CHAPTER 5

CONCLUSIONS AND DISCUSSIONS

5.1 Some Qualitative Conclusions

It is possible to arrive at some qualitative conclusions without having to obtain explicit forms of the correlation functions defined by the set of nonlinear equations (4.3a)-(4.3d). Take for instance, the effects of the correlation functions on the transition temperatures at which the sublattice magnetization vanishes can be discussed. The plural temperature term is used since the temperature at which the spin alignments on one sublattice disappears does not have to be the same as the temperature at which the spin alignments on the other disappears. The reason why the two temperatures need not be the same is that we have included into our Hamiltonian, eq. (3.1), terms allowing for intralattice interactions between A ion spins and between B ion spins. It is therefore possible for the thermal agitation of the spins to overcome the interlattice interaction energy and the intralattice exchange energy of only one of the sublattices, leaving the spins on the other sublattice still partially aligned.

If we label the temperatures at which the spin alignments on the two sublattices disappear, T_A and T_B, we have from eqs. (4.19a) and (4.19b) as follows

$$\lim_{\langle A^z\rangle \longrightarrow 0} k_B T_A \frac{\langle A^z\rangle}{N_A} \sum_{\underline{k}} \frac{1}{2E_+E_-}\left[(E_- - E_+)\frac{X}{\sqrt{X^2+4n}} + (E_+ + E_-)\right]$$

$$= \frac{1}{3} A(A+1), \tag{5.1}$$

$$\lim_{\langle B^z\rangle \longrightarrow 0} k_B T_B \frac{\langle B^z\rangle}{N_B} \sum_{\underline{k}} \frac{1}{2E_+E_-}\left[(E_+ - E_-)\frac{X}{\sqrt{X^2+4n}} + (E_+ + E_-)\right]$$

$$= \frac{1}{3} B(B+1), \tag{5.2}$$

where A and B are the spin quantum numbers of the magnetic ions on the two sublattices and where $E_\pm$, X and n are given by eqs. (3.34), (3.33) and (3.32), respectively. The symbol X is used in place of the energy term A of eq. (3.33) in order to avoid the possible confusion between the spin quantum number A and the energy term A. Substituting the definitions of $E_\pm$, X and n into eqs. (5.1) and (5.2), we get

$$\lim_{\langle A^z\rangle \longrightarrow 0} k_B T_A \frac{\langle A^z\rangle}{N_A} \sum_{\underline{k}} \left(\left[\left(\frac{N_A}{N_B}\right)^{1/2} J(0)\langle A^z\rangle - \hat{\epsilon}_B(\underline{k})\langle B^z\rangle \right] \right.$$

$$\times \left[\left(\left(\frac{N_A}{N_B}\right)^{1/2} J(0)\langle A^z\rangle - \tilde{\epsilon}_B(\underline{k})\langle B^z\rangle \right) \left(\left(\frac{N_B}{N_A}\right)^{1/2} J(0)\langle B^z\rangle - \tilde{\epsilon}_A(\underline{k})\langle A^z\rangle \right) \right.$$

$$- \left(J(\underline{k})\langle B^z\rangle + \alpha_A\langle A^z\rangle \frac{1}{\sqrt{N_A N_B}} \sum_{\underline{k}'} J(\underline{k}-\underline{k}')\langle A^- B^+\rangle_{\underline{k}'} \right)$$

$$\left. \left. \times \left(J(\underline{k})\langle A^z\rangle + \alpha_B\langle B^z\rangle \frac{1}{\sqrt{N_A N_B}} \sum_{\underline{k}'} J(\underline{k}-\underline{k}')\langle B^- A^+\rangle_{\underline{k}'} \right) \right]^{-1} \right) = \text{(cont.)}$$

$$= \frac{1}{3} A(A+1) \tag{5.3}$$

and

$$\lim_{\langle B^z\rangle \longrightarrow 0} k_B T_B \frac{\langle B^z\rangle}{N_B} \sum_{\underline{k}} \left(\left[\left(\frac{N_B}{N_A}\right)^{1/2} J(0)\langle B^z\rangle - \tilde{\epsilon}_A(\underline{k})\langle A^z\rangle \right] \right.$$

$$\times \left[\left(\frac{N_A}{N_B}\right)^{1/2} J(0)\langle A^z\rangle - \tilde{\epsilon}_B(\underline{k})\langle B^z\rangle \right] \left[\left(\frac{N_B}{N_A}\right)^{1/2} J(0)\langle B^z\rangle - \tilde{\epsilon}_A(\underline{k})\langle A^z\rangle \right]$$

$$- \left(J(\underline{k})\langle B^z\rangle + \alpha_A \langle A^z\rangle \frac{1}{\sqrt{N_A N_B}} \sum_{\underline{k}'} J(\underline{k}-\underline{k}')\langle A^- B^+\rangle_{\underline{k}'} \right)$$

$$\left. \times \left(J(\underline{k})\langle A^z\rangle + \alpha_B \langle B^z\rangle \frac{1}{\sqrt{N_A N_B}} \sum_{\underline{k}'} J(\underline{k}-\underline{k}')\langle B^- A^+\rangle_{\underline{k}'} \right) \right]^{-1} \right)$$

$$= \frac{1}{3} B(B+1), \tag{5.4}$$

where $\tilde{\epsilon}_A(a)$ and $\tilde{\epsilon}_B(b)$ are defined by eqs. (3.35a) and (3.35b):

$$\tilde{\epsilon}_A(\underline{k}) = 2\left[J'(\underline{k}') - J'(0) + \alpha_A' \frac{1}{N_A} \sum_{\underline{k}'} (J'(\underline{k}-\underline{k}') - J'(\underline{k}'))\langle A^- A^+\rangle_{\underline{k}'} \right]$$

$$- \alpha_A' \frac{1}{N_A} \sum_{\underline{k}'} J(\underline{k}')\langle A^- B^+\rangle_{\underline{k}'} .$$

$$\tilde{\epsilon}_B(\underline{k}) = 2\left[J''(\underline{k}') - J''(\ ) + \alpha_B' \frac{1}{N_B} \sum_{\underline{k}'} (J''(\underline{k}-\underline{k}') - J''(\underline{k}'))\langle B^- B^+\rangle_{\underline{k}'} \right]$$

$$- \alpha_B' \frac{1}{N_B} \sum_{\underline{k}'} J(\underline{k}')\langle B^- A^+\rangle_{\underline{k}'} .$$

If we set $\alpha_A' = \alpha_B' = 0$, the above two definitions become

$$\tilde{\epsilon}_A(\underline{k}) = 2(J'(\underline{k})-J'(0)), \qquad (5.5a)$$

$$\tilde{\epsilon}_B(\underline{k}) = 2(J''(\underline{k})-J''(0)). \qquad (5.5b)$$

