The Néel order for a frustrated antiferromagnetic Heisenberg model: beyond linear spin-wave theory

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Abstract

Within Dyson-Maleev (DM) transformation and self-consistent mean-field treatment, the Néel order/disorder transition is studied for an antiferromagnetic Heisenberg model which is defined on a square lattice with a nearest neighbour exchange $J_1$ and a next-nearest neighbour exchange $J_2$ along only one of the diagonals. It is found that the Néel order may exist up to $J_2/J_1 = 0.572$, beyond its classically stable regime. This result qualitatively improves that from linear spin-wave theory based on Holstein-Primakoff transformation.
The two-dimensional (2D) antiferromagnetic Heisenberg models have attracted great interest in recent years, partly because of the fact that the parent compounds of the high temperature superconducting materials are excellent realizations of quasi-2D quantum antiferromagnets \[1\]. While the unfrustrated Heisenberg model has been well understood, much attention has been paid to the frustrated models such as square-lattice nearest and next-nearest neighbour interaction (so called \(J_1-J_2\) model, triangular lattice model and Kagomé lattice model etc \[2\] \[4\]). In these frustrated Heisenberg models the property of the ground state, whether magnetically ordered or disordered, is a subject of considerable interest. For example, the \(J_1-J_2\) model takes on Néel order at small \(J_2/J_1\) and collinear order at large \(J_2/J_1\), which are separated by a region of disordered state \[4\].

Very recently a \(S = 1/2\) Heisenberg model, which defined on a square lattice with a nearest neighbour antiferromagnetic exchange \(J_1\) and a next-nearest neighbour exchange \(J_2\) along only one of the diagonals of the lattice as shown in Fig. 1, has been proposed \[8,9\]. Its Hamiltonian is written as

\[
H = J_1 \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + J_2 \sum_{\langle lm \rangle} \mathbf{S}_l \cdot \mathbf{S}_m ,
\]

where the notation \(\langle ij \rangle\) denote nearest neighbour bonds and \(\langle lm \rangle\) denote next-nearest neighbour bonds along only one diagonal. Topologically this model is equivalent to the Heisenberg antiferromagnet on an anisotropic triangular lattice \[10,11\]. In special cases \(J_2 = 0\), \(J_1 = J_2\) and \(J_1 = 0\), it will recover to unfrustrated square lattice model, isotropic triangular lattice model and decoupled spin chains, respectively. Therefore this model provides a way of interpolating between several well-known one and two-dimensional models. One can study the role of frustration in going from one to two dimensions. On the other hand, this model is of direct relevance to the magnetic phases of some quasi-2D organic superconductors. It has been argued that this model may describe the spin degree of freedom of the insulating phase of the layered molecular crystals \(\kappa-(\text{BEDT-TTF})_2\text{X}\) \[12\]. The parameter \(J_2/J_1\) for these materials is suggested to be \(\sim 0.3 - 1\) and the magnetic frustration will play an important role.
FIG. 1. The 2D square lattice with nearest neighbour interaction $J_1$ and next-nearest neighbour interaction $J_2$ along only one of the diagonals.

Classically, the ground state of the model can be derived straightforwardly as a function of the ratio $J_2/J_1$ if we assume that the spins lie in the $xz$ plane and are described by a spiral form $S_i = \hat{S}_i\theta_i = S_i(\sin\theta_i, 0, \cos\theta_i)$ as shown in Fig. 1. Here the angle $\theta_i = \mathbf{q} \cdot \mathbf{r}_i$ and the wavevector $\mathbf{q} = (q,q)$ defines a relative orientation of the spins. Minimization of the classical energy with respect to $q$ gives the result that the ground state take on Néel order (i.e., $q = \pi$) for $J_2/J_1 \leq 1/2, \text{ and spiral order with } q = \arccos(-J_1/2J_2)$ for $J_2/J_1 > 1/2$.

