

## Chapter 2

# SPIN WAVES IN FERROMAGNETS

### 2.1. Spin-Boson Transformation

The model Hamiltonian we will study in this chapter is the Heisenberg Hamiltonian for a ferromagnet, that is an exchange Hamiltonian with NN ferromagnetic interaction. The aim of the present section is to find a transformation from the spin operators to Bose “creation” and “destruction” operators which are essential to build a perturbative theory to infinite order. First of all, let us compare the matrix elements of the spin operators and the Bose operators. From the matrix elements given in Eqs. (1.3.6) and (1.3.7), one can write the spin operators  $S_i^+$ ,  $S_i^-$ ,  $S_i^z$  and  $S_i^2$  as square matrices of dimension  $(2S + 1)$ . For instance, for  $S = 1$ , one obtains

$$S_i^+ = \begin{pmatrix} 0 & \sqrt{2} & 0 \\ 0 & 0 & \sqrt{2} \\ 0 & 0 & 0 \end{pmatrix}, \quad S_i^- = \begin{pmatrix} 0 & 0 & 0 \\ \sqrt{2} & 0 & 0 \\ 0 & \sqrt{2} & 0 \end{pmatrix}, \quad S_i^z = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & -1 \end{pmatrix}$$

$$S_i^2 = \begin{pmatrix} 2 & 0 & 0 \\ 0 & 2 & 0 \\ 0 & 0 & 2 \end{pmatrix} = S(S+1) \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix}. \quad (2.1.1)$$

The Bose creation and destruction operators satisfy the commutation rules<sup>5</sup>

$$[a_i, a_j^+] = \delta_{i,j}, \quad [a_i, a_j] = [a_i^+, a_j^+] = 0. \quad (2.1.2)$$

In the *occupation number* representation where the operator number  $n_i = a_i^+ a_i$  is diagonal, one has

$$\langle n' | a_i^+ a_i | n \rangle = n \delta_{n,n'}, \quad n \geq 0. \quad (2.1.3)$$

The matrix elements of  $a_i$  e  $a_i^+$  are given by

$$\langle n' | a_i | n \rangle = \sqrt{n} \delta_{n',n-1}, \quad \langle n' | a_i^+ | n \rangle = \sqrt{n+1} \delta_{n',n+1}. \quad (2.1.4)$$

From the matrix elements (2.1.3) and (2.1.4), one sees that the Bose operators can be written as matrices of *infinite* dimension like

$$a_i = \begin{pmatrix} 0 & \sqrt{1} & 0 & \dots \\ 0 & 0 & \sqrt{2} & \dots \\ 0 & 0 & 0 & \dots \\ \vdots & \vdots & \vdots & \ddots \end{pmatrix}, \quad a_i^+ = \begin{pmatrix} 0 & 0 & 0 & \dots \\ \sqrt{1} & 0 & 0 & \dots \\ 0 & \sqrt{2} & 0 & \dots \\ \vdots & \vdots & \vdots & \ddots \end{pmatrix}$$

$$n_i = a_i^+ a_i = \begin{pmatrix} 0 & 0 & 0 & \dots \\ 0 & 1 & 0 & \dots \\ 0 & 0 & 2 & \dots \\ \vdots & \vdots & \vdots & \ddots \end{pmatrix}. \quad (2.1.5)$$

The great advantage of representing the spin operators  $S_i^\pm$ ,  $S_i^z$  in terms of Bose operators  $a_i^+$ ,  $a_i$  is that the commutation rules of the latter are *c*-numbers and this fact is crucial for building a series expansion of the Green function or propagator in a systematic way.<sup>13</sup> A spin-boson transformation has to satisfy the following requirements:

- (1) The transformation must be *hermitian* such that the conjugation relations are preserved. This means that the raising and lowering spin operators written in terms of creation and destruction boson operators must be hermitian conjugate of each other.
- (2) The transformation must be a *unitary* one in order to preserve the commutation rules between the spin operators given by Eq. (1.3.4) when the spin operators are expressed in terms of Bose operators for which the commutation rules are given by Eq. (2.1.2).
- (3) The transformation must satisfy the equality between the matrix elements of the spin operators between the states  $|Sm\rangle$  and the matrix elements of their bosonic representations between the states  $|n\rangle$ .

The choice of such a transformation is not unique. The first spin-boson transformation was proposed long time ago by Holstein and Primakoff<sup>14</sup> (HP)

$$S_i^+ = \sqrt{2S} \left( 1 - \frac{a_i^+ a_i}{2S} \right)^{\frac{1}{2}} a_i, \quad S_i^- = \sqrt{2S} a_i^+ \left( 1 - \frac{a_i^+ a_i}{2S} \right)^{\frac{1}{2}}, \quad S_i^z = S - a_i^+ a_i. \quad (2.1.6)$$

The HP transformation satisfies the properties (1)–(3) only if the restriction to the boson subspace with  $n_i \leq 2S$  is accounted for because of the presence of the square roots in Eq. (2.1.6). About twenty years later, a new transformation was proposed by Dyson<sup>15</sup> and Maleev<sup>16</sup> (DM) as follows:

$$S_i^+ = \sqrt{2S} \left( 1 - \frac{a_i^+ a_i}{2S} \right) a_i, \quad S_i^- = \sqrt{2S} a_i^+ \quad \text{and} \quad S_i^z = S - a_i^+ a_i \quad (2.1.7)$$

with the restriction  $n_i \leq 2S$  (physical states). The DM transformation does not satisfy requirement (1) since it is not hermitian as seen by Eq. (2.1.7). However, Dyson himself showed that even though the DM representation transforms the Heisenberg Hamiltonian into a non-hermitian boson Hamiltonian, the matrix elements of the Heisenberg Hamiltonian between the localized spin states are the same as the boson DM Hamiltonian between independent boson states, provided that only physical states are accounted for, that is  $n_i \leq 2S$  (kinematical interaction). Moreover, Dyson showed that the release of the restriction  $n_i \leq 2S$ , that is the involvement of the *unphysical* states with  $n_i \geq 2S$ , does not affect the power law contributions in the low temperature results. Indeed, the effect of the unphysical states is proved to be lesser than  $e^{-a \frac{T_c}{T}}$  where  $T_c$  is the Curie temperature and  $a$  is a numerical coefficient of the order of unity and independent of temperature.<sup>15</sup> A similar proof cannot be extended to the HP Hamiltonian.

## 2.2. Bosonic Approach to the Heisenberg Hamiltonian

The Heisenberg Hamiltonian in terms of the raising and lowering spin operators is given by Eq. (1.3.8). In the absence of the external magnetic field, Eq. (1.3.8) becomes

$$\mathcal{H} = -J \sum_{i,\delta} \left[ S_i^z S_{i+\delta}^z + \frac{1}{2} (S_i^+ S_{i+\delta}^- + S_i^- S_{i+\delta}^+) \right]. \quad (2.2.1)$$

