Lattice-coupled Antiferromagnet on Frustrated Lattices

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Lattice-coupled antiferromagnetic spin model is analyzed for a number of frustrated lattices: triangular, Kagomé, and pyrochlore. In triangular and Kagomé lattices where ground state spins are locally ordered, the spin-lattice interaction does not lead to a static deformation of the lattice. In the pyrochlore structure, spin-lattice coupling supports a picture of the hexagon spin cluster proposed in the recent experiment[5]. Through spin-lattice interaction a uniform contraction of the individual hexagons in the pyrochlore lattice can take place and reduce the exchange energy. Residual hexagon-hexagon interaction takes the form of a 3-states Potts model where the preferred directions of the spin-loop directors for nearby hexagons are mutually orthogonal.

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The continued interest in insulating antiferromagnet appears to have a twofold objective. One is the search for a novel type of quantum ground state, other than the conventional Néel-ordered state, particularly in two dimensions. A resonating-valence-bond ground state, if it should exist, is believed to lead naturally to a superconducting phase when doped with holes. In a separate development, understanding the nature of antiferromagnetic ground state defined on the frustrated lattice, even at the classical level, has been the focus of much theoretical as well as experimental activity.

In both these cases the fundamental Hamiltonian describing the spin interaction is

\[ H = \sum_{\langle ij \rangle} J_{ij} S_i \cdot S_j \quad (J_{ij} > 0) \quad (1) \]

defined for an appropriate set of bonds \( \langle ij \rangle \). The antiferromagnetic interaction for an insulating magnet is mediated by the superexchange process in which overlap of Wannier orbitals localized at different positions provides the necessary (virtual) hopping mechanism. As such, it is not surprising that the exchange energy \( J_{ij} \) should depend on the separation of orbitals. Existing experimental data suggests that \( J_{ij} \) falls off as 6-14th power of the separation.

Writing the equilibrium position of the ions by \( i \) and \( j \), and the displacement vector of each ion by \( u_i \) and \( u_j \), the exchange integral has the expansion in the small displacements

\[ J_{ij} = J(|i + u_i - j - u_j|) \approx J_0 - J_1 \hat{e}_{ij} \cdot (u_j - u_i) \quad (2) \]

where \( \hat{e}_{ij} = (j - i)/(|j - i|) \) is the unit vector, and \( J_0 \) and \( J_1 \) are positive constants. For an Einstein, or optical, phonon mode and ignoring the kinetic energy of the displacement, we arrive at the lattice-coupled spin model

\[ H = \sum_{\langle ij \rangle} (J_0 - J_1 \hat{e}_{ij} \cdot (u_j - u_i)) S_i \cdot S_j + \frac{K}{2} \sum_i u_i^2. \quad (3) \]

The purpose of this paper is to analyze the ground state of the model Hamiltonian, Eq. (3), for a number of frustrated lattices: triangular, Kagomé, and pyrochlore.

By rescaling the displacements, \( u_i \rightarrow u_i/\sqrt{K} \), \( J_1 \) is rescaled to \( J_1/\sqrt{K} \equiv \alpha \), while the overall energy scale is fixed by \( J_0 \), which is set to one. The reduced Hamiltonian has the form

\[ H = \sum_{\langle ij \rangle} S_i \cdot S_j - \alpha \sum_i u_i \cdot f_i + \frac{1}{2} \sum_i u_i^2 \quad (4) \]

with \( f_i = \sum_{j \in i} \hat{e}_{ij} S_i \cdot S_j \). We introduce the notation \( j \in i \) to indicate all bonds \( j \) that are exchange-coupled to site \( i \). Minimizing the energy gives the condition relating the lattice positions with the spins:

\[ u_i/\alpha = \langle f_i \rangle = \sum_{j \in i} \hat{e}_{ij} \langle S_i \cdot S_j \rangle. \quad (5) \]

