

## CHAPTER 3

### GREEN'S FUNCTIONS IN FERRIMAGNETISM

The double-time temperature dependent Green's function method has been applied to an isotropic two-sublattice ferrimagnet by Yablonskii<sup>40</sup> to obtain the ground and low temperature elementary excitations of the system in the spin wave approximation. The present work will be devoted to a system of normal spinel ferrimagnet using the Green's function method with the decoupling scheme different from that used in the Yablonskii's paper. The Green's functions pertaining spin waves for such a system will be developed in details in this chapter.

#### 3.1 The Normal Spinel Ferrites

The term ferrimagnetic was coined originally to describe the phenomenon associated with ferrites which are a kind of magnetic oxides having the usual chemical formula such as  $MO.Fe_2O_3$  where Fe yields a trivalent ion  $Fe^{3+}$  and M is a divalent ion, often Zn, Cd, Fe, Ni, Cu, Co, or Mg. Ferrites have the spinel crystal structure in which there are two types of sites of magnetic ions: one is tetrahedrally surrounded by four oxygen ions, and is called a tetrahedral site, or A site, whereas the other is surrounded octahedrally by six oxygen ions, and

is called an octahedral, or B, site. A unit cell of the spinel lattice has eight tetrahedral sites and sixteen octahedral sites. The eight tetrahedral sites form the A sublattice while the sixteen octahedral ones form the B sublattice. Since the following A sites are shared by four unit cells each,

$$\left(\frac{1}{2}, 0, \frac{1}{2}\right), \left(-\frac{1}{2}, 0, \frac{1}{2}\right), \left(0, \frac{1}{2}, \frac{1}{2}\right), \left(0, -\frac{1}{2}, \frac{1}{2}\right), \left(\frac{1}{2}, 0, -\frac{1}{2}\right), \left(-\frac{1}{2}, 0, -\frac{1}{2}\right), \\ \left(0, \frac{1}{2}, -\frac{1}{2}\right), \left(0, -\frac{1}{2}, -\frac{1}{2}\right), \left(\frac{1}{2}, -\frac{1}{2}, 0\right), \left(\frac{1}{2}, \frac{1}{2}, 0\right), \left(-\frac{1}{2}, \frac{1}{2}, 0\right) \text{ and } \left(-\frac{1}{2}, -\frac{1}{2}, 0\right),$$

they contribute altogether three possible occupation sites to the unit cell. The other five sites are at

$$(0, 0, 0), \left(\frac{1}{4}, \frac{1}{4}, \frac{1}{4}\right), \left(-\frac{1}{4}, \frac{1}{4}, \frac{1}{4}\right), \left(\frac{1}{4}, \frac{1}{4}, -\frac{1}{4}\right) \text{ and } \left(-\frac{1}{4}, -\frac{1}{4}, -\frac{1}{4}\right).$$

The sixteen octahedral sites of the B sublattice are located at

$$\left(\frac{3}{8}, \frac{1}{8}, \frac{1}{8}\right), \left(\frac{1}{8}, \frac{3}{8}, \frac{1}{8}\right), \left(\frac{1}{8}, \frac{1}{8}, \frac{3}{8}\right), \left(\frac{3}{8}, \frac{3}{8}, \frac{3}{8}\right), \\ \left(-\frac{3}{8}, -\frac{1}{8}, \frac{1}{8}\right), \left(-\frac{1}{8}, -\frac{3}{8}, \frac{1}{8}\right), \left(-\frac{1}{8}, -\frac{1}{8}, \frac{3}{8}\right), \left(-\frac{3}{8}, -\frac{3}{8}, \frac{3}{8}\right), \\ \left(-\frac{1}{8}, \frac{1}{8}, -\frac{3}{8}\right), \left(-\frac{1}{8}, \frac{3}{8}, -\frac{1}{8}\right), \left(-\frac{3}{8}, \frac{1}{8}, -\frac{1}{8}\right), \left(-\frac{3}{8}, \frac{3}{8}, -\frac{3}{8}\right), \\ \left(\frac{1}{8}, -\frac{1}{8}, -\frac{3}{8}\right), \left(\frac{1}{8}, -\frac{3}{8}, -\frac{1}{8}\right), \left(\frac{3}{8}, -\frac{1}{8}, -\frac{1}{8}\right) \text{ and } \left(\frac{3}{8}, -\frac{3}{8}, -\frac{3}{8}\right).$$

The diagram of tetrahedral and octahedral sites in the spinel structure is shown in Fig. 5.

The spinel structure is classified into two categories, normal spinel and inverse spinel, according to the distribution

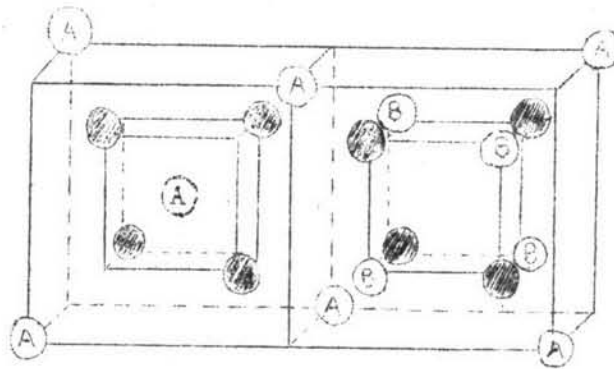


Fig. 5 A spinel structure. The small circles with A represent tetrahedral sites, the circles with B are for octahedral and the black circles indicate oxygen ions. Only two octants of a unit cell is shown. The other octants have either of these two structures and are arranged so that no two adjacent octants have the same configuration.

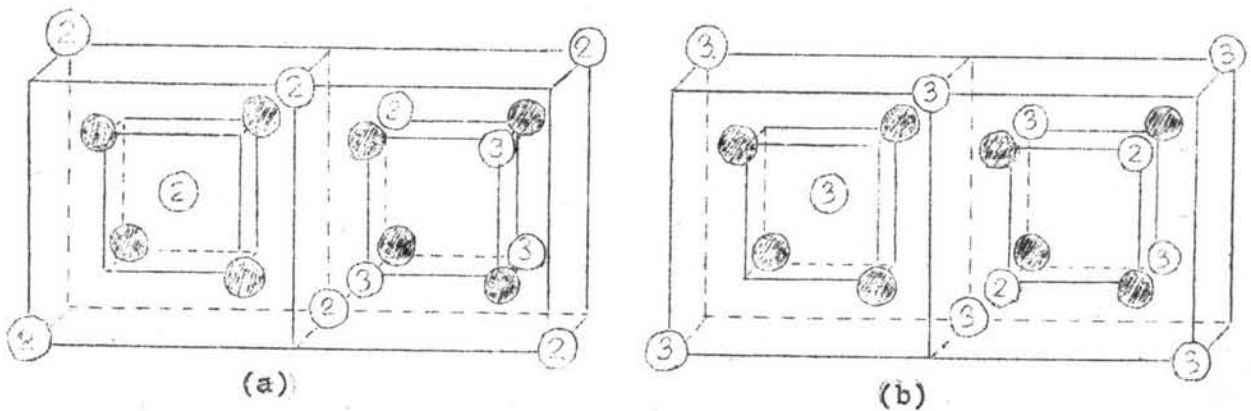


Fig. 6 Spinel structures. (a) Normal spinel structure and (b) inverse spinel structure. A small circle with the number 2 indicates a divalent ion and the one with the number 3 indicates a trivalent ion while a solid dot stands for an oxygen ion.

of divalent and trivalent magnetic ions on the two sites. Fig. 6 is a schematic illustration of these two types.

In a normal spinel the divalent metal ions occupy the tetrahedral sites and the trivalent ones occupy the octahedral sites;  $MgAl_2O_4$  is a typical example. On the other hand, if all of the tetrahedral sites are occupied by trivalent metal ions, while the octahedral sites are occupied half by divalent and half by trivalent ions, generally distributed at random, the structure is said to be an inverse spinel. Almost all of the simple spinels that are ferrimagnetic have the inverse arrangement, an example is magnetite,  $Fe_3O_4$ .

However, the present work will be confined to the simpler case of normal spinel, in which a unit cell is composed of eight divalent ions and sixteen trivalent ions occupy the A and B sublattices, respectively, at the various positions previously cited. The spins of the ions in the two sublattices are aligned so as to give a ferrimagnetic order between different sublattices.