Now, let us replace the spin raising and lowering operators by the boson creation and destruction operators according to the HP transformation given by Eq. (2.1.6) or to the DM transformation given by Eq. (2.1.7). Let us begin with the HP transformation and expand the square roots in the HP transformation in powers of  $a_i^+ a_i$  or even better, in a *normal ordered*<sup>17</sup> (NO) series expansion where all the creation operators  $a_i^+$  are on the left of all the destruction operators  $a_i$ . So the HP transformation process can be written as

$$\begin{aligned} S_i^+ &= \sqrt{2S} \left[ 1 - \frac{a_i^+ a_i}{4S} - \frac{(a_i^+ a_i)^2}{32S^2} \dots \right] a_i = \sqrt{2S} \left\{ 1 - \left( 1 - \sqrt{1 - \frac{1}{2S}} \right) a_i^+ a_i \right. \\ &\quad \left. + \left[ 1 - \sqrt{1 - \frac{1}{2S}} - \frac{1}{2} \left( 1 - \sqrt{1 - \frac{1}{S}} \right) \right] (a_i^+)^2 (a_i)^2 \dots \right\} a_i, \end{aligned} \quad (2.2.2)$$

$$\begin{aligned} S_i^- &= \sqrt{2S} a_i^+ \left[ 1 - \frac{a_i^+ a_i}{4S} - \frac{(a_i^+ a_i)^2}{32S^2} \dots \right] = \sqrt{2S} a_i^+ \left\{ 1 - \left( 1 - \sqrt{1 - \frac{1}{2S}} \right) a_i^+ a_i \right. \\ &\quad \left. + \left[ 1 - \sqrt{1 - \frac{1}{2S}} - \frac{1}{2} \left( 1 - \sqrt{1 - \frac{1}{S}} \right) \right] (a_i^+)^2 (a_i)^2 \dots \right\} \end{aligned} \quad (2.2.3)$$

and

$$S_i^z = S - a_i^+ a_i. \quad (2.2.4)$$

The advantage of the NO expansion is that no terms with a smaller number of boson operators are produced by the higher order terms: this is not true for the usual expansion in powers of  $(a_i^+ a_i)^n$ . For instance, the term with 4 operators corresponding to  $n = 2$  produces a NO term with 4 operators as well as a term with 2 operators: indeed,  $a_i^+ a_i a_i^+ a_i = a_i^+ a_i^+ a_i a_i + a_i^+ [a_i, a_i^+] a_i = a_i^+ a_i^+ a_i a_i + a_i^+ a_i$ . In the same way, the term with 6 operators ( $n = 3$ ) produces NO terms with 6, 4 and 2 operators and so on. The truncation of the series (2.2.2) and (2.2.3) to the first few terms is reliable for large spin  $S$  or most importantly, for small values of the average occupation number  $\langle a_i^+ a_i \rangle$ . The last condition is satisfied for any  $S$  at low temperature where the number of spin deviations is small and the restriction  $\langle n_i \rangle / 2S \leq 1$  may be neglected. Replacing the spin operators in the Hamiltonian (2.2.1) by Eqs. (2.2.2)–(2.2.4), one obtains

$$\mathcal{H}^{HP} = E_0^{HP} + \mathcal{H}_2^{HP} + \mathcal{H}_4^{HP} + \mathcal{H}_6^{HP} + \dots \quad (2.2.5)$$

where

$$E_0^{HP} = -zJS^2N, \quad (2.2.6)$$

$$\mathcal{H}_2^{HP} = JS \sum_{i,\delta} (a_i^+ a_i + a_{i+\delta}^+ a_{i+\delta} - a_{i+\delta}^+ a_i - a_i^+ a_{i+\delta}), \quad (2.2.7)$$

$$\begin{aligned} \mathcal{H}_4^{HP} = -J \sum_{i,\delta} & \left[ \frac{1}{2} (a_i^+ a_{i+\delta}^+ a_i a_{i+\delta} + \text{h.c.}) - S \left( 1 - \sqrt{1 - \frac{1}{2S}} \right) \right. \\ & \left. \times (a_{i+\delta}^+ a_i^+ a_i a_i + a_{i+\delta}^+ a_{i+\delta}^+ a_{i+\delta} a_{i+\delta} + \text{h.c.}) \right], \end{aligned} \quad (2.2.8)$$

$$\begin{aligned} \mathcal{H}_6^{HP} = -JS \sum_{i,\delta} & \left\{ \left[ 1 - \sqrt{1 - \frac{1}{2S}} - \frac{1}{2} \left( 1 - \sqrt{1 - \frac{1}{S}} \right) \right] (a_{i+\delta}^+ a_i^+ a_i^+ a_i a_i a_i \right. \\ & + a_{i+\delta}^+ a_{i+\delta}^+ a_{i+\delta}^+ a_{i+\delta} a_{i+\delta} a_i + \text{h.c.}) \\ & \left. + \left( 1 - \sqrt{1 - \frac{1}{2S}} \right)^2 (a_{i+\delta}^+ a_{i+\delta}^+ a_i^+ a_{i+\delta} a_i a_i + \text{h.c.}) \right\} \end{aligned} \quad (2.2.9)$$

where “h.c.” means “hermitian conjugate”. Remember that the hermitian conjugate of a product of operators can be written as  $(AB)^+ = B^+ A^+$ . Notice that the usual HP expansion occurring in the textbooks is obtained from Eq. (2.2.8) replacing  $S(1 - \sqrt{1 - \frac{1}{2S}})$  by the first few terms of its series expansion in powers of  $\frac{1}{S}$  that is by  $\frac{1}{4}(1 + \frac{1}{8S} + \frac{1}{32S^2})$  and from Eq. (2.2.9) replacing  $S[1 - \sqrt{1 - \frac{1}{2S}} - \frac{1}{2}(1 - \sqrt{1 - \frac{1}{S}})]$  by  $-\frac{1}{32S}(1 + \frac{3}{4S})$  and  $S(1 - \sqrt{1 - \frac{1}{2S}})^2$  by  $\frac{1}{16S}(1 + \frac{1}{4S})$ . The main point to be noticed using the HP transformation is the generation of an *infinite* number of terms when the Heisenberg Hamiltonian is transformed into the equivalent boson Hamiltonian. On the contrary, the DM transformation (2.1.7) leads to an equivalent

boson Hamiltonian consisting of only three terms given by

$$\mathcal{H}^{DM} = E_0^{DM} + \mathcal{H}_2^{DM} + \mathcal{H}_4^{DM} \quad (2.2.10)$$

where

$$E_0^{DM} = -zJS^2N, \quad (2.2.11)$$

$$\mathcal{H}_2^{DM} = JS \sum_{i,\delta} (a_i^+ a_i + a_{i+\delta}^+ a_{i+\delta} - a_{i+\delta}^+ a_i - a_i^+ a_{i+\delta}) \quad (2.2.12)$$

and

$$\mathcal{H}_4^{DM} = -J \sum_{i,\delta} \left[ \frac{1}{2} (a_i^+ a_{i+\delta}^+ a_i a_{i+\delta} + \text{h.c.}) - \frac{1}{2} (a_{i+\delta}^+ a_i^+ a_i a_i + a_i^+ a_{i+\delta}^+ a_{i+\delta} a_{i+\delta}) \right]. \quad (2.2.13)$$

The ground-state energy  $E_0^{DM}$  and the bilinear Hamiltonian  $\mathcal{H}_2^{DM}$  coincide with those obtained by the HP transformation [compare Eq. (2.2.11) with (2.2.6) and Eq. (2.2.12) with (2.2.7)]. The interaction Hamiltonian is now reduced to the *single* term  $\mathcal{H}_4^{DM}$  even though it is no longer hermitian as one can see from the second term of Eq. (2.2.13). In any case, this is not a serious problem because the DM boson Hamiltonian has been proven<sup>15</sup> to have the same matrix elements between the independent boson states as the original Heisenberg Hamiltonian between the spin states. Moreover, the kinematical interaction ( $n_i \leq 2S$ ) can be ignored since it enters only exponentially small terms in the temperature expansion.