Therefore in the case of $\alpha'_A = \alpha'_B = 0$, i.e., the results if Tyablikov decoupling is used, the equations which determine the transition temperatures become

$$\lim_{\langle A^z\rangle \to 0} k_B T_A \frac{\langle A^z\rangle}{N_A} \sum_{\underline{k}} \left[\left(\langle A^z\rangle \left(\frac{N_A}{N_B}\right)^{1/2} J(0) - \langle B^z\rangle 2(J''(\underline{k})-J''(0)) \right) \right.$$
$$\times \left(\langle A^z\rangle \left(\frac{N_A}{N_B}\right)^{1/2} J(0) - 2\langle B^z\rangle (J''(\underline{k})-J''(0)) \right)$$
$$\times \left(\langle B^z\rangle \left(\frac{N_B}{N_A}\right)^{1/2} J(0) - 2\langle A^z\rangle (J'(\underline{k})-J'(0)) \right)$$
$$\left. - \langle A^z\rangle\langle B^z\rangle J^2(\underline{k}) \right]^{-1} = \frac{1}{3} A(A+1), \qquad (5.6)$$

$$\lim_{\langle B^z\rangle \to 0} k_B T_B \frac{\langle B^z\rangle}{N_B} \sum_{\underline{k}} \left[\left(\langle B^z\rangle \left(\frac{N_B}{N_A}\right)^{1/2} J(0) - \langle A^z\rangle 2(J'(\underline{k})-J'(0)) \right) \right.$$
$$\times \left(\langle A^z\rangle \left(\frac{N_A}{N_B}\right)^{1/2} J(0) - 2\langle B^z\rangle (J''(\underline{k})-J''(0)) \right)$$
$$\times \left(\langle B^z\rangle \left(\frac{N_B}{N_A}\right)^{1/2} J(0) - 2\langle A^z\rangle (J'(\underline{k})-J'(0)) \right)$$
$$\left. - \langle A^z\rangle\langle B^z\rangle J^2(\underline{k}) \right]^{-1} = \frac{1}{3} B(B+1). \qquad (5.7)$$

If we make the substitutions, $A = S_1$, $B = S_2$, $\langle A^z \rangle = \sigma_1$, $\langle B^z \rangle = \sigma_2$, $J(\underline{k}) = J_3(\underline{k})$, $2J'(\underline{k}) = J_1(\underline{k})$, $2J''(\underline{k}) = J_2(\underline{k})$ and $N_A = N_B$, we would see that our results (eqs. (5.6) and (5.7)) are exactly the results obtained by Yablonskii[40].

The equations for determining the transition temperatures for $\alpha_A' \neq 0$ and $\alpha_B' \neq 0$, i.e., eqs. (5.3) and (5.4) can also be obtained from Yablonskii's equations if the following substitutions are made

$$J_3(\underline{k})\langle A^z \rangle \longrightarrow J(\underline{k})\langle A^z \rangle + \alpha_B' \langle B^z \rangle \frac{1}{\sqrt{N_A N_B}} \sum_{\underline{k}'} J(\underline{k}-\underline{k}')\langle B^- A^+ \rangle_{\underline{k}'}, \tag{5.8}$$

$$J_3(\underline{k})\langle B^z \rangle \longrightarrow J(\underline{k})\langle B^z \rangle + \alpha_A' \langle A^z \rangle \frac{1}{\sqrt{N_A N_B}} \sum_{\underline{k}'} J(\underline{k}-\underline{k}')\langle A^- B^+ \rangle_{\underline{k}'}, \tag{5.9}$$

$$J_1(\underline{k}) \longrightarrow J'(\underline{k}) + \alpha_A' \frac{1}{N_A} \sum_{\underline{k}'} J''(\underline{k}-\underline{k}')\langle A^- A^+ \rangle_{\underline{k}'}, \tag{5.10}$$

$$J_2(\underline{k}) \longrightarrow J''(\underline{k}) + \alpha_B' \frac{1}{N_B} \sum_{\underline{k}'} J'(\underline{k}-\underline{k}')\langle A^- A^+ \rangle_{\underline{k}'} \tag{5.11}$$

and the different in the number of sites in the two sublattices are taken into account. It therefore appears that the effect of Callen decoupling is to renormalize (or correct) the exchange interaction constants. The amount of correction depends on the spin quantum numbers since α_A' and α_B' are directly related

to the spin quantum numbers, i.e., $\alpha_S' = \frac{1}{2S^2}$, while the correlation functions are indirectly related to them.

If all the interactions are antiferromagnetic, the transition temperatures determined by eqs. (5.3) and (5.4) would in general be higher than those predicted by eqs. (5.6) and (5.7). Since α_S' becomes smaller as S increases, we would expect the difference between the two sets of transition temperatures to decrease.

5.2 Possible Future Work

The results obtained in this work are very complicated and do not give a clear description of the spin wave behaviors such as the energy spectrum, resonance susceptibility, magnetization and transition temperatures.

The trouble which, unfortunately, is extremely difficult to work out at present is due to two main causes. The first one is the correlation functions which play an important role in the determination of those behaviors mentioned above. These correlation functions appear in a set of nonlinear simultaneous equations (eqs. (4.3a)-(4.3d)) which can only be solved numerically. The other difficulty to obtain mathematically clear results is due to the lack of the evaluation of the summation over the wave vector $\underline{k}$ of the exchange integrals of the system. Some evalutions of those quantities have been made only for the simpler

structures such as, the simple cubic lattice, face-centered cubic lattice and body-centered cubic lattice whereas for the much more complicated structure of the spinel type, such an evaluation of those quantities has never been made at all.

Consequently, it should be noted that eventhough the present work does not give quantitative results, it predicts valuable qualitative ones which make it be possible to obtain complete results, both qualitative and quantitative, when the two problems are solved. However, this schedul needs about a year or more to be finished.

APPENDIX A

THE HEISENBERG HAMILTONIAN

Consider a system of two electrons subject to fields derived from similar potential functions, the Hamiltonian for the pairs is

$$\mathcal{H} = -\frac{\hbar^2}{2m}\nabla_1^2 - \frac{\hbar^2}{2m}\nabla_2^2 + V(r_1) + V(r_2) + \frac{e^2}{r_{12}} = \mathcal{H}_o + \mathcal{H}_{12}, \quad (1)$$

where the subscripts 1 and 2 refer to the two electrons, considered distinguishable and r_{12} is the separation of the two electrons. Neglecting the interaction between the two electrons, we get a wave equation of the form

$$\mathcal{H}_o = E_o \Psi, \quad (2)$$

which can be separated into two independent wave equations involving each electron. The possible solutions are

$$\Psi = \psi_i(1)\,\psi_j(2)$$

and

$$\Psi = \psi_i(2)\,\psi_j(1) \quad (3)$$

with $E_o = E_i + E_j$ for both cases.

However, the electrons are indistinguishable, so as a consequence the acceptable solutions are the linear combination of the wave functions of eq. (3) such as

$$\psi_S(1,2) = \frac{1}{\sqrt{2}}\left[\psi_i(1)\,\psi_j(2) + \psi_i(2)\,\psi_j(1)\right], \tag{4}$$

$$\psi_T(1,2) = \frac{1}{\sqrt{2}}\left[\psi_i(1)\,\psi_j(2) - \psi_i(2)\,\psi_j(1)\right], \tag{5}$$

where $\psi_S(1,2)$ is called symmetrical and $\psi_T(1,2)$ is called antisymmetrical. If we apply first order perturbation theory to calculate the effect of the interaction $\mathcal{H}_{12}$, we find

$$E = E_o + \int \psi_i^*(1)\,\psi_j^*(2)\,\mathcal{H}_{12}\,\psi_i(1)\,\psi_j(2)\; d\tau_1 d\tau_2 \tag{6}$$

$$= E_o + C_{ij}, \tag{7}$$

where $$C_{ij} = \int \psi_i^*(1)\,\psi_j^*(2)\,\mathcal{H}_{12}\,\psi_i(1)\,\psi_j(2)\; d\tau_1 d\tau_2 \tag{8}$$

is physically interpreted as the average Coulomb interaction of the two electrons in states i and j.

