Microscopic Theory of Nonlinear Spin Waves in Ferromagnets*

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Abstract Considering the attractive interaction between two magnons with opposite wave vectors in a Heisenberg ferromagnet, we propose the model of magnon-pairs, which is suitable for low-temperature environment. A dressed magnon is an energy quantum of the magnon-pairs whose energy is a monotonically increasing function of absolute temperature. Based on the model, we re-investigate the excitation mechanism and thermodynamic properties of the Heisenberg ferromagnet. The correction factor e(0) plays an important role in studying the low-temperature properties of a ferromagnet.

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1 Introduction

Ferromagnet has always attracted the attention of scientists because of it's extensive applications to superconductivity,^[1-2] magnetoelectronic devices^[3] and so on. In solid, the Heisenberg model of spin-spin interactions can be considered as the starting point for understanding the complex phenomena of a ferromagnet. It is well known that quasiparticles play a fundamental role in nature. In ferromagnet, elemental magnetic collective excitations (magnons) are essential for explaining magnetic ordering^[4] and electron spin dynamics.^[5] The magnons are of great importance also for modern spintronic devices.^[6-9]

Bloch first researched the magnons in the Heisenberg model of a simple ferromagnet. He had predicted the temperature dependency of the magnetization at low temperatures (Bloch's $T^{3/2}$ law). In Bloch's theory he assumed that the number of magnons is so small that the interaction between two or more magnons may be neglected. In fact, researchers have always ignored the interaction between the magnons and paid more attention to the interactions between magnons and phonons or other guasiparticles, [10-13] especially to the magnon phonon coupling^[14] whose research may date back to 1970s.^[15] The interactions between the magnons had been first considered by Dyson.^[16] After him, some other physicists also began to consider this issue.^[17-18] In Dyson's theory he defined two kinds of interactions: one is the kinematical interaction. The other is the dynamical interaction, which represents the nondiagonal part of the Hamiltonian in his basic set of states.

In 1957, Bardeen Cooper and Schrieffer (BCS) published the first truly microscopic theory of superconductivity. The theory was soon recognized to be correct in all the essential aspect, and to correctly explain a number of important experimental phenomena. We know an electron tends to pull the positive ions towards itself, so that it is surrounded by a region where the lattice is denser than usual. Other electrons will be drawn towards the region. It will look as if it was attracted towards the first electron. The magnons are the energy quanta of linear spin waves, which can also be considered to attract to each other like electrons. In this paper, we ignore the repulsive interaction and only consider the attractive interaction between two magnons with opposite wave vectors \mathbf{k} and $-\mathbf{k}$. The attractive interaction between the magnons leads to bound magnon-pairs. The dressed magnons are the energy quanta of magnon-pairs whose energy is a monotonically increasing function of absolute temperature. Based on the model, we re-investigate the excitation mechanism and some thermodynamic properties of the Heisenberg ferromagnet. We organize the paper as follows. In Sec. 2 we introduce our model. In Sec. 3 we investigate the properties of the dressed magnon. Further the thermal excitation process and thermodynamic properties of the system are also investigated. Finally, we make a summary and concluding remarks in Sec. 4.

2 Model and Theory

The ground state of a simple ferromagnet has all spins parallel. Consider N spins each of magnitude S with neighbor spins coupled by the Heisenberg interaction. The Hamiltonian of the spin system can be written as

$$H = -J \sum_{l,\delta} \hat{\boldsymbol{S}}_l \cdot \hat{\boldsymbol{S}}_{l+\delta} , \qquad (1)$$

where J is the exchange integral, \hat{S}_l is the angular momentum of the spin at site r_l and δ represents the distances of the nearest lattices.