So far, we have considered localized spin deviations but the spin waves are delocalized excitations as we have seen in Section 1.3. For this reason, let us define the spin wave or *magnon* creation and destruction operators by using the Fourier transforms of the corresponding localized operators (2.1.2):

$$a_{\mathbf{k}} = \frac{1}{\sqrt{N}} \sum_i e^{-i\mathbf{k} \cdot \mathbf{r}_i} a_i, \quad a_{\mathbf{k}}^+ = \frac{1}{\sqrt{N}} \sum_i e^{i\mathbf{k} \cdot \mathbf{r}_i} a_i^+ \quad (2.2.14)$$

whose commutation rules, as a direct consequence of Eq. (2.1.2), are given by

$$[a_{\mathbf{k}}, a_{\mathbf{k}'}^+] = \delta_{\mathbf{k}, \mathbf{k}'}, \quad [a_{\mathbf{k}}, a_{\mathbf{k}'}] = [a_{\mathbf{k}}^+, a_{\mathbf{k}'}^+] = 0. \quad (2.2.15)$$

Replacing the magnon operators (2.2.14) into the HP boson Hamiltonian (2.2.5), the spin wave or magnon Hamiltonian becomes

$$\mathcal{H}_2^{HP} = zJS \sum_{\mathbf{k}} [(1 - \gamma_{\mathbf{k}}) + \text{c.c.}] a_{\mathbf{k}}^+ a_{\mathbf{k}} \quad (2.2.16)$$

where “c.c.” means “complex conjugate”: for instance, the complex conjugate of  $1 - \gamma_{\mathbf{k}}$  is  $1 - \gamma_{\mathbf{k}}^*$  with

$$\gamma_{\mathbf{k}} = \frac{1}{z} \sum_{\delta} e^{i\mathbf{k} \cdot \delta} \quad (2.2.17)$$

where  $z$  is the coordination number. The Hamiltonian (2.2.16) of non-interacting spin waves is called “harmonic” or “bilinear” Hamiltonian. The magnon-magnon

interaction Hamiltonian is given by

$$\mathcal{H}_4^{HP} = -\frac{zJ}{N} \sum_{\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3, \mathbf{k}_4} \delta_{\mathbf{k}_1+\mathbf{k}_2, \mathbf{k}_3+\mathbf{k}_4} v_{\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3, \mathbf{k}_4}^{HP} a_{\mathbf{k}_1}^+ a_{\mathbf{k}_2}^+ a_{\mathbf{k}_3} a_{\mathbf{k}_4} \quad (2.2.18)$$

with

$$v_{\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3, \mathbf{k}_4}^{HP} = \frac{1}{2} \gamma_{\mathbf{k}_1 - \mathbf{k}_3} - S \left( 1 - \sqrt{1 - \frac{1}{2S}} \right) (\gamma_{\mathbf{k}_1} + \gamma_{\mathbf{k}_2}) + \text{c.c.} \quad (2.2.19)$$

and

$$\begin{aligned} \mathcal{H}_6^{HP} = & -\frac{zJ}{N^2} \sum_{\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3, \mathbf{k}_4, \mathbf{k}_5, \mathbf{k}_6} \\ & \times \delta_{\mathbf{k}_1+\mathbf{k}_2+\mathbf{k}_3, \mathbf{k}_4+\mathbf{k}_5+\mathbf{k}_6} w_{\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3, \mathbf{k}_4, \mathbf{k}_5, \mathbf{k}_6}^{HP} a_{\mathbf{k}_1}^+ a_{\mathbf{k}_2}^+ a_{\mathbf{k}_3}^+ a_{\mathbf{k}_4} a_{\mathbf{k}_5} a_{\mathbf{k}_6} \end{aligned} \quad (2.2.20)$$

where

$$\begin{aligned} w_{\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3, \mathbf{k}_4, \mathbf{k}_5, \mathbf{k}_6}^{HP} = & S \left( 1 - \sqrt{1 - \frac{1}{2S}} \right)^2 \gamma_{\mathbf{k}_1+\mathbf{k}_2-\mathbf{k}_4} \\ & + S \left[ 1 - \sqrt{1 - \frac{1}{2S}} - \frac{1}{2} \left( 1 - \sqrt{1 - \frac{1}{S}} \right) \right] (\gamma_{\mathbf{k}_1} + \gamma_{\mathbf{k}_6}) + \text{c.c.} \end{aligned} \quad (2.2.21)$$

To obtain Eqs. (2.2.16)–(2.2.21), the following relationship has been used

$$\frac{1}{N} \sum_i e^{i(\mathbf{k}_1+\mathbf{k}_2-\mathbf{k}_3-\mathbf{k}_4) \cdot \mathbf{r}_i} = \delta_{\mathbf{k}_1+\mathbf{k}_2, \mathbf{k}_3+\mathbf{k}_4}. \quad (2.2.22)$$

Replacing the magnon operators (2.2.14) into the DM Hamiltonian (2.2.10), one obtains

$$\mathcal{H}_2^{DM} = zJS \sum_{\mathbf{k}} [(1 - \gamma_{\mathbf{k}}) + \text{c.c.}] a_{\mathbf{k}}^+ a_{\mathbf{k}} \quad (2.2.23)$$

and

$$\mathcal{H}_4^{DM} = -\frac{zJ}{N} \sum_{\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3, \mathbf{k}_4} \delta_{\mathbf{k}_1+\mathbf{k}_2-\mathbf{k}_3-\mathbf{k}_4} v_{\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3, \mathbf{k}_4}^{DM} a_{\mathbf{k}_1}^+ a_{\mathbf{k}_2}^+ a_{\mathbf{k}_3} a_{\mathbf{k}_4} \quad (2.2.24)$$

where

$$v_{\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3, \mathbf{k}_4}^{DM} = \frac{1}{2} [(\gamma_{\mathbf{k}_1-\mathbf{k}_3} - \gamma_{\mathbf{k}_1}) + \text{c.c.}]. \quad (2.2.25)$$

The magnon-magnon interaction potential  $v^{DM}$  (dynamical interaction) goes to zero in the long wavelength limit [see Eq. (2.2.25)] so that the interaction between the spin waves of small wavevector becomes negligible. This is not the case for the interaction potential in the HP Hamiltonian as one can see from Eqs. (2.2.19) and (2.2.21). However, it was proven<sup>18</sup> that the results obtained from the boson HP Hamiltonian (2.2.16)–(2.2.21) reduce to those obtained from the DM Hamiltonian (2.2.23)–(2.2.25) if a grouping of all terms of the same order in  $\frac{1}{S}$  is performed.

Note that the function  $\gamma_{\mathbf{k}}$  is real for lattices with inversion symmetry where for each spin at the site  $i + \delta$  exists a spin located at  $i - \delta$ . In particular, for all Bravais lattices we have  $\gamma_{\mathbf{k}} = \gamma_{-\mathbf{k}} = \gamma_{\mathbf{k}}^*$  so that everywhere, the sign “c.c.” can be neglected provided that the spin wave potentials are multiplied by a factor 2.

