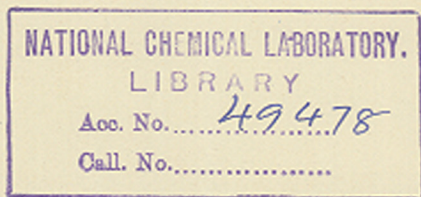


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AWARDED Ph.D. DEGREE



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## S Y N O P S I S

Since the Heisenberg's theory of a ferromagnet and the Bloch's idea of spin waves which represent the excitations of a magnetic system, the magnetically ordered systems have stimulated considerable interest among research workers. Among the three main types of ordered systems, namely, ferro-, ferri- and antiferromagnetics, we are interested in the ferrimagnetic crystals in the present dissertation. The simplest ferrimagnetic system consists of two simple cubic sublattices which interpenetrate to form a body-centred cubic lattice. The spins of the atoms on sublattice A are opposite to and different in magnitude from the spins of the atoms on sublattice B.

Firstly, we give a general discussion of the exchange coupling processes and other elementary excitations such as the lattice vibrations and a review of the two different methods of treating the magnetically ordered system, i.e., the spin wave method and the Green function method. Then we calculate the spin wave spectrum of a ferrimagnet and the phonon-magnon interaction terms. By calculating the transition probabilities and the energy transfer from the phonon system to the magnon system, we evaluate the phonon-magnon relaxation time. The interaction terms obtained above are used to calculate the low temperature thermal conductivity of ferrimagnets. This is found to agree well with the experimental

data available for some systems. The spin wave spectrum, is also calculated by using the more rigorous Green function method, which further gives the temperature dependence of magnetization of a ferrimagnet.

Finally, we consider the scattering of neutrons by magnons. The magnon system, considered to be in equilibrium, is excited due to the interaction with the neutrons, thereby causing scattering of neutrons in different directions. In this interaction, since we have considered only ~~critical~~ *orbital* transitions, only two magnon processes are considered. The scattering cross sections are evaluated in a few different cases. The thesis concludes with a discussion of different results obtained and the merits and demerits of the different methods used.

CHAPTER - 1

## INTRODUCTION

In recent years, considerable interest has grown in the physical properties of magnetic compounds. Our main interest lies in the crystalline solids which contain an assembly of atoms or ions with permanent magnetic moments. Although the importance of the exchange and correlation phenomena related to certain spin dependent properties is fully realized, these effects have not been properly understood.

The fundamental concept of spin exchange emanates from Heisenberg's theory in 1928. This theory is an application of the Heitler-London's celebrated theory of the chemical bond and proved to be a brilliant step in the explanation of ferromagnetism. In this model a certain number of unpaired electrons are assumed to be present in each atom which are regularly spaced in a crystal. The electronic spins are assumed to be localized at each lattice site. It was shown in Heisenberg's theory that an exchange effect leading to a strong coupling between the electronic spins would be caused by ordinary Coulomb interactions among electrons when proper cognizance of the exclusion principle is taken into account.

This, in effect, is a many electron problem and, in 1929, two methods were suggested to deal with it: the



determinantal method developed by Slater<sup>1</sup> and the spin operator method proposed by Dirac.<sup>2</sup> In this approach, Dirac showed that the spin dependent energy for interacting electrons ~~can be written~~ can be written as<sup>3-5</sup>

$$H_{ex} = - \sum_{i,j} J_{ij} \cdot \underline{S}_i \cdot \underline{S}_j, \quad (1.1)$$

where  $\underline{S}_i$  is the spin operator for the electron in the  $i^{th}$  orbital. Let  $\psi_a$  and  $\psi_b$  be the localized atomic orbitals being respectively the solutions of

$$\left. \begin{aligned} \left( \frac{p_i^2}{2m} - \frac{ze^2}{r_{ia}} \right) \psi_a(i) &= E_a \psi_a(i), \\ \left( \frac{p_j^2}{2m} - \frac{ze^2}{r_{jb}} \right) \psi_b(j) &= E_b \psi_b(j), \end{aligned} \right\} \quad (1.2)$$

where  $p_i^2/2m$ ,  $p_j^2/2m$  are the kinetic energy operators for the electrons  $i$  and  $j$ ,  $m$  is the mass of electron,  $ze$  is the charge of the ion core and  $-\frac{ze^2}{r_{ia}}$ ,  $-\frac{ze^2}{r_{jb}}$  are the potential energies. Here  $r_{ia}$  is the distance of the electron  $i$  from the ion  $a$  and  $r_{jb}$  that of the electron  $j$  from the ion  $b$ .  $E_{a,b}$  are the corresponding energies. In terms of these wave functions, the exchange integral, in the general case, is given by

$$J_{ij} = \left\langle \frac{ab}{r_{ij}} \frac{e^2}{ba} \right\rangle - 2S_{ab} \langle a/V/b \rangle. \quad (1.3)$$

Here  $V = -\frac{ze^2}{r_{ib}} - \frac{ze^2}{r_{ja}}$

and  $S_{ab} = \langle a/b \rangle = \int \psi_a^* \psi_b d\tau.$

If we are dealing with orthogonal orbitals, the overlap integral  $S_{ab} = 0$  and only the first term in (1.3) survives. This term is always positive being the self energy of the overlap charge  $e\psi_a^*(i)\psi_b(i)$ . This favours ferromagnetism ( the triplet state ). However, if the orbitals are non-orthogonal, the sign of  $J_{ij}$  would depend on whether the first or the second term in (1.3) dominates and when the second term dominates the antiferromagnetic coupling ( the singlet state ) is favoured. This state was actually found to exist in most of the insulators. In this type of crystals the total magnetic moment is zero due to the anti-parallel alignment.

Another significant development in ordered magnetic systems is the concept of ferrimagnetism due to Néel in 1948. According to Néel's idea, ferrimagnets also have sublattices with anti-parallel spins but differ from antiferromagnets in that the resultant magnetic moment is not zero as a result of one or more of the following factors: (i) unequal spins, (ii) unequal g factors, (iii) unequal number of

sites in the different sublattices. These substances constitute an important class of magnetic materials and occur in various forms.