However, the electron wave functions are not only functions of spatial coordinates but also of the spin. Taking into consideration the Pauli exclusion principle, which requires that the total wave functions be antisymmetric with respect to the

exchange of space and spin coordinates of the two electrons, the appropriate total wave functions are then

$$\Psi_I = \psi_S(1,2)\Phi_T(1,2), \tag{9}$$

$$\Psi_{II} = \psi_T(1,2)\Phi_S(1,2), \tag{10}$$

where $\psi_S(1,2)$ and $\psi_T(1,2)$ are symmetrical and antisymmetrical wave functions which involve only the spatial coordinates, respectively, whereas $\Phi_T(1,2)$ and $\Phi_S(1,2)$ are antisymmetrical and symmetrical spin wave functions, respectively. These wave functions, are, respectively,

$$\Psi_I = \frac{1}{\sqrt{2}}\left[\psi_i(1)\psi_j(2) + \psi_i(2)\psi_j(1)\right]\Phi_o, \tag{11}$$

$$\Psi_{II} = \frac{1}{\sqrt{2}}\left[\psi_i(1)\psi_j(2) - \psi_i(2)\psi_j(1)\right]\Phi_1, \tag{12}$$

where Φ_o represents the spin function in which the electron spins are antiparallel ($S = 0$, singlet state), whereas Φ_1 represents the spin function in which the spins are parallel ($S = 1$, triplet states). If we recalculate the first order perturbation contribution to the energy, we find that

$$E_I = E_o + C_{ij} + J_{ij}, \tag{13}$$

$$E_{II} = E_o + C_{ij} - J_{ij}, \tag{14}$$

where $$J_{ij} = \int \psi_i^*(1)\psi_j^*(2)\mathcal{H}_{12}\psi_i(2)\psi_j(1)\, d\tau_1 d\tau_2 \tag{15}$$

is the exchange energy of the two electrons in states i and j.

It is useful to express the energies of the two states (eq. (11) and eq. (12)) in terms of the electron spins. The two electrons may be considered to be coupled via their spins and this coupling must be proportional to the eigenvalues of the scalar product $\underline{s}_i \cdot \underline{s}_j$. If we write

$$\mathcal{H}_{12} = C_{ij} - \frac{1}{2} J_{ij}(1+4\underline{s}_i \cdot \underline{s}_j) \tag{16}$$

as the Hamiltonian for the spins $\underline{s}_i$ and $\underline{s}_j$ of the electrons in states i and j, then the eigenvalue of $\mathcal{H} = \mathcal{H}_o + \mathcal{H}_{12}$ of the two states are

$$E = E_o + C_{ij} - \frac{1}{2} J_{ij} - 2 J_{ij}\, \underline{s}_i \cdot \underline{s}_j \tag{17}$$

which coincide with eq. (13) and eq. (14). For, consider the identity

$$\underline{s}_i \cdot \underline{s}_j = \frac{1}{2}\left[(\underline{s}_i + \underline{s}_j)^2 - \underline{s}_i^2 - \underline{s}_j^2\right] = \frac{1}{2}\left[S^2 - \frac{3}{2}\right] \tag{18}$$

which shows that the eigenvalue of $\underline{s}_i \cdot \underline{s}_j$ is $-3/4$ for the singlet state ($S = 0$, spins antiparallel), and $1/4$ for the triplet states ($S = 1$, spins parallel). In this sense, the exchange force is equivalent to an interaction between the spins of electrons in states i and j.

In problems concerning ferromagnetism usually only the spin dependent term of eq. (17) is of interest. If it is a good approximation to assume that the electrons carrying magnetism in a crystal have localized states on the atom, then it is resonable to think that their exchange effect can be express as

$$\mathcal{H}_H = -2 \sum_{i,j} J_{ij} \underline{S}_i \cdot \underline{S}_j \,. \qquad (19)$$

This is precisely what Heisenberg assumed in his theory of ferromagnetism and is called the Heisenberg Hamiltonian which we have also used to describe our system under investigation.

APPENDIX B

THE GREEN'S FUNCTION

The definition and some properties of the Green's function used to study magnetism are given in this appendix.

The double-time retarded temperature dependent Green's function is defined by the relationship:

$$\langle\langle A(t);B(t')\rangle\rangle = -i\theta(t-t')\left\langle\left[A(t),B(t')\right]_{\eta}\right\rangle , \qquad (1)$$

where $$A(t) = e^{i\mathcal{H}t} A(0) e^{-i\mathcal{H}t}$$

and $$B(t') = e^{i\mathcal{H}t'} B(0) e^{-i\mathcal{H}t'} \qquad (2)$$

are the Heisenberg operators satisfy the following relations

$$i\hbar \frac{d}{dt} A(t) = \left[A(t), \mathcal{H}\right] = A(t)\mathcal{H} - \mathcal{H}A(t),$$

$$i\hbar \frac{d}{dt} B(t') = \left[B(t'), \mathcal{H}\right] = B(t')\mathcal{H} - \mathcal{H}B(t'), \qquad (3)$$

and $$\left[A(t),B(t')\right]_{\eta} = A(t)B(t') - \eta B(t')A(t), \qquad (4)$$

where $\eta = 1$ if $A(t)$ and $B(t')$ are represented in terms of the Bose operators while $\eta = -1$ if they are represented in terms

of the Fermi operators.

The symbol $\langle \ldots \rangle$ denotes, as usual, a statistical averaging and the use of the Hamiltonian .

$$\theta(t-t') = \begin{cases} 1, & t > t' \\ 0, & t < t' \end{cases} \tag{5}$$

is a step function.

Using eq. (5) and since the time derivative of the step function is a delta function $\delta(t-t')$, we obtain the equation of motion for the Green's function $\langle\langle A(t);B(t')\rangle\rangle$ as

$$i\hbar \frac{d}{dt}\langle\langle A(t);B(t')\rangle\rangle = \delta(t-t')\langle [A(t),B(t')]_{\eta}\rangle + \langle\langle [A(t),\mathcal{H}];B(t')\rangle\rangle . \tag{6}$$

The above equation is called the Green's differential equation, and as a consequence the function $\langle\langle A(t);B(t')\rangle\rangle$ is called the Green's function.

Introducing the Fourier transform of the Green's function as

$$\langle\langle A;B\rangle\rangle_E = \frac{1}{2\pi}\int_{-\infty}^{\infty} \langle\langle A(t);B(t')\rangle\rangle\, e^{i\omega(t-t')}\, d(t-t') \tag{7}$$

with the inverse transform

$$\langle\langle A(t);B(t')\rangle\rangle = \int_{-\infty}^{\infty} \langle\langle A;B\rangle\rangle_E\, e^{-iE(t-t')}\, dE \tag{8}$$

then the Green's differential equation (eq. (6)) becomes

$$E\langle\langle A;B\rangle\rangle_E = \frac{1}{2\pi}\langle[A,B]_\eta\rangle + \langle\langle[A,\mathcal{H}];B\rangle\rangle_E. \qquad (9)$$

In our problems under investigation of spin waves, the double-time retarded temperature dependent Green's function is used with $A(t)$ and $B(t')$ are represented in terms of Boson particles owing to the fact that spin waves or magnons may be thought of as being Boson particles. Hence, eq. (9) as applied to our case becomes

$$E\langle\langle A;B\rangle\rangle_E = \frac{1}{2\pi}\langle[A,B]\rangle + \langle\langle[A,\mathcal{H}];B\rangle\rangle_E \qquad (10)$$

where $[A,B] = AB - BA$.

Equation (10) is the equation of motion we have used in our problems.

Since the right hand side of eq. (10) contains higher order Green's functions, one obtains a hierarchy for the equation of motion which, in practice, must be truncated by introducing some approximations in order to close the system of equations.