Let us conclude this section by writing the boson representation of the anisotropic Hamiltonians given by Eqs. (1.7.1)–(1.7.3):

$$\mathcal{H}_H = -h \sum_i S_i^z = -hSN + h \sum_{\vec{k}} a_{\vec{k}}^+ a_{\vec{k}}, \quad (2.2.26)$$

$$\begin{aligned} \mathcal{H}_D = -D \sum_i (S_i^z)^2 = -DS^2N + D(2S-1) \sum_{\vec{k}} a_{\vec{k}}^+ a_{\vec{k}} \\ - \frac{D}{N} \sum_{\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3, \mathbf{k}_4} \delta_{\mathbf{k}_1 + \mathbf{k}_2, \mathbf{k}_3 + \mathbf{k}_4} a_{\mathbf{k}_1}^+ a_{\mathbf{k}_2}^+ a_{\mathbf{k}_3} a_{\mathbf{k}_4} \end{aligned} \quad (2.2.27)$$

and

$$\begin{aligned} \mathcal{H}_K = -K \sum_{i, \delta} S_i^z S_{i+\delta}^z = -zKS^2N + 2zKS \sum_{\vec{k}} a_{\vec{k}}^+ a_{\vec{k}} \\ - \frac{zK}{N} \sum_{\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3, \mathbf{k}_4} \delta_{\mathbf{k}_1 + \mathbf{k}_2, \mathbf{k}_3 + \mathbf{k}_4} \gamma_{\mathbf{k}_1 - \mathbf{k}_3} a_{\mathbf{k}_1}^+ a_{\mathbf{k}_2}^+ a_{\mathbf{k}_3} a_{\mathbf{k}_4}. \end{aligned} \quad (2.2.28)$$

Notice that the anisotropic Hamiltonians (2.2.26)–(2.2.28) are functions of the  $z$  component of the spin operators so that they are the same in both HP and DM representations. This is no longer true for easy-plane anisotropic Hamiltonians ( $D, K < 0$ ) for which the lowering and raising spin operators enter the Hamiltonians (2.2.27) and (2.2.28) since in this case, the magnetization lies in the  $xy$  easy-plane.

As a summary, we stress that the DM transformation minimizes both the kinematical and the dynamical interaction in the limit of long wavelengths. The rigorous proof, however, is restricted to isotropic ferromagnets with NN exchange interaction even if it is reasonably believed that a similar conclusion can hold for other magnetic systems like antiferromagnets, ferrimagnets, helimagnets and anisotropic magnetic systems. In many actual calculations, both the kinematical and dynamical interactions are neglected (harmonic approximation) since only the low temperature region ( $T \ll T_c$ ) is considered.

### 2.3. Harmonic Approximation

The harmonic approximation consists of keeping only the bilinear boson Hamiltonian: as one can see from Eqs. (2.2.16) and (2.2.23), the harmonic approximation is independent of the HP or DM transformation: in the harmonic approximation, the magnetic system is reduced to an ideal gas of non-interacting magnons whose Hamiltonian reads

$$\mathcal{H}_0 = \mathcal{H}_2^{HP} = \mathcal{H}_2^{DM} = \sum_{\vec{k}} \hbar \omega_{\vec{k}} a_{\vec{k}}^+ a_{\vec{k}} \quad (2.3.1)$$

where

$$\hbar\omega_{\mathbf{k}} = 2zJS(1 - \gamma_{\mathbf{k}}) = 2JS \sum_{\boldsymbol{\delta}} (1 - \cos \mathbf{k} \cdot \boldsymbol{\delta}). \quad (2.3.2)$$

Expanding Eq. (2.3.2) in powers of the wavevector  $\mathbf{k}$ , one sees that the energy spectrum (2.3.2) vanishes quadratically for  $k \rightarrow 0$ . For a SC lattice ( $z = 6$ ) for which  $\boldsymbol{\delta} = (\pm a, 0, 0)$ ,  $(0, \pm a, 0)$  and  $(0, 0, \pm a)$ , where  $a$  is the side of the cube, the energy spectrum Eq. (2.3.2) reads

$$\hbar\omega_{\mathbf{k}} = 12JS \left[ 1 - \frac{1}{3}(\cos k_x a + \cos k_y a + \cos k_z a) \right]. \quad (2.3.3)$$

For a body centered cubic (BCC) lattice ( $z = 8$ ) for which  $\boldsymbol{\delta} = (\pm \frac{a}{2}, \pm \frac{a}{2}, \pm \frac{a}{2})$ , the energy spectrum reads

$$\hbar\omega_{\mathbf{k}} = 16JS \left( 1 - \cos \frac{k_x a}{2} \cos \frac{k_y a}{2} \cos \frac{k_z a}{2} \right). \quad (2.3.4)$$

For a face centered cubic (FCC) lattice ( $z = 12$ ) for which  $\boldsymbol{\delta} = (\pm \frac{a}{2}, \pm \frac{a}{2}, 0)$ ,  $(\pm \frac{a}{2}, 0, \pm \frac{a}{2})$  and  $(0, \pm \frac{a}{2}, \pm \frac{a}{2})$ , the energy spectrum reads

$$\hbar\omega_{\mathbf{k}} = 24JS \left[ 1 - \frac{1}{3} \left( \cos \frac{k_x a}{2} \cos \frac{k_y a}{2} + \cos \frac{k_x a}{2} \cos \frac{k_z a}{2} + \cos \frac{k_y a}{2} \cos \frac{k_z a}{2} \right) \right]. \quad (2.3.5)$$

In the limit of long wavelength, all cubic lattices yield energy spectra which are vanishing as  $\hbar\omega_{\mathbf{k}} \rightarrow 2JS(ak)^2$ . The energy of the uniform mode ( $k = 0$ ) vanishes in agreement to the equivalent non-relativistic Goldstone theorem<sup>19</sup> that works for Hamiltonians with short-range interactions, continuous symmetry (the 3D rotation group in the present case) and a ground state with a broken symmetry (the ferromagnetic ground state). It is worthwhile noticing that the energy spectrum (2.3.2) is an *exact* eigenvalue of the ferromagnetic Heisenberg Hamiltonian (2.2.1) belonging to the eigenstate with a delocalized spin deviation with momentum  $\mathbf{k}$  as one can see by comparing the spectrum (2.3.2) with the spectrum given by Eq. (1.3.21) obtained from an exact calculation on the Heisenberg Hamiltonian.

In presence of an external magnetic field, of a single-ion easy-axis anisotropy and of an exchange anisotropy, the energy spectrum becomes

$$\hbar\omega_{\mathbf{k}} = 2zJS(1 - \gamma_{\mathbf{k}}) + h + D(2S - 1) + 2zKS \quad (2.3.6)$$

as one can obtain from Eqs. (2.2.26)–(2.2.28). The main effect of these types of easy-axis anisotropy consists of moving the magnon spectrum upwards rigidly with the appearance of a gap at  $\mathbf{k} = 0$ . We stress that the harmonic approximation, which neglects any interaction between magnons, is expected to be a good representation of a ferromagnet at low temperature where the magnons can be treated as an ideal Bose gas. Obviously, since the number of magnons is not conserved (as well as in the case of a “phonon gas”) but determined by the temperature of the thermal bath, the chemical potential of the magnons is zero and any Bose-Einstein condensation is prevented.



## 2.4. Low Temperature Thermodynamic Functions

The low temperature thermodynamic functions of a ferromagnet may be obtained from the harmonic Hamiltonian (2.3.1). The granpartition function<sup>20</sup> reads

$$\begin{aligned} \mathcal{Q} &= e^{-\beta E_0} \text{Tr}(e^{-\beta \mathcal{H}_0}) = e^{-\beta E_0} \sum_{\{n_{\mathbf{k}}\}} e^{-\beta \sum_{\mathbf{k}} \hbar \omega_{\mathbf{k}} n_{\mathbf{k}}} \\ &= e^{-\beta E_0} \prod_{\mathbf{k}} \sum_{n_{\mathbf{k}}=0}^{\infty} e^{-\beta \hbar \omega_{\mathbf{k}} n_{\mathbf{k}}} = e^{-\beta E_0} \prod_{\mathbf{k}} \frac{1}{1 - e^{-\beta \hbar \omega_{\mathbf{k}}}} \end{aligned} \quad (2.4.1)$$