The simplest ferrites are represented by the formula  $A(B_2)O_4$ , where A is a divalent metal ion such as  $Mn^{2+}$ ,  $Co^{2+}$ ,  $Ni^{2+}$ ,  $Fe^{2+}$ , etc. and B is a trivalent metal ion (e.g.,  $Fe^{3+}$ ). These compounds are known to exist in the spinel structure.<sup>6,7</sup> The A sites are tetrahedrally coordinated by four oxygen ions whereas the B ions are surrounded octahedrally by six oxygens. There are eight formula units in one unit cell with the cubic symmetry  $Fd\bar{3}m-O_h^7$ . In the inverse spinel, the A site ions and half of the B site ions exchange positions, the formula being represented by  $B(AB)O_4$ . Another structure of interest is the general garnet structure which can be denoted by the formula<sup>8,9</sup>  $\{A_3\} [B_2] (C_3) O_{12}$ . A unit cell of this structure also contains eight formula units and all the metal ions are in highly special positions:  $24A^{2+}$  ions in the c positions dodecahedrally surrounded by oxygen ions,  $16B^{3+}$  ions in 'a' sites surrounded octahedrally by oxygen ions and  $24C^{4+}$  ions in 'd' sites each at the centre of a tetrahedron of oxygen ions.

These magnetic materials (ferro-, antiferro- and ferrimagnetic) have a definite spin ordering in the ground state which is realized only at the absolute zero of temperature. If the temperature is slightly increased,

excitations in the spin system will be produced over the ground state. The physical properties of a system are governed by these low lying excitations. Thus, if we know the energy spectrum of the elementary excitations of a particular system, we can have a fairly clear idea of its physical behaviour. Two important methods have been developed to treat such problems: (a) the spinwave method, and (b) the Green function method.

#### A. MAGNONS:

Historically the concept of spin waves was introduced by Bloch in 1932. He conceived of a spin wave as a single spin reversal in an otherwise ordered system, which due to the strong exchange interactions, does not remain localized but is coherently distributed over the crystal lattice. He was able to show that the low-energy excited states of a ferromagnet would be of this character. In this theory, the interactions between spin waves are neglected.

An altogether new technique was suggested by Holstein and Primakoff<sup>10</sup> (HP) to include the spin wave interactions which was followed by a rigorous and satisfying treatment by Dyson.<sup>11</sup> They successfully defined a set of coordinates which have the appearance of the spin wave amplitudes and which describe accurately the quantum state of the system. Thus, let us consider a body centered cubic ferromagnet. Let  $S$  be the magnitude of the spin and  $\underline{S}_i$  the spin

operator for the site 1. Then it is convenient to introduce the new operators in terms of the x, y and z components of the operator  $S_1$  by

$$\left. \begin{aligned} S_1^+ &= S_1^x + i S_1^y, \\ \hat{n}_1 &= S - S_1^z. \end{aligned} \right\} \quad (1.4)$$

The eigenstate of the operators  $\hat{n}_1$  and  $S_1^z$  is written as

$$\Psi_{n_1} \dots n_1 \dots n_N = \Psi_{n_1}, \quad (1.5)$$

the corresponding eigenvalues being  $n_1$  and  $m_1$  respectively.  $m_1$  takes the values  $S, S-1, \dots, -S$  and  $n_1$ , which takes the integral values,  $0, 1, \dots, 2S$ , obviously represents the difference between the z component of the spin at the 1<sup>th</sup> site and its maximum value. This is known as the spin deviation.

The operators defined in (1.4) have the properties

$$\left. \begin{aligned} S_1^+ \Psi_{n_1} &= (2S)^{\frac{1}{2}} \left( 1 - \frac{n_1-1}{2S} \right)^{\frac{1}{2}} (n_1)^{\frac{1}{2}} \Psi_{n_1-1}, \\ S_1^- \Psi_{n_1} &= (2S)^{\frac{1}{2}} (n_1+1)^{\frac{1}{2}} \left( 1 - \frac{n_1}{2S} \right)^{\frac{1}{2}} \Psi_{n_1+1}, \end{aligned} \right\} \quad (1.6)$$

$$\hat{n}_1 \Psi_{n_1} = n_1 \Psi_{n_1}$$

and satisfy the commutation relations

$$\left. \begin{aligned} [S_l^z, S_m^\pm] &= \pm S_l^\pm \delta_{lm}, \\ [S_l^\pm, S_m^\mp] &= 2S_l^z \delta_{lm}, \end{aligned} \right\} \quad (1.7)$$

where  $\delta_{lm}$  is the Kronecker  $\delta$ -function.

In handling most of the problems concerning spin wave interactions, it is expedient to work in the second quantization formalism or the number operator formalism. Thus we conceive of the quasi-particles associated with the spin waves and denote by  $a^+$  and  $a$  the creation and the destruction operators respectively for these quasi-particles. These operators are defined by

$$\left. \begin{aligned} a_l^+ \psi_{n_l} &= (n_l + 1)^{\frac{1}{2}} \psi_{n_l+1}, \\ a_l \psi_{n_l} &= (n_l)^{\frac{1}{2}} \psi_{n_l-1}. \end{aligned} \right\} \quad (1.8)$$