Now consider the excited state of the system, it can be obtained by offering some energy to the spin system, for

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example, increasing the temperature of the system. When a magnetic ion gets some energy from the outer, the angular momentum of the spin of it will deviate from the equilibrium position. Further, the spin deviation keeps moving about through the lattice in a wave-like manner, owing to the exchange interaction of all magnetic ions. If we introduce the creation and annihilation operators \hat{a}_l^{\dagger} and \hat{a}_l of the spin deviation at site \mathbf{r}_l and write them as

$$\hat{a}_{l} = \sqrt{N} \sum_{\boldsymbol{k}} \exp(\mathrm{i}\boldsymbol{k} \cdot \boldsymbol{r}_{l}) \hat{b}_{\boldsymbol{k}} ,$$
$$\hat{a}_{l}^{\dagger} = \sqrt{N} \sum_{\boldsymbol{k}} \exp(-\mathrm{i}\boldsymbol{k} \cdot \boldsymbol{r}_{l}) \hat{b}_{\boldsymbol{k}}^{\dagger} , \qquad (2)$$

using the Holstein–Primakoff transformation^[19]

$$S_{+} = (\sqrt{2S - \hat{a}^{\dagger} \hat{a}})\hat{a},$$

$$S_{-} = \hat{a}^{\dagger}(\sqrt{2S - \hat{a}^{\dagger} \hat{a}}), \quad S^{z} = S - \hat{a}^{\dagger} \hat{a}, \quad (3)$$

where \hat{S}_{\pm} is the spin-raising and spin-lowering operators and satisfies the relation $\hat{S}_{\pm} = \hat{S}_x \pm i \hat{S}_y$, the Hamiltonian of the system can be obtained in terms of \hat{b}_k^{\dagger} and \hat{b}_k

$$H = E_0 + \sum_{\boldsymbol{k}} \hbar w_{\boldsymbol{k}} \hat{b}^{\dagger}_{\boldsymbol{k}} \hat{b}_{\boldsymbol{k}}$$
$$+ \sum_{\boldsymbol{k}, \boldsymbol{k}', \boldsymbol{q}} V_{\boldsymbol{k}, \boldsymbol{k}'}(\boldsymbol{q}) \hat{b}^{\dagger}_{\boldsymbol{k}-\boldsymbol{q}} \hat{b}^{\dagger}_{\boldsymbol{k}'+\boldsymbol{q}} \hat{b}_{\boldsymbol{k}'} \hat{b}_{\boldsymbol{k}}, \qquad (4)$$

where $E_0 = -JS^2Nz$ is the energy of the ground state of the system. z is the number of the nearest neighbors of an ion. The operators \hat{b}_{k}^{\dagger} and \hat{b}_{k} are the creation and annihilation operators of the linear spin-wave mode which is quantized in unit

$$\hbar w_{\mathbf{k}} = 2JSz(1 - r_{\mathbf{k}}), \qquad (5)$$

where $r_{\mathbf{k}}$ is defined by

$$r_{\boldsymbol{k}} = \frac{1}{z} \sum_{\delta \neq 0} \exp(\mathrm{i} \boldsymbol{k} \cdot \delta)$$

The energy quanta $\hbar w_k$ of linear spin waves are called free magnons, which are bosons. For simplicity, we always call them magnons. In fact, the ground state of the system can be regarded as a Bose–Einstein condensation (BEC) configuration of the magnons. We know the system of magnons can be described in the context of the grand canonical ensemble. One can easily evaluate the total number of magnons

$$N = \sum_{i} \frac{1}{\exp\{(\varepsilon_i - \mu)/k_B T\} - 1},$$
(6)

which can be written as the sum of the average occupation numbers

$$\bar{n}_i = \frac{1}{\exp\{(\varepsilon_i - \mu)/k_B T\} - 1} \tag{7}$$

of each single-particle state. For the lowest single-particle energy ε_0 , when $T \to 0$ K the occupation number

$$N_0 \equiv \bar{n}_0 = \frac{1}{\exp\{(\varepsilon_0 - \mu)/k_B T\} - 1}$$
(8)

becomes increasingly large. At T = 0 K, the number of magnons in the condensate becomes macroscopic,

all magnons occur the lowest single-particle energy state which is the zero-momentum state. As a result, the ground state of a simple ferromagnet has all spins parallel. At low temperature, long-wave condition $|\mathbf{k} \cdot \delta| \ll 1$ can be taken. For $|\mathbf{k} \cdot \delta| \ll 1$

$$\hbar w_{\boldsymbol{k}} \approx 2JSa^2 \boldsymbol{k}^2 \,, \tag{9}$$

for all three cubic lattices, where a is the lattice constant. The third term in Eq. (4) describes the magnon-magnon interaction. The interaction matrix element is given by

$$V_{k,k'}(q) = -\frac{Jz}{2N} (2r_{k-q-k'} - r_{k-q} - r_{k'}).$$
(10)

