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Goldstone modes and quantum gaps in the square frustrated Heisenberg antiferromagnet

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Abstract. The ground state configuration of the two-dimensional (2D) Heisenberg model is a topic to which much theoretical effort is being applied with particular emphasis on the frustration effects caused by exchange competition between spins of different neighbouring shells. In this respect a central topic is the effect of quantum fluctuations on the soft modes present in the simple spin wave dispersion curve. A rigorous theorem based on the Bogoliubov inequality implies that a zero energy excitation must exist at the helix wavevector $\boldsymbol{Q}$ when long-range order is present. Recently it has been suggested that only the soft mode at $\boldsymbol{k} = 0$ survives quantum fluctuations in the square frustrated Heisenberg antiferromagnet, so that the Goldstone mode at $\boldsymbol{k} = \boldsymbol{Q}$ could be recovered only by looking for an excitation different from the single-particle-like excitation. We show that this result is a spurious consequence of neglecting second-order perturbation contributions which are of the same power in $1/S$ as the first-order perturbation contribution accounted for evaluating the magnon self-energy. Indeed we show that a delicate balance of these contributions restores the Goldstone mode at the helix wavevector $\boldsymbol{Q}$ and substantially reduces the value of the quantum gaps that replace the *accidental* soft modes of the simple spin wave spectrum.

1. Introduction

The loosely packed two-dimensional (2D) Heisenberg antiferromagnet with nearest neighbour (NN) coupling and $S > \frac{1}{2}$ exhibits long-range order (LRO) at zero temperature [1] and well grounded arguments support the existence of LRO at $T = 0$ even for $S = \frac{1}{2}$ [2]. Exchange competition between spins of different shells of neighbours prevents exact approaches, but certainly introduces a rich manifold of spin configurations (see for instance [3]) and the existence of a *spin liquid* phase in the vicinity of *magic* lines in the parameter space is currently accepted [4]. In particular we consider the so called 3N antiferromagnetic 2D Heisenberg model on a square lattice where the NN spins are coupled antiferromagnetically while next nearest neighbours (NNN) and third nearest neighbours (TNN) can be either ferromagnetically or antiferromagnetically coupled. In the classical approximation [5] four minimum energy configurations exist at zero temperature: AF, AF_1, H_1 and H_2. The AF configuration is the usual two-sublattice antiferromagnetic configuration characterized by the wavevector $\boldsymbol{Q} = (\pi, \pi)$. (From now on we assume a unit lattice constant.) The AF_1 configuration consists of ferromagnetic lines of spins alternating antiferromagnetically with $\boldsymbol{Q} = (\pi, 0)$. The H_1 configuration is a helix with $\boldsymbol{Q} = (\pi, \arccos[(2j_2-1)/4j_3])$

where $j_2 = J_2/J_1$ and $j_3 = J_3/J_1$, J_1, J_2 and J_3 being the NN, NNN, TNN spin coupling, respectively. H_2 is characterized by

$$Q = \left[\arccos\left(\frac{-1}{2j_2+4j_3}\right), \arccos\left(\frac{-1}{2j_2+4j_3}\right)\right].$$

The AF–H_1 and AF–H_2 phase boundaries are given by $1 - 2j_2 - 4j_3 = 0$ with $j_2 < \frac{1}{2}$, while the H_1–H_2 phase boundary is $j_2 = 2j_3$ with $j_3 > \frac{1}{8}$. This line is an infinite degeneration line because all helix wavevectors satisfying the condition $\cos Q_x + \cos Q_y = -1/4j_3$ minimize the energy of the model in the classical approximation. The H_1–AF_1 phase boundary is $1 - 2j_2 + 4j_3 = 0$ with $j_2 > \frac{1}{2}$ and the AF–AF_1 boundary is $j_2 = \frac{1}{2}$ with $j_3 < 0$. Notice that the AF_1 phase consists of two interpenetrating antiferromagnetic sublattices of NNN spins which are completely decoupled so that the angle between them is arbitrary. The zero temperature phase diagram in the parameter space is shown in figure 1 of [5].

