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Ground states of a system of classical spins on an anisotropic triangular lattice and the spin-liquid problem in $NiGa_2S_4$ and $FeGa_2S_4$ compounds

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It is shown that the ground states of a system of classical spins on an anisotropic triangular lattice with interactions within an elementary triangular plaquette can be constructed by minimizing the energy of a single plaquette. Even in the case when all three angles between plaquette spins are different, there exist five global ground-state configurations with equal energies. The most complex of these is an incommensurate four-sublattice conical spiral structure. Our results may shed some light on the experimentally observed spin-liquid-like disorder in $NiGa_2S_4$ and $FeGa_2S_4$ where a four-sublattice spin structure was observed.

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When, in 1936, Néel announced a discovery of a new magnetic state of matter, antiferromagnetism, many physicists were skeptical about this. They based their arguments on quantum properties of spin. So Landau argued that quantum fluctuations lead to mutual spin flips ($|\uparrow\downarrow\rangle \leftrightarrow |\downarrow\uparrow\rangle$) and a disordered Néel's state. However, some time later, many antiferromagnets had been discovered experimentally, and a Néel's period in the physics of magnetism had began and lasted until 1972, when Anderson returned to the above-mentioned arguments and suggested the existence of a so-called spin liquid in a strongly frustrated system, a Heisenberg model on a triangular lattice [1]. Although it was soon shown that neighboring spins in a triangular lattice order at 120°, an intensive search for this new magnetic state of matter that can exist even at the $T = 0$ limit began both theoretically and experimentally (see recent reviews [2,3] and Ref. [4]). And, at last, a spin-liquid state was discovered on a frustrated triangular lattice [5] and, 12 years later, on a more frustrated kagome lattice [6].

At the present, many materials are known where a spin-liquid phase is believed to exist. Almost all these materials are spin-$\frac{1}{2}$ compounds [2,3]. However, it is assumed that a quantum spin liquid can also exist in systems of spins greater than $\frac{1}{2}$. In 2005, Nakatsuji *et al.* announced the existence of a spin-liquid phase in a spin-1 compound, $NiGa_2S_4$, where magnetic atoms of nickel are arranged in layers representing a rare example of an ideal triangular lattice [7–9]. Moreover, the coupling between magnetic atoms of different layers is much less than the coupling within a layer.

Nakatsuji *et al.* identified an interesting four-sublattice spin structure in $NiGa_2S_4$. A similar spin structure and magnetic properties were found in $FeGa_2S_4$ where iron atoms carry spin-2 [7]. In our opinion, this fact suggests that the spin-liquid properties of these compounds can be explained on the basis of a classical spin model. It should be noted here that, in addition to a quantum spin liquid, there is also a classical spin liquid that was found in the so-called spin-ice compounds [10] and in some theoretical models (see Ref. [11] and references therein).

Ground states of a system of classical spins on an isotropic triangular lattice were analyzed in detail in Refs. [12,13] (see also references therein). However, the isotropic triangular lattice, due to the existence of mechanical stress and various defects, can lose its symmetry. Therefore, it is of interest to analyze the ground states of a system of classical spins on an anisotropic triangular lattice. Performing this analysis, we have found some interesting spin arrangements, including a complex four-sublattice order, similar to that given in Refs. [7–9].

Let us first consider an anisotropic triangular plaquette with classical spins (unit 3-vectors) at its vertices and with linear (K, L, and M) and biquadratic (A, B, and C) pairwise interactions between spins (per one plaquette) [Fig. 1(a)]. The Hamiltonian of such a spin system can be written as a sum over all the triangular plaquettes,

$$H = \sum_{\triangle_i}[K\vec{S}_{i1}\cdot\vec{S}_{i2} + L\vec{S}_{i2}\cdot\vec{S}_{i3} + M\vec{S}_{i1}\cdot\vec{S}_{i3} - A(\vec{S}_{i1}\cdot\vec{S}_{i2})^2 - B(\vec{S}_{i2}\cdot\vec{S}_{i3})^2 - C(\vec{S}_{i1}\cdot\vec{S}_{i3})^2], \tag{1}$$

where $\vec{S}_{i1}$, $\vec{S}_{i2}$, and $\vec{S}_{i3}$ are spins at three vertices of the ith triangular plaquette.