where  $\beta = \frac{1}{k_B T}$ ,  $E_0 = -zJS^2N$  is the ground-state energy and  $\hbar \omega_{\mathbf{k}}$  is the energy spectrum of spin waves given by Eq. (2.3.2) in the isotropic case and by Eq. (2.3.6) in the anisotropic case. The trace (Tr) of the density matrix is performed over the states number  $\{|n_{\mathbf{k}}\rangle\}$  that are eigenstates of  $\mathcal{H}_0$  with eigenvalues  $\sum_{\mathbf{k}} \hbar \omega_{\mathbf{k}} n_{\mathbf{k}}$ . The free energy is directly connected to the grancanonical partition function

$$\begin{aligned} F &= -k_B T \ln \mathcal{Q} = E_0 + k_B T \sum_{\mathbf{k}} \ln(1 - e^{-\beta \hbar \omega_{\mathbf{k}}}) \\ &= E_0 + k_B T N \frac{v_c}{(2\pi)^3} \int_{\text{BZ}} d^3 \mathbf{k} \ln(1 - e^{-\beta \hbar \omega_{\mathbf{k}}}) \\ &= E_0 - k_B T N \sum_{n=1}^{\infty} \frac{1}{n} \frac{v_c}{(2\pi)^3} \int_{\text{BZ}} d^3 \mathbf{k} e^{-n\beta \hbar \omega_{\mathbf{k}}} \end{aligned} \quad (2.4.2)$$

where  $v_c$  is the volume of the unit cell (for instance,  $v_c = \frac{a^3}{m}$  with  $m = 1, 2, 4$  for SC, BCC, FCC lattices, respectively) and BZ indicates the first Brillouin zone. In Eq. (2.4.2), the series expansion of the logarithmic function  $\ln(1 - x) = -\sum_{n=1}^{\infty} \frac{x^n}{n}$  has been used. At low temperature, only magnons of low energy can be excited so that the magnon energy spectrum (2.3.2) can be replaced by its expansion for long wavelengths

$$\hbar \omega_{\mathbf{k}} = 2JS \sum_{\delta} (\mathbf{k} \cdot \boldsymbol{\delta})^2$$

that reduces to  $\hbar \omega_{\mathbf{k}} = 2JS(ak)^2$  for all cubic lattices. Then the free energy (2.4.2) becomes

$$\begin{aligned} F &\simeq E_0 - k_B T N \sum_{n=1}^{\infty} \frac{1}{n} \frac{1}{8\pi^3 m} \int_{\text{BZ}} d^3 \mathbf{q} e^{-2n\beta JS q^2} \\ &\simeq E_0 - k_B T N \frac{1}{8\pi^3 m} \sum_{n=1}^{\infty} \frac{1}{n} \int_0^{\infty} q^2 dq \int_0^{\pi} \sin \theta d\theta \int_0^{2\pi} d\phi e^{-2n\beta JS q^2} \\ &= E_0 - k_B T N \frac{1}{2\pi^2 m} \sum_{n=1}^{\infty} \frac{1}{n} \int_0^{\infty} q^2 dq e^{-2n\beta JS q^2} \end{aligned} \quad (2.4.3)$$

where  $\mathbf{q} = a\mathbf{k}$ . Due to the parabolic  $q$ -dependence of the magnon spectrum in Eq. (2.4.3), we have assumed spherical coordinates and replaced the original BZ with a sphere whose radius is sent to infinity. As we will see later, the higher powers in  $q$  of the magnon spectrum and the actual lattice structure give rise to higher powers in the free energy temperature expansion. Making use of the relationship<sup>3</sup>

$$\int_0^\infty dx x^2 e^{-px^2} = \frac{1}{4p} \sqrt{\frac{\pi}{p}},$$

the free energy becomes

$$F = E_0 - k_B T N \zeta\left(\frac{5}{2}\right) \frac{1}{m} \left(\frac{k_B T}{8\pi JS}\right)^{\frac{3}{2}} \quad (2.4.4)$$

where  $\zeta(\frac{5}{2}) = 1.34149$  is the Riemann function<sup>4</sup>  $\zeta(p) = \sum_{n=1}^\infty n^{-p}$ . As one can see, the first term in the temperature expansion of the free energy is proportional to  $T^{\frac{5}{2}}$ . In the presence of uniaxial anisotropy, the uniform mode has a gap  $\hbar\omega_o = h + D(2S-1) + 2zKS$  and the free energy is obtained from Eq. (2.4.4) after replacing  $\zeta(\frac{5}{2})$  by the function  $Z_{\frac{5}{2}}(\beta\hbar\omega_o)$  where  $Z_p(x) = \sum_{n=1}^\infty n^{-p} e^{-nx}$ . In this case, the free energy shows an exponential dependence on the temperature instead of a power law. Indeed, at low temperature ( $k_B T \ll \hbar\omega_o$ ), the function  $Z_{\frac{5}{2}}(\beta\hbar\omega_o)$  may be approximated by the first term of the series so that  $Z_{\frac{5}{2}}(\beta\hbar\omega_o) \sim e^{-\beta\hbar\omega_o}$ . From the free energy, all the other thermodynamic functions can be obtained. In particular, the entropy is given by

$$S = -\frac{\partial F}{\partial T} = 5k_B N \zeta\left(\frac{5}{2}\right) \frac{1}{2m} \left(\frac{k_B T}{8\pi JS}\right)^{\frac{3}{2}}, \quad (2.4.5)$$

the internal energy is then obtained from the relationship  $U = F + TS$  or directly from the free energy derivative as

$$U = -T^2 \frac{\partial}{\partial T} \left(\frac{F}{T}\right) = E_0 + 12\pi JS N \zeta\left(\frac{5}{2}\right) \frac{1}{m} \left(\frac{k_B T}{8\pi JS}\right)^{\frac{5}{2}}. \quad (2.4.6)$$

The same result could have been obtained from the equation  $U = \sum_{\mathbf{k}} \hbar\omega_{\mathbf{k}} \langle a_{\mathbf{k}}^+ a_{\mathbf{k}} \rangle$  where the average number of magnons is evaluated as follows

$$\begin{aligned} \langle a_{\mathbf{k}}^+ a_{\mathbf{k}} \rangle &= \frac{\text{Tr}(a_{\mathbf{k}}^+ a_{\mathbf{k}} e^{-\beta \sum_{\mathbf{q}} \hbar\omega_{\mathbf{q}} a_{\mathbf{q}}^+ a_{\mathbf{q}}})}{\text{Tr}(e^{-\beta \sum_{\mathbf{q}} \hbar\omega_{\mathbf{q}} a_{\mathbf{q}}^+ a_{\mathbf{q}}})} = \frac{\sum_{n_{\mathbf{k}}=0}^\infty n_{\mathbf{k}} e^{-\beta \hbar\omega_{\mathbf{k}} n_{\mathbf{k}}}}{\sum_{n_{\mathbf{k}}=0}^\infty e^{-\beta \hbar\omega_{\mathbf{k}} n_{\mathbf{k}}}} \\ &= -\frac{1}{\beta \hbar \omega_{\mathbf{k}}} \frac{\partial}{\partial \omega_{\mathbf{k}}} \ln \left( \sum_{n_{\mathbf{k}}=0}^\infty e^{-\beta \hbar\omega_{\mathbf{k}} n_{\mathbf{k}}} \right) \\ &= \frac{1}{\beta \hbar \omega_{\mathbf{k}}} \frac{\partial}{\partial \omega_{\mathbf{k}}} \ln (1 - e^{-\beta \hbar\omega_{\mathbf{k}}}) = \frac{1}{e^{\beta \hbar\omega_{\mathbf{k}}} - 1} \end{aligned} \quad (2.4.7)$$

leading to the average occupation number of the Bose-Einstein statistics. The heat capacity is given by

$$C = \frac{\partial U}{\partial T} = 15k_B N \zeta\left(\frac{5}{2}\right) \frac{1}{4m} \left(\frac{k_B T}{8\pi JS}\right)^{\frac{3}{2}}. \quad (2.4.8)$$

For an isotropic insulating ferromagnet, the heat capacity at low temperature is expected to behave like  $C = A_{\text{mag}} T^{\frac{3}{2}} + B_{\text{vib}} T^3$  where the first term is the magnetic contribution and the second one is the vibrational contribution. A convenient way to separate the magnetic from the vibrational contribution is to draw the quantity  $CT^{-\frac{3}{2}}$  as function of  $T^{\frac{3}{2}}$ . In this way, one obtains a straight line whose slope  $B_{\text{vib}}$  gives the coefficient of the vibrational contribution while the intercept at  $T = 0$  gives the coefficient  $A_{\text{mag}}$  of the magnetic contribution.