Finally, it should be noted that since the Green's function $\langle\langle A;B\rangle\rangle$ depends linearly on each of the operator arguments, A or B, therefore

$$\langle\langle(\alpha_1 A_1+\alpha_2 A_2;B)\rangle\rangle = \alpha_1\langle\langle A_1;B\rangle\rangle + \alpha_2\langle\langle A_2;B\rangle\rangle. \qquad (11)$$

This property of the Green's function is used to obtain eqs. (3.17a)-(3.17d).

APPENDIX C

CALCULATIONS OF THE EXCHANGE INTEGRALS IN THE NORMAL SPINEL STRUCTURE

In this appendix, the exchange integrals $J(\underline{k})$, $J'(\underline{k})$ and $J''(\underline{k})$ as defined in section 3.2.2 are calculated referring to the positions of ions in the normal spinel structure as described in section 3.1.

1. Calculation of $J(\underline{k})$

According to eq. (3.6a), we have

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

or we can write

$$J(\underline{k}) = \frac{1}{\sqrt{N_A N_B}} \sum_i J_i(\underline{k}), \qquad (2)$$

where

$$J_i(\underline{k}) = \sum_j J_{ij} \, e^{i\underline{k}\cdot(\underline{r}_j - \underline{r}_i)}. \qquad (3)$$

The reason for making this representation is that not all the sites in a sublattice are equivalent. While all the sites within one sublattice are similarly coordinated, the surrounding sites do not form the same space group. If we consider the group of B sites surrounding an A site, we will find that the set of B sites forms a space group which is the inversion of the space group formed the set of B sites surrounding some of the other A sites. This property will be demonstrated in the following paragraph.

Assuming nearest neighbour approximation with the isotropic exchange integral J, i.e., $J_{ij} = J$ for nearest neighbour interactions and $J_{ij} = 0$, otherwise, eq. (3) can be put into the form

$$J_i(\underline{k}) = J \sum_{j=1}^{z} e^{i\underline{k}\cdot(\underline{r}_j - \underline{r}_i)}, \qquad (4)$$

where z is the number of nearest neighbours in the B sublattice of the ith atom in the A sublattice.

Consider the ion in the A sublattice located at (0,0,0), the twelve nearest neighbours in the B sublattice of this ion are at the following positions (see Fig. 6 (a)):

$\left(\frac{3}{8},\frac{1}{8},\frac{1}{8}\right)a$ $\qquad$ $\left(-\frac{1}{8},\frac{1}{8},-\frac{3}{8}\right)a$

$\left(\frac{1}{8},\frac{3}{8},\frac{1}{8}\right)a$ $\qquad$ $\left(-\frac{1}{8},\frac{3}{8},-\frac{1}{8}\right)a$

(continued)

$$\left(\frac{1}{8},\frac{1}{8},\frac{3}{8}\right)a \qquad \left(-\frac{3}{8},\frac{1}{8},-\frac{1}{8}\right)a$$

$$\left(-\frac{3}{8},-\frac{1}{8},\frac{1}{8}\right)a \qquad \left(\frac{1}{8},-\frac{1}{8},-\frac{3}{8}\right)a$$

$$\left(-\frac{1}{8},-\frac{3}{8},\frac{1}{8}\right)a \qquad \left(\frac{1}{8},-\frac{3}{8},-\frac{1}{8}\right)a$$

$$\left(-\frac{1}{8},-\frac{1}{8},\frac{3}{8}\right)a \qquad \left(\frac{3}{8},-\frac{1}{8},-\frac{1}{8}\right)a, \qquad (5)$$

where a is the lattice constant.

Taking $\underset{\sim}{r}_1 = (0,0,0)a$ and $\underset{\sim}{r}_j$ as defined in eq. (5), we get

$$J_1(\underset{\sim}{k}) = J\Big[e^{i\underset{\sim}{k}\cdot\left(\frac{3}{8},\frac{1}{8},\frac{1}{8}\right)a} + e^{i\underset{\sim}{k}\cdot\left(\frac{1}{8},\frac{3}{8},\frac{1}{8}\right)a} + e^{i\underset{\sim}{k}\cdot\left(\frac{1}{8},\frac{1}{8},\frac{3}{8}\right)a}$$

$$+ e^{i\underset{\sim}{k}\cdot\left(-\frac{3}{8},-\frac{1}{8},\frac{1}{8}\right)a} + e^{i\underset{\sim}{k}\cdot\left(-\frac{1}{8},-\frac{3}{8},\frac{1}{8}\right)a} + e^{i\underset{\sim}{k}\cdot\left(-\frac{1}{8},-\frac{1}{8},\frac{3}{8}\right)a}$$

$$+ e^{i\underset{\sim}{k}\cdot\left(-\frac{1}{8},\frac{1}{8},-\frac{3}{8}\right)a} + e^{i\underset{\sim}{k}\cdot\left(-\frac{1}{8},\frac{3}{8},-\frac{1}{8}\right)a} + e^{i\underset{\sim}{k}\cdot\left(-\frac{3}{8},\frac{1}{8},-\frac{1}{8}\right)a}$$

$$+ e^{i\underset{\sim}{k}\cdot\left(\frac{1}{8},-\frac{1}{8},-\frac{3}{8}\right)a} + e^{i\underset{\sim}{k}\cdot\left(\frac{1}{8},-\frac{3}{8},-\frac{1}{8}\right)a} + e^{i\underset{\sim}{k}\cdot\left(\frac{3}{8},-\frac{1}{8},-\frac{1}{8}\right)a}\Big]. \qquad (6)$$

The A ion at $\left(\frac{1}{4},-\frac{1}{4},\frac{1}{4}\right)a$ has twelve nearest neighbour B ions located at

$$\left(\frac{1}{8},\frac{1}{8},\frac{3}{8}\right)a \qquad \left(-\frac{1}{8},-\frac{1}{8},\frac{3}{8}\right)a \qquad \left(\frac{1}{8},-\frac{1}{8},\frac{5}{8}\right)a$$

$$\left(\frac{3}{8},\frac{1}{8},\frac{1}{8}\right)a \qquad \left(-\frac{1}{8},-\frac{3}{8},\frac{1}{8}\right)a \qquad \left(\frac{3}{8},-\frac{1}{8},\frac{5}{8}\right)a$$

(continued)

$$\left(\frac{1}{8},-\frac{5}{8},\frac{1}{8}\right)a \qquad \left(\frac{5}{8},-\frac{1}{8},\frac{1}{8}\right)a \qquad \left(\frac{1}{8},-\frac{3}{8},-\frac{1}{8}\right)a$$

$$\left(\frac{3}{8},-\frac{5}{8},\frac{3}{8}\right)a \qquad \left(\frac{5}{8},-\frac{3}{8},\frac{3}{8}\right)a \qquad \left(\frac{3}{8},-\frac{1}{8},-\frac{1}{8}\right)a. \qquad (7)$$

Taking $\underline{r}_2 = \left(\frac{1}{4},-\frac{1}{4},\frac{1}{4}\right)a$ and $\underline{r}_j$ as defined in eq. (7), we get the corresponding distances $(\underline{r}_j-\underline{r}_2)$ as follows:

$$\left(-\frac{1}{8},\frac{3}{8},\frac{1}{8}\right)a \qquad \left(-\frac{3}{8},\frac{1}{8},\frac{1}{8}\right)a \qquad \left(-\frac{1}{8},\frac{1}{8},\frac{3}{8}\right)a$$

$$\left(\frac{1}{8},\frac{3}{8},-\frac{1}{8}\right)a \qquad \left(-\frac{3}{8},-\frac{1}{8},-\frac{1}{8}\right)a \qquad \left(\frac{1}{8},-\frac{1}{8},\frac{3}{8}\right)a$$

$$\left(-\frac{1}{8},-\frac{3}{8},-\frac{1}{8}\right)a \qquad \left(\frac{3}{8},\frac{1}{8},-\frac{1}{8}\right)a \qquad \left(-\frac{1}{8},-\frac{1}{8},-\frac{3}{8}\right)a$$

$$\left(\frac{1}{8},-\frac{3}{8},\frac{1}{8}\right)a \qquad \left(\frac{3}{8},-\frac{1}{8},\frac{1}{8}\right)a \qquad \left(\frac{1}{8},\frac{1}{8},-\frac{3}{8}\right)a. \qquad (8)$$