An interesting question about this model concerns the effect of quantum fluctuations on the soft modes of the magnon spectrum obtained in the classical approximation ($S \to \infty$). These soft modes are located not only at $k = 0$ and $k = \pm Q$ where Q is the helix wavevector, but even at those wavevectors that belong to the star of Q [6]. Recent calculations [7] based on an extended Schwinger boson approach verified the persistence of soft modes at $k = \pm Q$ for antiferromagnetic configurations (AF or AF_1), whereas in the helical configuration (for instance H_1) all soft modes except the soft mode at $k = 0$ are replaced by quantum gaps. If the latter statement was true, one should conclude that the single-particle-like excitation does not exhibit Goldstone modes at $k = \pm Q$ which have to be preserved for reasons of symmetry [8] and one should look for these Goldstone modes in other multi-particle excitations [7]. However we prove that the removal of the soft mode in the spin wave spectrum at the helix wavevector is due to neglecting quantum corrections arising from repeated scattering by the three bose operator potentials, which gives contributions of the same order in $1/S$ as the single scattering by the four bose operator potential [8]. Indeed a rigorous theorem based on the Bogoliubov inequality implies that the Heisenberg helimagnets with LRO must have zero energy excitations at the helix wavevector [8]. Before and after this theorem was proved an apparent lifting of the helix soft mode was found in the axial NNN Heisenberg (ANNNH) model [9, 10], a quantum version of the well known ANNNI model of Fisher and Selke [11]. However, it was realized [8, 12] that the lifting of the soft mode at $k = \pm Q$ is an artifact of the Hartree–Fock [9], RPA [10] and variational [9] techniques that neglect the contribution of the three operator potential appearing in the bosonic equivalent Hamiltonian that one obtains by the usual spin–boson transformation. Indeed the Hartree–Fock, RPA and variational approaches consist of keeping an average of the interaction potential so that at this order the cubic potential gives no contribution. In [8] we have shown and in [12] we have confirmed that the magnon at the helix wavevector is just the Goldstone mode of the ANNNH model. Here we show that the same holds for the 3N model for which quantum corrections have been recently evaluated [7] leading to the wrong conclusion that the magnon soft mode at the helix wavevector is removed. As for the *accidental* soft modes at the wavevectors of the star of Q we confirm that quantum fluctuations remove them in agreement with [7], but the value of these quantum gaps is severely reduced if the three operator potential is accounted for correctly.

2. Mathematical theory

Let us focus on the antiferromagnetic phase AF_1 with $Q = (0, \pi)$. In this case as well as for any collinear configuration the three operator potential vanishes, so that the only quantum correction within the first order in $1/S$, is the Hartree–Fock correction that leads to the following magnon spectrum $\hbar\omega_k^{\mathrm{I}}$

$$(\hbar\omega_k^{\mathrm{I}})^2 = (\hbar\omega_k)^2 \left[1 + \frac{1}{S}\left(1 - \frac{1}{N}\sum_q \frac{A_q}{\hbar\omega_q}\right)\right] - \frac{1}{S}\left[A_k \frac{1}{N}\sum_q \hbar\omega_q - \frac{1}{N}\sum_q D_{k-q}\frac{1}{2}\left(S_k\sqrt{\frac{S_q}{D_q}} + D_k\sqrt{\frac{D_q}{S_q}}\right)\right] \qquad (1)$$

where

$$A_k = \sum_\delta 2J_\delta S[\cos Q\cdot\delta - \tfrac{1}{2}(1 + \cos Q\cdot\delta)\cos k\cdot\delta] \qquad (2)$$

$$B_k = -\sum_\delta J_\delta S \cos k\cdot\delta(1 - \cos Q\cdot\delta) \qquad (3)$$