### 3.2 The Heisenberg Hamiltonian of the System

The system under consideration is a two sublattice Heisenberg spin system of a ferrimagnetic normal spinel structure, having strong isotropic antiferromagnetic exchange interactions between spins situated on different sublattices in addition to weak isotropic (ferromagnetic or antiferromagnetic)

exchange interactions between spins on the same sublattice, and having uniaxial crystal field single ion type anisotropy. In the application of a uniform static external magnetic field  $\underline{H}$  along the anisotropic axis (taken as the z axis), the Hamiltonian of the system is given by

$$\begin{aligned} \mathcal{H} = & -\mu_B H \left[ g_A \sum_i \Lambda_i^z + g_B \sum_j B_j^z \right] - \sum_{i,j} J_{ij} \underline{\Lambda}_i \cdot \underline{B}_j - \sum_{i,i'} J'_{ii'} \underline{\Lambda}_i \cdot \underline{\Lambda}_{i'} \\ & - \sum_{j,j'} J''_{jj'} \underline{B}_j \cdot \underline{B}_{j'} - D \left[ \sum_i (\Lambda_i^z)^2 + \sum_j (B_j^z)^2 \right], \end{aligned} \quad (3.1)$$

where  $\Lambda$  and  $B$  are localized spin angular momentum operators on the A and B sublattices, and  $i, i'$  and  $j, j'$  are sites on the A and B sublattices, respectively. The constants  $g_A, g_B, \mu_B$  and  $D$  are the splitting factors of the A, B spins, Bohr magneton and anisotropy constant, respectively.  $J_{ij}$  is the exchange integral for the intersublattice Heisenberg interaction while  $J'_{ii'}$  and  $J''_{jj'}$  are, respectively, the exchange integrals for the intrasublattice Heisenberg interactions of the A and B sublattices. The exchange integrals are assumed to obey the following relations:

$$\begin{aligned} J_{ij} = J_{ji} < 0, \quad J'_{ii'} \neq J''_{jj'}, \quad J'_{ii} = 0 = J''_{jj} \\ \text{and } \left| \sum_j J_{ij} \right| \gg \left| \sum_j J''_{jj'} \right| \text{ or } \left| \sum_i J'_{ii'} \right|. \end{aligned} \quad (3.2)$$

It is easily obtained that

$$\underline{A} \cdot \underline{B} = \frac{1}{2} (\underline{A}^+ \underline{B}^- + \underline{A}^- \underline{B}^+) + \underline{A}^Z \underline{B}^Z, \quad (3.3)$$

where, as usual  $\underline{A}^\pm = \underline{A}^X \pm i \underline{A}^Y$  and  $\underline{B}^\pm = \underline{B}^X \pm i \underline{B}^Y$ .

Using the commutation relations for spin operators such as

$\underline{A}_1^X, \underline{A}_1^Y = i \underline{A}_1^Z \delta_{11}$ , and cyclic permutations, we can get from eq. (3.3) the relation:

$$\underline{A}_1^+ \underline{A}_1^- = \underline{A}_1^- \underline{A}_1^+. \quad (3.4)$$

Making use of eqs. (3.3) and (3.4), the original Hamiltonian (3.1) becomes

$$\begin{aligned} \mathcal{H} &= -\mu_B H \left[ \epsilon_A \sum_i \underline{A}_i^Z + \epsilon_B \sum_j \underline{B}_j^Z \right] - \sum_{i,j} J_{ij} \left[ \frac{1}{2} (\underline{A}_i^+ \underline{B}_j^- + \underline{A}_i^- \underline{B}_j^+) + \underline{A}_i^Z \underline{B}_j^Z \right] \\ &\quad - \sum_{i,i'} J_{ii'}^I (\underline{A}_i^- \underline{A}_{i'}^+ + \underline{A}_i^Z \underline{A}_{i'}^Z) - \sum_{j,j'} J_{jj'}^II (\underline{B}_j^+ \underline{B}_{j'}^- + \underline{B}_j^Z \underline{B}_{j'}^Z) \\ &\quad - D \left[ \sum_i (\underline{A}_i^Z)^2 + \sum_j (\underline{B}_j^Z)^2 \right]. \end{aligned} \quad (3.5)$$

### 3.2.1 The Fourier transform of the exchange integrals

The Fourier transform of the exchange integrals may be defined as follows

$$J(\underline{k}) = \frac{1}{\sqrt{N_A N_B}} \sum_i \sum_j J_{ij} e^{-i \underline{k} \cdot (\underline{r}_i - \underline{r}_j)}, \quad (3.6a)$$

$$J'(\underline{k}) = \frac{1}{N_A} \sum_i \sum_{i'} J'_{ii'} e^{-i\underline{k} \cdot (\underline{r}_i - \underline{r}_{i'})}, \quad (3.6b)$$

$$J''(\underline{k}) = \frac{1}{N_B} \sum_j \sum_{j'} J''_{jj'} e^{-i\underline{k} \cdot (\underline{r}_j - \underline{r}_{j'})}, \quad (3.6c)$$

with their corresponding inverses:

$$J_{ij} = \frac{1}{\sqrt{N_A N_B}} \sum_{\underline{k}} J(\underline{k}) e^{i\underline{k} \cdot (\underline{r}_i - \underline{r}_j)}, \quad (3.6a')$$

$$J'_{ii'} = \frac{1}{N_A} \sum_{\underline{k}} J'(\underline{k}) e^{i\underline{k} \cdot (\underline{r}_i - \underline{r}_{i'})}, \quad (3.6b')$$

$$J''_{jj'} = \frac{1}{N_B} \sum_{\underline{k}} J''(\underline{k}) e^{i\underline{k} \cdot (\underline{r}_j - \underline{r}_{j'})}, \quad (3.6c')$$

where  $N_A$  and  $N_B$  are the number of ions in the A and B sublattices, respectively. In a unit cell of the normal spinel structure in which  $N_A = 8$  and  $N_B = 16$ , assuming nearest neighbour approximation, we have

$$\begin{aligned} J(\underline{k}) = 2\sqrt{2} J & \left[ \cos \frac{\sqrt{3}}{8} k_x a \cos \frac{1}{8} k_y a \cos \frac{1}{8} k_z a \right. \\ & + \cos \frac{1}{8} k_x a \cos \frac{\sqrt{3}}{8} k_y a \cos \frac{1}{8} k_z a \\ & \left. + \cos \frac{1}{8} k_x a \cos \frac{1}{8} k_y a \cos \frac{\sqrt{3}}{8} k_z a \right], \end{aligned} \quad (3.7a)$$

$$J'(\underline{k}) = 4J' \left[ \cos \frac{1}{4} k_x a \cos \frac{1}{4} k_y a \cos \frac{1}{4} k_z a \right], \quad (3.7b)$$

$$\begin{aligned}
J''(\underline{k}) = 2J'' & \left[ \cos \frac{1}{4}k_x a \cos \frac{1}{4}k_y a + \cos \frac{1}{4}k_y a \cos \frac{1}{4}k_z a \right. \\
& + \cos \frac{1}{4}k_z a \cos \frac{1}{4}k_x a + \sin \frac{1}{4}k_x a \sin \frac{1}{4}k_y a \\
& \left. - \sin \frac{1}{4}k_y a \sin \frac{1}{4}k_z a + \sin \frac{1}{4}k_z a \sin \frac{1}{4}k_x a \right], \quad (3.7c)
\end{aligned}$$

where  $J$ ,  $J'$  and  $J''$  are the isotropic exchange integrals for the interaction between spin in the A and B sublattices, within the A sublattice and within the B sublattice, respectively, and  $a$  is the lattice constant.

The detail calculations of  $J(\underline{k})$ ,  $J'(\underline{k})$  and  $J''(\underline{k})$  as obtained in the above equations are shown in Appendix C.

### 3.3 The Green's Functions

#### 3.3.1 The equations of motion

For our problems under investigation, the calculations of the following Green's functions are needed

$$\begin{aligned}
\langle\langle A_1^+; e^{aA_1^Z} A_1^- \rangle\rangle_E, & \quad \langle\langle A_1^+; e^{bB_1^Z} B_1^- \rangle\rangle_E, \\
\langle\langle B_1^+; e^{aA_1^Z} A_1^- \rangle\rangle_E, & \quad \langle\langle B_1^+; e^{bB_1^Z} B_1^- \rangle\rangle_E, \quad (3.8)
\end{aligned}$$

where  $a$  and  $b$  are parameters and  $l$  and  $l'$  are arbitrary numbers.



The reason for using these Green's functions will be explained in Chapter 4.