Classical antiferromagnet on a triangular lattice has a ground state characterized by a 120° angle between a pair of adjacent spins. Long-range order is established at the mean-field level. When Eq. (4) is applied to the ground state spin arrangements of the triangular antiferromagnet, one finds \( u_i = 0 \). Unrestricted numerical solution of Eq. (4) together with the classical mean-field equations for the spin average \( \langle S_i \rangle \) also yields \( u_i = 0 \), consistent with a preliminary Monte Carlo simulation of the model Eq. (3). Thus, at the classical level, the ground state of the classical antiferromagnet on a triangular lattice is unaffected by the coupling to lattice. Employing the acoustic phonon model, \( (K/2) \sum_{\langle ij \rangle} (u_i - u_j)^2 \), alters the mean-field equation to \( \sum_{j \in i} (u_i - u_j)/\alpha = \langle f_i \rangle \), which also gives zero for \( u_i \). The effective Hamiltonian after “integrating out” the displacement \( u_i \),

\[ H_{eff} = \sum_{\langle ij \rangle} S_i \cdot S_j - \frac{1}{2} \alpha^2 \sum_i f_i^2, \quad (6) \]
can be expanded around its classical minimum by use of the Holstein-Primakoff (HP) theory. Up to quadratic order in the HP bosons,

$$H_{\text{eff}} = E_0 + \frac{z S^2}{2} \sum_k \left[ \omega_k \left( b_k^+ b_k + \frac{1}{2} \right) - \frac{A_k}{2} \right]$$

(7)

with

$$A_k = 1 + \frac{\gamma_k}{2}, \quad B_k = \frac{3 \gamma_k}{4},$$

$$E_0 = -\frac{1}{4} N z S^2, \quad \omega_k = \sqrt{A_k^2 - (2 B_k)^2},$$

where $\gamma_k = \frac{1}{2} \sum_{i(\tau)} e^{i k (r_j - r_i)}$, $z = 6$ is the coordination number for triangular lattice, and $N$ is the number of sites. There are no terms in the effective Hamiltonian proportional to $\alpha^2$ up to quadratic order in the HP bosons, and the “vacuum” of the Hamiltonian remains unaltered after the introduction of spin-lattice coupling.

Average of $f_i$ can be worked out within the quadratic theory, yielding

$$\langle f_i \rangle = \left[ \frac{S^2}{2} - \frac{S}{N} \sum_k \left( \omega_k \left( n_B(k) + \frac{1}{2} \right) - \frac{A_k}{2} \right) \right] \sum_{j \in i} \hat{e}_{ji} = 0$$

(8)

for the triangular lattice, and $n_B(k)$ is the boson occupation number of energy $\omega_k$. Therefore, small quantum or thermal fluctuation fails to produce a lattice distortion, or “spin-Peierls effects” as it is often known in the literature. The higher-order terms in the HP bosons may have interesting consequences, including a spontaneous distortion of the lattice, and shall be considered in the future.

Classical ground state of the antiferromagnet on the Kagomé lattice is defined by the condition $\sum_{i \in \tau} \vec{S}_i = 0$ where $\tau$ is the basic building block and $S_i$ is the total spin of the $i$-th site. This leads to the well-known ground-state degeneracy of order $2^N$, where $N$ is the number of triangles in the lattice. Nevertheless, ground-state spin-spin correlation $S_i \cdot S_j$ equals $-1/2$ for all nearest-neighbor pairs $(ij)$. Although there is no long-range order, the spins are locally ordered, with a coherence length of one lattice spacing. The spin ordering patterns for triangular and Kagomé lattices are summarized in Fig.1.

The r.h.s. of Eq.(6) gives zero for the Kagomé lattice, as in the triangular lattice, because the local spin structures are the same in both lattices, $S_i \cdot S_j = -1/2$. HP boson analysis of the Kagomé lattice also reveals the absence of static lattice distortion, whether with optical or acoustic phonons. Our findings may be summarized as follows: In the triangular lattice where spins are long-range ordered, and the Kagomé lattice which has a local spin ordering without the long-range order, spin-lattice coupling fails to produce the static lattice distortion, or the spin-Peierls effect.