Comparing (1.6) and (1.8) we obtain

$$\left. \begin{aligned} S_l^+ &= (2S)^{\frac{1}{2}} \left( 1 - \frac{a_l^+ a_l}{2S} \right)^{\frac{1}{2}} a_l, \\ S_l^- &= (2S)^{\frac{1}{2}} a_l^+ \left( 1 - \frac{a_l^+ a_l}{2S} \right)^{\frac{1}{2}}, \end{aligned} \right\} \quad (1.9)$$

$$\hat{n}_l = a_l^+ a_l = S - S_l^z.$$

Using (1.7) we can obtain the commutation relations for these operators

$$[a_l, a_m^+] = \delta_{lm} \quad (1.10)$$

The exchange Hamiltonian for a ferromagnet is given by (1.1). This can now be written in terms of the new operators as

$$\begin{aligned} H_{\text{ex}} &= - \sum_{i,j} J_{ij} \underline{S}_i \cdot \underline{S}_j \\ &= - \sum_{i,j} J_{ij} \left\{ S_i^z S_j^z + \frac{1}{2} (S_i^+ S_j^- + S_i^- S_j^+) \right\} \end{aligned}$$

and assuming that  $a_i^+ \cdot a_i < 2S$ , we can expand the brackets in equation (1.9) and obtain

$$\begin{aligned} H_{\text{ex}} &= \text{Const.} + \sum_{i,j} J_{ij} S (a_i^+ a_i + a_j^+ a_j - a_i a_j^+ - a_i^+ a_j) \\ &\quad + \text{higher order terms.} \end{aligned} \quad (1.11)$$

Now a spin deviation or disturbance will not remain localized at a particular site  $R_1$ , but will move throughout the crystal like a wave due to the strong exchange forces.<sup>12</sup> If there is only one spin wave present, it was shown by Bloch and later by Dyson<sup>11</sup> that this will be an exact eigenstate of the Hamiltonian. However, if there are more than one spin

waves present in the lattice, interactions between them will come into play and the Hamiltonian will no longer be diagonal. The Hamiltonian in this case splits up into two parts; one quadratic and the other containing higher order terms in the operators  $a$  and  $a^+$ .

We will now introduce the Fourier transforms to the reciprocal space by

$$\left. \begin{aligned} a_i &= \frac{1}{\sqrt{N}} \sum_{\underline{\lambda}} e^{i \underline{\lambda} \cdot \underline{R}_i} a_{\underline{\lambda}}, \\ a_i^+ &= \frac{1}{\sqrt{N}} \sum_{\underline{\lambda}} e^{-i \underline{\lambda} \cdot \underline{R}_i} a_{\underline{\lambda}}^+, \end{aligned} \right\} \quad (1.12)$$

where  $N$  is the number of sites in the crystal and  $\underline{\lambda}$  the wave vector of the quasi-particle, now called as magnon. By using the periodic boundary condition in the form of the equation

$$\sum_i e^{i \underline{\lambda} \cdot \underline{R}_i} = N \Delta(\underline{\lambda}), \quad (1.13)$$

where  $\Delta(\underline{\lambda}) = 0$  for  $\underline{\lambda} \neq 0$ ,  $\Delta(\underline{\lambda}) = 1$  for  $\underline{\lambda} = 0$ , we obtain the inverse transformations



$$\left. \begin{aligned}
 a_{\underline{\lambda}} &= \frac{1}{\sqrt{N}} \sum_{\mathbf{i}} e^{-i \underline{\lambda} \cdot \mathbf{R}_{\mathbf{i}}} a_{\mathbf{i}}, \\
 a_{\underline{\lambda}}^{\dagger} &= \frac{1}{\sqrt{N}} \sum_{\mathbf{i}} e^{i \underline{\lambda} \cdot \mathbf{R}_{\mathbf{i}}} a_{\mathbf{i}}^{\dagger}.
 \end{aligned} \right\} (1.14)$$

The operators  $a_{\underline{\lambda}}$  and  $a_{\underline{\lambda}}^{\dagger}$  satisfy the commutation rules which can be found from (1.10)

$$[a_{\underline{\lambda}}, a_{\underline{\lambda}'}^{\dagger}] = \delta_{\underline{\lambda}\underline{\lambda}'}, \quad (1.15)$$

all other commutators being zero. From these commutation relations, it is clear that magnons are bosons. In terms of these operators, the diagonal part of the Hamiltonian takes the form

$$H_0 = \sum_{\underline{\lambda}} \hbar \omega_{\underline{\lambda}} \left( a_{\underline{\lambda}}^{\dagger} a_{\underline{\lambda}} + \frac{1}{2} \right), \quad (1.16)$$

where  $\hbar \omega_{\underline{\lambda}}$  is the energy of the magnon with wave vector  $\underline{\lambda}$  and takes different expressions for different systems. For a cubic ferromagnet, this takes the form<sup>13,14</sup>

$$\hbar \omega_{\underline{\lambda}} = 2 JSz (1 - \gamma_{\underline{\lambda}}), \quad (1.17)$$

where  $J$  is the exchange integral  $J_{\mathbf{i}\mathbf{j}}$  for nearest neighbours,  $z$  is their number and

$$\gamma_{\underline{\lambda}} = \frac{1}{z} \sum_{\mathbf{h}} e^{i \underline{\lambda} \cdot \mathbf{R}_{\mathbf{h}}} \quad (1.18)$$

where  $\mathbf{R}_{\mathbf{h}}$  is the vector to the nearest neighbour.

### B. PHONONS:

Thus we have seen how the spin system at finite temperatures can be likened to an assembly of spin waves. The elementary excitations of such a system are called magnons. In crystal, there exists another elementary excitation, namely, the lattice waves. Lattice waves can be pictured as follows: the ions in a crystal are never quiescent; they execute small oscillations about their equilibrium positions. But, since an ion is strongly coupled to its neighbouring ions by elastic and other inter-ionic forces, the small oscillation spreads in the form of a disturbance through the crystal. This leads to a collective motion of the ions which gives rise to very important thermodynamic effects and produces interactions with other entities such as electrons and spin waves.