Using eq. (2.59), the equations of motion for these functions are

$$E \langle\langle A_1^+; e^{aA_1^Z}, A_1^- \rangle\rangle_E = \frac{1}{2\pi} \langle [A_1^+, e^{aA_1^Z}, A_1^-] \rangle + \langle\langle [A_1^+, \mathcal{H}]; e^{aA_1^Z}, A_1^- \rangle\rangle_E, \quad (3.9a)$$

$$E \langle\langle A_1^+; e^{bB_1^Z}, B_1^- \rangle\rangle_E = \frac{1}{2\pi} \langle [A_1^+, e^{bB_1^Z}, B_1^-] \rangle + \langle\langle [A_1^+, \mathcal{H}]; e^{bB_1^Z}, B_1^- \rangle\rangle_E, \quad (3.9b)$$

$$E \langle\langle B_1^+; e^{aA_1^Z}, A_1^- \rangle\rangle_E = \frac{1}{2\pi} \langle [B_1^+, e^{aA_1^Z}, A_1^-] \rangle + \langle\langle [B_1^+, \mathcal{H}]; e^{aA_1^Z}, A_1^- \rangle\rangle_E, \quad (3.9c)$$

$$E \langle\langle B_1^+; e^{bB_1^Z}, B_1^- \rangle\rangle_E = \frac{1}{2\pi} \langle [B_1^+, e^{bB_1^Z}, B_1^-] \rangle + \langle\langle [B_1^+, \mathcal{H}]; e^{bB_1^Z}, B_1^- \rangle\rangle_E. \quad (3.9d)$$

Introduce the commutation relations:

$$[A_1^+, e^{aA_1^Z}, A_1^-] = \Theta_A(a) \delta_{11}, \quad (3.10a)$$

$$[B_1^+, e^{bB_1^Z}, B_1^-] = \Theta_B(b) \delta_{11}, \quad (3.10b)$$

$$\text{and} \quad [A_1^+, e^{bB_1^Z}, B_1^-] = [B_1^+, e^{aA_1^Z}, A_1^-] = 0, \quad (3.10c)$$

where we can see from eqs. (3.10a) and (3.10b) that

$$\Theta_A(a) = [\Lambda^+, e^{a\Lambda^Z} \Lambda^-], \quad (3.11a)$$

$$\Theta_B(b) = [B^+, e^{bB^Z} B^-]. \quad (3.11b)$$

Calculation of the commutators defining  $\Theta_A(a)$  and  $\Theta_B(b)$  in eqs. (3.11a) and (3.11b) yields, respectively,<sup>32</sup>

$$\Theta_A(a) = A(A+1)(e^{-a}-1)\langle e^{a\Lambda^Z} \rangle + (e^{-a}+1)\langle e^{a\Lambda^Z} \Lambda^Z \rangle - (e^{-a}-1)\langle e^{a\Lambda^Z} (\Lambda^Z)^2 \rangle, \quad (3.12a)$$

$$\Theta_B(b) = B(B+1)(e^{-b}-1)\langle e^{bB^Z} \rangle + (e^{-b}+1)\langle e^{bB^Z} B^Z \rangle - (e^{-b}-1)\langle e^{bB^Z} (B^Z)^2 \rangle. \quad (3.12b)$$

By using eqs. (3.10a)-(3.10c), eqs. (3.9a)-(3.9d) become

$$\mathbb{E} \langle \langle \Lambda_1^+; e^{a\Lambda_1^Z}, \Lambda_1^- \rangle \rangle_{\mathbb{E}} = \frac{1}{2\pi} \Theta_A(a) \delta_{11} + \langle \langle [\Lambda_1^+, \mathcal{H}] ; e^{a\Lambda_1^Z}, \Lambda_1^- \rangle \rangle_{\mathbb{E}}, \quad (3.13a)$$

$$\mathbb{E} \langle \langle \Lambda_1^+; e^{bB_1^Z}, B_1^- \rangle \rangle_{\mathbb{E}} = \langle \langle [\Lambda_1^+, \mathcal{H}] ; e^{bB_1^Z}, B_1^- \rangle \rangle_{\mathbb{E}}, \quad (3.13b)$$

$$\mathbb{E} \langle \langle B_1^+; e^{a\Lambda_1^Z}, \Lambda_1^- \rangle \rangle_{\mathbb{E}} = \langle \langle [B_1^+, \mathcal{H}] ; e^{a\Lambda_1^Z}, \Lambda_1^- \rangle \rangle_{\mathbb{E}}, \quad (3.13c)$$

$$E \langle\langle B_1^+; e^{bB_1^Z}; B_1^- \rangle\rangle_E = \frac{1}{2\mathcal{H}} \mathcal{E}_B(b) \delta_{11}, + \langle\langle [B_1^+, \mathcal{H}]; e^{bB_1^Z}; B_1^- \rangle\rangle_E. \quad (3.13d)$$

3.3.1a The commutation relation of the spin operators with the Hamiltonian: The commutators  $[A_1^+, \mathcal{H}]$  and  $[B_1^+, \mathcal{H}]$  which appear in the four equations of motion are calculated using the spin commutation relations:

$$\begin{aligned} [A_1^+, A_1^Z] &= -A_1^+ \delta_{11}, & [B_1^+, B_1^Z] &= -B_1^+ \delta_{11}, \\ [A_1^-, A_1^Z] &= A_1^- \delta_{11}, & [B_1^-, B_1^Z] &= B_1^- \delta_{11}, \\ [A_1^+, A_1^-] &= 2A_1^Z \delta_{11}, & [B_1^+, B_1^-] &= 2B_1^Z \delta_{11}, \end{aligned} \quad (3.14)$$

and the commutator property

$$[A, BC] = [A, B]C + B[A, C]. \quad (3.15)$$

Hence, we get

$$\begin{aligned} [A_1^+, \mathcal{H}] &= \mathcal{E}_A \mu_B \mathcal{H} A_1^+ - \sum_j J_{j1} B_j^+ A_1^Z + \sum_j J_{j1} B_j^Z A_1^+ \\ &\quad - 2 \sum_i J_{i1}^i A_i^+ A_1^Z + 2 \sum_i J_{i1}^i A_i^Z A_1^+ \\ &\quad + D A_1^+ A_1^Z + D A_1^Z A_1^+, \end{aligned} \quad (3.16a)$$

$$\begin{aligned} [B_1^+, \mathcal{H}] &= \mathcal{E}_B \mu_B \mathcal{H} B_1^+ - \sum_i J_{i1} A_i^+ B_1^Z + \sum_i J_{i1} A_i^Z B_1^+ \\ &\quad - 2 \sum_j J_{j1}^j B_j^+ B_1^Z + 2 \sum_j J_{j1}^j B_j^Z B_1^+ \\ &\quad + D B_1^+ B_1^Z + D B_1^Z B_1^+. \end{aligned} \quad (3.16b)$$

Substituting eqs. (3.16a) and (3.16b) into the equations of motion (3.9a)-(3.9d), we get

$$\begin{aligned}
 E \langle \langle A_{11}^+; e^{aA_{11}^Z}; A_{11}^- \rangle \rangle_E &= \frac{1}{2\pi} \Theta_A(a) \delta_{111} + \mathcal{E}_{A^+B} \langle \langle A_{11}^+; e^{aA_{11}^Z}; A_{11}^- \rangle \rangle_E \\
 &- \sum_j J_{ij} \langle \langle B_j^+ A_{11}^Z; e^{aA_{11}^Z}; A_{11}^- \rangle \rangle_E \\
 &+ \sum_j J_{j1} \langle \langle B_j^Z A_{11}^+; e^{aA_{11}^Z}; A_{11}^- \rangle \rangle_E \\
 &- 2 \sum_i J_{i1}^i \langle \langle A_{i1}^+ A_{11}^Z; e^{aA_{11}^Z}; A_{11}^- \rangle \rangle_E \\
 &+ 2 \sum_i J_{i1}^i \langle \langle A_{i1}^Z A_{11}^+; e^{aA_{11}^Z}; A_{11}^- \rangle \rangle_E \\
 &+ D \langle \langle A_{11}^+ A_{11}^Z; e^{aA_{11}^Z}; A_{11}^- \rangle \rangle_E + D \langle \langle A_{11}^Z A_{11}^+; e^{aA_{11}^Z}; A_{11}^- \rangle \rangle_E,
 \end{aligned} \tag{3.17a}$$

$$\begin{aligned}
 E \langle \langle A_{11}^+; e^{bB_{11}^Z}; B_{11}^- \rangle \rangle_E &= \mathcal{E}_{A^+B} \langle \langle A_{11}^+; e^{bB_{11}^Z}; B_{11}^- \rangle \rangle_E \\
 &- \sum_j J_{j1} \langle \langle B_j^+ A_{11}^Z; e^{bB_{11}^Z}; B_{11}^- \rangle \rangle_E \\
 &+ \sum_j J_{j1} \langle \langle B_j^Z A_{11}^+; e^{bB_{11}^Z}; B_{11}^- \rangle \rangle_E \\
 &- 2 \sum_i J_{i1}^i \langle \langle A_{i1}^+ A_{11}^Z; e^{bB_{11}^Z}; B_{11}^- \rangle \rangle_E \\
 &+ 2 \sum_i J_{i1}^i \langle \langle A_{i1}^Z A_{11}^+; e^{bB_{11}^Z}; B_{11}^- \rangle \rangle_E \\
 &+ D \langle \langle A_{11}^+ A_{11}^Z; e^{bB_{11}^Z}; B_{11}^- \rangle \rangle_E + D \langle \langle A_{11}^Z A_{11}^+; e^{bB_{11}^Z}; B_{11}^- \rangle \rangle_E,
 \end{aligned} \tag{3.17b}$$