The quantum model has also been studied numerically and analytically. The series expansions were adopted by Zheng et al. [8] for numerical calculation. It was found that the Néel order persists up to $J_2/J_1 = 0.7$. In the region $0.7 \leq J_2/J_1 \leq 0.9$ there is no magnetic order and for larger values of $J_2/J_1$ there is incommensurate or spiral order. It is interesting to note that the Néel order exists beyond its classical result ($J_2/J_1 = 0.5$). Analytically the standard and simple linear spin-wave theory (LSW) based on Holstein-Primakoff (HP) transformation was used by Merino et al. [9] to discuss the possible ordered and disordered states. It is helpful to repeat some details here for later discussion. First, for convenience the spin at each site is rotated along its reference direction characterized by the angle $\theta_i$. Such a rotation may be accomplished by the following transformation for spin operators

$$S_i^x = \sin \theta_i \hat{S}_i^x + \cos \theta_i \hat{S}_i^z$$
$$S_i^y = \hat{S}_i^y$$
$$S_i^z = \cos \theta_i \hat{S}_i^z - \sin \theta_i \hat{S}_i^x$$

For new spins $\hat{\mathbf{S}}$, the reference ground state becomes ferromagnetic. The rotated Hamiltonian

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becomes

\[ H = J_1 \sum_{ij} \left[ \cos \theta_{ij} (\hat{S}_i^z \hat{S}_j^z + \hat{S}_i^y \hat{S}_j^y) + \sin \theta_{ij} (\hat{S}_i^z \hat{S}_j^z - \hat{S}_i^x \hat{S}_j^x) + \hat{S}_i^y \hat{S}_j^y \right] \\
+ J_2 \sum_{lm} \left[ \cos \theta_{lm} (\hat{S}_l^x \hat{S}_m^x + \hat{S}_l^z \hat{S}_m^z) + \sin \theta_{lm} (\hat{S}_l^z \hat{S}_m^x - \hat{S}_l^x \hat{S}_m^z) + \hat{S}_l^y \hat{S}_m^y \right] , \]  

with \( \theta_{ij} = \theta_i - \theta_j = q, \theta_{lm} = \theta_l - \theta_m = 2q \). Then with HP transformation the above Hamiltonian with only quadratic terms kept may be diagonalized in the momentum space. The final dispersion relation for the spin excitation is

\[ \omega_k = \sqrt{\left\{ [J(k + q) + J(k - q)]/2 - J(q) \right\} [J(k) - J(q)]} \]

\[ \omega_k = \sqrt{\left\{ [J(k - q) + J(k + q)]/2 - J(q) \right\} [J(k) - J(q)]} \]

with \( J(k) = J_1 (\cos k_x + \cos k_y) + J_2 \cos (k_x + k_y) \). By calculation of the magnetization \( \langle \hat{S}_i^z \rangle \) (see also the dashed line in Fig. 2 later) it was suggested by the authors in Ref. [9] that a possible Néel order/disorder transition happens at the point \( J_2/J_1 \simeq 0.5 \). However, we want to point out the inherent limitation for LSW theory here. From the spectrum \( \omega_k \), it is easy to find that, in order to ensure the argument in the square root always positive in the whole Brillouin zone, the parameter \( q \) has to be set as \( \pi \) (characterizing Néel order) in the region \( J_2/J_1 < 0.5 \), but must not be set as \( \pi \) in the region \( J_2/J_1 > 0.5 \). This means that within LSW theory the Néel ordered state can never appear beyond its classically stable region, which is in contrast with the numerical result [8]. As also pointed out by the authors in Ref. [9] themselves, the interaction between spin waves becomes very large at the transition point and it may lead to a completely different picture for the states. So it is very necessary to go beyond the LSW theory to see how the above result will be modified, which is the purpose of this paper. Instead of HP transformation, the Dyson-Maleev (DM) transformation will be adopted to avoid \( 1/S \) expansion. It has been also recognized that the DM transformation must be preferred if one needs really to go beyond LSW theory within a perturbation scheme [13]. In the treatment of the so called \( J_1-J_2 \) model, it has been shown that the spin-wave theory based on DM transformation gives perfectly consistent result as that from numerical calculation; much better than that from LSW theory [8].