Therefore, when we are considering all the interactions, we should write the total Hamiltonian, instead of equation (1.1), by

$$H = H_L + H_{el} , \quad (1.19)$$

where  $H_L$  is the lattice Hamiltonian and  $H_{el}$  is the electron Hamiltonian including the two-body interactions of all types. Let us denote by  $\underline{p}_{l,b}$  and  $\underline{h}_{l,b}$  the momentum and the displacement of the  $b$ -th ion in the  $l$ -th unit cell ( here  $\underline{l}$  also denotes the vector to the unit cell from a fixed origin and  $\underline{b}$  the vector to the ion from a fixed ion in the cell ). Then  $H_L$  is explicitly given by

$$H_L = \frac{1}{2} \sum_{l,b} \left( \frac{1}{m_b} \right) p_{l,b} \cdot p_{l,b} + V, \quad (1.20)$$

where  $m_b$  is the mass of the b-th ion in the unit cell. The two terms in (1.20) represent the kinetic and the potential energies of the ions respectively. Now we expand the potential energy in Taylor series as

$$V = V_0 + \frac{1}{2} \sum_{l,b} \sum_{l',b'} \hbar_{l,b} \cdot \left[ \frac{\partial^2 V}{\partial \hbar_{l,b} \partial \hbar_{l',b'}} \right] \cdot \hbar_{l',b'} + \dots \quad (1.21)$$

Substituting this in (1.20), we get

$$H_L = \frac{1}{2} \sum_{l,b} \left( \frac{1}{m_b} \right) p_{l,b} \cdot p_{l,b} + \frac{1}{2} \sum_{l,b} \sum_{l',b'} \hbar_{l,b} \cdot \left[ \frac{\partial^2 V}{\partial \hbar_{l,b} \partial \hbar_{l',b'}} \right] \cdot \hbar_{l',b'} + \dots \quad (1.22)$$

Here also it is convenient to introduce second quantization by defining<sup>15</sup>

$$\left. \begin{aligned} \hbar_{l,b} &= -i \left( \frac{\hbar}{2Nm_b} \right)^{\frac{1}{2}} \sum_{q,p} e_{qbp} w_{qp}^{\frac{1}{2}} (b_{qp}^+ - b_{-qp}) e^{-iq \cdot \mathbf{r}_l} \\ p_{l,b} &= \left( \frac{\hbar m_b}{2N} \right)^{\frac{1}{2}} \sum_{q,p} e_{qbp} w_{qp}^{\frac{1}{2}} (b_{qp} + b_{-qp}^+) e^{iq \cdot \mathbf{r}_l} \end{aligned} \right\} \quad (1.23)$$

where  $\underline{q}$  and  $p$  are the wave vector and the branch number of the lattice wave respectively,  $\underline{e}_{\underline{q}p}$  the polarization vector and  $\omega_{\underline{q}p}$  the frequency of the lattice wave. In terms of the new operators, the Hamiltonian (1.22) becomes

$$H_L = \sum_{\underline{q}p} \hbar \omega_{\underline{q}p} (b_{\underline{q}p}^{\dagger} b_{\underline{q}p} + \frac{1}{2}) + \text{higher order terms,} \quad (1.24)$$

with  $N_{\underline{q}p} = b_{\underline{q}p}^{\dagger} b_{\underline{q}p}$  the occupation number operator which has only positive integers for its eigenvalues. From the analogy of (1.24) with (1.16) we can interpret the lattice vibrational field as consisting of a system of noninteracting particles called 'phonons', each having an energy  $\hbar \omega_{\underline{q}p}$ . The operator  $b_{\underline{q}p}^{\dagger}$  creates a phonon of wave vector  $\underline{q}$  in branch  $p$  and  $b_{\underline{q}p}$  destroys such a phonon. The eigenfunctions of the Hamiltonian (1.24) can be denoted in the number representation by  $|\dots N_{\underline{q}p} \dots\rangle$  and the above operators then have the properties

$$\left. \begin{aligned} b_{\underline{q}p}^{\dagger} |\dots N_{\underline{q}p} \dots\rangle &= (N_{\underline{q}p} + 1)^{\frac{1}{2}} |\dots N_{\underline{q}p} + 1 \dots\rangle, \\ b_{\underline{q}p} |\dots N_{\underline{q}p} \dots\rangle &= (N_{\underline{q}p})^{\frac{1}{2}} |\dots N_{\underline{q}p} - 1 \dots\rangle. \end{aligned} \right\} \quad (1.25)$$

They also satisfy the boson commutation relations

$$[b_{\underline{q}p}, b_{\underline{q}'p'}^{\dagger}] = \delta_{\underline{q}\underline{q}'} \delta_{pp'} \quad (1.26)$$

C. THE GREEN'S FUNCTION METHOD:

The spin wave method described in the first few pages has proved to be of great use in describing the behaviour of the ordered magnetic systems. It gives a very clear account of the low temperature excitations of the system and also gives expressions for magnetization which agree well with the experimental observations. But the main drawback of this method is that it is restricted to the low temperature region since the spin waves lose their meaning as the temperature rises. At temperatures near the Curie point, the molecular field method has been used to find the magnetization and susceptibility, etc. However, in this method, the correlation between the transverse components of the spins is ignored partly or wholly and consequently it fails at low temperatures as well as above the Curie point. For these reasons and in an attempt to formulate one single theory which is valid throughout the temperature range, the Green function method, which was developed for problems in field theory, was extended to statistical mechanics and magnetism.