Pyrochlore lattice is distinct from the previous two cases in the sense that spins do not order even locally. Ground state manifold of classical spins on the pyrochlore lattice is defined by the condition $\sum_{i \in \tau} \vec{S}_i = 0$ where the basic building block $\tau$ is a tetrahedron, for all the tetrahedrons forming the lattice. The requirement is not sufficient to determine $S_i \cdot S_j$ uniquely for nearest-neighbor sites, hence the claim that local spin ordering is missing in the pyrochlore lattice.

Interaction energy $-(\alpha^2/2) \sum_i f_i^2$ is minimized for $f_i^2$ maximum at each site. This selects the collinear spins (all spins parallel or antiparallel to each other) as the preferred ground state. The collinear spin patterns and the associated lattice distortion were analyzed in details in Ref.12 following the pioneering work on the spin-Peierls effect in the pyrochlore structure in Ref.12.

Recently, neutron scattering data on the pyrochlore compound ZnCr$_2$O$_4$ revealed a very interesting picture of the spin dynamics. According to Ref.13, each non-overlapping hexagon embedded inside the pyrochlore lattice has six spins form a collinear, antiferromagnetic cluster, which are energetically decoupled from those of other hexagonal clusters. The resulting block spins are chrested “spin-loop directors”, or directors for short, in Ref.13. While this picture is intuitively appealing, no quantitative justification for the formation, and the stability of, such a hexagonal spin cluster appears to exist to date.

In the rest of the paper we show that spin-lattice coupling can aid in the formation of a hexagonal cluster suggested in Ref.13. We argue that each hexagon can shrink - uniform contraction without breaking $C_6$ symmetry - to maximize the gain in exchange energy within the cluster. Spins within each hexagon is antiferromagnetically ordered, at the classical level. The contraction imposes a constraint, via Eq.(9), that the directors of nearby hexagons be mutually orthogonal, thus lifting the huge degeneracy of the original ground state manifold.

We first analyze the case of a single hexagonal antiferromagnetic chain coupled to the lattice as in Eq.(9). The classical ground state of the effective Hamiltonian is given by the staggered spin, $S^z_i = \pm S_{\hat{z}}$. The staggered spins give $u_i$ all pointing inward to the center of the hexagon, as given by

FIG. 1: Ground-state spin configuration of the triangular and Kagomé lattice. $A, B, C$ refer to three orientations of the spin.
\[
\langle f_i \rangle = \left[ S(S+1) - \frac{S}{6} \sum_k \omega_k \left( n_B(k) + \frac{1}{2} \right) \right] \sum_{j \in i} \hat{e}_{ji}
\]

in the HP analysis. Here \( \omega_k \) is the dispersion of a single hexagonal unit: \( \omega_k = 2|\sin k|, k = 2\pi (\text{integer})/6 \). The orientation of the antiferromagnetic spins defines the spin-loop director of Ref. \(^{13}\).

Taking individual hexagon as a structural unit, the pyrochlore lattice is built up of four different types of non-overlapping hexagons (defined A, B, C, and D), depending on the orientation of the face of a hexagon. The four directions correspond also to the normal of the four faces of a tetrahedron. Planes perpendicular to each orientation defines a Kagomé lattice embedded in a pyrochlore structure. Taking one such Kagomé plane as the basal plane, spanned by \( \hat{e}_1 \) and \( \hat{e}_2 \), and the vector \( \hat{e}_3 \) connecting two nearby basal planes, the A-type hexagons are given the coordinates \((3m, 2n, p)\), or \(3m\hat{e}_1 + 2n\hat{e}_2 + p\hat{e}_3\), for integers \(m, n, \) and \(p\) (Fig. 2). The other three types are sandwiched between two nearby Kagomé planes that are shown in Fig. 2. Coordinates of a B-type hexagon, for example, is defined by those of the A-type lying nearest to it. The remaining hexagons are then each located at \((B; 3m, 2n+1, p)\), \((C;3m-1, n, 2p+1)\), and \((D;3m-1, n, 2p)\). An A-type hexagon at \((3m, 2n, p)\) is neighbor by four B-type hexagons located at \((3m, 2n+1, p-1)\), \((3m, 2n-1, p-1)\), \((3m, 2n+1, p)\) and \((3m, 2n-1, p)\). Two other hexagons, of type C and D, surround the A-type hexagon, too. Such “connectivity” of a given hexagon type to other hexagons can be worked out, for all hexagon types.