$$\begin{aligned}
E \langle \langle B_1^+; c^{a\Lambda_1^Z}, \Lambda_1^- \rangle \rangle_E &= C_{B^+B}^{\mu H} \langle \langle B_1^+; c^{a\Lambda_1^Z}, \Lambda_1^- \rangle \rangle_E \\
&- \sum_i J_{i1} \langle \langle \Lambda_i^+ B_1^Z; c^{a\Lambda_1^Z}, \Lambda_1^- \rangle \rangle_E \\
&+ \sum_i J_{i1} \langle \langle \Lambda_i^Z B_1^+; c^{a\Lambda_1^Z}, \Lambda_1^- \rangle \rangle_E \\
&- 2 \sum_j J_{j1}'' \langle \langle B_j^+ B_1^Z; c^{a\Lambda_1^Z}, \Lambda_1^- \rangle \rangle_E \\
&+ 2 \sum_j J_{j1}'' \langle \langle B_j^Z B_1^+; c^{a\Lambda_1^Z}, \Lambda_1^- \rangle \rangle_E \\
&+ \Gamma \langle \langle B_1^+ B_1^Z; c^{a\Lambda_1^Z}, \Lambda_1^- \rangle \rangle_E + D \langle \langle B_1^Z B_1^+; c^{a\Lambda_1^Z}, \Lambda_1^- \rangle \rangle_E,
\end{aligned}
\tag{3.17c}$$

$$\begin{aligned}
E \langle \langle B_1^+; c^{bB_1^Z}, B_1^- \rangle \rangle_E &= \frac{1}{2\pi} \Theta_B(b) \delta_{11} + C_{D^+B}^{\mu H} \langle \langle B_1^+; c^{bB_1^Z}, B_1^- \rangle \rangle_E \\
&- \sum_i J_{i1} \langle \langle \Lambda_i^+ B_1^Z; c^{bB_1^Z}, B_1^- \rangle \rangle_E \\
&+ \sum_i J_{i1} \langle \langle \Lambda_i^Z B_1^+; c^{bB_1^Z}, B_1^- \rangle \rangle_E \\
&- 2 \sum_j J_{j1}'' \langle \langle B_j^+ B_1^Z; c^{bB_1^Z}, B_1^- \rangle \rangle_E \\
&+ 2 \sum_j J_{j1}'' \langle \langle B_j^Z B_1^+; c^{bB_1^Z}, B_1^- \rangle \rangle_E \\
&+ D \langle \langle B_1^+ B_1^Z; c^{bB_1^Z}, B_1^- \rangle \rangle_E + D \langle \langle B_1^Z B_1^+; c^{bB_1^Z}, B_1^- \rangle \rangle_E.
\end{aligned}
\tag{3.17d}$$

In order to explicitly solve the above four equations of motion, the remaining problem is to express the higher order Green's functions on the right-hand side in terms of lower order Green's functions.

### 3.3.2 The Callen decoupling approximation

Owing to the fact that the Tyablikov decoupling as defined in eq. (2.61) does not give the correct description for the behaviors of the system at low temperatures<sup>30,31</sup>, a new decoupling scheme should be used instead. Here the Callen decoupling approximation, which gives the results valid through the entire temperature range, will be employed.

In the case of general spin,  $A_i^Z$  can be written in either of the following forms

$$A_i^Z = A(A+1) - (A_i^Z)^2 - A_i^- A_i^+, \quad (3.18)$$

$$A_i^Z = \frac{1}{2} (A_i^+ A_i^- - A_i^- A_i^+). \quad (3.19)$$

Neglecting the fluctuations of  $(A_i^Z)^2$ , Callen<sup>32</sup> developed a decoupling such as

$$\langle\langle A_i^Z B_j^+; C \rangle\rangle \longrightarrow \langle A^Z \rangle \langle\langle B_j^+; C \rangle\rangle - \alpha \langle A_i^- B_j^+ \rangle \langle\langle A_i^+; C \rangle\rangle, \quad (3.20)$$

where  $\alpha$  is the fractional contribution of the identity (3.18) and  $(1-\alpha)$  is the contribution of the identity (3.19) to this result. The value of  $\alpha$  for general spin  $S$  is determined by Callen as

$$\alpha = (1/2S)\langle S^2 \rangle / S. \quad (3.21)$$

It should be noted that even though the fluctuations of  $(A_i^z)^2$  are taken into account, the same result is obtained. Therefore the Callen decoupling excludes the effects of crystal field anisotropy.

Consequently, the D terms in the original Hamiltonian are dropped, and hence, the equations of motion (3.17a)-(3.17d) do not contain the D terms.

For later comparison, we note that Yablonskii decoupled his Green's functions by the decoupling

$$\langle\langle AB; C \rangle\rangle = \langle A \rangle \langle\langle B; C \rangle\rangle + \langle B \rangle \langle\langle A; C \rangle\rangle.$$

### 3.3.3 The matrix form of the Green's functions

Consider the equation of motion for the Green's function  $\langle\langle A_1^+; e^{aA_2^z} A_1^- \rangle\rangle_E$ , i.e., eq. (3.17a). Apply the Callen decoupling approximation given by eq. (3.20) to the higher order terms on the right hand side of the equation of motion, hence, those terms are decoupled as follows;

$$\begin{aligned} \langle\langle B_j^+ A_1^z; e^{a\Lambda_1^z} A_1^- \rangle\rangle_E &= \langle\Lambda^z\rangle \langle\langle B_j^+; e^{a\Lambda_1^z} A_1^- \rangle\rangle_E \\ &\quad - \alpha_A \langle\langle A_1^- B_j^+ \rangle\rangle \langle\langle A_1^+; e^{a\Lambda_1^z} A_1^- \rangle\rangle_E, \end{aligned} \quad (3.22a)$$

$$\begin{aligned} \langle\langle B_j^z A_1^+; e^{a\Lambda_1^z} A_1^- \rangle\rangle_E &= \langle B^z \rangle \langle\langle A_1^+; e^{a\Lambda_1^z} A_1^- \rangle\rangle_E \\ &\quad - \alpha_B \langle\langle B_j^- A_1^+ \rangle\rangle \langle\langle B_j^+; e^{a\Lambda_1^z} A_1^- \rangle\rangle_E, \end{aligned} \quad (3.22b)$$

$$\begin{aligned} \langle\langle A_i^+ A_1^z; e^{a\Lambda_1^z} A_1^- \rangle\rangle_E &= \langle\Lambda^z\rangle \langle\langle A_i^+; e^{a\Lambda_1^z} A_1^- \rangle\rangle_E \\ &\quad - \alpha_A \langle\langle A_1^- A_i^+ \rangle\rangle \langle\langle A_1^+; e^{a\Lambda_1^z} A_1^- \rangle\rangle_E, \end{aligned} \quad (3.11c)$$

$$\begin{aligned} \langle\langle A_i^z A_1^+; e^{a\Lambda_1^z} A_1^- \rangle\rangle_E &= \langle\Lambda^z\rangle \langle\langle A_1^+; e^{a\Lambda_1^z} A_1^- \rangle\rangle_E \\ &\quad - \alpha_A \langle\langle A_i^- A_1^+ \rangle\rangle \langle\langle A_1^+; e^{a\Lambda_1^z} A_1^- \rangle\rangle_E. \end{aligned} \quad (3.22d)$$