This method is particularly useful owing to the simplicity in its formulation and interpretation and when combined with the spectral representations it provides a powerful tool for attacking various types of problems. The first application of the Green function method to problems of non-relativistic solid state theory<sup>16</sup> was in 1955 and was thereafter developed by a number of authors in connection with different statistical problems. All these authors,

however, restricted their discussions to the case  $T = 0$ . Matsubara<sup>17</sup>, for the first time, attempted to produce a method suited to finite temperatures; he, however, considered only time independent Green functions. It was not long after this that a complete generalization of this method was achieved by several workers, some of which are mentioned in the references.<sup>18-20</sup>

The Green functions can be defined in a variety of ways depending on the problem at hand, though the method of treating them is more or less the same. Thus, if  $A(x)$  and  $B(x')$  be any two operators, where  $x$  contains the spatial coordinate  $\underline{x}$  and the time coordinate  $t$ , then we can write the following definitions for the Green functions.<sup>22</sup>

$$\left. \begin{aligned} G_R(x, x') &= i\theta(t) \langle A(x)B(x') \rangle \equiv \langle\langle A(x); B(x') \rangle\rangle_R, \\ G_R^+(x, x') &= i\theta(t) \langle [A(x), B(x')] \rangle_+ \equiv \langle\langle A(x); B(x') \rangle\rangle_R^+, \end{aligned} \right\} \quad (1.27)$$

$$\left. \begin{aligned} G_A(x, x') &= -i\theta(-t) \langle A(x)B(x') \rangle \equiv \langle\langle A(x); B(x') \rangle\rangle_A, \\ G_A^+(x, x') &= -i\theta(-t) \langle [A(x), B(x')] \rangle_+ \equiv \langle\langle A(x); B(x') \rangle\rangle_A^+, \end{aligned} \right\} \quad (1.28)$$

$$G_C(x, x') = i \langle T A(x)B(x') \rangle \equiv \langle\langle A(x); B(x') \rangle\rangle_C. \quad (1.29)$$

Here  $\theta(t) = \begin{cases} 0, & \text{if } t < 0, \\ 1, & \text{if } t > 0. \end{cases}$

For any operator  $P$ ,  $\langle P \rangle$  denotes the average over a grand canonical ensemble,

$$\langle P \rangle = \frac{\text{Tr} ( P e^{-H/k_B T} )}{\text{Tr} ( e^{-H/k_B T} )} = Z^{-1} \text{Tr} ( P e^{-\beta H} ),$$

where  $H$  is the Hamiltonian,  $k_B$  the Boltzmann's constant and  $T$  the temperature. The different Green functions  $G_r$ ,  $G_a$  and  $G_c$  are known as 'retarded', 'advanced' and 'causal' Green functions respectively. The function of our interest is  $G_r^+$  and we shall denote it only by  $G$  in the following.

It is expedient to introduce the spectral function  $J_{BA}(\epsilon)$  which is the Fourier transform of the correlation function of the two operators  $A(t)$  and  $B(0)$  ( we have arbitrarily put  $t' = 0$  ) by

$$\langle B(0)A(t) \rangle e^{-\alpha|t|} = \int_{-\infty}^{\infty} J_{BA}(\epsilon) e^{-i\epsilon t} d\epsilon$$

$$= Z^{-1} \text{Tr} ( e^{-\beta H} B e^{iHt} A e^{-iHt} ) e^{-\alpha|t|}, \quad (1.30)$$

where we have introduced an infinitesimal convergence factor  $\alpha$  in order to ensure the convergence of the Fourier transform and in the Heisenberg representation,

$$A(t) = e^{iHt} A e^{-iHt}.$$

The inverse transform of (1.30) is

$$J_{BA}(\epsilon) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \langle B(0)A(t) \rangle e^{i\epsilon t - \alpha|t|} dt. \quad (1.31)$$

Continuing the functions (1.31) and (1.32)

Now we change the order of operation as follows:

$$\begin{aligned} \langle B(0)A(t) \rangle &= Z^{-1} \text{Tr} ( e^{-\beta H} B e^{iHt} A e^{-iHt} ) \\ &= Z^{-1} \text{Tr} ( e^{iHt} A e^{-iHt} e^{-\beta H} B ) \\ &= Z^{-1} \text{Tr} ( e^{-\beta H} e^{iH(t-i\beta)} A e^{-iH(t-i\beta)} B ) \\ &= \langle A(t-i\beta) B(0) \rangle \end{aligned}$$

then

$$J_{BA}(\epsilon) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \langle A(t-i\beta) B(0) \rangle e^{i\epsilon t - \alpha|t|} dt.$$

We can now distort the path of integration in the complex  $t$  plane by the analyticity condition and obtain

$$\begin{aligned} J_{BA}(\epsilon) &= \frac{e^{-\beta\epsilon}}{2\pi} \int_{-\infty+i\beta}^{\infty+i\beta} \langle A(s)B(0) \rangle e^{i\epsilon s - \alpha|s|} ds \\ &= \frac{e^{-\beta\epsilon}}{2\pi} \int_{-\infty}^{\infty} \langle A(t)B(0) \rangle e^{i\epsilon t - \alpha|t|} dt. \end{aligned}$$



Hence

$$e^{\beta E} J_{BA}(E) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \langle A(t)B(0) \rangle e^{iEt - \alpha|t|} dt. \quad (1.32)$$

Combining equations (1.31) and (1.32) we have

$$(e^{\beta E} \pm 1) J_{BA}(E) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \langle [A(t), B(0)]_{\pm} \rangle e^{iEt - \alpha|t|} dt. \quad (1.33)$$

Since  $t$  varies from  $-\infty$  to  $\infty$  this integral splits up into two parts in the following form:

$$(e^{\beta E} \pm 1) J_{BA}(E) = \frac{1}{2\pi i} \left[ \int_{-\infty}^{\infty} \langle\langle A(t); B(0) \rangle\rangle_{\pm} e^{i(E+i\alpha)t} dt + \int_{-\infty}^{\infty} \langle\langle A(t); B(0) \rangle\rangle_{\mp} e^{i(E-i\alpha)t} dt \right] \\ = \frac{1}{i} \left[ \langle\langle A; B \rangle\rangle_{E+i\alpha} - \langle\langle A; B \rangle\rangle_{E-i\alpha} \right], \quad (1.33)$$

where the Fourier transforms are defined by

$$\langle\langle A; B \rangle\rangle_{E+i\alpha} = \frac{1}{2\pi} \int_{-\infty}^{\infty} \langle\langle A(t); B(0) \rangle\rangle_{\pm} e^{i(E+i\alpha)t} dt, \\ \langle\langle A; B \rangle\rangle_{E-i\alpha} = \frac{1}{2\pi} \int_{-\infty}^{\infty} \langle\langle A(t); B(0) \rangle\rangle_{\mp} e^{i(E-i\alpha)t} dt. \quad (1.34)$$

The relation (1.33) is very important for the calculation of many properties. The equation of motion for the Green functions follows from the definitions (1.27) and (1.28),

$$i \frac{d}{dt} \langle\langle A(t); B(0) \rangle\rangle_{r,a} = -\chi(t) \langle[A, B]_{\pm}\rangle + \langle\langle [A(t), H]_{\pm}; B(0) \rangle\rangle_{r,a}, \quad (1.35)$$

where  $\delta(t)$  is the Dirac's  $\delta$ -function.

