J_0$ along legs and $J_1$ and $J_2$ along rungs (Fig. 3). The lattice can be partitioned into identical two-triangle clusters. Each $J_1$- and $J_2$-bond belongs to two such clusters; therefore, one should minimize the energy of a two-triangle cluster with the interactions $J_0$, $J_1/2$, and $J_2/2$. The $J_0 < 0$ case is trivial: there is no frustration. Consider the $J_0 > 0$ case. Let the angle between the vectors at the ends of the $J_0$-bond be fixed and equal to $2\phi$ ($0 \leq \phi \leq \pi/2$). To minimize the energy, the angles between neighboring spins joint by the $J_i$-bonds ($i = 1, 2$) should be equal to $\phi$ if $J_i < 0$ or to $\pi - \phi$ if $J_i > 0$. The energy per cluster is

$$E_{gs} = J_0 \cos 2\phi - (|J_1| + |J_2|) \cos \phi.$$  

(3)

FIG. 3: (Color online) Left: A generalized triangular lattice and a spin configuration on it for $J_1 < 0$ and $J_2 > 0$ ($J_0 > 0$, $|J_1| + |J_2| \leq 4J_0$). A two-triangle cluster is shown by bold lines. Right: Square of spiral phases for the generalized triangular lattice. As the border of the square is approached, $\phi = \arccos \frac{|J_1| + |J_2|}{4J_0}$ tends to zero and the spiral phases continuously evolve to their collinear limits.

The angle $\phi$ that minimizes this energy is given by

$$\cos \phi = \frac{|J_1| + |J_2|}{4J_0}.$$  

(4)

If $|J_1| + |J_2| \leq 4J_0$, then the spin ordering is spiral and determined by the angle $\phi$ and by the signs of $J_1$ and $J_2$, otherwise all the spins are collinear. In accordance with the signs of $J_1$ and $J_2$, there are four spiral phases. The square of spiral phases is shown in Fig. 3 (right panel). One of these is depicted in Fig. 3 (left panel). As the border of the square is approached, $\phi$ tends to zero and the spiral phases continuously evolve to their collinear limits where the spins belonging to the same leg have the same direction, although $J_0 > 0$. If $J_2 = J_1 = J_0 > 0$, then we have a well-known 120°-phase on the triangular lattice.

Let us pass to a 3D zigzag ladder lattice, shown in Fig. 4. We refer to it as a generalized hollandite lattice. The ground states of a classical Heisenberg model on the hollandite lattice (dark cyan and green rungs are identical) were studied in Ref. [21]. There one can find an overview of magnetic compounds with this lattice (see also Ref. [22]). Since the hollandite lattice is not a Bravais one, a straightforward application of the Luttinger-Tisza method is impossible; therefore, the authors used a modification of it to find the ground-state energy; they also performed numerical simulations. The cluster method makes it possible to solve this ground-state problem in a very simple and clear way.

The generalized hollandite lattice is composed of three types of zigzag ladders. Let the strengths of coupling between neighboring spins along legs of all the ladders be $J_0$ (blue bonds) and $J_1$ (red bonds), $J_2$ (green bonds), and $J_3$ (dark cyan bonds) along their rungs (Fig. 4). It is easy to see that this lattice can be partitioned into identical three-triangle clusters shown in Fig. 4.

Each $J_1$-, $J_2$-, and $J_3$-bond belongs to two such clusters; therefore, one should minimize the energy of a three-
triangle cluster with the interactions \( J_0, J_1/2, J_2/2, \) and \( J_3/2 \). In the \( J_0 < 0 \) case, there is no frustration. Consider the \( J_0 > 0 \) case. Let the angle between the spins at the ends of the \( J_0 \)-bond be \textit{fixed} and equal to \( 2\phi \) \((0 \leq \phi \leq \pi/2)\). To minimize the energy, the angles between neighboring spins joint by the \( J_i \)-bonds \((i = 1-3)\) should be equal to \( \phi \) if \( J_i < 0 \) or to \( \pi - \phi \) if \( J_i > 0 \) (see Fig. 1). The energy per cluster (or per site) equals to

\[
E_{gs} = J_0 \cos 2\phi - (|J_1| + |J_2| + |J_3|) \cos \phi. \tag{5}
\]

The angle \( \phi \) that minimizes the energy is given by

\[
\cos \phi = \frac{|J_1| + |J_2| + |J_3|}{4J_0}. \tag{6}
\]

It is clear that all the spins on the lattice should be coplanar that is parallel to a plane but this plane can be arbitrary. If the plane, the orientation of one arbitrarily chosen spin, and the chirality are fixed, then the distribution of angles completely determines the ground-state spin configuration of the lattice at fixed couplings.

Each set of signs of the interactions \( J_1, J_2, \) and \( J_3 \) \((J_0 > 0)\) corresponds to a spiral phase and its collinear limit. Since there are eight sets of signs, there are also eight spiral phases and eight collinear limits. The octahedron of spiral phases and two spiral orderings with their collinear limits are shown in Fig. 5.