The magnetization is given by

$$M = -\frac{\partial F}{\partial H} = g\mu_B \left\langle \sum_i S_i^z \right\rangle = g\mu_B \left( SN - \sum_{\mathbf{k}} \langle a_{\mathbf{k}}^+ a_{\mathbf{k}} \rangle \right) \quad (2.4.9)$$

or using Eq. (2.4.7),

$$\begin{aligned} M &= M_0 \left[ 1 - \frac{1}{mS} \frac{v_c}{(2\pi)^3} \int_{\text{BZ}} d^3\mathbf{k} \frac{1}{e^{\beta\hbar\omega_{\mathbf{k}}} - 1} \right] \\ &= M_0 \left[ 1 - \frac{1}{mS} \sum_{n=1}^{\infty} \frac{v_c}{(2\pi)^3} \int_{\text{BZ}} d^3\mathbf{k} e^{-n\beta\hbar\omega_{\mathbf{k}}} \right] \end{aligned} \quad (2.4.10)$$

where  $M_0 = g\mu_B SN$  is the saturation magnetization at  $T = 0$ . In the low temperature limit, that is for  $\hbar\omega_{\mathbf{k}} \simeq h + 2JS(ak)^2$ , one obtains

$$\begin{aligned} M &\simeq M_0 \left[ 1 - \frac{1}{mS} \sum_{n=1}^{\infty} e^{-n\frac{h}{k_B T}} \frac{1}{2\pi^2} \int_0^{\infty} q^2 dq e^{-2n\beta JS q^2} \right] \\ &\times M_0 \left[ 1 - \frac{1}{mS} \left( \frac{k_B T}{8\pi JS} \right)^{\frac{3}{2}} Z_{\frac{3}{2}} \left( \frac{h}{k_B T} \right) \right] \end{aligned} \quad (2.4.11)$$

For  $h = 0$ , since  $Z_{\frac{3}{2}}(0) = \zeta(\frac{3}{2}) = 2.61238$ , the spontaneous magnetization  $M_s = M(T, H = 0)$  deviates from its saturation value following the  $T^{\frac{3}{2}}$ -law found long time ago by Bloch.<sup>21</sup> The spontaneous magnetization is the order parameter of the ferromagnetic phase since it is non-zero in the ordered phase and vanishes in the (disordered) paramagnetic phase.

The susceptibility is given by

$$\chi = \frac{\partial M}{\partial H} \simeq \frac{(g\mu_B)^2 N}{m8\pi JS} \sqrt{\frac{k_B T}{8\pi JS}} Z_{\frac{1}{2}} \left( \frac{h}{k_B T} \right). \quad (2.4.12)$$

Equation (2.4.12) is obtained by taking advantage of the equality  $Z'_{\frac{3}{2}}(x) = -Z_{\frac{1}{2}}(x)$ .

For weak magnetic field ( $h \ll k_B T$ ), one obtains

$$\chi = \sqrt{\pi} \frac{(g\mu_B)^2 N}{m(8\pi JS)^{\frac{3}{2}}} \frac{k_B T}{\sqrt{h}}$$

since  $Z_{\frac{1}{2}}(x) \simeq \sqrt{\frac{\pi}{x}}$ , for  $x \rightarrow 0$ . For  $h \rightarrow 0$ , the susceptibility diverges at any finite temperature. This divergence is not surprising because in the ordered phase of the isotropic ferromagnet, any infinitesimal magnetic field is sufficient to direct the macroscopic magnetization along the field. In other words, the system reacts to the magnetic field with an infinite response. In a ferromagnet with an easy-axis anisotropy ( $D, K > 0$ ), the divergence in the susceptibility disappears. Indeed, for small anisotropy ( $\hbar\omega_o \ll k_B T$ ), the susceptibility becomes

$$\chi \simeq \sqrt{\pi} \frac{(g\mu_B)^2 N}{m(8\pi JS)^{\frac{3}{2}}} \frac{k_B T}{(\hbar\omega_o)^{\frac{1}{2}}} \quad (2.4.13)$$

while for low temperature but large anisotropy ( $\hbar\omega_o \gg k_B T$ ), the susceptibility shows an exponential behavior

$$\chi \simeq \frac{(g\mu_B)^2 N}{m(8\pi JS)} \left( \frac{k_B T}{8\pi JS} \right)^{\frac{1}{2}} e^{-\beta\hbar\omega_o}. \quad (2.4.14)$$

Before concluding this section, we evaluate some of the higher order corrections in the temperature expansion of the thermodynamic functions entered by the actual  $\mathbf{k}$ -dependence of the magnon spectrum (2.3.2). To do this, we consider the SC magnon spectrum (2.3.3) and we replace it in the free energy (2.4.2) obtaining

$$\begin{aligned} F &= E_0 - k_B T N \sum_{n=1}^{\infty} \frac{e^{-12n\beta JS}}{n} \left( \frac{1}{2\pi} \int_{-\pi}^{\pi} dq_x e^{4n\beta JS \cos q_x} \right)^3 \\ &= E_0 - k_B T N \sum_{n=1}^{\infty} \frac{e^{-12n\beta JS}}{n} [I_0(4n\beta JS)]^3 \end{aligned} \quad (2.4.15)$$

where  $E_0 = -6JS^2 N$  and  $I_0(z) = \frac{1}{2\pi} \int_{-\pi}^{\pi} d\theta e^{z \cos \theta}$  is the modified Bessel function of the first kind and of order zero.<sup>4</sup> In the low temperature limit ( $4\beta JS \gg 1$ ), we use the asymptotic expansion of  $I_0(z)$

$$I_0(z) = \frac{e^z}{\sqrt{2\pi z}} \left( 1 + \frac{1}{8z} + \frac{9}{128z^2} + \dots \right) \quad (2.4.16)$$

leading to the free energy expansion

$$\begin{aligned} F &= E_0 - k_B T N \left[ \zeta\left(\frac{5}{2}\right) \left( \frac{k_B T}{8\pi JS} \right)^{\frac{3}{2}} + \frac{3\pi}{4} \zeta\left(\frac{7}{2}\right) \left( \frac{k_B T}{8\pi JS} \right)^{\frac{5}{2}} \right. \\ &\quad \left. + \frac{33\pi^2}{32} \zeta\left(\frac{9}{2}\right) \left( \frac{k_B T}{8\pi JS} \right)^{\frac{7}{2}} \right] \end{aligned} \quad (2.4.17)$$

where<sup>4</sup>  $\zeta(\frac{5}{2}) = 1.34149$ ,  $\zeta(\frac{7}{2}) = 1.12673$ ,  $\zeta(\frac{9}{2}) = 1.05471$ . This result could also be obtained by expanding the magnon spectrum up to  $k^6$ . For the SC lattice, one obtains

$$\hbar\omega_{\mathbf{k}} = 2JS(ak)^2 - \frac{1}{6}JSa^4(k_x^4 + k_y^4 + k_z^4) + \frac{1}{180}JSa^6(k_x^6 + k_y^6 + k_z^6). \quad (2.4.18)$$