The Fourier transform of this equation yields:

$$E \langle\langle A; B \rangle\rangle_E = -\frac{1}{2\pi} \langle[A, B]_{\pm}\rangle + \langle\langle [A, H]_{\pm}; B \rangle\rangle_E \quad (1.36)$$

Since the spectral functions are related to  $\langle\langle A; B \rangle\rangle_{E \pm i\alpha}$ , it is required to know only the Fourier transform of the Green function. It is also seen that the Fourier transformed equation of motion is the same whether we start from the advanced or the retarded Green function.

The study carried out in the present work is based on these two methods; the spin wave method and the Green function method. We have obtained the spin wave spectrum of a ferrimagnet and the phonon-magnon relaxation time. The matrix elements for the phonon-magnon interaction and the transition probabilities are used to calculate the thermal conductivity of ferrimagnetic insulators at low temperatures. The spin wave spectrum is then calculated by Green function method which also gives the temperature dependence of magnetization. Finally, we have considered the interaction

of neutrons with magnetically ordered crystals. The neutron-magnon interaction terms are formulated and the neutron scattering cross sections are calculated for two-magnon Raman processes including only orbital transition of the crystal atoms.

CHAPTER - 2

SPIN WAVE METHOD FOR FERRIMAGNETISMA. Spin wave spectrum:

The method developed by Holstein and Primakoff and outlined in chapter 1 is useful for calculating the low energy excitations of the spin system, i.e., the spin wave spectrum. We consider a ferrimagnetic system with the body centred cubic structure. The two constituent sublattices are denoted by A and B. Let two different types of magnetic ions be situated at the lattice points of both the sublattices A and B having a few unpaired d or f electrons in addition to the closed shell. Let the spins be  $S_A$  and  $S_B$  and the anisotropy fields at the two sites,  $H_A$  and  $H_B$ . The total Hamiltonian of such a system can be written as

$$H = H_L + H_{el} + H_Z + H_{an} + H_c, \quad (2.1)$$

where  $H_L$  is the lattice Hamiltonian and  $H_{el}$  is the one-electron Hamiltonian

$$H_{el} = \sum_i \frac{P_i^2}{2m} + \sum_{i,n} U(\underline{r}_i - \underline{R}_n), \quad (2.2)$$

$H_c$  is the electron-electron Coulomb interaction of the form

$$H_c = \sum_{i \neq j} \frac{1}{2} \frac{e^2}{r_{ij}} = \frac{1}{2} \sum_{i \neq j} \epsilon_{ij};$$

$H_Z$  is the Zeeman term and  $H_{an}$  is the anisotropy energy. In (2.2),  $p_1^2/2m$  is the kinetic energy of the  $i$ -th electron and  $U(\underline{r}_1 - \underline{R}_n)$  is the potential energy at  $\underline{r}_1$  due to the ion core at  $\underline{R}_n$ . For the present, we neglect the spin-orbit interaction, dipolar interaction between magnetic ions and the anharmonic terms in lattice vibrations. A particular  $i$ -th electron will be acted upon by the potential due to all the ions of the crystal or the crystal field potential

$$V = \sum_{m \neq 1} U(\underline{r}_1 - \underline{R}_m). \quad (2.3)$$

Then, for the  $i$ -th electron, the Hamiltonian can be rewritten as

$$H_{el}^{(i)} = \frac{p_1^2}{2m} + U(\underline{r}_1 - \underline{R}_1^0) + V_0 + H', \quad (2.3)$$

with  $V = V_0 + H'$ , so that  $V_0$  is the static crystal field potential and  $H'$  represents the first and higher order terms in the Taylor series expansion of  $V$  in terms of the displacement  $\delta \underline{R}_n$

$$H' = \sum_{\underline{h}} \left( \frac{\partial V}{\partial \underline{R}_n} \right)_0 \cdot \delta \underline{R}_n + \frac{1}{2} \sum_{\underline{h}, \underline{h}'} \delta \underline{R}_n \cdot \left( \frac{\partial^2 V}{\partial \underline{R}_n \cdot \partial \underline{R}_{n'}} \right) \cdot \delta \underline{R}_{n'} + \dots \quad (2.4)$$

where  $\underline{R}_n$  is the perturbed ionic position and  $\delta \underline{R}_n$  obeys the following rules

$$\text{with } \underline{R}_n = \sum_i \underline{R}_{1k}^0 - \underline{R}_{m^0} \quad \delta \underline{R}_n = \delta \underline{R}_{ik}$$

Also,  $U(\underline{r}_1 - \underline{R}_1^0)$  represents the electrostatic potential acting on the electron when the ion core to which the electron belongs is in its equilibrium position.