Let α, β, and γ be angles between the spins ($0 \le \alpha,\beta, \gamma \le \pi$). These angles should satisfy the following inequalities,

$$\begin{aligned} \alpha + \beta + \gamma &\le 2\pi, \\ -\alpha + \beta + \gamma &\ge 0, \\ \alpha - \beta + \gamma &\ge 0, \\ \alpha + \beta - \gamma &\ge 0. \end{aligned} \tag{2}$$

The solution of this set of inequalities is the tetrahedron, shown in Fig. 1(b). If at least one of the inequalities becomes an equality, then the vectors are coplanar. This corresponds to a point on the surface of the tetrahedron. The coupling energy between the spins of the plaquette shown in Fig. 1(a) reads

$$E = K\cos\alpha + L\cos\beta + M\cos\gamma - A\cos^2\alpha - B\cos^2\beta - C\cos^2\gamma. \tag{3}$$

If the energy attains its minimum in an intrinsic point of the tetrahedron [Fig. 1(b)], then this minimum is determined from the following conditions (partial derivatives are equal to zero),

$$\begin{aligned} \frac{\partial E}{\partial\alpha} &= (-K + 2A\cos\alpha)\sin\alpha = 0, \\ \frac{\partial E}{\partial\beta} &= (-L + 2B\cos\beta)\sin\beta = 0, \\ \frac{\partial E}{\partial\gamma} &= (-M + 2C\cos\gamma)\sin\gamma = 0. \end{aligned} \tag{4}$$

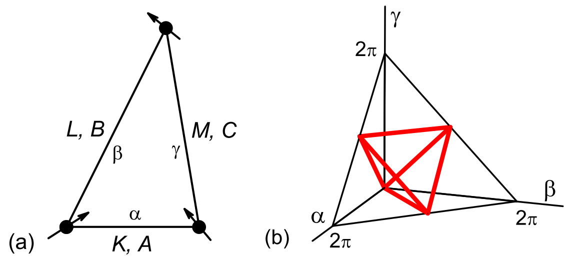

FIG. 1. (a) An elementary triangular plaquette of an anisotropic triangular lattice with three spins at its vertices and pairwise interactions between neighboring spins (linear and biquadratic per one plaquette). (b) Tetrahedron of values for angles α, β, and γ between spins of the plaquette.

We have from this set of equations

$$\cos\alpha = \frac{K}{2A}, \quad \cos\beta = \frac{L}{2B}, \quad \cos\gamma = \frac{M}{2C}. \tag{5}$$

Let us consider the case where all the three angles α, β, and γ are different. How, when having a local ground state, that is, a ground state of a plaquette, can one construct the global ground state of an infinite lattice? For all the triangular plaquettes on the lattice, the angles between corresponding pairs of spins should be α, β, and γ.

The solutions of the equations

$$\vec{a}\cdot\vec{b} = \vec{c}\cdot\vec{d} = \cos\beta, \quad \vec{a}\cdot\vec{c} = \vec{b}\cdot\vec{d} = \cos\alpha \tag{6}$$

(where all the vectors are unit 3-vectors) are the following vectors $\vec{d}_1$ and $\vec{d}_2$ (Fig. 2),

$$\vec{d}_1 = 2\frac{\vec{a}\cdot(\vec{c}-\vec{b})}{(\vec{b}-\vec{c})^2}(\vec{b}-\vec{c}) + \vec{a} = \frac{\cos\alpha - \cos\beta}{1-\cos\gamma}(\vec{b}-\vec{c}) + \vec{a}, \tag{7}$$

$$\vec{d}_2 = 2\frac{\vec{a}\cdot(\vec{b}+\vec{c})}{(\vec{b}+\vec{c})^2}(\vec{b}+\vec{c}) - \vec{a} = \frac{\cos\alpha + \cos\beta}{1+\cos\gamma}(\vec{b}+\vec{c}) - \vec{a}. \tag{8}$$

Transformation (7) (vectors $\vec{b}$ and $\vec{c}$ being fixed) changes the chirality: If the triplet of vectors $\vec{a}$, $\vec{b}$, and $\vec{c}$ is right (left) handed, that is, $\vec{a}\cdot[\vec{b}\times\vec{c}] > 0\,(<0)$, then the triplet of vectors $\vec{d}_1$, $\vec{c}$, and $\vec{b}$ is left (right) handed, that is, $\vec{d}_1\cdot[\vec{c}\times\vec{b}] < 0\,(>0)$. The cone with elements $\vec{a}$, $\vec{b}$, and $\vec{c}$ is invariant under