In this work we will not discuss the possible spiral state for large \( J_2/J_1 \), but focus
on the Néel order/disorder transition in the intermediate $J_2/J_1$ region, especially on the problem whether the Néel order may appear beyond its classical region of stability which is one of the most interesting topics for this model. Technically, the Hamiltonian under DM transformation have terms as high as sixth order when spiral state is considered (i.e., for general $q$), which are relatively complicated to treat. When only Néel state is considered the transformed Hamiltonian has no term higher than fourth order, which can be easily treated by mean-field (MF) theory or perturbation theory. Explicitly, one may apply the DM transformation onto the original Hamiltonian (1); or begin with the rotated Hamiltonian (2) by setting $\theta_{ij} = \pi$, $\theta_{lm} = 2\pi$ and then use the DM transformation for A and B two sublattices in the following form:

$$\hat{S}^+ = (1 - a_i^\dagger a_i) a_i, \quad \hat{S}^- = a_i^\dagger, \quad \hat{S}_z^+ = 1/2 - a_i^\dagger a_i$$

$$\hat{S}^+ = b_j^\dagger (1 - b_j^\dagger b_j), \quad \hat{S}_z^- = b_j, \quad \hat{S}^- = 1/2 - b_j^\dagger b_j,$$

where $a(a^\dagger)$, $b(b^\dagger)$ are bosonic operators for sublattices A and B, respectively. Then the Hamiltonian (2) is transformed into

$$H = -J_1 \sum_{\langle ij \rangle} (1/2 - a_i^\dagger a_i)(1/2 - b_j^\dagger b_j) + [(1 - a_i^\dagger a_i)a_i b_j + a_i^\dagger b_j (1 - b_j^\dagger b_j)]/2$$

$$+ J_2 \{ \sum_{\langle lm \rangle \in A} (1/2 - a_i^\dagger a_i)(1/2 - a_m^\dagger a_m) + [(1 - a_i^\dagger a_i)a_i a_m + (1 - a_m^\dagger a_m)a_m a_i^\dagger]/2 + \sum_{\langle lm \rangle \in B} a \rightarrow b \}.$$ (3)

To diagonalize the above Hamiltonian, we treat the quartic terms with a self-consistent MF theory. For those terms which could be decoupled in two ways, we will combine the two kinds of decoupling form together through a weight factor $\lambda$ as introduced by Chu and Shen [14]. For example, the term like $a_i^\dagger a_i b_j b_j$ will be decoupled in the way

$$a_i^\dagger a_i b_j b_j \simeq \lambda[a_i^\dagger a_i b_j^\dagger b_j + a_i^\dagger a_i b_j^\dagger b_j] + (1 - \lambda)[a_i^\dagger a_i b_j b_j + a_i^\dagger b_j^\dagger (a_i^\dagger a_i) - \langle a_i^\dagger b_j^\dagger a_i b_j \rangle].$$

The parameter $0 \leq \lambda \leq 1$ reflects the competition between two decoupling ways. Its value may be decided by minimization of the energy, which was found to be 1/2 in Ref. [14] for unfrustrated lattice; or it is required to be equal to 1/2 in order to keep the symmetry of the Hamiltonian before and after decoupling [15]. We will take the value $\lambda = 1/2$ throughout
our calculations. With definition of several parameters: 

\[ u = \langle a_i^\dagger a_i \rangle = \langle b_i^\dagger b_i \rangle, \quad v = \langle a_i^\dagger b_j^\dagger \rangle = \langle a_i b_j \rangle, \quad w = \langle a_i^\dagger a_m \rangle = \langle b_l^\dagger b_m \rangle, \]

we may obtain a quadratic Hamiltonian in the momentum space:

\[
H = \sum_k \left[ C_k (a_k^\dagger a_k + b_k^\dagger b_k) + E_k (a_k b_{-k} + a_k^\dagger b_{-k}^\dagger) \right] + \text{const},
\]

\[
C_k = 2J_1(1 - u + v) - J_2(1 - u + w)[1 - \cos(k_x + k_y)],
\]

\[
E_k = -J_1(1 - u + v)(\cos k_x + \cos k_y),
\]

\[
\text{const} = \frac{N}{2} [J_1(-1 + 2u^2 + 2v^2 - 4uv) + J_2(1 - 2u^2 - 2w^2 + 4uw)/2],
\]

where the summation is over half of the original Brillouin zone and \( N \) is total number of lattice sites. Under a Bogoliubov transformation

\[
a_k = \cosh \lambda_k \bar{a}_k + \sinh \lambda_k \bar{b}_{-k}^\dagger
\]