Obviously, it can be negative, which represents the attractive interaction between the magnons. The presence of the attractive interaction has an inherent mechanism. It leads to bound magnon-pairs which are stable only if the two magnons have opposite wave vectors \mathbf{k} and $-\mathbf{k}$. At low temperatures, we can ignore the repulsive interaction between the magnons and rewrite the Hamiltonian as

$$H = E_0 + \sum_{\boldsymbol{k}} \hbar w_{\boldsymbol{k}} \hat{b}_{\boldsymbol{k}} \hat{b}_{\boldsymbol{k}}^{\dagger} + \sum_{\boldsymbol{k},\boldsymbol{k}'} V_{\boldsymbol{k},\boldsymbol{k}'} \hat{b}_{\boldsymbol{k}'}^{\dagger} \hat{b}_{-\boldsymbol{k}'}^{\dagger} \hat{b}_{\boldsymbol{k}} \hat{b}_{-\boldsymbol{k}} , \quad (11)$$

here,

$$V_{k,k'} = -\frac{Jz}{2N} (2r_{k+k'} - r_k - r_{k'}).$$
(12)

At low temperatures, taking the reasonable approximation $\mathbf{r}_{\mathbf{k}+\mathbf{k}'} \approx r_{\mathbf{k}}r_{\mathbf{k}'}$ and noting $2r_{\mathbf{k}} \approx r_{\mathbf{k}}$, Eq. (12) becomes approximately

$$V_{\boldsymbol{k},\boldsymbol{k}'} \approx -\frac{\hbar w_{\boldsymbol{k}} \hbar w_{\boldsymbol{k}'}}{8NJS^2 z}, \qquad (13)$$

where $\hbar w_{\mathbf{k}} \approx 2JSa^2 \mathbf{k}^2$ and \mathbf{k} takes all possible values of the \mathbf{k} space.

Within the framework of a mean-field theory^[20]

$$\hat{b}_{\boldsymbol{k}}^{\dagger}\hat{b}_{-\boldsymbol{k}}^{\dagger}\approx\langle\hat{b}_{\boldsymbol{k}}^{\dagger}\hat{b}_{-\boldsymbol{k}}^{\dagger}\rangle_{T},\quad\hat{b}_{\boldsymbol{k}}\hat{b}_{-\boldsymbol{k}}\approx\langle\hat{b}_{\boldsymbol{k}}\hat{b}_{-\boldsymbol{k}}\rangle_{T},\qquad(14)$$

the Hamiltonian of the system becomes approximately

$$H \approx \frac{1}{2} \sum_{\boldsymbol{k}} \hbar w_{\boldsymbol{k}} (b^{\dagger}_{\boldsymbol{k}} b_{\boldsymbol{k}} + b^{\dagger}_{-\boldsymbol{k}} b_{-\boldsymbol{k}})$$

+
$$\sum_{\boldsymbol{k},\boldsymbol{k}'} V_{\boldsymbol{k},\boldsymbol{k}'} \{b^{\dagger}_{\boldsymbol{k}'} b^{\dagger}_{-\boldsymbol{k}'} \langle \hat{b}^{\dagger}_{\boldsymbol{k}} \hat{b}^{\dagger}_{-\boldsymbol{k}} \rangle_{T}$$