These distances are the negative of the distances given by the set of distances of eq. (5). The set of eq. (7) of B lattice sites forms a reflection of the group formed by the set of eq. (5). Hence, we obtain

$$J_2(\underline{k}) = J\left[e^{i\underline{k}\cdot\left(\frac{1}{8},\frac{3}{8},\frac{1}{8}\right)a} + e^{i\underline{k}\cdot\left(\frac{1}{8},\frac{3}{8},-\frac{1}{8}\right)a} + e^{i\underline{k}\cdot\left(-\frac{1}{8},-\frac{3}{8},-\frac{1}{8}\right)a}\right.$$

$$+ e^{i\underline{k}\cdot\left(\frac{1}{8},-\frac{3}{8},\frac{1}{8}\right)a} + e^{i\underline{k}\cdot\left(-\frac{3}{8},\frac{1}{8},\frac{1}{8}\right)a} + e^{i\underline{k}\cdot\left(-\frac{3}{8},-\frac{1}{8},-\frac{1}{8}\right)a}$$

$$+ e^{i\underline{k}\cdot\left(\frac{3}{8},\frac{1}{8},-\frac{1}{8}\right)a} + e^{i\underline{k}\cdot\left(\frac{3}{8},-\frac{1}{8},\frac{1}{8}\right)a} + e^{i\underline{k}\cdot\left(-\frac{1}{8},\frac{1}{8},\frac{3}{8}\right)a}$$

$$\left. + e^{i\underline{k}\cdot\left(\frac{1}{8},-\frac{1}{8},\frac{3}{8}\right)a} + e^{i\underline{k}\cdot\left(-\frac{1}{8},-\frac{1}{8},-\frac{3}{8}\right)a} + e^{i\underline{k}\cdot\left(\frac{1}{8},\frac{1}{8},-\frac{3}{8}\right)a}\right].$$

(9)

Adding eqs. (6) and (9) gives

$$J_1(\underline{k})+J_2(\underline{k}) = J\Big[2\cos \underline{k}\cdot\Big(\frac{3}{8},\frac{1}{8},\frac{1}{8}\Big)a + 2\cos \underline{k}\cdot\Big(\frac{1}{8},\frac{3}{8},\frac{1}{8}\Big)a$$
$$+ 2\cos \underline{k}\cdot\Big(\frac{1}{8},\frac{1}{8},\frac{3}{8}\Big)a + 2\cos \underline{k}\cdot\Big(-\frac{3}{8},-\frac{1}{8},\frac{1}{8}\Big)a$$
$$+ 2\cos \underline{k}\cdot\Big(-\frac{1}{8},-\frac{3}{8},\frac{1}{8}\Big)a + 2\cos \underline{k}\cdot\Big(-\frac{1}{8},-\frac{1}{8},\frac{3}{8}\Big)a$$
$$+ 2\cos \underline{k}\cdot\Big(-\frac{1}{8},\frac{1}{8},-\frac{3}{8}\Big)a + 2\cos \underline{k}\cdot\Big(-\frac{1}{8},\frac{3}{8},-\frac{1}{8}\Big)a$$
$$+ 2\cos \underline{k}\cdot\Big(-\frac{3}{8},\frac{1}{8},-\frac{1}{8}\Big)a + 2\cos \underline{k}\cdot\Big(\frac{1}{8},-\frac{1}{8},-\frac{3}{8}\Big)a$$
$$+ 2\cos \underline{k}\cdot\Big(\frac{1}{8},-\frac{3}{8},-\frac{1}{8}\Big)a + 2\cos \underline{k}\cdot\Big(\frac{3}{8},-\frac{1}{8},-\frac{1}{8}\Big)a\Big], \quad (10)$$

where we have used the identity $\cos x = \frac{e^{ix}+e^{-ix}}{2}$.

Repeating the same steps of derivation, we get

$$J_3(\underline{k}) = J_4(\underline{k}) = J_5(\underline{k}) = J_2(\underline{k}), \quad (11)$$

where we take $\underline{r}_3 = \Big(-\frac{1}{4},\frac{1}{4},\frac{1}{4}\Big)a$, $\underline{r}_4 = \Big(\frac{1}{4},\frac{1}{4},-\frac{1}{4}\Big)a$ and $\underline{r}_5 = \Big(-\frac{1}{4},-\frac{1}{4},-\frac{1}{4}\Big)a$.

Also we get

$$J_6(\underline{k}) = J_7(\underline{k}) = J_8(\underline{k}) = \cdots\cdots\cdots = J_{17}(\underline{k}) = \frac{1}{4} J_1(\underline{k}), \quad (12)$$

where $\underline{r}_6$, $\underline{r}_7$, $\underline{r}_8$, ..., $\underline{r}_{17}$ are the rest of the A sites, each of which is occupied by 1/4 ion.

Therefore we obtain

$$\sum_i J_i(\underline{k}) = J_1(\underline{k})+J_2(\underline{k})+J_3(\underline{k})+\cdots\cdots\cdots+J_{17}(\underline{k})$$

$$= J_1(\underline{k})+J_2(\underline{k}) + 3\,J_2(\underline{k}) + \frac{12}{4}\,J_1(\underline{k})$$

$$= J_1(\underline{k})+J_2(\underline{k}) + 3(J_1(\underline{k})+J_2(\underline{k}))$$

$$= 4(J_1(\underline{k})+J_2(\underline{k})). \qquad (13)$$

Substituting eq. (10) into eq. (13) gives

$$\begin{aligned}\sum_i J_i(\underline{k}) = 4\times 2J\Big[&\cos\Big(\tfrac{3}{8}k_x+\tfrac{1}{8}k_y+\tfrac{1}{8}k_z\Big)a + \cos\Big(\tfrac{1}{8}k_x+\tfrac{3}{8}k_y+\tfrac{1}{8}k_z\Big)a\\ &+\cos\Big(\tfrac{1}{8}k_x+\tfrac{1}{8}k_y+\tfrac{3}{8}k_z\Big)a + \cos\Big(-\tfrac{3}{8}k_x-\tfrac{1}{8}k_y+\tfrac{1}{8}k_z\Big)a\\ &+\cos\Big(-\tfrac{1}{8}k_x-\tfrac{3}{8}k_y+\tfrac{1}{8}k_z\Big)a + \cos\Big(-\tfrac{1}{8}k_x-\tfrac{1}{8}k_y+\tfrac{3}{8}k_z\Big)a\\ &+\cos\Big(-\tfrac{1}{8}k_x+\tfrac{1}{8}k_y-\tfrac{3}{8}k_z\Big)a + \cos\Big(-\tfrac{1}{8}k_x+\tfrac{3}{8}k_y-\tfrac{1}{8}k_z\Big)a\\ &+\cos\Big(-\tfrac{3}{8}k_x+\tfrac{1}{8}k_y-\tfrac{1}{8}k_z\Big)a + \cos\Big(\tfrac{1}{8}k_x-\tfrac{1}{8}k_y-\tfrac{3}{8}k_z\Big)a\\ &+\cos\Big(\tfrac{1}{8}k_x-\tfrac{3}{8}k_y-\tfrac{1}{8}k_z\Big)a + \cos\Big(\tfrac{3}{8}k_x-\tfrac{1}{8}k_y-\tfrac{1}{8}k_z\Big)a\Big]. \qquad (14)\end{aligned}$$