\[
b_{-k}^\dagger = \sinh \lambda_k \bar{a}_k + \cosh \lambda_k \bar{b}_{-k}^\dagger
\]

with \( \tanh 2\lambda_k = -E_k/C_k \), the Hamiltonian (4) may be diagonalized into

\[
H = \sum_k \left[ \bar{\omega}_k (\bar{a}_k^\dagger \bar{a}_k + \bar{b}_{-k}^\dagger \bar{b}_{-k}^\dagger + 1) - C_k \right] + \text{const}
\]

with the excitation spectrum \( \bar{\omega}_k = \sqrt{C_k^2 - E_k^2} \). Correspondingly the self-consistent equations for \( u, \ v, \ w \) are expressed as

\[
u = \frac{1}{N} \sum_k \frac{E_k \cos k_x}{\sqrt{C_k^2 - E_k^2}},
\]

\[
w = \frac{1}{N} \sum_k \frac{C_k \cos(k_x + k_y)}{\sqrt{C_k^2 - E_k^2}}.
\]

The magnetization is simply given by \( m = 1/2 - u \) and the ground state energy is \( E_0 = \sum_k (\bar{\omega}_k - C_k) + \text{const} \).
FIG. 2. The magnetization $m$ calculated from our treatment (solid line) and that from LSW theory (dashed line) as a function of $J_2/J_1$. It goes to zero at about $J_2/J_1 = 0.499$ within LSW theory, but up to $J_2/J_1 = 0.572$ within our treatment.

FIG. 3. The self-consistent results for the parameters $v$ and $w$.

We show the numerical results for the above self-consistent equations in Figs. 2 and 3. The magnetization $m$ (derived from the parameter $u$) as a function of $J_2/J_1$ is given by the solid line in Fig. 2, which is the main result in this paper. It is found that the magnetization does not vanish until $J_2/J_1 \simeq 0.572$, which gives the Néel order/disorder transition point. As comparison, the result from LSW theory is plotted by the dashed line; the magnetization goes to zero immediately before the classical value $J_2/J_1 = 0.5$, see also Ref. [11]. As expected, the current result is closer to the numerical one from series expansions and most importantly, it qualitatively improves the result from LSW theory. As we discussed before, the LSW theory is impossible to deduce a Néel ordered state beyond $J_2/J_1 = 0.5$. In our treatment the interaction between spin waves is actually partly considered, then the fact that Néel order may exist beyond its classically stable region is shown. It is quite possible that
the transition point will shift to larger value if the residual interaction between spin waves is included. This may be also hinted from Fig. 3, where the parameter $v$, which represents the antiferromagnetic correlation between the two original nearest-neighbour spins, is large near the transition point.

For completeness, the ground state energy in the region $0 < J_2/J_1 < 0.57$ is also shown in Fig. 4 by the solid line, which is close to the result from LSW theory (the dashed line). In the small $J_2/J_1$ region the energy calculated here is a little lower than that of LSW theory, and becomes a little higher with increase of $J_2/J_1$. For $J_2/J_1 > 0.5$ the energy within LSW theory is derived from spiral state; there is a cusp at point $J_2/J_1 = 0.5$ [9]. In the current case the state is still Néel ordered and the energy changes smoothly.

![Graph](image)

**FIG. 4.** The ground state energy in unit of $NJ_1$ calculated from our treatment (solid line) and that from LSW theory (dashed line) as a function of $J_2/J_1$ (see text).

In summary, within Dyson-Maleev (DM) transformation and self-consistent mean-field treatment, we have studied an antiferromagnetic Heisenberg model on a square lattice which includes a nearest neighbour exchange $J_1$ and a next-nearest neighbour exchange $J_2$ along only one of the diagonals. This model should be of direct relevance to some layered organic superconductors. In this work we focus on the discussion of Néel order/disorder transition for not large $J_2/J_1$. It is found that the Néel order may exist up to $J_2/J_1 = 0.572$, beyond its classically stable regime. This property is consistent with numerical finding from series expansions, which is one of the most interesting features for this model. Especially, because the interaction between spin waves is partly considered in our treatment, the result derived here qualitatively improves that from LSW theory based on Holstein-Primakoff transforma-
tion. It is certainly necessary to continue this work to study the large $J_2/J_1$ region where a spiral order will appear, so that a whole phase diagram may be constructed.

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