+
$$\hat{b}_{\boldsymbol{k}} \hat{b}_{-\boldsymbol{k}} \langle \hat{b}^{\dagger}_{\boldsymbol{k}'} \hat{b}^{\dagger}_{-\boldsymbol{k}'} \rangle_{T} - \langle \hat{b}_{\boldsymbol{k}} \hat{b}_{-\boldsymbol{k}} \rangle_{T} \langle \hat{b}^{\dagger}_{\boldsymbol{k}'} \hat{b}^{\dagger}_{-\boldsymbol{k}'} \rangle_{T}, \qquad (15)$$

where the energy of the ground state of the system is ignored. Introducing the definitions

$$\Delta = \sum_{\boldsymbol{k}} \alpha \hbar w_{\boldsymbol{k}} \langle \hat{b}_{\boldsymbol{k}} \hat{b}_{-\boldsymbol{k}} \rangle_T , \quad \Delta^* = \sum_{\boldsymbol{k}} \alpha \hbar w_{\boldsymbol{k}} \langle \hat{b}_{\boldsymbol{k}}^{\dagger} \hat{b}_{-\boldsymbol{k}}^{\dagger} \rangle_T , \quad (16)$$

the Hamiltonian of the system has the form

$$H = \frac{1}{2} \sum_{\mathbf{k}} h w_{\mathbf{k}} (\hat{b}^{\dagger}_{\mathbf{k}} \hat{b}_{\mathbf{k}} + \hat{b}^{\dagger}_{-\mathbf{k}} \hat{b}_{-\mathbf{k}}) + \Delta \sum_{\mathbf{k}} \hbar w_{\mathbf{k}} (\hat{b}^{\dagger}_{\mathbf{k}} \hat{b}^{\dagger}_{-\mathbf{k}} + \hat{b}_{\mathbf{k}} \hat{b}_{-\mathbf{k}}) - \Delta^2 / \alpha , \qquad (17)$$

where Δ satisfies the relation $\Delta = \Delta^* = |\Delta|$ and α is given by $\alpha = -1/8NJS^2z$. Bogoliubov's method of making a canonical transformation of the set, $\hat{b}^{\dagger}_{\mathbf{k}}$ and $\hat{b}_{\mathbf{k}}$, to new creation and annihilation operators is suitable for our model. For simplicity let us drop the spin indices and write

$$\hat{b}_{\boldsymbol{k}} = \mu_{\boldsymbol{k}} \hat{c}_{\boldsymbol{k}} - \nu_{\boldsymbol{k}} \hat{c}^{\dagger}_{-\boldsymbol{k}} \hat{b}^{\dagger}_{\boldsymbol{k}} = \mu_{\boldsymbol{k}} \hat{c}^{\dagger}_{\boldsymbol{k}} - \nu_{\boldsymbol{k}} \hat{c}_{-\boldsymbol{k}} ,$$
$$\hat{b}_{-\boldsymbol{k}} = \mu_{\boldsymbol{k}} \hat{c}_{-\boldsymbol{k}} - \nu_{\boldsymbol{k}} \hat{c}^{\dagger}_{\boldsymbol{k}} \hat{b}^{\dagger}_{-\boldsymbol{k}} = \mu_{\boldsymbol{k}} \hat{c}^{\dagger}_{-\boldsymbol{k}} - \nu_{\boldsymbol{k}} \hat{c}_{\boldsymbol{k}} , \qquad (18)$$

where the coefficients $\mu_{\boldsymbol{k}}$ and $\nu_{\boldsymbol{k}}$ satisfy the following equation

$$\mu_{k}^{2} - \nu_{k}^{2} = 1, \quad \mu_{k}^{2} + \nu_{k}^{2} = \mu_{k}\nu_{k}/\Delta.$$
 (19)

Substituting the canonical transformation (18) into Eq. (17), the Hamiltonian of the system is diagonalized into

$$H = E_s + \sum_{\boldsymbol{k}} \hbar w_{\boldsymbol{k}} e(T) \hat{c}^{\dagger}_{\boldsymbol{k}} \hat{c}_{\boldsymbol{k}} , \qquad (20)$$

where E_s represents a metastable configuration and $\hbar w_{\mathbf{k}} e(T)$ are the energy quanta of elementary excitations of the system. The result has a deep physical meaning. It shows that the original system of interacting particles can be described in terms of a Hamitonian of independent quasi-particles having energy $\hbar w_{\mathbf{k}} e(T)$ and whose annihilation and creation operators are given, respectively, by $\hat{c}_{\mathbf{k}}$ and $\hat{c}_{\mathbf{k}}^{\dagger}$. We call the energy quanta $\hbar w_{\mathbf{k}} e(T)$ dressed magnons. The dressed magnons are also bosons.