$$\begin{aligned}\sum_i J_i(\underline{k}) = 8J\Big[&\Big[\cos\Big(\tfrac{3}{8}k_x+\Big(\tfrac{1}{8}k_y+\tfrac{1}{8}k_z\Big)\Big)a + \cos\Big(\tfrac{3}{8}k_x-\Big(\tfrac{1}{8}k_y+\tfrac{1}{8}k_z\Big)\Big)a\Big]\\ &+\Big[\cos\Big(\tfrac{1}{8}k_x+\Big(\tfrac{3}{8}k_y+\tfrac{1}{8}k_z\Big)\Big)a + \cos\Big(\tfrac{1}{8}k_x-\Big(\tfrac{3}{8}k_y+\tfrac{1}{8}k_z\Big)\Big)a\Big]\\ &+\Big[\cos\Big(\tfrac{1}{8}k_x+\Big(\tfrac{1}{8}k_y+\tfrac{3}{8}k_z\Big)\Big)a + \cos\Big(\tfrac{1}{8}k_x-\Big(\tfrac{1}{8}k_y+\tfrac{3}{8}k_z\Big)\Big)a\Big]\\ &+\Big[\cos\Big(-\tfrac{3}{8}k_x+\Big(\tfrac{1}{8}k_y-\tfrac{1}{8}k_z\Big)\Big)a + \cos\Big(-\tfrac{3}{8}k_x-\Big(\tfrac{1}{8}k_y-\tfrac{1}{8}k_z\Big)\Big)a\Big]\\ &+\Big[\cos\Big(-\tfrac{1}{8}k_x+\Big(\tfrac{3}{8}k_y-\tfrac{1}{8}k_z\Big)\Big)a + \cos\Big(-\tfrac{1}{8}k_x-\Big(\tfrac{3}{8}k_y-\tfrac{1}{8}k_z\Big)\Big)a\Big]\\ &+\Big[\cos\Big(-\tfrac{1}{8}k_x+\Big(\tfrac{1}{8}k_y-\tfrac{3}{8}k_z\Big)\Big)a + \cos\Big(-\tfrac{1}{8}k_x-\Big(\tfrac{1}{8}k_y-\tfrac{3}{8}k_z\Big)\Big)a\Big]\Big]. \qquad (15)\end{aligned}$$

Using the identities: $\cos(A+B) = \cos A \cos B - \sin A \sin B$

$\cos(A-B) = \cos A \cos B + \sin A \sin B$

$\cos(A+B) + \cos(A-B) = 2 \cos A \cos B,$

we get

$$\sum_i J_i(\underline{k}) = 8J\Big[2\cos\tfrac{3}{8}k_x a\cos\Big(\tfrac{1}{8}k_y+\tfrac{1}{8}k_z\Big)a + 2\cos\tfrac{1}{8}k_x a\cos\Big(\tfrac{3}{8}k_y+\tfrac{1}{8}k_z\Big)a + 2\cos\tfrac{1}{8}k_x a\cos\Big(\tfrac{1}{8}k_y+\tfrac{3}{8}k_z\Big)a + 2\cos\tfrac{3}{8}k_x a\cos\Big(\tfrac{1}{8}k_y-\tfrac{1}{8}k_z\Big)a + 2\cos\tfrac{1}{8}k_x a\cos\Big(\tfrac{3}{8}k_y-\tfrac{1}{8}k_z\Big)a + 2\cos\tfrac{1}{8}k_x a\cos\Big(\tfrac{1}{8}k_y-\tfrac{3}{8}k_z\Big)a\Big]. \tag{16}$$

$$\sum_i J_i(\underline{k}) = 16J\Big[\cos\tfrac{3}{8}k_x a\Big[\cos\Big(\tfrac{1}{8}k_y+\tfrac{1}{8}k_z\Big)a + \cos\Big(\tfrac{1}{8}k_y-\tfrac{1}{8}k_z\Big)a\Big] + \cos\tfrac{1}{8}k_x a\Big[\cos\Big(\tfrac{3}{8}k_y+\tfrac{1}{8}k_z\Big)a + \cos\Big(\tfrac{3}{8}k_y-\tfrac{1}{8}k_z\Big)a\Big] + \cos\tfrac{1}{8}k_x a\Big[\cos\Big(\tfrac{1}{8}k_y+\tfrac{3}{8}k_z\Big)a + \cos\Big(\tfrac{1}{8}k_y-\tfrac{3}{8}k_z\Big)a\Big]\Big]. \tag{17}$$

$$\sum_i J_i(\underline{k}) = 32J\Big[\cos\tfrac{3}{8}k_x a\cos\tfrac{1}{8}k_y a\cos\tfrac{1}{8}k_z a + \cos\tfrac{1}{8}k_x a\cos\tfrac{3}{8}k_y a\cos\tfrac{1}{8}k_z a + \cos\tfrac{1}{8}k_x a\cos\tfrac{1}{8}k_y a\cos\tfrac{3}{8}k_z a\Big]. \tag{18}$$

Substituting eq. (18) into eq. (2) gives

$$J(\underline{k}) = \frac{1}{\sqrt{N_A N_B}}\,32J\Big[\cos\tfrac{3}{8}k_x a\cos\tfrac{1}{8}k_y a\cos\tfrac{1}{8}k_z a + \cos\tfrac{1}{8}k_x a\cos\tfrac{3}{8}k_y a\cos\tfrac{1}{8}k_z a + \cos\tfrac{1}{8}k_x a\cos\tfrac{1}{8}k_y a\cos\tfrac{3}{8}k_z a\Big]. \tag{19}$$

In a unit cell of the normal spinel structure, $N_A = 8$ and $N_B = 16$, therefore

$$J(\underline{k}) = 2\sqrt{2}\, J\Big[\cos\tfrac{3}{8}k_x a \cos\tfrac{1}{8}k_y a \cos\tfrac{1}{8}k_z a + \cos\tfrac{1}{8}k_x a \cos\tfrac{3}{8}k_y a \cos\tfrac{1}{8}k_z a + \cos\tfrac{1}{8}k_x a \cos\tfrac{1}{8}k_y a \cos\tfrac{3}{8}k_z a\Big] \qquad (20)$$

which we put into eq. (3.7a).

2. Calculation of $J'(\underline{k})$

According to eq. (3.6b), we have

$$J'(\underline{k}) = \frac{1}{N_A} \sum_i \sum_{i'} J'_{ii'}\, e^{i\underline{k}\cdot(\underline{r}_{i'}-\underline{r}_i)} \qquad (21)$$

or
$$J'(\underline{k}) = \frac{1}{N_A} \sum_i J'_i(\underline{k}), \qquad (22)$$

where
$$J'_i(\underline{k}) = \sum_{i'} J'_{ii'}\, e^{i\underline{k}\cdot(\underline{r}_{i'}-\underline{r}_i)}. \qquad (23)$$

Assuming nearest neighbour approximation with the isotropic exchange integral J', eq. (23) becomes

$$J'_i(\underline{k}) = J' \sum_{i'=1}^{z} e^{i\underline{k}\cdot(\underline{r}_{i'}-\underline{r}_i)}, \qquad (24)$$

where z is the number of nearest neighbours in the A sublattice of the ith ion.