$$S_k = A_k + B_k \qquad (4)$$

$$D_k = A_k - B_k \qquad (5)$$

and

$$\hbar\omega_k = \sqrt{S_k D_k}. \qquad (6)$$

For example we choose $j_3 = 0$ and $j_2 > \frac{1}{2}$ to give the antiferromagnetic configuration AF_1 with $Q = (0, \pi)$. Note that the classical magnon spectrum given by equation (6) shows soft modes at $k = (0,0)$, $k = Q = (0, \pm\pi)$, $k = Q' = (\pm\pi, 0)$, $k = (\pm\pi, \pm\pi)$. It is straightforward to verify that the magnon energy including the quantum correction given by equation (1) is strictly zero for $k = (0,0)$ and $k = Q = (0, \pm\pi)$ which are the Goldstone modes related to the rotational invariance of the Heisenberg Hamiltonian. However for $k = Q' = (\pm\pi, 0)$ one obtains

$$(\hbar\omega_{Q'}^I)^2 = -\frac{1}{S}(4J_1S)^2(2j_2 - 1)\frac{1}{N}\sum_q 2(\cos q_x + \cos q_y) \times \left[\frac{\cos q_x - \cos q_y + 2j_2(1 - \cos q_x \cos q_y)}{\cos q_x + \cos q_y + 2j_2(1 + \cos q_x \cos q_y)}\right]^{1/2}. \qquad (7)$$

We have computed this quantum gap for selected values of j_2 and found

$$\hbar\omega_{Q'}^I = 4|J_1|\sqrt{S}G \qquad (8)$$

where $G = 0.374$, 0.447, 0.490, 0.520, 0.542 for $j_2 = 0.6$, 0.7, 0.8, 0.9, 1.0, respectively. An analogous calculation for $k = (\pm\pi, \pm\pi)$ gives identical numerical values. These results agree with equations (5.6) and (5.7) of [7] where a mean field decoupling scheme of the Heisenberg Hamiltonian realized by an extended Schwinger boson approach is used.

We now evaluate the effect of quantum fluctuations on the soft modes present in the classical spin wave spectrum at wavevectors belonging to the star of the classical helix wavevector $Q = (\pi, \arccos[(2j_2 - 1)/4j_3])$ characterizing the H_1 configuration. For this helical configuration the removal of *all* soft modes in the spin wave spectrum except the uniform mode, has been obtained as a consequence of quantum fluctuations (see equation (5.21) of [7]). Conversely we prove that the soft mode at the helix wavevector survives quantum fluctuations, so that it has to be recognized as the Goldstone mode which must exist because of the rotational invariance of the Heisenberg Hamiltonian [8]. At the same time the accidental classical soft modes are removed. We stress that the crucial point is the correct expansion in $1/S$.

The magnon spectrum including *all* quantum corrections of order $1/S$ is given by

$$(\hbar\omega_k')^2 = (\hbar\omega_k^{\mathrm{I}})^2 - (\hbar\omega_k^{\mathrm{II}})^2 \tag{9}$$

where

$$(\hbar\omega_k^{\mathrm{II}})^2 = -2\hbar\omega_k[\hbar\Sigma_1(k) + \hbar\Sigma_2(k)] \tag{10}$$

The generic expression of $\hbar\omega_k^{\mathrm{I}}$ given by equation (1) is the magnon spectrum including the first-order perturbation contribution coming from the four boson operator potential (Hartree–Fock approximation). For the 3N Heisenberg antiferromagnet one has

$$\begin{aligned}(\hbar\omega_k^{\mathrm{I}})^2 = (4J_1S)^2 \Bigg\{ & s_k d_k \left[1 + \frac{1}{S}\left(1 - \frac{1}{2N}\sum_q \left(\sqrt{\frac{s_q}{d_q}} + \sqrt{\frac{d_q}{s_q}}\right)\right)\right] \\ & + s_k \frac{1}{2NS} \sum_q [-\cos Q \cos q_x(1 - \cos k_x) + \cos q_y(1 - \cos k_y) \\ & + 2j_2 \cos Q \cos q_x \cos q_y (1 - \cos k_x \cos k_y) \\ & - j_3 \cos 2Q \cos 2q_x(1 - \cos 2k_x) - j_3 \cos 2q_y(1 - \cos 2k_y)]\sqrt{\frac{s_q}{d_q}} \\ & + d_k \frac{1}{2NS} \sum_q [-\cos q_x(1 - \cos Q \cos k_x) - \cos q_y(1 + \cos k_y) \\ & - 2j_2 \cos q_x \cos q_y (1 + \cos Q \cos k_x \cos k_y) \\ & - j_3 \cos 2q_x(1 - \cos 2Q \cos 2k_x) - j_3 \cos 2q_y(1 - \cos 2k_y)]\sqrt{\frac{d_q}{s_q}} \Bigg\}\end{aligned} \tag{11}$$