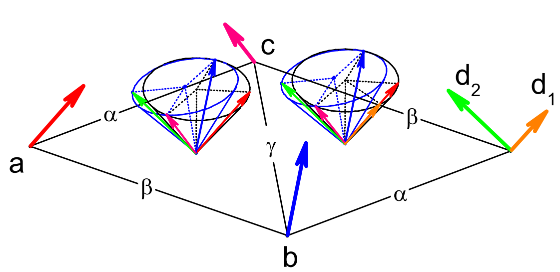

FIG. 2. Two ways of constructing the third spin of the triangular plaquette (vectors $\vec{d}_1$ and $\vec{d}_2$), if two other spins ($\vec{b}$ and $\vec{c}$) are given, as well as the angles (α, β, and γ) between the spin pairs of the plaquette.

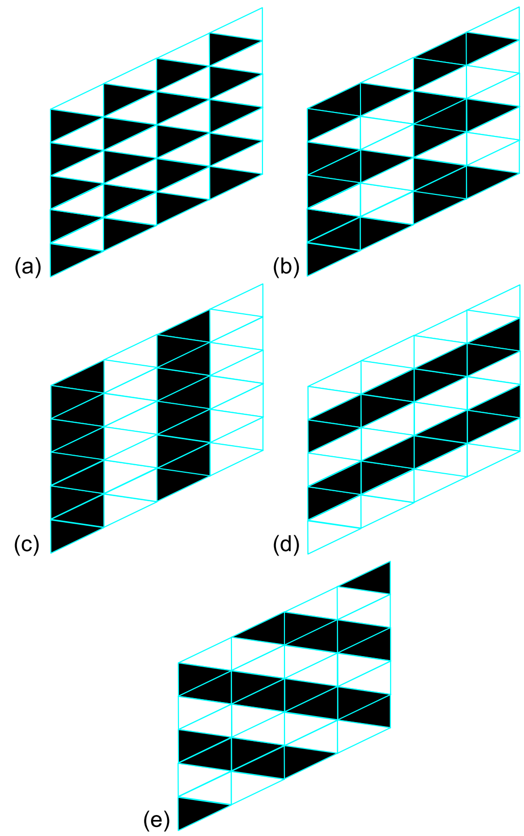

FIG. 3. Three types of structures which are possible for a system of classical Heisenberg spins on an anisotropic triangular lattice with interactions within an elementary triangular plaquette: (a) simple conical spiral structure; (b) four-sublattice conical spiral structure; (c)–(e) two-sublattice conical spiral structures (see Figs. 4–6). Black (white) color of a triangular plaquette means that the triplet of vectors at its vertices is right (left) handed (or vice versa).

transformation (6), that is, the vector $\vec{d}_1$ is also an element of this cone.

Transformation (8) does not change the chirality: If the triplet of vectors $\vec{a}$, $\vec{b}$, and $\vec{c}$ is right (left) handed, then the triplet $\vec{d}_2$, $\vec{c}$, and $\vec{b}$ is also right (left) handed.

So, the global spin configuration of the triangular lattice is fully determined by an arbitrary pair of neighboring spins and by the chirality for each plaquette. Only one condition should be satisfied: *Two plaquettes which are mutually symmetric with respect to their common lattice site should have different chirality* (see the Appendix for a proof). It follows immediately from this that only three types of global chirality configurations for the lattice are possible (see Fig. 3).

Examples of spin configurations for possible chirality configurations of plaquettes are shown in Figs. 4–6. The simplest among them is that which corresponds to Fig. 3(a). This configuration is shown in Fig. 4. It is a simple conical spiral structure where, passing from one site to the neighboring one along the same direction on the lattice, spin rotates to the same angle on the surface of a cone.