The ion in the A sublattice located at (0,0,0) has four nearest neighbours in the same sublattice located at

$$\left(\tfrac{1}{4},-\tfrac{1}{4},\tfrac{1}{4}\right)a \qquad \left(-\tfrac{1}{4},\tfrac{1}{4},\tfrac{1}{4}\right)a$$

$$\left(\tfrac{1}{4},\tfrac{1}{4},-\tfrac{1}{4}\right)a \qquad \left(-\tfrac{1}{4},-\tfrac{1}{4},-\tfrac{1}{4}\right)a. \tag{25}$$

Taking $\underline{r}_1 = (0,0,0)a$ and $\underline{r}_i$ as defined in eq. (25), we get

$$J_1'(\underline{k}) = J'\left[e^{i\underline{k}\cdot\left(\frac{1}{4},-\frac{1}{4},\frac{1}{4}\right)a} + e^{i\underline{k}\cdot\left(-\frac{1}{4},\frac{1}{4},\frac{1}{4}\right)a} + e^{i\underline{k}\cdot\left(\frac{1}{4},\frac{1}{4},-\frac{1}{4}\right)a} + e^{i\underline{k}\cdot\left(-\frac{1}{4},-\frac{1}{4},-\frac{1}{4}\right)a}\right]. \tag{26}$$

The A ion at $\left(\tfrac{1}{4},-\tfrac{1}{4},\tfrac{1}{4}\right)a$ has four nearest neighbour A ions located at

$$(0,0,0)a \qquad \left(\tfrac{1}{2},0,\tfrac{1}{2}\right)a$$

$$\left(0,-\tfrac{1}{2},\tfrac{1}{2}\right)a \qquad \left(\tfrac{1}{2},-\tfrac{1}{2},0\right)a. \tag{27}$$

Taking $\underline{r}_2 = \left(\tfrac{1}{4},-\tfrac{1}{4},\tfrac{1}{4}\right)a$ and $\underline{r}_i$, as defined in eq. (27) then the corresponding distances $(\underline{r}_i,-\underline{r}_2)$ are

$$-\left(\tfrac{1}{4},\tfrac{1}{4},-\tfrac{1}{4}\right)a \qquad \left(\tfrac{1}{4},\tfrac{1}{4},\tfrac{1}{4}\right)a$$

$$-\left(\tfrac{1}{4},-\tfrac{1}{4},\tfrac{1}{4}\right)a \qquad \left(\tfrac{1}{4},-\tfrac{1}{4},-\tfrac{1}{4}\right)a, \tag{28}$$

which are negative to the distances given by eq. (25). Therefore we get

$$J_2'(\underline{k}) = J'\left[e^{i\underline{k}\cdot\left(\frac{1}{4},\frac{1}{4},-\frac{1}{4}\right)a} + e^{i\underline{k}\cdot\left(\frac{1}{4},\frac{1}{4},\frac{1}{4}\right)a} + e^{i\underline{k}\cdot\left(-\frac{1}{4},-\frac{1}{4},\frac{1}{4}\right)a} + e^{i\underline{k}\cdot\left(\frac{1}{4},-\frac{1}{4},-\frac{1}{4}\right)a}\right]. \quad (29)$$

Combining together eqs. (26) and (29), we get

$$J_1'(\underline{k})+J_2'(\underline{k}) = J'\left[2\cos\,\underline{k}\cdot\left(\frac{1}{4},-\frac{1}{4},\frac{1}{4}\right)a + 2\cos\,\underline{k}\cdot\left(-\frac{1}{4},\frac{1}{4},\frac{1}{4}\right)a + 2\cos\,\underline{k}\cdot\left(\frac{1}{4},\frac{1}{4},-\frac{1}{4}\right)a + 2\cos\,\underline{k}\cdot\left(-\frac{1}{4},-\frac{1}{4},-\frac{1}{4}\right)a\right]. \quad (30)$$

We can also get

$$J_3'(\underline{k}) = J_4'(\underline{k}) = J_5'(\underline{k}) = J_2'(\underline{k}), \quad (31)$$

where $\underline{r}_3 = \left(-\frac{1}{4},\frac{1}{4},\frac{1}{4}\right)a$, $\underline{r}_4 = \left(\frac{1}{4},\frac{1}{4},-\frac{1}{4}\right)a$ and $\underline{r}_5 = \left(-\frac{1}{4},-\frac{1}{4},-\frac{1}{4}\right)a$,

and

$$J_6'(\underline{k}) = J_7'(\underline{k}) = \cdots\cdots\cdots = J_{17}'(\underline{k}) = \frac{1}{4}\,J_1'(\underline{k}), \quad (32)$$

where $\underline{r}_6$, $\underline{r}_7$, ..., $\underline{r}_{17}$ are the rest of the A sites, each of which is **occupied by** 1/4 ion.

Hence, we obtain

$$\begin{aligned}\sum_i J_i'(\underline{k}) &= J_1'(\underline{k})+J_2'(\underline{k})+J_3'(\underline{k})+\cdots\cdots\cdots+J_{17}'(\underline{k}) \\ &= J_1'(\underline{k})+J_2'(\underline{k}) + 3J_2'(\underline{k}) + \frac{12}{4}\,J_1'(\underline{k}) \\ &= J_1'(\underline{k})+J_2'(\underline{k}) + 3(J_1'(\underline{k})+J_2'(\underline{k})) \\ &= 4(J_1'(\underline{k})+J_2'(\underline{k})). \end{aligned} \quad (33)$$

Substituting eq. (30) into eq. (33), we get

$$\sum_i J_i'(\underline{k}) = 2 \cdot 4J' \left[\left[\cos\left(\tfrac{1}{4}k_x + \left(\tfrac{1}{4}k_y - \tfrac{1}{4}k_z\right)\right) a + \cos\left(\tfrac{1}{4}k_x - \left(\tfrac{1}{4}k_y - \tfrac{1}{4}k_z\right)\right) a \right] + \left[\cos\left(-\tfrac{1}{4}k_x + \left(\tfrac{1}{4}k_y + \tfrac{1}{4}k_z\right)\right) a + \cos\left(-\tfrac{1}{4}k_x - \left(\tfrac{1}{4}k_y + \tfrac{1}{4}k_z\right)\right) a \right] \right]. \tag{34}$$

$$\sum_i J_i'(\underline{k}) = 8J' \left[2\cos \tfrac{1}{4}k_x a \cos\left(\tfrac{1}{4}k_y - \tfrac{1}{4}k_z\right) a + 2\cos \tfrac{1}{4}k_x a \cos\left(\tfrac{1}{4}k_y + \tfrac{1}{4}k_z\right) a \right]. \tag{35}$$

$$\sum_i J_i'(\underline{k}) = 16J' \left[\cos \tfrac{1}{4}k_x a \left[\cos\left(\tfrac{1}{4}k_y - \tfrac{1}{4}k_z\right) a + \cos\left(\tfrac{1}{4}k_y + \tfrac{1}{4}k_z\right) a \right] \right]. \tag{36}$$

$$\sum_i J_i'(\underline{k}) = 32J' \left[\cos \tfrac{1}{4}k_x a \cos \tfrac{1}{4}k_y a \cos \tfrac{1}{4}k_z a \right]. \tag{37}$$

Substituting eq. (37) into eq. (22) gives

$$J'(\underline{k}) = \frac{1}{N_A} 32J' \left[\cos \tfrac{1}{4}k_x a \cos \tfrac{1}{4}k_y a \cos \tfrac{1}{4}k_z a \right]$$

$$= 4J' \left[\cos \tfrac{1}{4}k_x a \cos \tfrac{1}{4}k_y a \cos \tfrac{1}{4}k_z a \right]. \tag{38}$$

The above equation is what we put into eq. (3.7b).