where $s_q = S_q/(4|J_1|S)$ and $d_q = D_q/(4|J_1|S)$ are given by

$$\begin{aligned}s_q = {} & 1 - \cos Q + 2j_2 \cos Q - j_3(1 + \cos 2Q) \\ & + \cos q_x + \cos q_y + 2j_2 \cos q_x \cos q_y + j_3(\cos 2q_x + \cos 2q_y)\end{aligned} \tag{12}$$

$$\begin{aligned}d_q = {} & 1 - \cos Q + 2j_2 \cos Q - j_3(1 + \cos 2Q) + \cos Q \cos q_x - \cos q_y \\ & - 2j_2 \cos Q \cos q_x \cos q_y + j_3(\cos 2Q \cos 2q_x + \cos 2q_y).\end{aligned} \tag{13}$$

$\Sigma_1(k)$ and $\Sigma_2(k)$ appearing in equation (10) are the contributions to the self-energy coming from the second-order perturbation theory applied to the three operator potential. The generic expressions of $\Sigma_1(k)$ and $\Sigma_2(k)$ are explicitly given by equations (34) and (35) of [8]. These two contributions lead to a quantum correction of the self-energy which is of the same order in $1/S$ as the first-order perturbation contribution coming from the four operator potential. Note that if one neglects $\hbar\omega_k^{\mathrm{II}}$ in equation (9), the remaining Hartree–Fock spectrum $\hbar\omega_k^{\mathrm{I}}$ implies the existence of quantum gaps both at $k = \pm Q = \pm(Q, \pi)$ and at $k = \pm Q' = \pm(\pi, Q)$. Indeed equation (11) evaluated at $k = \pm Q$ gives

$$(\hbar\omega_{\pm Q}^{\mathrm{I}})^2 = (4J_1S)^2 \frac{1}{2S} G_1 d_Q \sin^2 Q \tag{14a}$$

where

$$G_1 = \frac{1}{N}\sum_q (-\cos q_x - 2j_2 \cos q_x \cos q_y - 4j_3 \cos^2 Q \cos 2q_x)\sqrt{d_q/s_q} \tag{14b}$$

and for $k = \pm Q'$ one has

$$(\hbar\omega_{\pm Q'}^{\mathrm{I}})^2 = (4J_1S)^2 \frac{1}{2S} G_1' d_{Q'}(1 + \cos Q) \tag{15a}$$

where

$$\begin{aligned} G_1' = \frac{1}{N}\sum_q [&-\cos q_x - \cos q_y - 2j_2(1 - \cos Q)\cos q_x \cos q_y \\ &- 2j_3(1 - \cos Q)(\cos 2q_x + \cos 2q_y)]\sqrt{d_q/s_q}. \end{aligned} \tag{15b}$$

In equations (14a) and (15a) d_Q and $d_{Q'}$ are given by

$$d_Q = 2 - \cos Q(1 - \cos Q) + 2j_2 \cos Q(1 + \cos Q) - j_3 \cos 2Q(1 - \cos 2Q) \tag{16}$$

and

$$d_{Q'} = 1 - 3\cos Q + 2j_2 \cos Q(1 + \cos Q) - j_3(1 - \cos 2Q). \tag{17}$$

Note that *gaps* (14a) and (15a) coincide with those in equation (5.21) of [7] obtained by a mean field treatment of the Heisenberg Hamiltonian realized by an extended Schwinger boson approach. In contrast we stress that the complete and correct magnon spectrum is given by equation (9). The cubic potential contribution to the self-energy evaluated at $k = \pm Q$ reads