Let us define the wave function  $\phi_{a\sigma}$  as the solution of

$$\left[ \frac{P^2}{2m} + U(\underline{r} - \underline{R}_1^0) + V_0 \right] \phi_{a\sigma} = \epsilon_{a\sigma} \phi_{a\sigma} \quad (2.5)$$

The eigenstates of the perturbed Hamiltonian (2.3) are

$$\Psi_{a\sigma} = \phi_{a\sigma} + \sum_{\alpha} \frac{H'_{\alpha a} \phi_{\alpha\sigma}}{(\epsilon_{\alpha} - \epsilon_a)} + \dots \quad (2.6)$$

where the excited states are indicated by the Greek letters. The wave functions  $\phi_{a\sigma}$  are orthogonal to each other. We now introduce the number operators  $A_{a\sigma}$  and  $A_{a\sigma}^+$  for the state  $\Psi_{a\sigma}$ . It is then possible to write the total Hamiltonian in the occupation number representation as

$$H = H_L + H_Z + H_{an} + \sum_{a,\sigma} \epsilon'_{a\sigma} A_{a\sigma}^+ A_{a\sigma} + \frac{1}{2} \sum_{abcd} \sum_{\sigma\sigma'} A_{a\sigma}^+ A_{b\sigma'}^+ \langle a\sigma, b\sigma' / \epsilon_{12} / c\sigma, d\sigma' \rangle A_{c\sigma} A_{d\sigma'} \quad (2.7)$$

where  $\epsilon'_{a\sigma}$  is the perturbed electron energy and the operators obey the following rules

$$A_i A_k^+ + A_k^+ A_i = \delta_{ik},$$

$$A_i^+ A_i = N_i,$$

where the subscripts i, k include both the orbital index a, b ... and the spin index  $\sigma, \sigma'$ . For non-degenerate orbital states, the following relations hold between these operators and the spin operators:

$$A_a^+(+)A_a(+) + A_a^+(-)A_a(-) = 1,$$

$$N_a(+) - N_a(-) = 2S_{az},$$

$$A_a^+(+)A_a(-) = S_a^+,$$

$$A_a^+(-)A_a(+) = S_a^-.$$

Now the ~~third~~ <sup>last</sup> term of (2.7) can be expanded by using (2.6) for the state functions, so that it becomes

$$\frac{1}{2^2} \sum_{abcd} \sum_{\sigma\sigma'} A_{a\sigma}^+ A_{b\sigma'}^+ \left\langle \left( \phi_a + \sum_{\alpha} \frac{H'_{a\alpha} \phi_{\alpha}}{\Delta E_{\alpha}} \right)_{\sigma} \left( \phi_b + \sum_{\beta} \frac{H'_{b\beta} \phi_{\beta}}{\Delta E_{\beta}} \right)_{\sigma'} \right. \\ \left. / \epsilon_{12} / \left( \phi_c + \sum_{\gamma} \frac{H'_{c\gamma} \phi_{\gamma}}{\Delta E_{\gamma}} \right)_{\sigma} \left( \phi_d + \sum_{\delta} \frac{H'_{d\delta} \phi_{\delta}}{\Delta E_{\delta}} \right)_{\sigma'} \right\rangle A_{c\sigma} A_{d\sigma'} \quad (2.8)$$

This expression contains the phonon-magnon interaction in various orders.



To carry out summation over  $\delta, \delta'$ , we allow them to take (+) and (-) values. The operator part becomes

$$\sum_{\delta \delta'} A_{a\delta}^+ A_{b\delta'}^+ A_{b\delta} A_{a\delta'}$$

$$= N_a(+)\mathbb{N}_b(+) + S_a^+ S_b^- + S_a^- S_b^+ + N_a(-)\mathbb{N}_b(-)$$

$$= \frac{1}{2} (1 + 4 S_a \cdot S_b)$$

We consider the first order term involving an excited orbital

$$\sum_{a,b} \sum_{\alpha} \frac{1}{2} \langle \phi_{\alpha}^{(1)} \phi_b^{(2)} / \epsilon_{12} / \phi_b^{(1)} \phi_a^{(2)} \rangle \frac{H'_{a\alpha}}{\Delta E_{\alpha}} (1 + 4 S_a \cdot S_b)$$

$$= \sum_{a,b} \sum_{\alpha} \frac{1}{4 \Delta E_{\alpha}} \langle \phi_{\alpha}^{(1)} \phi_b^{(2)} / \epsilon_{12} / \phi_b^{(1)} \phi_a^{(2)} \rangle \langle \phi_{\alpha} / H' / \phi_a \rangle$$

$$\cdot (1 + 4 S_a \cdot S_b)$$

Using the expansion (2.4) for  $H'$  upto first order, this becomes

$$\sum_{\text{hab}\alpha} \frac{1}{4 \Delta E_{\alpha}} \frac{\langle \phi_{\alpha} / \frac{\partial V}{\partial h} / \phi_a \rangle}{\Delta E_{\alpha}} \cdot \delta_{\text{hab}} (S_a \cdot S_b) \quad (2.9)$$

where we have dropped the constant factors, independent of spins.

The total Hamiltonian can then be separated into three parts, the lattice part, the pure magnon part and the phonon-magnon interaction terms. Thus, for a ferri-magnet,<sup>23</sup>

$$H = H_L + H_m + H_{int}$$

where

$$H_L = \sum_{\mathbf{q}} \hbar \omega_{\mathbf{q}} (b_{\mathbf{q}}^{\dagger} b_{\mathbf{q}} + \frac{1}{2}), \quad (2.10)$$

$$H_m = 2J_{AB} \sum_{\text{nei}} \underline{S}_{1A} \cdot \underline{S}_{mB} - (H+H_A) \sum_{\beta} S_{\beta zA} - (H-H_B) \sum_{\beta} S_{\beta zB}, \quad (2.11)$$

$$H_{int} = \sum_{\text{lmh}} 4 \left[ \frac{eJ(\mathbf{R}_h) \cdot \partial \mathbf{R}_h}{\hbar} \right] (\underline{S}_{1A} \cdot \underline{S}_{mB}) \quad (2.12)$$

with

$$J_{AB} = \left\langle \frac{\phi_1^{(1)} \phi_m^{(2)}}{\epsilon_{12}} / \frac{\phi_m^{(1)} \phi_1^{(2)}}{\epsilon_{12}} \right\rangle, \quad (2.11a)$$

$$eJ(\mathbf{R}_h) = \sum_{\alpha} \frac{\left\langle \frac{\phi_{\alpha} \phi_m / \epsilon_{12} / \phi_m \phi_1}{\Delta E_{\alpha}} \right\rangle \left\langle \frac{\phi_{\alpha} / \partial \mathbf{R}_h / \phi_1}{\Delta E_{\alpha}} \right\rangle}{\Delta E_{\alpha}} \quad (2.12a)$$