Performing an integration over a sphere instead of over the actual cubic cell and assuming the radius of the sphere goes to infinity, the half-integer powers of the series are recovered. However, the coefficients of the series are not the same. Indeed, the first two coefficients are unchanged whereas the third one gives  $-\frac{\pi^2}{4}$  instead of the correct value  $\frac{33\pi^2}{32}$ . This means that the SC lattice BZ may be treated as a sphere up to the order  $k^4$ . The cubic structure of the BZ becomes important only at the order  $k^6$ . Even worse is the result for the BCC and FCC lattices where the actual structure affects the expansion coefficients at the order  $T^{\frac{7}{2}}$  coming from the  $k^4$ -term of the spectrum. In any case, the coefficient of the main term of the expansion is correctly given by the parabolic approximation of the spectrum ( $k^2$ -term), ignoring any actual structure of the cell. Notice that the higher semi-integer powers of the temperature series expansion given in Eq. (2.4.17) are introduced as a result of the actual magnon spectrum, not by the magnon-magnon interaction which is neglected in the harmonic approximation. The series expansion for the internal energy and for the heat capacity of a SC lattice read

$$U = E_0 + k_B T N \left[ \frac{3}{2} \zeta \left( \frac{5}{2} \right) \left( \frac{k_B T}{8\pi JS} \right)^{\frac{3}{2}} + \frac{15\pi}{8} \zeta \left( \frac{7}{2} \right) \left( \frac{k_B T}{8\pi JS} \right)^{\frac{5}{2}} + \frac{231\pi^2}{64} \zeta \left( \frac{9}{2} \right) \left( \frac{k_B T}{8\pi JS} \right)^{\frac{7}{2}} \right] \quad (2.4.19)$$

and

$$\begin{aligned} C &= k_B N \left[ \frac{15}{4} \zeta \left( \frac{5}{2} \right) \left( \frac{k_B T}{8\pi JS} \right)^{\frac{3}{2}} + \frac{105\pi}{16} \zeta \left( \frac{7}{2} \right) \left( \frac{k_B T}{8\pi JS} \right)^{\frac{5}{2}} + \frac{2079\pi^2}{128} \zeta \left( \frac{9}{2} \right) \left( \frac{k_B T}{8\pi JS} \right)^{\frac{7}{2}} \right] \\ &= \frac{15}{4} k_B N \left[ 1.34149 \left( \frac{k_B T}{8\pi JS} \right)^{\frac{3}{2}} + 6.19452 \left( \frac{k_B T}{8\pi JS} \right)^{\frac{5}{2}} + 45.0865 \left( \frac{k_B T}{8\pi JS} \right)^{\frac{7}{2}} \right], \end{aligned} \quad (2.4.20)$$

respectively. The series expansion for the magnetization can be obtained from Eq. (2.4.10), giving

$$\begin{aligned} M &= M_0 \left[ 1 - \frac{1}{S} \sum_{n=1}^{\infty} e^{-12n\beta JS} \left( \frac{1}{2\pi} \int_{-\pi}^{\pi} dq_x e^{4n\beta JS \cos q_x} \right)^3 \right] \\ &= M_0 \left\{ 1 - \frac{1}{S} \sum_{n=1}^{\infty} e^{-12n\beta JS} [I_0(4n\beta JS)]^3 \right\}. \end{aligned} \quad (2.4.21)$$

At low temperature ( $k_B T \ll 4JS$ ), the asymptotic expansion (2.4.16) can be used to obtain

$$\begin{aligned}
 M &= M_0 \left\{ 1 - \frac{1}{S} \left[ \zeta \left( \frac{3}{2} \right) \left( \frac{k_B T}{8\pi JS} \right)^{\frac{3}{2}} + \frac{3\pi}{4} \zeta \left( \frac{5}{2} \right) \left( \frac{k_B T}{8\pi JS} \right)^{\frac{5}{2}} \right. \right. \\
 &\quad \left. \left. + \frac{33\pi^2}{32} \zeta \left( \frac{7}{2} \right) \left( \frac{k_B T}{8\pi JS} \right)^{\frac{7}{2}} \right] \right\} \\
 &= M_0 \left\{ 1 - \frac{1}{S} \left[ 2.61238 \left( \frac{k_B T}{8\pi JS} \right)^{\frac{3}{2}} + 3.16081 \left( \frac{k_B T}{8\pi JS} \right)^{\frac{5}{2}} \right. \right. \\
 &\quad \left. \left. + 11.4679 \left( \frac{k_B T}{8\pi JS} \right)^{\frac{7}{2}} \right] \right\}. \tag{2.4.22}
 \end{aligned}$$

In Eq. (2.4.22), the corrections to the  $T^{\frac{3}{2}}$ -law obtained by Bloch<sup>21</sup> are due to the actual dispersion relation of the magnon spectrum. The first correction entered by the magnon-magnon interaction is proportional to  $T^4$  as it will be shown in Section 3.6.

## 2.5. Application to Quasi-2D and Quasi 1D-models

In order to understand how the thermodynamic functions are modified in low dimensional magnetic systems, we evaluate the thermodynamic quantities for a tetragonal (T) lattice where the  $c$ -edge of the unit cell may be chosen as  $c \gg a$  or  $c \ll a$ . For a T ferromagnet, the exchange interaction between the 4 NN spins in the basal  $ab$  plane is assumed to be  $J > 0$  while the exchange interaction between the 2 NN spins along the  $c$  axis is assumed to be  $J' > 0$ . According to  $c \gg a$  or  $c \ll a$ , one takes  $J \gg J'$  or  $J \ll J'$ , respectively. In the former case, we have a staking of ferromagnetic planes weakly coupled to each other (quasi-2D ferromagnet), in the latter case, we have weakly coupled ferromagnetic chains (quasi-1D ferromagnet). For a T lattice, the spectrum (2.3.3) becomes

$$\hbar\omega_{\mathbf{k}} = 8JS \left[ 1 - \frac{1}{2}(\cos k_x a + \cos k_y a) \right] + 4J'S(1 - \cos k_z c). \tag{2.5.1}$$

Obviously, for  $c = a$  and  $J = J'$ , the T lattice reduces to the SC lattice and the results of the previous section are recovered. For a T lattice, the free energy can be obtained from Eq. (2.4.2), giving

$$F = E_0 - k_B T N \sum_{n=1}^{\infty} \frac{e^{-n(8J+4J')S\beta}}{n} [I_0(4n\beta JS)]^2 I_0(4n\beta J'S) \tag{2.5.2}$$

where  $E_0 = -(4J+2J')S^2 N$ . At very low temperature  $k_B T \ll 4JS, 4J'S$ , one recovers the 3D result. Indeed, in this temperature range ( $4\beta JS, 4\beta J'S \gg 1$ ), one can

use the asymptotic expansion (2.4.16), obtaining

$$F = E_0 - k_B T N \left[ \zeta \left( \frac{5}{2} \right) \left( \frac{J}{J'} \right)^{\frac{1}{2}} \left( \frac{k_B T}{8\pi J S} \right)^{\frac{3}{2}} \dots \right]. \quad (2.5.3)$$

As one can see, the free energy shows a  $T^{\frac{5}{2}}$ -dependence similar to that obtained for a SC lattice. The SC result is recovered for  $J = J'$  as expected. The internal energy and the heat capacity of the T lattice become

$$U = E_0 + k_B T N \left[ \frac{3}{2} \zeta \left( \frac{5}{2} \right) \left( \frac{J}{J'} \right)^{\frac{1}{2}} \left( \frac{k_B T}{8\pi J S} \right)^{\frac{3}{2}} \dots \right] \quad (2.5.4)$$

and

$$C = k_B N \left[ \frac{15}{4} \zeta \left( \frac{5}{2} \right) \left( \frac{J}{J'} \right)^{\frac{1}{2}} \left( \frac{k_B T}{8\pi J S} \right)^{\frac{3}{2}} \dots \right], \quad (2.5.5)$$

respectively. The typical  $T^{\frac{3}{2}}$  contribution to the heat capacity of a 3D ferromagnet is recovered at very low temperature.