The spin structure that corresponds to Fig. 3(b) is the most complex one. It is shown in Fig. 5. This is a four-sublattice conical spiral configuration. The structure on each sublattice

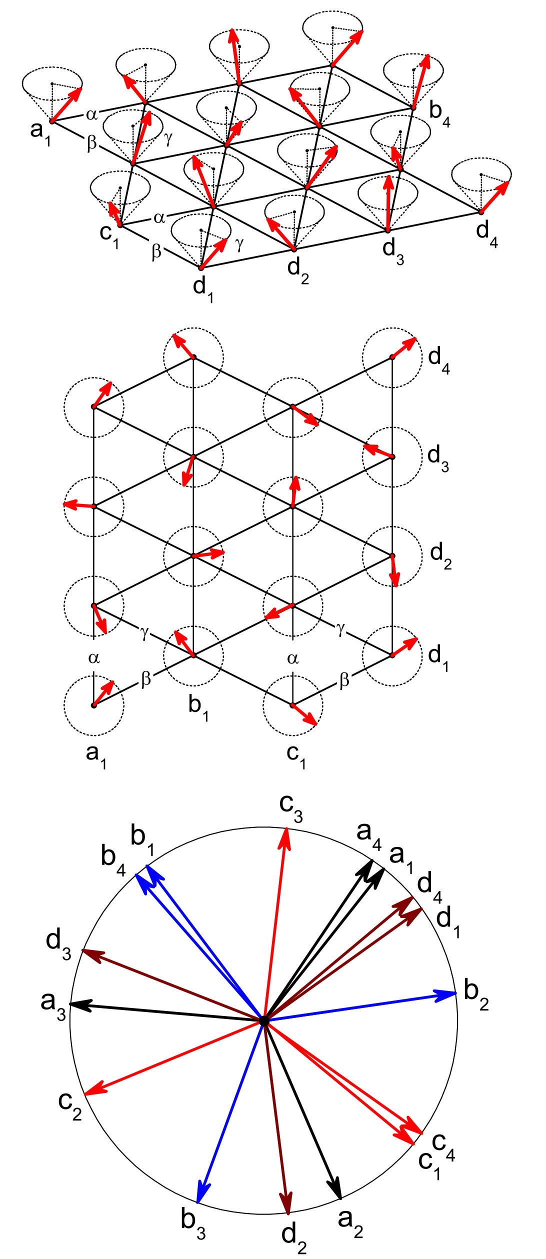

FIG. 4. Conical spiral structure that corresponds to Fig. 3(a). Side and top views are shown. In the lower panel, all the spins are depicted on the same cone (top view).

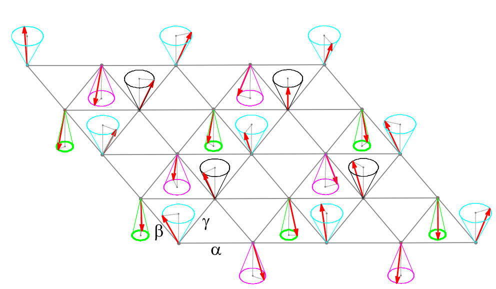

FIG. 5. Four-sublattice spin configuration that corresponds to Fig. 3(b). The cones for different sublattices are depicted in different colors. Within each sublattice, the spin structure is a simple spiral conical structure (see Fig. 4) but on a triangular lattice with doubled lattice periods. The axes of all the cones are parallel.

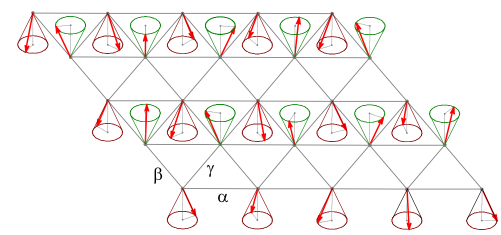

FIG. 6. Two-sublattice spin configuration that corresponds to Figs. 3(c)–3(e). The axis vectors of the cones for different sublattices are antiparallel.

is a simple conical spiral structure (see Fig. 4) on a triangular lattice with doubled lattice periods. The cones for each sublattice are different in the general case, but, as one can easily prove using Eqs. (7) and (8), their axes are parallel.

Figures 3(c)–3(e) correspond to two-sublattice structures. The structure on each sublattice is a simple conical spiral structure as on a square lattice (Fig. 6). The axis vectors of the cones for different sublattices are antiparallel.

To conclude, the ground states of the system of classical spins on an anisotropic triangular lattice with interactions within an elementary triangular plaquette can be constructed by minimizing the energy for a single plaquette. If all the three angles between spins of the plaquette are different, then there are three types of global ground-state structures. The most complex among these is an incommensurate four-sublattice structure. A similar spin arrangement was observed experimentally in $NiGa_2S_4$ and $FeGa_2S_4$ magnetic materials [7–9]. The liquidlike spin disorder in these compounds may be a result of a complex domain structure where there are domains of different types, since all of them have an equal energy.