$$(\hbar\omega_{\pm Q}^{\mathrm{II}})^2 = (4J_1S)^2 \frac{1}{2S} (G_2 + G_3) d_Q \sin^2 Q \tag{18a}$$

where

$$\begin{aligned} G_2 = \frac{1}{N}\sum_q c_1^2 &\sqrt{\frac{s_q(s_q - c_2\sin^2 Q + c_3 \sin Q \cos Q)}{d_q(d_q + c_1 \sin Q)}} \\ &\times \left[\sqrt{s_q d_q} + \sqrt{(s_q - c_2 \sin^2 Q + c_3 \sin Q \cos Q)(d_q + c_1 \sin Q)}\right]^{-1} \end{aligned} \tag{18b}$$

and

$$G_3 = \frac{1}{N}\sum_q \frac{c_1(c_2 \sin Q - c_3 \cos Q)}{\sqrt{s_q d_q} + \sqrt{(s_q - c_2 \sin^2 Q + c_3 \sin Q \cos Q)(d_q + c_1 \sin Q)}}. \tag{18c}$$

The cubic contribution evaluated at $k = \pm Q'$ reads

$$(\hbar\omega^{\mathrm{II}}_{\pm Q'})^2 = (4J_1 S)^2 \frac{1}{2S}(G'_2 + G'_3) d_{Q'} \sin^2 Q \tag{19a}$$

where

$$G'_2 = \frac{1}{N}\sum_q c_1^2 \sqrt{\frac{s_q(s_q - f_2(1+\cos Q) - f_3 \sin Q)}{d_q(d_q + g_2(1+\cos Q) + g_3 \sin Q)}} \left[\sqrt{s_q d_q} \right.$$
$$\left. + \sqrt{(s_q - f_2(1+\cos Q) - f_3 \sin Q)(d_q + g_2(1+\cos Q) + g_3 \sin Q)}\right]^{-1} \tag{19b}$$

and

$$G'_3 = \frac{1}{N}\sum_q c_1(c'_2 - c'_3 \sin Q) \left[\sqrt{s_q d_q} \right.$$
$$\left. + \sqrt{(s_q - f_2(1+\cos Q) - f_3 \sin Q)(d_q + g_2(1+\cos Q) + g_3 \sin Q)}\right]^{-1} \tag{19c}$$

where

$$c_1 = \sin q_x(1 - 2j_2 \cos q_y + 4j_3 \cos Q \cos q_x) \tag{20}$$

$$c_2 = \cos q_x(1 + 2j_2 \cos q_y) + 4j_3 \cos^2 Q \cos 2q_x \tag{21}$$

$$c_3 = \sin q_x(1 + 2j_2 \cos q_y + 4j_3 \cos 2Q \cos q_x) \tag{22}$$

$$c'_2 = \sin q_x(1 - 2j_2 \cos Q \cos q_y - 4j_3 \cos Q \cos q_x) \tag{23}$$

$$c'_3 = 2j_2 \sin q_x \sin q_y \tag{24}$$

$$f_2 = \cos q_x + \cos q_y + 2j_2(1 - \cos Q)\cos q_x \cos q_y$$
$$+ 2j_3(1 - \cos Q)(\cos 2q_x + \cos 2q_y) \tag{25}$$

$$f_3 = \sin q_y(1 - 2j_2 \cos Q \cos q_x - 4j_3 \cos Q \cos q_y) \tag{26}$$

$$g_2 = -\cos q_x + \cos q_y + 2j_3(1 - \cos Q)(\cos 2q_x - \cos 2q_y) \tag{27}$$

$$g_3 = \sin q_y(1 - 2j_2 \cos q_x + 4j_3 \cos Q \cos q_x). \tag{28}$$

We have performed the numerical evaluation of the magnon energy at $k = \pm Q$ and $k = \pm Q'$ given by equations (14a), (18a) and (15a), (19a), respectively, for $j_3 = 0.1$ and selected values of j_2. At $k = \pm Q$ the Hartree–Fock contribution results give