Substitute, eqs. (3.22a)-(3.22d) into eq. (3.17a) in which the D terms are dropped, then introduce the Fourier transform of the Green's functions to the reciprocal space as follows

$$\langle\langle A_k^+; e^{a\Lambda^z} A_k^- \rangle\rangle_E = \frac{1}{N_A} \sum_l \sum_{l'} e^{ik \cdot (r_l - r_{l'})} \langle\langle A_l^+; e^{a\Lambda_1^z} A_{l'}^- \rangle\rangle_E, \quad (3.23a)$$

$$\langle\langle A_k^+; e^{bB^z} B_k^- \rangle\rangle_E = \frac{1}{\sqrt{N_A N_B}} \sum_l \sum_{l'} e^{ik \cdot (r_l - r_{l'})} \langle\langle A_l^+; e^{bB_1^z} B_{l'}^- \rangle\rangle_E, \quad (3.23b)$$



$$\langle\langle B_k^+; e^{a\Lambda_k^z} \Lambda_k^- \rangle\rangle_E = \frac{1}{\sqrt{N_A N_B}} \sum_{\underline{1}} \sum_{\underline{1}'} e^{i\mathbf{k} \cdot (\underline{r}_1 - \underline{r}_{1'})} \langle\langle B_{\underline{1}}^+; e^{a\Lambda_{\underline{1}}^z} \Lambda_{\underline{1}}^- \rangle\rangle_E, \quad (3.23c)$$

$$\langle\langle B_k^+; e^{bB_k^z} B_k^- \rangle\rangle_E = \frac{1}{N_B} \sum_{\underline{1}} \sum_{\underline{1}'} e^{i\mathbf{k} \cdot (\underline{r}_1 - \underline{r}_{1'})} \langle\langle B_{\underline{1}}^+; e^{bB_{\underline{1}}^z} B_{\underline{1}}^- \rangle\rangle_E, \quad (3.23d)$$

where their corresponding inverses are

$$\langle\langle A_{\underline{1}}^+; e^{a\Lambda_{\underline{1}}^z} \Lambda_{\underline{1}}^- \rangle\rangle_E = \frac{1}{N_A} \sum_{\underline{k}} e^{-i\mathbf{k} \cdot (\underline{r}_1 - \underline{r}_{1'})} \langle\langle A_{\underline{k}}^+; e^{a\Lambda_{\underline{k}}^z} \Lambda_{\underline{k}}^- \rangle\rangle_E, \quad (3.23a')$$

$$\langle\langle A_{\underline{1}}^+; e^{bB_{\underline{1}}^z} B_{\underline{1}}^- \rangle\rangle_E = \frac{1}{\sqrt{N_A N_B}} \sum_{\underline{k}} e^{-i\mathbf{k} \cdot (\underline{r}_1 - \underline{r}_{1'})} \langle\langle A_{\underline{k}}^+; e^{bB_{\underline{k}}^z} B_{\underline{k}}^- \rangle\rangle_E, \quad (3.23b')$$

$$\langle\langle B_{\underline{1}}^+; e^{a\Lambda_{\underline{1}}^z} \Lambda_{\underline{1}}^- \rangle\rangle_E = \frac{1}{\sqrt{N_A N_B}} \sum_{\underline{k}} e^{-i\mathbf{k} \cdot (\underline{r}_1 - \underline{r}_{1'})} \langle\langle B_{\underline{k}}^+; e^{a\Lambda_{\underline{k}}^z} \Lambda_{\underline{k}}^- \rangle\rangle_E, \quad (3.23c')$$

$$\langle\langle B_{\underline{1}}^+; e^{bB_{\underline{1}}^z} B_{\underline{1}}^- \rangle\rangle_E = \frac{1}{N_B} \sum_{\underline{k}} e^{-i\mathbf{k} \cdot (\underline{r}_1 - \underline{r}_{1'})} \langle\langle B_{\underline{k}}^+; e^{bB_{\underline{k}}^z} B_{\underline{k}}^- \rangle\rangle_E. \quad (3.23d')$$

In addition, the Fourier transform of the correlation functions and their corresponding inverses are defined as follows;

$$\langle\langle A^- A^+ \rangle\rangle_{\underline{k}} = \frac{1}{N_A} \sum_{\underline{1}} \sum_{\underline{1}'} e^{-i\mathbf{k} \cdot (\underline{r}_1 - \underline{r}_{1'})} \langle\langle A_{\underline{1}}^- A_{\underline{1}}^+ \rangle\rangle, \quad (3.24a)$$

$$\langle\langle A^- B^+ \rangle\rangle_{\underline{k}} = \frac{1}{\sqrt{N_A N_B}} \sum_{\underline{1}} \sum_{\underline{1}'} e^{-i\mathbf{k} \cdot (\underline{r}_1 - \underline{r}_{1'})} \langle\langle A_{\underline{1}}^- B_{\underline{1}}^+ \rangle\rangle, \quad (3.24b)$$

$$\langle\langle B^- A^+ \rangle\rangle_{\underline{k}} = \frac{1}{\sqrt{N_A N_B}} \sum_{\underline{1}} \sum_{\underline{1}'} e^{-i\mathbf{k} \cdot (\underline{r}_1 - \underline{r}_{1'})} \langle\langle B_{\underline{1}}^- A_{\underline{1}}^+ \rangle\rangle, \quad (3.24c)$$

$$\langle\langle B^- B^+ \rangle\rangle_{\underline{k}} = \frac{1}{N_B} \sum_{\underline{1}} \sum_{\underline{1}'} e^{-i\mathbf{k} \cdot (\underline{r}_1 - \underline{r}_{1'})} \langle\langle B_{\underline{1}}^- B_{\underline{1}}^+ \rangle\rangle, \quad (3.24d)$$

and

$$\langle A_1^- A_1^+ \rangle = \frac{1}{N_A} \sum_{\underline{k}} e^{i\underline{k} \cdot (\underline{r}_1 - \underline{r}_1')} \langle A^- A^+ \rangle_{\underline{k}}, \quad (3.24a')$$

$$\langle A_1^- B_1^+ \rangle = \frac{1}{\sqrt{N_A N_B}} \sum_{\underline{k}} e^{i\underline{k} \cdot (\underline{r}_1 - \underline{r}_1')} \langle A^- B^+ \rangle_{\underline{k}}, \quad (3.24b')$$

$$\langle B_1^- A_1^+ \rangle = \frac{1}{\sqrt{N_A N_B}} \sum_{\underline{k}} e^{i\underline{k} \cdot (\underline{r}_1 - \underline{r}_1')} \langle B^- A^+ \rangle_{\underline{k}}, \quad (3.24c')$$

$$\langle B_1^- B_1^+ \rangle = \frac{1}{N_B} \sum_{\underline{k}} e^{i\underline{k} \cdot (\underline{r}_1 - \underline{r}_1')} \langle B^- B^+ \rangle_{\underline{k}}, \quad (3.24d')$$

where  $N_A$  and  $N_B$  are the number of ions in the A and B sublattices, respectively. Using the Fourier transform of the Green's functions and the correlation function of eqs. (3.23a)-(3.23d') and (3.24a)-(3.24d') as well as the Fourier transform of the exchange integrals defined in eqs. (3.6a)-(3.6c'), the equation of motion for the function  $\langle\langle A_1^+; e^{a_1^z} A_1^- \rangle\rangle_E$  becomes

$$\begin{aligned} & \left[ E - \epsilon_A^z N_B H + \left( \frac{N_B}{N_A} \right)^{1/2} J(0) \langle B^z \rangle + 2(J'(k) - J'(0)) \langle A^z \rangle \right. \\ & \left. - \frac{1}{N_A} \alpha_A \sum_{\underline{k}'} J(\underline{k}) \langle A^- B^+ \rangle_{\underline{k}'} + \frac{1}{N_A} 2\alpha_A \sum_{\underline{k}'} (J'(k - \underline{k}') - J'(k')) \langle A^- A^+ \rangle_{\underline{k}'} \right] \\ & \times \langle\langle A_k^+; e^{a_1^z} A_k^- \rangle\rangle_E = \frac{1}{2\pi} \Theta_A(a) - \left[ J(\underline{k}) \langle A^z \rangle \right. \\ & \left. + \frac{1}{\sqrt{N_A N_B}} \alpha_B \sum_{\underline{k}'} J(\underline{k} - \underline{k}') \langle B^- A^+ \rangle_{\underline{k}'} \right] \langle\langle B_k^+; e^{a_1^z} A_k^- \rangle\rangle_E. \end{aligned} \quad (3.25)$$