For quasi-2D ferromagnets ( $J' \ll J$ ) and for an intermediate temperature range  $4J'S \ll k_B T \ll 4JS$  ( $4\beta JS \gg 1 \gg 4\beta J'S$ ), the asymptotic expansion for  $I_0(4\beta JS)$  is appropriate but the series expansion<sup>4</sup>

$$I_0(z) = 1 + \frac{1}{4}z^2 + \frac{1}{64}z^4 \dots \quad (2.5.6)$$

has to be used for  $I_0(4\beta J'S)$ . Then the free energy (2.5.2) becomes

$$\begin{aligned} F &= E_0 - k_B T N \left[ Z_2(4\beta J'S) \left( \frac{k_B T}{8\pi J S} \right) \dots \right] \\ &\simeq E_0 - k_B T N \zeta(2) \left( \frac{k_B T}{8\pi J S} \right) \dots \end{aligned} \quad (2.5.7)$$

where<sup>4</sup>  $\zeta(2) = \frac{\pi^2}{6} = 1.64493$ . The internal energy and heat capacity read

$$U \simeq E_0 - k_B T N \zeta(2) \left( \frac{k_B T}{8\pi J S} \right) \dots \quad (2.5.8)$$

and

$$C \simeq 2k_B N \zeta(2) \left( \frac{k_B T}{8\pi J S} \right) \dots, \quad (2.5.9)$$

respectively. As one can see, the temperature dependence of the magnetic heat capacity of a quasi-2D ferromagnet is linear in  $T$  so that one should expect a crossover between the  $T^{3/2}$ -law at very low temperature and a linear  $T$ -law at intermediate temperatures. The window of the linear contribution of the heat capacity vs temperature is related to the ratio  $J'/J$ . For typical quasi-2D magnets ( $J'/J \sim 10^{-3} - 10^{-5}$  and  $J/k_B \sim 10 - 100$  K), the 3D-behaviour is restricted to temperatures  $T \lesssim 1$  K while the linear dependence may be extended from  $T \lesssim 1$  K to  $T \lesssim 100$  K. Obviously, in the experiment, the elastic contribution (phonons) proportional to  $T^{\frac{3}{2}}$  cannot be separated from the magnetic one. However, after drawing the

function  $T^{-1}C$  versus  $T^{\frac{1}{2}}$ , we may distinguish between the magnetic contribution (intercept at the origin) and the elastic contribution (slope of the straight line).

For quasi-1D ferromagnets ( $J' \gg J$ ) in the intermediate temperature range  $4JS \ll k_B T \ll 4J'S$  ( $4\beta JS \ll 1 \ll 4\beta J'S$ ), the asymptotic expansion (2.4.16) is used for  $I_0(4\beta J'S)$  and the series expansion (2.5.6) for  $I_0(4\beta JS)$  so that one obtains

$$\begin{aligned} F &= E_0 - k_B T N Z_{\frac{3}{2}}(8\beta JS) \left( \frac{k_B T}{8\pi J'S} \right)^{\frac{1}{2}} \cdots \\ &\simeq E_0 - k_B T N \zeta \left( \frac{3}{2} \right) \left( \frac{k_B T}{8\pi J'S} \right)^{\frac{1}{2}} \cdots \end{aligned} \quad (2.5.10)$$

The internal energy and heat capacity of a quasi-1D ferromagnet read

$$U \simeq E_0 + k_B T N \frac{1}{2} \zeta \left( \frac{3}{2} \right) \left( \frac{k_B T}{8\pi J'S} \right)^{\frac{1}{2}} \cdots \quad (2.5.11)$$

and

$$C \simeq \frac{3}{4} k_B N \zeta \left( \frac{3}{2} \right) \left( \frac{k_B T}{8\pi J'S} \right)^{\frac{1}{2}} \cdots, \quad (2.5.12)$$

respectively. A  $T^{\frac{1}{2}}$ -behaviour is expected for the heat capacity of a quasi-1D ferromagnet at intermediate temperatures.

Finally, let us consider the magnetization of the T ferromagnet. From Eq. (2.4.10), one has

$$M = M_0 \left\{ 1 - \frac{1}{S} \sum_{n=1}^{\infty} e^{-n(8J+4J')\beta S} [I_0(4n\beta JS)]^2 I_0(4n\beta J'S) \right\}. \quad (2.5.13)$$

For temperatures such that  $k_B T \ll 4JS$ ,  $4J'S$  ( $4\beta JS, 4\beta J'S \gg 1$ ), the 3D-behaviour, that is the  $T^{\frac{3}{2}}$ -law of Bloch, is recovered

$$M = M_0 \left[ 1 - \frac{1}{S} \zeta \left( \frac{3}{2} \right) \left( \frac{J}{J'} \right)^{\frac{1}{2}} \left( \frac{k_B T}{8\pi JS} \right)^{\frac{3}{2}} \cdots \right]. \quad (2.5.14)$$

For a quasi-2D ferromagnet ( $J' \ll J$ ) at intermediate temperatures ( $4J'S \ll k_B T \ll 4JS$ ), one obtains

$$M = M_0 \left[ 1 + \frac{k_B T}{8\pi JS^2} \ln(1 - e^{-\frac{4J'S}{k_B T}}) \right] \simeq M_0 \left( 1 - \frac{k_B T}{8\pi JS^2} \ln \frac{k_B T}{4J'S} \right). \quad (2.5.15)$$

where  $\sum_{n=1}^{\infty} n^{-1} e^{-xn} = -\ln(1 - e^{-x})$  has been used. In a quasi-2D ferromagnet, the magnetization is strongly depressed at intermediate temperature: this fact reflects the absence of long range order (LRO) at any finite temperature in a pure 2D Heisenberg ferromagnet ( $J' = 0$ ). The absence of LRO in 2D systems with continuous symmetry and short-range interaction is rigorously proven by the Mermin-Wagner theorem.<sup>22</sup>



For a quasi-1D ferromagnet ( $J' \gg J$ ) at intermediate temperatures ( $4JS \ll k_B T \ll 4J'S$ ), one obtains

$$M = M_0 \left[ 1 - \frac{1}{S} Z_{\frac{1}{2}}(8\beta JS) \left( \frac{k_B T}{8\pi J'S} \right) \right] \simeq M_0 \left[ 1 - \frac{\pi}{S} \left( \frac{J'}{J} \right)^{\frac{1}{2}} \left( \frac{k_B T}{8\pi J'S} \right)^{\frac{3}{2}} \right] \quad (2.5.16)$$

since  $Z_{\frac{1}{2}}(x) = \sum_{n=1}^{\infty} n^{-\frac{1}{2}} e^{-xn} \simeq \sqrt{\frac{\pi}{x}} + O(1)$  for  $x \ll 1$ . In the quasi-1D ferromagnet, the deviation from the saturation value of the magnetization may be very strong since for many actual compounds, one has  $(J'/J)^{\frac{1}{2}} \sim 10^2$ .