APPENDIX

To prove that two plaquettes which are mutually symmetric with respect to their common lattice site have different chirality, let us find all the possible chirality configurations of the hexagon shown in the Fig. 7. Let vectors $\vec{a}_1$, $\vec{a}_2$, and $\vec{b}_2$ be specified and

$$\begin{aligned} \vec{a}_1 \cdot \vec{a}_2 &= \cos\alpha = x, \\ \vec{a}_1 \cdot \vec{b}_2 &= \cos\beta = y, \\ \vec{a}_2 \cdot \vec{b}_2 &= \cos\gamma = z. \end{aligned} \tag{A1}$$

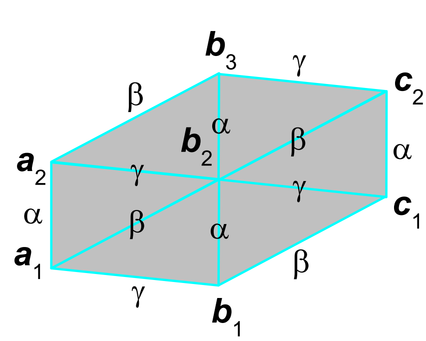

FIG. 7. A hexagon on an anisotropic triangular lattice with unit 3-vectors $\vec{a}_1$, $\vec{a}_2$, $\vec{b}_1$, $\vec{b}_2$, $\vec{c}_1$, and $\vec{c}_2$ at its vertices. Angles between neighboring vectors are indicated.

Then [see Eqs. (7) and (8)],

$$\vec{b}_1 = \frac{z + (-1)^{\beta_1} x}{1 + (-1)^{\beta_1} y}[\vec{a}_1 + (-1)^{\beta_1}\vec{b}_2] - (-1)^{\beta_1}\vec{a}_2,$$
$$\vec{b}_3 = \frac{x + (-1)^{\beta_3} y}{1 + (-1)^{\beta_3} z}[\vec{b}_2 + (-1)^{\beta_3}\vec{a}_2] - (-1)^{\beta_3}\vec{a}_1,$$
$$\vec{c}_1 = \frac{y + (-1)^{\gamma_1} z}{1 + (-1)^{\gamma_1} x}[\vec{b}_1 + (-1)^{\gamma_1}\vec{b}_2] - (-1)^{\gamma_1}\vec{a}_1,$$
$$\vec{c}_2 = \frac{y + (-1)^{\gamma_2} z}{1 + (-1)^{\gamma_2} x}[\vec{b}_2 + (-1)^{\gamma_2}\vec{b}_3] - (-1)^{\gamma_2}\vec{a}_2, \qquad \text{(A2)}$$

where $\beta_1,\beta_3,\gamma_1,\gamma_2 = 0,1$.

Taking into account these equations and the condition $\vec{c}_1 \cdot \vec{c}_2 = \cos\alpha = x$, we obtain (using, for instance, MAPLE software, since the expression for the scalar product is rather cumbersome) only four (from 16) possible sets of values for β_1, β_3, γ_1, and γ_2:

$$\beta_1 = 0, \quad \beta_3 = 0, \quad \gamma_1 = 1, \quad \gamma_2 = 1,$$
$$\beta_1 = 0, \quad \beta_3 = 1, \quad \gamma_1 = 0, \quad \gamma_2 = 0,$$
$$\beta_1 = 1, \quad \beta_3 = 0, \quad \gamma_1 = 0, \quad \gamma_2 = 0,$$
$$\beta_1 = 1, \quad \beta_3 = 1, \quad \gamma_1 = 1, \quad \gamma_2 = 1. \qquad \text{(A3)}$$

Zero means that the chirality does not change and 1 means a change in chirality. It follows from this that two plaquettes which are mutually symmetric with respect to their common lattice site should have different chirality and, therefore, only eight chirality configurations of the hexagon are possible (see Fig. 8).

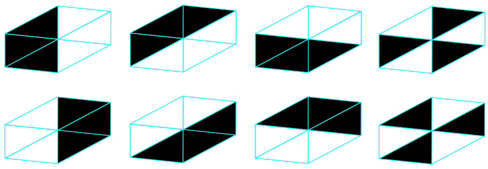

FIG. 8. Eight chirality configurations for a hexagon which are possible for a system of classical Heisenberg spins on a anisotropic triangular lattice with interactions within an elementary triangular plaquette. Black (white) color of a triangular plaquette means that the triplet of vectors at its vertices is right (left) handed (or vice versa).

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