In the derivation of eq.(3.25), we have to take the Fourier transform of the terms on the right hand side of the equation of motion. The detailed calculations of some of those terms will be presented here:

$$\begin{aligned}
 \text{the 4th term} &= \frac{1}{N_A} \mathcal{O}_A \sum_j \sum_l \sum_{l'} J_{jl} \langle A_l^- B_j^+ \rangle \langle \langle A_l^+ ; e^{aA_l^z} A_l^- \rangle \rangle_E e^{ik \cdot (r_l - r_{l'})} \\
 &= \frac{1}{N_A} \cdot \frac{1}{N_A N_B} \cdot \frac{1}{N_A} \mathcal{O}_A \sum_j \sum_l \sum_{l'} \sum_{k_1} \sum_{k_2} \sum_{k_3} J(k_1) \\
 &\quad \times e^{ik_1 \cdot (r_j - r_{l'})} \langle A_l^- B_j^+ \rangle_{k_2} e^{ik_2 \cdot (r_l - r_j)} \langle \langle A_{k_3}^+ ; e^{aA_{k_3}^z} A_{k_3}^- \rangle \rangle_E \\
 &\quad \times e^{ik_3 \cdot (r_l - r_{l'})} e^{ik \cdot (r_l - r_{l'})} \\
 &= \frac{1}{N_A N_B} \cdot \frac{1}{N_A} \mathcal{O}_A \sum_j \sum_l \sum_{k_1} \sum_{k_2} \sum_{k_3} J(k_1) e^{ik_1 \cdot (r_j - r_{l'})} \\
 &\quad \times \langle A_l^- B_j^+ \rangle_{k_2} e^{ik_2 \cdot (r_l - r_j)} \langle \langle A_{k_3}^+ ; e^{aA_{k_3}^z} A_{k_3}^- \rangle \rangle_E \\
 &\quad \times e^{i(k-k_3) \cdot r_{l'}} \delta(k-k_3),
 \end{aligned}$$

where we have used the fact that  $\frac{1}{N_A} \sum_{l'} e^{-i(k-k_3) \cdot r_{l'}} = \delta(k-k_3)$ .

$$\begin{aligned}
 \text{the 4th term} &= \frac{1}{N_A N_B} \cdot \frac{1}{N_A} \mathcal{O}_A \sum_j \sum_l \sum_{k_1} \sum_{k_2} J(k_1) e^{ik_1 \cdot (r_j - r_{l'})} \\
 &\quad \times \langle A_l^- B_j^+ \rangle_{k_2} e^{ik_2 \cdot (r_l - r_j)} \langle \langle A_k^+ ; e^{aA_k^z} A_k^- \rangle \rangle_E.
 \end{aligned}$$

$$\begin{aligned} \text{the 4th term} &= \frac{1}{N_A N_B} \alpha_A \sum_j \sum_{\underline{k}_1} \sum_{\underline{k}_2} J(\underline{k}_1) \langle \Lambda^{-B^+} \rangle_{\underline{k}_2} \langle \langle \Lambda_{\underline{k}}^+; e^{a\Lambda^Z} \Lambda_{\underline{k}}^- \rangle \rangle_E \\ &\quad \times e^{i(\underline{k}_1 - \underline{k}_2) \cdot \underline{r}_j} \delta(\underline{k}_1 - \underline{k}_2) \end{aligned}$$

$$\begin{aligned} &= \frac{1}{N_A N_B} \alpha_A \sum_j \sum_{\underline{k}_1} J(\underline{k}_1) \langle \Lambda^{-B^+} \rangle_{\underline{k}_1} \langle \langle \Lambda_{\underline{k}}^+; e^{a\Lambda^Z} \Lambda_{\underline{k}}^- \rangle \rangle_E \\ &= \frac{1}{N_A} \alpha_A \sum_{\underline{k}_1} J(\underline{k}_1) \langle \Lambda^{-B^+} \rangle_{\underline{k}_1} \langle \langle \Lambda_{\underline{k}}^+; e^{a\Lambda^Z} \Lambda_{\underline{k}}^- \rangle \rangle_E \\ &= \frac{1}{N_A} \alpha_A \sum_{\underline{k}'} J(\underline{k}') \langle \Lambda^{-B^+} \rangle_{\underline{k}'} \langle \langle \Lambda_{\underline{k}}^+; e^{a\Lambda^Z} \Lambda_{\underline{k}}^- \rangle \rangle_E. \end{aligned}$$

$$\begin{aligned} \text{the 10th term} &= \frac{1}{N_A} 2\alpha_A \sum_i \sum_l \sum_{l'} J'_{il} \langle \Lambda_i^- \Lambda_l^+ \rangle \langle \langle \Lambda_i^+; e^{a\Lambda^Z} \Lambda_l^- \rangle \rangle_E \\ &\quad \times e^{i\underline{k} \cdot (\underline{r}_l - \underline{r}_l')} \end{aligned}$$

$$\begin{aligned} &= \frac{1}{N_A} \left( \frac{1}{N_A} \right)^3 2\alpha_A \sum_i \sum_l \sum_{l'} \sum_{\underline{k}_1} \sum_{\underline{k}_2} \sum_{\underline{k}_3} J'(\underline{k}_1) \\ &\quad \times e^{i\underline{k}_1 \cdot (\underline{r}_i - \underline{r}_l)} \langle \Lambda_i^- \Lambda_l^+ \rangle_{\underline{k}_2} e^{i\underline{k}_2 \cdot (\underline{r}_i - \underline{r}_l)} \langle \langle \Lambda_{\underline{k}_3}^+; e^{a\Lambda^Z} \Lambda_{\underline{k}_3}^- \rangle \rangle_E \\ &\quad \times e^{i\underline{k}_3 \cdot (\underline{r}_l - \underline{r}_i)} e^{i\underline{k} \cdot (\underline{r}_l - \underline{r}_l')} \end{aligned}$$

$$\begin{aligned} &= \frac{1}{N_A} \left( \frac{1}{N_A} \right)^2 2\alpha_A \sum_i \sum_l \sum_{\underline{k}_1} \sum_{\underline{k}_2} \sum_{\underline{k}_3} J'(\underline{k}_1) e^{i\underline{k}_1 \cdot (\underline{r}_i - \underline{r}_l)} \\ &\quad \times \langle \Lambda_i^- \Lambda_l^+ \rangle_{\underline{k}_2} e^{i\underline{k}_2 \cdot (\underline{r}_i - \underline{r}_l)} \langle \langle \Lambda_{\underline{k}_3}^+; e^{a\Lambda^Z} \Lambda_{\underline{k}_3}^- \rangle \rangle_E \\ &\quad \times e^{-i\underline{k}_3 \cdot \underline{r}_i} e^{i\underline{k} \cdot \underline{r}_l} \delta(\underline{k} - \underline{k}_3). \end{aligned}$$

$$\begin{aligned}
\text{the 10th term} &= \frac{1}{N_A} \left( \frac{1}{N_A} \right)^2 2\alpha_A \sum_i \sum_l \sum_{\underline{k}_1} \sum_{\underline{k}_2} J^i(\underline{k}_1) e^{i\underline{k}_1 \cdot (\underline{r}_i - \underline{r}_1)} \\
&\quad \times \langle \underline{A}^- \underline{A}^+ \rangle_{\underline{k}_2} e^{i\underline{k}_2 \cdot (\underline{r}_i - \underline{r}_1)} \langle \langle \underline{A}_k^+; e^{a\Lambda^z} \underline{A}_k^- \rangle \rangle_E e^{i\underline{k} \cdot (\underline{r}_1 - \underline{r}_1)} \\
&= \frac{1}{N_A} \left( \frac{1}{N_A} \right)^2 2\alpha_A \sum_i \sum_{\underline{k}_1} \sum_{\underline{k}_2} J^i(\underline{k}_1) \langle \underline{A}^- \underline{A}^+ \rangle_{\underline{k}_2} \langle \langle \underline{A}_k^+; e^{a\Lambda^z} \underline{A}_k^- \rangle \rangle_E \\
&\quad \times e^{-i(\underline{k} - \underline{k}_1 - \underline{k}_2) \cdot \underline{r}_i} \delta(\underline{k} - \underline{k}_1 - \underline{k}_2) \\
&= \frac{1}{N_A} \left( \frac{1}{N_A} \right)^2 2\alpha_A \sum_i \sum_{\underline{k}_2} J^i(\underline{k} - \underline{k}_2) \langle \underline{A}^- \underline{A}^+ \rangle_{\underline{k}_2} \langle \langle \underline{A}_k^+; e^{a\Lambda^z} \underline{A}_k^- \rangle \rangle_E \\
&= \frac{1}{N_A} 2\alpha_A \sum_{\underline{k}_2} J^i(\underline{k} - \underline{k}_2) \langle \underline{A}^- \underline{A}^+ \rangle_{\underline{k}_2} \langle \langle \underline{A}_k^+; e^{a\Lambda^z} \underline{A}_k^- \rangle \rangle_E \\
\text{or} &= \frac{1}{N_A} 2\alpha_A \sum_{\underline{k}'} J^i(\underline{k} - \underline{k}') \langle \underline{A}^- \underline{A}^+ \rangle_{\underline{k}'} \langle \langle \underline{A}_k^+; e^{a\Lambda^z} \underline{A}_k^- \rangle \rangle_E.
\end{aligned}$$