3 Results and Discussions

The dressed magnons are also the energy quanta of magnon-pairs of the spin system, which carry all the information of the interaction between the magnons. The energy of a dressed magnon is determined by the correction factor e(T) which is a function of temperature T. Substituting the Bogoliubov's transformation given in Eq. (18) into Eq. (16), the correction factor e(T) can be reduced as

$$e(T) = -\alpha \sum_{\boldsymbol{k}} \hbar w_{\boldsymbol{k}} (1 + 2\langle \hat{c}_{\boldsymbol{k}}^{\dagger} \hat{c}_{\boldsymbol{k}} \rangle_T) \,. \tag{21}$$

Since the dressed magnons obey Bose–Einstein statistics one can obtain the average number of the dressed magnons excited at temperature T with wave vector \mathbf{k}

$$\langle \hat{c}_{\boldsymbol{k}}^{\dagger} \hat{c}_{\boldsymbol{k}} \rangle_T = \frac{1}{\exp\{\hbar w_{\boldsymbol{k}} e(T)/k_B T\} - 1}, \qquad (22)$$

where k_B is the Boltzmann constant. At T = 0 K, there is no thermal excitation, so the correction factor e(0) can be written as

$$e(0) = -\alpha \sum_{k} \hbar w_{k} , \qquad (23)$$

which is very important for us to analyze the lowtemperature properties of a ferromagnet. In fact, at T = 0 K the frequency of magnons can be confined within the effective Debye cutoff frequency, which is given by $\hbar w_D^* \approx 6.58 \times 10^2$ meV. Casting Eq. (23) into integral form, we can obtain

$$e(0) = -\frac{2}{5}\alpha\beta(\hbar w_D^*)^{5/2}, \qquad (24)$$

where β is given by

$$\beta = \frac{V}{4\pi^2} \left(\frac{1}{2JSa^2}\right)^{3/2}$$

At low temperatures, $e(T) \approx e(0)$ and $\hbar w_{\mathbf{k}} \approx 2JSa^2 \mathbf{k}^2$, Eq. (22) can be written approximately as

$$\langle \hat{c}_{\boldsymbol{k}}^{\dagger} \hat{c}_{\boldsymbol{k}} \rangle_T \approx \frac{1}{\exp\{2JSa^2 \boldsymbol{k}^2 e(0)/k_B T\} - 1}$$
 (25)

Obviously, the energy of dressed magnons is a monotonically increasing function of temperature T, which is equal to the energy of magnons at the critical temperature T_0 . In fact the critical temperature indeed has a deep physical meaning. The repulsive interaction between the magnons can be regarded as the result of random motion of magnons. The strength of it depends on the effective kinetic energy of magnons. The attractive interaction comes from the lattice deformation. At low temperatures, the effective kinetic energy of magnons is very small. As a result, we can only consider the attractive interaction between two magnons with opposite wave vectors \boldsymbol{k} and -k. The attractive interaction leads to bound magnonpairs. The repulsive interaction strengthens as the temperature rises. At critical temperature T_0 , the magnons will escape the bondage of attractive interaction and the repulsive interaction between the magnons are dominant. Consequently, at critical temperature T_0 the magnon-pairs disappear and all dressed magnons turn into magnons. Substituting $e(T_0) = 1$ into Eq. (18) we know the critical temperature T_0 satisfies the equation

$$1 = -\alpha \sum_{\boldsymbol{k}} \hbar w_{\boldsymbol{k}} \coth \hbar w_{\boldsymbol{k}} / 2k_{\scriptscriptstyle B} T_0 \,. \tag{26}$$