3. Calculation of $J''(\underline{k})$

According to eq. (3.6c), we have

$$J''(\underline{k}) = \frac{1}{N_B} \sum_j \sum_{j'} J''_{jj'} e^{i\underline{k}\cdot(\underline{r}_j - \underline{r}_j)}, \tag{39}$$

or $$J''(\underline{k}) = \frac{1}{N_B} \sum_j J''_j(\underline{k}), \tag{40}$$

where $$J''_j(\underline{k}) = \sum_{j'} J''_{jj'} \, e^{i\underline{k}\cdot(\underline{r}_j, -\underline{r}_j)}. \tag{41}$$

Assuming nearest neighbour approximation with the isotropic exchange integral J'', eq. (41) becomes

$$J''_j(\underline{k}) = J'' \sum_{j'=1}^{z} e^{i\underline{k}\cdot(\underline{r}_j, -\underline{r}_j)}, \tag{42}$$

where z is the number of nearest neighbours in the B sublattice of the jth ion in that sublattice.

Consider the B ion located at $\left(\frac{3}{8},\frac{1}{8},\frac{1}{8}\right)a$ having six nearest neighbours in the same sublattice at the following positions:

$$\left(\tfrac{3}{8},\tfrac{3}{8},\tfrac{3}{8}\right)a \qquad \left(\tfrac{5}{8},-\tfrac{1}{8},\tfrac{1}{8}\right)a$$

$$\left(\tfrac{1}{8},\tfrac{1}{8},\tfrac{3}{8}\right)a \qquad \left(\tfrac{3}{8},-\tfrac{1}{8},-\tfrac{1}{8}\right)a$$

$$\left(\tfrac{1}{8},\tfrac{3}{8},\tfrac{1}{8}\right)a \qquad \left(\tfrac{5}{8},\tfrac{1}{8},-\tfrac{1}{8}\right)a. \tag{43}$$

Taking $\underline{r}_1 = \left(\frac{3}{8},\frac{1}{8},\frac{1}{8}\right)a$ and $\underline{r}_j$, as defined in eq. (43), the corresponding distances $(\underline{r}_j, -\underline{r}_1)$ are

$$\left(0,\tfrac{1}{4},\tfrac{1}{4}\right)a \qquad \left(\tfrac{1}{4},-\tfrac{1}{4},0\right)a$$

$$\left(-\tfrac{1}{4},0,\tfrac{1}{4}\right)a \qquad \left(0,-\tfrac{1}{4},-\tfrac{1}{4}\right)a$$

$$\left(-\tfrac{1}{4},\tfrac{1}{4},0\right)a \qquad \left(\tfrac{1}{4},0,-\tfrac{1}{4}\right)a. \tag{44}$$

Substituting eq. (44) into eq. (42) gives

$$J_1''(\underline{k}) = J''\Big[e^{i\underline{k}\cdot\left(0,\frac{1}{4},\frac{1}{4}\right)a} + e^{i\underline{k}\cdot\left(-\frac{1}{4},0,\frac{1}{4}\right)a} + e^{i\underline{k}\cdot\left(-\frac{1}{4},\frac{1}{4},0\right)a} + e^{i\underline{k}\cdot\left(\frac{1}{4},-\frac{1}{4},0\right)a} + e^{i\underline{k}\cdot\left(0,-\frac{1}{4},-\frac{1}{4}\right)a} + e^{i\underline{k}\cdot\left(\frac{1}{4},0,-\frac{1}{4}\right)a}\Big]. \tag{45}$$

Similarly we can get

$$J_2''(\underline{k}) = J_3''(\underline{k}) = \cdots\cdots\cdots\cdots = J_{16}''(\underline{k}) = J_1''(\underline{k}), \tag{46}$$

where $\underline{r}_2$, $\underline{r}_3$, ..., $\underline{r}_{16}$ are the remaining B sites in a unit cell of the spinel structure.

Hence, we obtain

$$\sum_j J_j''(\underline{k}) = 16J_1''(\underline{k})$$

$$= 16J''\Big[2\cos\,\underline{k}\cdot\left(0,\tfrac{1}{4},\tfrac{1}{4}\right)a + 2\cos\,\underline{k}\cdot\left(-\tfrac{1}{4},0,\tfrac{1}{4}\right)a + 2\cos\,\underline{k}\cdot\left(-\tfrac{1}{4},\tfrac{1}{4},0\right)a\Big]. \tag{47}$$

$$\sum_j J_j''(\underline{k}) = 32J''\Big[\cos\left(\tfrac{1}{4}k_y + \tfrac{1}{4}k_z\right)a + \cos\left(-\tfrac{1}{4}k_x + \tfrac{1}{4}k_z\right)a + \cos\left(-\tfrac{1}{4}k_x + \tfrac{1}{4}k_y\right)a\Big]. \tag{48}$$

$$\sum_j J_j''(\underline{k}) = 32J''\Big[\cos\tfrac{1}{4}k_y a\cos\tfrac{1}{4}k_z a - \sin\tfrac{1}{4}k_y a\sin\tfrac{1}{4}k_z a + \cos\tfrac{1}{4}k_x a\cos\tfrac{1}{4}k_z a + \sin\tfrac{1}{4}k_x a\sin\tfrac{1}{4}k_z a + \cos\tfrac{1}{4}k_x a\cos\tfrac{1}{4}k_y a + \sin\tfrac{1}{4}k_x a\sin\tfrac{1}{4}k_y a\Big]. \tag{49}$$

$$\sum_j J''_j(k) = 32J'' \Big[\cos \tfrac{1}{4}k_x a \cos \tfrac{1}{4}k_y a + \cos \tfrac{1}{4}k_y a \cos \tfrac{1}{4}k_z a + \cos \tfrac{1}{4}k_z a \cos \tfrac{1}{4}k_x a + \sin \tfrac{1}{4}k_x a \sin \tfrac{1}{4}k_y a - \sin \tfrac{1}{4}k_y a \sin \tfrac{1}{4}k_z a + \sin \tfrac{1}{4}k_z a \sin \tfrac{1}{4}k_x a \Big]. \quad (50)$$

Substituting eq. (50) into eq. (40), we get

$$J''(k) = \frac{1}{N_B} 32J'' \Big[\cos \tfrac{1}{4}k_x a \cos \tfrac{1}{4}k_y a + \cos \tfrac{1}{4}k_y a \cos \tfrac{1}{4}k_z a + \cos \tfrac{1}{4}k_z a \cos \tfrac{1}{4}k_x a + \sin \tfrac{1}{4}k_x a \sin \tfrac{1}{4}k_y a - \sin \tfrac{1}{4}k_y a \sin \tfrac{1}{4}k_z a + \sin \tfrac{1}{4}k_z a \sin \tfrac{1}{4}k_x a \Big]. \quad (51)$$

$$J''(k) = 2J'' \Big[\cos \tfrac{1}{4}k_x a \cos \tfrac{1}{4}k_y a + \cos \tfrac{1}{4}k_y a \cos \tfrac{1}{4}k_z a + \cos \tfrac{1}{4}k_z a \cos \tfrac{1}{4}k_x a + \sin \tfrac{1}{4}k_x a \sin \tfrac{1}{4}k_y a - \sin \tfrac{1}{4}k_y a \sin \tfrac{1}{4}k_z a + \sin \tfrac{1}{4}k_z a \sin \tfrac{1}{4}k_x a \Big]. \quad (52)$$

The eq. (52) is what we put into eq. (3.7c).