The cluster shown in Fig. 4 gives the ground-state orderings of classical Heisenberg spins on many other zigzag ladder lattices (see Fig. 6). Eq. (6) and the octahedron of spiral phases (Fig. 5) are common for all these lattices. Hexagonal zigzag ladder lattices similar to those shown in Figs. 6(a) and 6(b) have been identified in many magnetic compounds \cite{13-20}.

It is also interesting and instructive to consider a generalized triangular lattice composed of three types of zigzag ladder (Fig. 7, left panel). This lattice cannot be partitioned into identical two-triangle clusters. Three types of two-triangle clusters have to be considered. It can be shown that a single-angle spiral ordering of classical Heisenberg spins is also possible in some regions of the parameter space of this model. Let us distribute the energy of each rung between two triangular plaquettes which share it in the manner shown in Fig. 7 (left panel). Coefficients \( \alpha_i \) for this distribution can be arbitrary but should satisfy the conditions \( 0 \leq \alpha_i \leq 1 \).

The energies of two-triangle clusters can be written as

\[
\begin{align*}
E_{12} &= J_0 \cos 2\phi_{12} - 2(\alpha_1|J_1| + (1 - \alpha_2)|J_2|) \cos \phi_{12}, \\
E_{23} &= J_0 \cos 2\phi_{23} - 2(\alpha_2|J_2| + (1 - \alpha_3)|J_3|) \cos \phi_{23}, \\
E_{31} &= J_0 \cos 2\phi_{31} - 2(\alpha_3|J_3| + (1 - \alpha_1)|J_1|) \cos \phi_{31}.
\end{align*}
\]

The condition for the minimum of these energies along with the condition \( \phi_{12} = \phi_{23} = \phi_{31} = \phi \) lead to the set of equations

\[
\begin{align*}
\alpha_1|J_1| + (1 - \alpha_2)|J_2| &= \alpha_2|J_2| + (1 - \alpha_3)|J_3|, \\
\alpha_3|J_3| + (1 - \alpha_1)|J_1| &= \frac{|J_1| + |J_2| + |J_3|}{3}.
\end{align*}
\]

The angle \( \phi \) that minimizes the energies is then given by

\[
\cos \phi = \frac{|J_1| + |J_2| + |J_3|}{6J_0}. \tag{9}
\]

It is easy to show that the region in \((J_1/J_0, J_2/J_0, J_3/J_0)\)-space, where Eqs. (8) and the inequalities \( 0 \leq \alpha_i \leq 1 \) are satisfied, is the polyhedral cone determined by six vectors: \((0, 2, 1), (0, 1, 2), (1, 0, 2), (2, 0, 1), (2, 1, 0), \) and \((1, 2, 0)\) in the first octant (Fig. 7, right panel) and similar polyhedral cones in other octants (not shown in the figure). The single-angle spiral phases exist in the parts of this cone which...
FIG. 6: (Color online) (a)- (d) Some hexagonal zigzag ladder lattices. (e) A zigzag ladder lattice based on the truncated tri-hexagonal tiling 4.6.12. The lattice (c) is a disordered mixture of the lattices (a) and (b). All these lattices are composed of identical clusters shown in Fig. 4. Cyan and black circles denote sites over and under the plane of the figure, respectively. The legs of zigzag ladders are perpendicular to the plane of the figure.

FIG. 7: (Color online) Left: A generalized triangular lattice, composed of three types of zigzag ladders, and three types of two-triangle clusters into which it can be partitioned. Right: Hexagonal pyramid (in red) of a single-angle spiral phase on this lattice for $J_i \geq 0$ ($i = 0$-$3$).

FIG. 8: (Color online) A hexagonal zigzag ladder lattice realized in the $S = \frac{3}{2}$ compound $\beta$-$CaCr_2O_4$ [18, 19] (however, the spin ordering observed in this compound is more complicated than obtained here). In contrast to the lattices shown in Figs. 6(a)-6(d), this lattice can be partitioned only into two types of three-triangle clusters (Fig. 8, right panel). The ground-state energies for these clusters are given by

$$E_1 = J_0 \cos 2\phi_1 - (2\alpha|J_2| + 2\beta|J_3| + |J_4|) \cos \phi_1,$$
$$E_2 = J_0 \cos 2\phi_2 - [|J_1| + 2(1 - \alpha)|J_2| + 2(1 - \beta)|J_3|] \cos \phi_2.$$ (10)

Minimizing these energies with respect to $\phi_1 = \phi_2 = \phi$, we obtain

$$\cos \phi = \frac{|J_1| + 2|J_2| + 2|J_3| + |J_4|}{8J_0}.$$ (11)

The conditions $0 \leq \alpha \leq 1$ and $0 \leq \beta \leq 1$ lead to the inequality

$$||J_4| - |J_1|| \leq 2(|J_2| + |J_3|).$$ (12)

This inequality along with the inequality $|J_1| + 2|J_2| + 2|J_3| + |J_4| \leq 8J_0$ determine the regions for existence of single-angle spiral phases.

To summarize, we have rigorously proven that, in systems of classical Heisenberg spins on zigzag (triangular) ladder lattices, there exist incommensurate ground-state spiral orderings (phases) characterized by a single angle and by the signs of the interactions between neighboring...
spins (along ladder rungs). We propose to name these orderings (phases) “single-angle spiral orderings (phases).”

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