The other three equations of motion for the functions  $\langle \langle \underline{A}_1^+; e^{bB_1^z} \underline{B}_1^- \rangle \rangle_E$ ,  $\langle \langle \underline{B}_1^+; e^{a\Lambda_1^z} \underline{A}_1^- \rangle \rangle_E$  and  $\langle \langle \underline{B}_1^+; e^{bB_1^z} \underline{B}_1^- \rangle \rangle_E$  may be reduced to give their explicit solutions in the similar form to eq. (3.25) by following the same steps of derivations. The resulting four equations can be combined to give the one matrix equation such as

$$\begin{aligned}
& \begin{pmatrix} E-E_1 & 0 \\ 0 & E-E_2 \end{pmatrix} \begin{pmatrix} \langle\langle A_k^+; e^{a\Lambda^Z} A_k^- \rangle\rangle_E & \langle\langle A_k^+; e^{bB^Z} B_k^- \rangle\rangle_E \\ \langle\langle B_k^+; e^{a\Lambda^Z} A_k^- \rangle\rangle_E & \langle\langle B_k^+; e^{bB^Z} B_k^- \rangle\rangle_E \end{pmatrix} \\
& = \begin{pmatrix} \frac{1}{2\pi} \Theta_A(a) & 0 \\ 0 & \frac{1}{2\pi} \Theta_B(b) \end{pmatrix} \\
& + \begin{pmatrix} -J_1 \langle\langle B_k^+; e^{a\Lambda^Z} A_k^- \rangle\rangle_E & -J_1 \langle\langle B_k^+; e^{bB^Z} B_k^- \rangle\rangle_E \\ -J_2 \langle\langle A_k^+; e^{a\Lambda^Z} A_k^- \rangle\rangle_E & -J_2 \langle\langle A_k^+; e^{bB^Z} B_k^- \rangle\rangle_E \end{pmatrix},
\end{aligned} \tag{3.26}$$

$$\begin{aligned}
\text{where } E_1 &= \varepsilon_A \mu_B H + \left(\frac{N_B}{N_A}\right)^{\frac{1}{2}} J(0) \langle B^Z \rangle - 2(J'(\underline{k}) - J'(0)) \langle \Lambda^Z \rangle \\
& + \frac{1}{N_A} \alpha_A \sum_{\underline{k}'} J(\underline{k}') \langle \Lambda^- B^+ \rangle_{\underline{k}'} \\
& - \frac{1}{N_A} 2\alpha_A \sum_{\underline{k}'} (J'(\underline{k}-\underline{k}') - J'(\underline{k}')) \langle \Lambda^- \Lambda^+ \rangle_{\underline{k}'}, \tag{3.27a}
\end{aligned}$$

$$\begin{aligned}
E_2 &= \varepsilon_B \mu_B H + \left(\frac{N_A}{N_B}\right)^{\frac{1}{2}} J(0) \langle \Lambda^Z \rangle - 2(J''(\underline{k}) - J''(0)) \langle B^Z \rangle \\
& + \frac{1}{N_B} \alpha_B \sum_{\underline{k}'} J(\underline{k}') \langle B^- \Lambda^+ \rangle_{\underline{k}'} \\
& - \frac{1}{N_B} 2\alpha_B \sum_{\underline{k}'} (J''(\underline{k}-\underline{k}') - J''(\underline{k}')) \langle B^- B^+ \rangle_{\underline{k}'}, \tag{3.27b}
\end{aligned}$$

$$J_1 = J(\underline{k}) \langle \Lambda^Z \rangle + \frac{1}{\sqrt{N_A N_B}} \alpha_B \sum_{\underline{k}'} J(\underline{k}-\underline{k}') \langle B^- \Lambda^+ \rangle_{\underline{k}'}, \tag{3.28a}$$

$$J_2 = J(\underline{k}) \langle B^Z \rangle + \frac{1}{\sqrt{N_A N_B}} \alpha_A \sum_{\underline{k}'} J(\underline{k}-\underline{k}') \langle \Lambda^- B^+ \rangle_{\underline{k}'}. \tag{3.28b}$$

Solving the matrix equation (3.26), we finally obtain the matrix form of the Green's functions of the system as

$$\begin{pmatrix} \langle\langle \Lambda_k^+; e^{a\Lambda^z} \Lambda_k^- \rangle\rangle_E & \langle\langle \Lambda_k^+; e^{bB^z} B_k^- \rangle\rangle_E \\ \langle\langle B_k^+; e^{a\Lambda^z} \Lambda_k^- \rangle\rangle_E & \langle\langle B_k^+; e^{bB^z} B_k^- \rangle\rangle_E \end{pmatrix} = \frac{1}{(E-E_1)(E-E_2) - (J_1)(J_2)} \begin{pmatrix} E-E_2 & -J_1 \\ -J_2 & E-E_1 \end{pmatrix} \begin{pmatrix} \frac{1}{2\pi} \Theta_{\Lambda}(a) & 0 \\ 0 & \frac{1}{2\pi} \Theta_B(b) \end{pmatrix}, \quad (3.29)$$

where  $E_1$ ,  $E_2$  and  $J_1$ ,  $J_2$  are defined in eqs. (3.27a), (3.27b) and (3.28a), (3.28b), respectively.

Consequently, the matrix form of the Green's functions (eq. (3.26)) can be written out into the following four equations:

$$\langle\langle \Lambda_k^+; e^{a\Lambda^z} \Lambda_k^- \rangle\rangle_E = \frac{(2\pi)^{-1} \Theta_{\Lambda}(a)(E-E_2)}{(E-E_1)(E-E_2) - (J_1)(J_2)}, \quad (3.30a)$$

$$\langle\langle \Lambda_k^+; e^{bB^z} B_k^- \rangle\rangle_E = \frac{(2\pi)^{-1} \Theta_B(b)(J_1)}{(E-E_1)(E-E_2) - (J_1)(J_2)}, \quad (3.30b)$$

$$\langle\langle B_k^+; e^{a\Lambda^z} \Lambda_k^- \rangle\rangle_E = \frac{(2\pi)^{-1} \Theta_{\Lambda}(a)(J_2)}{(E-E_1)(E-E_2) - (J_1)(J_2)}, \quad (3.30c)$$

$$\langle\langle B_k^+; e^{bB^z} B_k^- \rangle\rangle_E = \frac{(2\pi)^{-1} \Theta_B(b)(E-E_1)}{(E-E_1)(E-E_2) - (J_1)(J_2)}. \quad (3.30d)$$

By rearranging the terms in the definitions of  $E_1, E_2,$   
 $J_1$  and  $J_2$ , we get

$$\langle\langle A_k^+; e^{a\Lambda^z} A_k^- \rangle\rangle_E = \frac{1}{2\pi} \Theta_A(a) \left[ \frac{\Lambda + \sqrt{\Lambda^2 + 4n}}{2\sqrt{\Lambda^2 + 4n}} \cdot \frac{1}{E - E_+} - \frac{\Lambda - \sqrt{\Lambda^2 + 4n}}{2\sqrt{\Lambda^2 + 4n}} \cdot \frac{1}{E - E_-} \right],$$

$$\langle\langle A_k^+; e^{bB^z} B_k^- \rangle\rangle_E = \frac{\Theta_B(b)}{2\pi} \frac{J(\underline{k}) \langle \Lambda^z \rangle - \langle B^z \rangle \frac{\alpha_B}{\sqrt{N_A N_B}} \sum_{\underline{k}'} J(\underline{k} - \underline{k}') \langle B^- A^+ \rangle_{\underline{k}'}}{2\sqrt{\Lambda^2 + 4n}} \times \left[ \frac{1}{E - E_-} - \frac{1}{E - E_+} \right], \quad (3.31b)$$

$$\langle\langle B_k^+; e^{a\Lambda^z} A_k^- \rangle\rangle_E = \frac{\Theta_A(a)}{2\pi} \frac{J(\underline{k}) \langle B^z \rangle - \langle \Lambda^z \rangle \frac{\alpha_A}{\sqrt{N_A N_B}} \sum_{\underline{k}'} J(\underline{k} - \underline{k}') \langle A^- B^+ \rangle_{\underline{k}'}}{2\sqrt{\Lambda^2 + 4n}} \times \left[ \frac{1}{E - E_-} - \frac{1}{E - E_+} \right], \quad (3.31c)$$

$$\langle\langle B_k^+; e^{bB^z} B_k^- \rangle\rangle_E = \frac{1}{2\pi} \Theta_B(b) \left[ \frac{\Lambda + \sqrt{\Lambda^2 + 4n}}{2\sqrt{\Lambda^2 + 4n}} \cdot \frac{1}{E - E_-} - \frac{\Lambda - \sqrt{\Lambda^2 + 4n}}{2\sqrt{\Lambda^2 + 4n}} \cdot \frac{1}{E - E_+} \right],$$