$$\hbar\omega^{\mathrm{I}}_{\pm Q} = 4|J_1|\sqrt{S} G^{\mathrm{I}}_Q \tag{29}$$

where $G_Q^{\rm I} = 0.407$, 0.920, 0.727, 0.682, 0.410 for $j_2 = 0.31$, 0.4, 0.5, 0.6, 0.69, respectively. Note that the existence of the $\rm H_1$ phase is restricted to $0.3 < j_2 < 0.7$ for $j_3 = 0.1$. Within the limits of numerical accuracy (10^{-4} in our calculations) the cubic potential contribution of equation (18a) exactly cancels the Hatree–Fock contribution. In this way the Goldstone mode is restored.

At $\boldsymbol{k} = \pm\boldsymbol{Q}'$ the Hartree–Fock contribution is

$$\hbar\omega^{\rm I}_{\pm\boldsymbol{Q}'} = 4|J_1|\sqrt{S}G^{\rm I}_{\boldsymbol{Q}'} \tag{30}$$

where $G^{\rm I}_{Q'} = 0.405$, 1.075, 0.823, 0.463, 0.565 for $j_3 = 0.1$ and $j_2 = 0.31$, 0.4, 0.5, 0.6, 0.69, respectively. In this case the cubic potential contribution is

$$\hbar\omega^{\rm II}_{\pm\boldsymbol{Q}'} = 4|J_1|\sqrt{S}G^{\rm II}_{\boldsymbol{Q}'} \tag{31}$$

where $G^{\rm II}_{Q'} = 0.381$, 0.921, 0.697, 0.377, 0.272 for $j_3 = 0.1$ and $j_2 = 0.31$, 0.4, 0.5, 0.6, 0.69, respectively. It is clear that quantum gaps replace the accidental soft mode of the simple spin wave spectrum. Indeed the magnon energy is

$$\hbar\omega'_{\pm\boldsymbol{Q}'} = 4|J_1|\sqrt{S}G'_{\boldsymbol{Q}'} \tag{32}$$

where $G'_{Q'} = 0.138$, 0.555, 0.437, 0.268, 0.495 for $j_3 = 0.1$ and $j_2 = 0.31$, 0.4, 0.5, 0.6, 0.69, respectively.

The quantum nature of such gaps can be experimentally distinguished from gaps entered by planar anisotropy. Indeed planar anisotropy also raises the magnon energy at the helix wavevector whereas quantum fluctuations do not destroy that soft mode. Moreover it should be noticed that the planar anisotropy in real compounds is usually much weaker than the exchange energy whereas the quantum gaps described here are clearly of the order of the exchange coupling. An inelastic neutron scattering experiment can easily select between gaps of a quantum nature and customary gaps caused by anisotropy.

3. Summary

We have shown that the soft mode at the helix wavevector $\boldsymbol{Q}$ in the magnon dispersion curve survives quantum fluctuations so that the Goldstone mode related to the rotational invariance of the Heisenberg Hamiltonian has to be identified with the single-particle-like excitation at $\boldsymbol{k} = \boldsymbol{Q}$ even for the square frustrated antiferromagnet when LRO is present. On the other hand the existence of LRO at zero temperature in such a model may be questionable only where a signal in this sense can be found. This is the case of the AF–$\rm H_1$, AF–$\rm H_2$, and $\rm H_1$–$\rm H_2$ boundary lines on which or, perhaps in their vicinity, the peculiar softening of the normal mode frequencies obtained in classical approximation seems to indicate the destruction of LRO [4]. As for the internal regions where the AF, $\rm AF_1$, $\rm H_1$ and $\rm H_2$ phases exist, LRO is expected. Possible change in the helix wavevector caused by quantum fluctuations [13] is of order $1/S$ and this should affect the magnon energy only at orders higher than $1/S$. This higher order correction is outside the scope of the present calculation. We stress that any physical inference based on the spurious removal of the Goldstone modes in the magnon spectrum is meaningless.

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