The total number of dressed magnons excited at temperature $T \ (T \ll T_c)$ have the form

$$\sum_{\boldsymbol{k}} \langle \hat{c}_{\boldsymbol{k}}^{\dagger} \hat{c}_{\boldsymbol{k}} \rangle_T = \gamma T^{3/2} \,, \tag{27}$$

where T_c is the Curie temperature and γ is given by

$$\gamma = \left\{ \frac{V}{4\pi^2} \left(\frac{k_B}{2SJa^2} \right)^{3/2} \int_0^\infty \frac{x^{1/2}}{e^x - 1} dx \right\} e(0)^{-3/2}.$$
 (28)

Now consider the low-temperature magnetization of the system, it should decrease as the temperature increases according to the formula

$$NS\left(\frac{M(0) - M(T)}{M(0)}\right) = \sum_{\boldsymbol{k}} \langle \hat{c}_{\boldsymbol{k}}^{\dagger} \hat{c}_{\boldsymbol{k}} \rangle_T = \gamma T^{3/2}, \quad (29)$$

where M(0) is the saturation magnetization at T = 0 K. Comparing with Bloch's $T^{3/2} \, \text{law}^{[21]}$

$$NS\left(\frac{M(0) - M(T)}{M(0)}\right) = \left\{\frac{\gamma}{e(0)^{-3/2}}\right\} T^{3/2}, \qquad (30)$$

which has not considered the interaction between the magnons, we can find our result adds a correction factor $e(0)^{-3/2}$. The correction factor comes from the attractive interactions between the magnons. Taking Fe crystal for example, the parameters of it are as follow: S = 1, z = 8, and $J = 11.9 \text{ meV}.^{[22]}$ The correct factor of it is obtained as e(0) = 0.64. According to Eqs. (29) and (30), the relation between the fractional change of magnetization $\Delta M/M(0)$ and temperature T is shown in Fig. 1. From Fig. 1, we can find that the influence of magnon-magnon

interaction on the magnetization of the system becomes more obvious as the temperature rises.



Fig. 1 The fractional change of magnetization of Fe crystal as functions of temperature: the solid line refers to our conclusion, which considers the interactions between the magnons, the dotted line is Bloch's conclusion which ignores the interactions.

The additional energy caused by the thermal excitation of the system is

$$\Delta E(T) = \sum_{\mathbf{k}} \hbar w_{\mathbf{k}} e(T) \langle \hat{c}_{\mathbf{k}}^{\dagger} \hat{c}_{\mathbf{k}} \rangle_{T} \,. \tag{31}$$

At low temperature the additional energy can be reduced as

$$\Delta E(T) = \frac{V}{4\pi^2} \left(\frac{1}{2JSa^2 e(0)}\right)^{3/2} \Gamma\left(\frac{5}{2}\right) \zeta\left(\frac{5}{2}\right) (k_B T)^{5/2} . \tag{32}$$

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The low-temperature specific heat capacity of the system can be obtained from Eq. (29), which has the form

$$c_m = \frac{\partial \Delta E(T)}{\partial T} = \zeta T^{3/2} , \qquad (33)$$

where ζ is given by

$$\zeta = \frac{5V}{8\pi^2} \left(\frac{1}{2JSa^2 e(0)}\right)^{3/2} \Gamma\left(\frac{5}{2}\right) \zeta\left(\frac{5}{2}\right)^{5/2} k_B^{5/2}$$

It should be noted that our model is not suitable for ferromagnets with low Curie temperature, like EuO. For them, the repulsive interactions between the magnons can not be ignored at low temperature.

4 Conclusion

In summary, considering the attractive interaction between two magnons with opposite wave vectors \mathbf{k} and $-\mathbf{k}$, we establish the magnon-pair model for Heisenberg ferromagnet. We call the energy quanta of the magnon-pairs dressed magnons whose energy monotonically increases as the temperature rises. At critical temperature T_0 , the magnon-pairs disappear and the dressed magnons all turn into magnons. For $T > T_0$, the repulsive interaction between the magnons is dominant. At low temperatures, based on the model, we also investigate the fractional change of magnetization and specific heat capacity of the system. It is shown that our results add a correction factor $e(0)^{-3/2}$, comparing the results without considering the interactions between the magnons.

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