(3.31d)

where  $\alpha_A = \frac{\alpha_A}{\langle \Lambda^z \rangle}$ ,  $\alpha_B = \frac{\alpha_B}{\langle B^z \rangle}$ ,

$$n = \left[ J(\underline{k}) \langle B^z \rangle + \alpha_A' \langle \Lambda^z \rangle \frac{1}{\sqrt{N_A N_B}} \sum_{\underline{k}'} J(\underline{k} - \underline{k}') \langle A^- B^+ \rangle_{\underline{k}'} \right] \times \left[ J(\underline{k}) \langle \Lambda^z \rangle + \alpha_B' \langle B^z \rangle \frac{1}{\sqrt{N_A N_B}} \sum_{\underline{k}'} J(\underline{k} - \underline{k}') \langle B^- A^+ \rangle_{\underline{k}'} \right], \quad (3.32)$$



$$A = \frac{1}{2} \left[ \left( \frac{N_B}{N_A} \right)^{\frac{1}{2}} J(0) \langle B^z \rangle - \left( \frac{N_A}{N_B} \right)^{\frac{1}{2}} J(0) \langle \Lambda^z \rangle - \tilde{\epsilon}_A(\underline{k}) \langle \Lambda^z \rangle + \tilde{\epsilon}_B(\underline{k}) \langle B^z \rangle \right], \quad (3.33)$$

and where 
$$E_{\pm} = a \pm b, \quad (3.34a)$$

$$a = \frac{1}{2} (\mathcal{E}_A + \mathcal{E}_B) \mu_B^H + \frac{1}{2} \left[ \left( \frac{N_B}{N_A} \right)^{\frac{1}{2}} J(0) \langle B^z \rangle - \left( \frac{N_A}{N_B} \right)^{\frac{1}{2}} J(0) \langle \Lambda^z \rangle - \tilde{\epsilon}_A(\underline{k}) \langle \Lambda^z \rangle - \tilde{\epsilon}_B(\underline{k}) \langle B^z \rangle \right], \quad (3.34b)$$

$$b = \frac{1}{2} \left[ \left( (\mathcal{E}_A - \mathcal{E}_B) \mu_B^H + \left( \frac{N_B}{N_A} \right)^{\frac{1}{2}} J(0) \langle B^z \rangle - \left( \frac{N_A}{N_B} \right)^{\frac{1}{2}} J(0) \langle \Lambda^z \rangle - \tilde{\epsilon}_A(\underline{k}) \langle \Lambda^z \rangle + \tilde{\epsilon}_B(\underline{k}) \langle B^z \rangle \right)^2 + 4n \right]^{\frac{1}{2}}, \quad (3.34c)$$

$$\begin{aligned} \tilde{\epsilon}_A(\underline{k}) = 2 & \left[ J^i(\underline{k}) - J^i(0) + \alpha'_A \frac{1}{N_A} \sum_{\underline{k}'} (J^i(\underline{k}-\underline{k}') - J^i(\underline{k}')) \langle \Lambda^- \Lambda^+ \rangle_{\underline{k}'} \right] \\ & - \alpha'_A \frac{1}{N_A} \sum_{\underline{k}'} J(\underline{k}') \langle \Lambda^- B^+ \rangle_{\underline{k}'}, \quad (3.35a) \end{aligned}$$

$$\begin{aligned} \tilde{\epsilon}_B(\underline{k}) = 2 & \left[ J^{ii}(\underline{k}) - J^{ii}(0) + \alpha'_B \frac{1}{N_B} \sum_{\underline{k}'} (J^{ii}(\underline{k}-\underline{k}') - J^{ii}(\underline{k}')) \langle B^- B^+ \rangle_{\underline{k}'} \right] \\ & - \alpha'_B \frac{1}{N_B} \sum_{\underline{k}'} J(\underline{k}') \langle B^- \Lambda^+ \rangle_{\underline{k}'}, \quad (3.35b) \end{aligned}$$

The correlation functions appearing in  $\tilde{\epsilon}_A(\underline{k})$  and  $\tilde{\epsilon}_B(\underline{k})$  are obtained from the Green's functions of eqs. (3.31a)-(3.31d) for  $a = b = 0$ . By the use of eqs. (3.12a) and (3.12b) for

$a = b = 0$ , we have

$$\Theta_A(0) = 2\langle A^z \rangle, \quad (3.36a)$$

$$\Theta_B(0) = 2\langle B^z \rangle, \quad (3.36b)$$

### 3.4 Comparison with Other Works

It is interesting to note that what happens in case of antiferromagnetism when  $\langle A^z \rangle = -\langle B^z \rangle$  and  $N_A = N_B$ . Since there is no distinction between the interactions between spin up ions and between spin down ions, the intrasublattice coupling must be the same, i.e.,  $J^i(\underline{k}) = J^{ii}(\underline{k})$ . With these equalities, we find (in terms of the z components of the B sublattice spins)

$$\begin{aligned} \Theta_A(0) &= -\Theta_B(0), \\ \tilde{\epsilon}_A(\underline{k}) &= \tilde{\epsilon}_B(\underline{k}), \end{aligned} \quad (3.37a)$$

and 
$$E_{\pm} = \mu_B H \pm \frac{1}{2} \left[ A^2 + 4n \right]^{1/2}, \quad (3.37b)$$

where 
$$A = \langle B^z \rangle \left[ J(0) + \tilde{\epsilon}_B(\underline{k}) \right], \quad (3.37c)$$

$$\begin{aligned} n &= -\langle B^z \rangle^2 \left[ J(\underline{k}) - \infty \frac{1}{N} \sum_{\underline{k}'} J(\underline{k}-\underline{k}') \langle A^- B^+ \rangle_{\underline{k}'} \right] \\ &\quad \times \left[ J(\underline{k}) - \infty \frac{1}{N} \sum_{\underline{k}'} J(\underline{k}-\underline{k}') \langle A^- B^+ \rangle_{\underline{k}'}^* \right]. \end{aligned} \quad (3.37d)$$

In terms of the z component of the A sublattice spins, we get (in place of eqs. (3.37b), (3.37c) and (3.37d))

$$E_{\pm} = g_{B}^{\mu} H_{\pm} \mp \frac{1}{2} \left[ \Lambda^2 + 4n \right]^{\frac{1}{2}}, \quad (3.37b')$$

$$\Lambda = - \langle \Lambda^z \rangle \left[ J(0) + \tilde{\epsilon}_{\Lambda}(\underline{k}) \right], \quad (3.37c')$$

$$n = - \langle \Lambda^z \rangle^2 \left[ J(\underline{k}) - \alpha \frac{1}{N} \sum_{\underline{k}'} J(\underline{k}-\underline{k}') \langle \Lambda^z B^+ \rangle_{\underline{k}'} \right] \\ \times \left[ J(\underline{k}) - \alpha \frac{1}{N} \sum_{\underline{k}'} J(\underline{k}-\underline{k}') \langle \Lambda^z B^+ \rangle_{\underline{k}'}^* \right] \quad (3.37d')$$

Substituting eqs. (3.37b), (3.37c) and (3.37d) into eq. (3.31a) and eqs. (3.37b'), (3.37c') and (3.37d') into eq. (3.31d), we find that the Green's function for the A sublattice,  $\langle \langle \Lambda_{\underline{k}}^+; e^{a\Lambda^z} \Lambda_{\underline{k}'}^- \rangle \rangle_E$  is the Green's function for the B sublattice,  $\langle \langle B_{\underline{k}}^+; e^{bB^z} B_{\underline{k}'}^- \rangle \rangle_E$ . If we set  $\alpha_{\Lambda}^{\pm} = \alpha_{B}^{\pm} = \alpha = 0$  and recover Tyablikov's decoupling scheme, our results for  $\langle \langle \Lambda_{\underline{k}}^+; \Lambda_{\underline{k}'}^- \rangle \rangle_E$  and  $\langle \langle B_{\underline{k}}^+; B_{\underline{k}'}^- \rangle \rangle_E$  are similar to those obtained by Barry<sup>36</sup>, except that Barry keeps the anisotropic field term which we dropped.

Our results<sup>41</sup> would give a more complicated spin wave

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<sup>41</sup>In order to obtain the energy spectrum, a set of simultaneous nonlinear equations would have to be solved. See section 4.1.

energy spectrum than that obtained by Yablonskii. The reason that Yablonskii was able to obtain a clear cut expression for the energy spectrum can be traced to the decoupling procedure used. While Yablonskii's scheme yields clear cut results, there appears to be no physical justification for the scheme he used. One must decide which is preferable, make a mathematical approximation which is physically unjustifiable but which yields a mathematically clear answer or make an approximation which is physically justified but which leads to a mathematical dead end.