A unit cell has a \(2 \times 2 \times 3\) structure with eight hexagons, two of each orientation. Each hexagon is surrounded by six nearest-neighboring hexagons as shown in Fig. 3. According to the previous discussion, a single hexagon undergoes a uniform contraction and has six collinear, antiferromagnetic spins. Requiring such hexagon contraction throughout the whole pyrochlore lattice, the r.h.s. of Eq. (4) imposes the condition that the directors of the nearest-neighbor hexagons be orthogonal (Fig. 3). A self-consistency requirement on the hexagons is therefore that the network of hexagons be “colored” in one of three colors, say R, G, and B, with no two neighboring hexagons having the same color. In other words, the relative orientations of the directors satisfy an antiferromagnetic 3-state Potts model

\[
H_{3SP} = J_{eff} \sum_{\langle IJ \rangle} \delta(d_I, d_J)
\]

for the nearest-neighbour hexagons, \( \langle IJ \rangle \), and their respective directors, \((d_I, d_J)\), where \(d_I, d_J\) takes on R, G, or B. We find, through explicit construction, that such coloring of the hexagon units can indeed be realized for a pyrochlore lattice. The strength of coupling between the directors \(J_{eff}\) depends on the spin-lattice interaction \(\alpha\), and also on the level of quantum fluctuation within a hexagonal cluster. A severe quantum fluctuation leads to the reduction of \(\langle S_i \cdot S_j \rangle\) for \(i, j\) belonging to different hexagons, and an effectively weaker \(\alpha\) in Eq. (5). In the extreme limit \(J_{eff} \to 0\) the hexagons are completely decoupled from each other, but more generally a residual hexagon-hexagon interaction of order \(J_{eff}\) lifts the degeneracy and leads to a band of excitation spectra.

So far the distortion mode is discussed in terms of the hexagon as a unit. Now, let us consider the distortion of an isolated tetrahedron. For a single tetrahedron, there are six vibrational modes: singlet \(A_1\), a doublet \(E\), and a triplet \(T_2\). When the pyrochlore lattice assumes the hexagonal distortion we discuss, the resulting distortion for an isolated tetrahedron is not tetragonal \((E)\) as discussed in Ref. 11, but is a linear combination of a doublet \(E\) and a triplet \(T_2\) (Fig. 4).

In conclusion, lattice-coupled antiferromagnetic Heisenberg spin model on a variety of frustrated lattices is analyzed. For a triangular and Kagomé lattice where the ground state spin structures are locally rigid,
no spin-Peierls effect arises. Pyrochlore spins have a continuous manifold of ground states and a lack of local rigidity of spins, and spin-lattice coupling easily induces a lattice distortion leading to a reduction of the ground state degeneracy. A particular pattern of such lattice distortion, namely a hexagon contraction, is proposed and analyzed, following the suggestion of Ref.13. Each hexagonal cluster takes advantage of the lattice deformation to reduce its size and maximize the exchange energy within the cluster. The hexagonal ‘protectorate’ of Ref.13 is thus obtained. Interaction between nearby hexagons is mediated by the spin-lattice condition, Eq. (5), and leads to mutually orthogonal directors of nearby hexagons. In broader perspective, the spin-lattice interaction provides a channel for the self-organization of spins, which helps relieve the frustration inherent in the underlying microscopic Hamiltonian.

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