In the above, l and m run over the magnetic ions of sublattices, A and B respectively. H is the external magnetic field

assumed to be along the  $z$  direction and  $g_{A,B}$  the  $g$ -factors for the two sublattices. In point of fact, our  $J_{AB}$  and  ${}^0J(\underline{R}_h)$  contain in addition to the Heisenberg direct processes as in (2.11a) and (2.12a), the superexchange or indirect processes also. For example the superexchange transfer process can be written as

$$J_{AB} \rightarrow \frac{\langle 1 / T / m \rangle \langle m / T / 1 \rangle}{U},$$

$${}^0J(\underline{R}_h) \rightarrow \sum_{\alpha} \langle 1 / V' / \alpha \rangle \frac{\langle \alpha / T / m \rangle \langle m / T / 1 \rangle}{U}$$

with  $V' = \partial V / \partial \underline{R}_h$ ;  $T$  represents the electron transfer operator and  $U$  is the corresponding energy denominator.

The terms in (2.11) contain the exchange, Zeeman and anisotropic interactions of the spin system and will give us the spin wave spectrum. The term (2.12) represents the spin-lattice interaction. At finite temperatures, the lattice is in a vibrational state producing changes in the interatomic distance  $\underline{R}_h$ . The dependence of the exchange integral on the interatomic distance  $\underline{R}_h$  causes the spin-lattice interaction.

Now we define the spin deviation operators for the two sublattices

$$S_{1A}^+ = S_{1xA} + iS_{1yA} = (2S_A)^{\frac{1}{2}} \left(1 - \frac{n_1}{2S_A}\right)^{\frac{1}{2}} a_1,$$

$$S_{1A}^- = S_{1xA} - iS_{1yA} = (2S_A)^{\frac{1}{2}} a_1^+ \left(1 - \frac{n_1}{2S_A}\right)^{\frac{1}{2}}, \quad (2.13)$$

$$S_A - S_{1zA} = a_1^+ a_1 = n_1;$$

$$S_{mB}^+ = (2S_B)^{\frac{1}{2}} d_m^+ \left(1 - \frac{n_m}{2S_B}\right)^{\frac{1}{2}},$$

$$S_{mB}^- = (2S_B)^{\frac{1}{2}} \left(1 - \frac{n_m}{2S_B}\right)^{\frac{1}{2}} d_m, \quad (2.14)$$

$$S_B + S_{mzB} = d_m^+ d_m = n_m.$$

Here  $a_1^+$ ,  $a_1$  and  $d_m^+$ ,  $d_m$  are the creation and annihilation operators for the spin deviations at sites 1 and m respectively,  $n_{1,m}$  the spin deviation operator with positive integers for its eigenvalues.

Since the spin deviations do not remain localized at a particular site but manifest themselves and spread over the entire crystal lattice as an assembly of spin waves with different wavelengths, it is advisable to define the Fourier transforms of the above operators. Thus

$$\begin{aligned}
 a_{\underline{\lambda}} &= \left(\frac{2}{N}\right)^{\frac{1}{2}} \sum_{\underline{1}} \exp(i\underline{\lambda} \cdot \underline{R}_{\underline{1}}^0) a_{\underline{1}}, \\
 a_{\underline{\lambda}}^{\dagger} &= \left(\frac{2}{N}\right)^{\frac{1}{2}} \sum_{\underline{1}} \exp(-i\underline{\lambda} \cdot \underline{R}_{\underline{1}}^0) a_{\underline{1}}^{\dagger}, \\
 d_{\underline{\lambda}} &= \left(\frac{2}{N}\right)^{\frac{1}{2}} \sum_{\underline{m}} \exp(-i\underline{\lambda} \cdot \underline{R}_{\underline{m}}^0) d_{\underline{m}}, \\
 d_{\underline{\lambda}}^{\dagger} &= \left(\frac{2}{N}\right)^{\frac{1}{2}} \sum_{\underline{m}} \exp(i\underline{\lambda} \cdot \underline{R}_{\underline{m}}^0) d_{\underline{m}}^{\dagger},
 \end{aligned} \tag{2.15}$$

In terms of these operators, the spin wave Hamiltonian (2.2) becomes

$$\begin{aligned}
 H_m &= 2J_{AB} (S_A S_B)^{\frac{1}{2}} \sum_{\underline{\lambda}} z \gamma_{\underline{\lambda}} (a_{\underline{\lambda}} d_{\underline{\lambda}} + a_{\underline{\lambda}}^{\dagger} d_{\underline{\lambda}}^{\dagger}) \\
 &+ 2J_{AB} \sum_{\underline{\lambda}} z (S_A d_{\underline{\lambda}}^{\dagger} d_{\underline{\lambda}} + S_B a_{\underline{\lambda}}^{\dagger} a_{\underline{\lambda}}) \\
 &+ (H + H_A) \mathcal{E}_A \int_{\beta} \sum_{\underline{\lambda}} a_{\underline{\lambda}}^{\dagger} a_{\underline{\lambda}} - (H + H_B) \mathcal{E}_B \int_{\beta} \sum_{\underline{\lambda}} d_{\underline{\lambda}}^{\dagger} d_{\underline{\lambda}},
 \end{aligned}$$

where  $\gamma_{\underline{\lambda}}$  is defined in (1.18). It is seen that this Hamiltonian is not diagonal in terms of the spin wave operators. In order to diagonalize this part, we make use of the following canonical transformation<sup>24</sup>

$$\begin{aligned}
 a_{\underline{\lambda}} &= \alpha_{\underline{\lambda}} \cos h \theta_{\underline{\lambda}} + \beta_{\underline{\lambda}}^{\dagger} \sin h \theta_{\underline{\lambda}}, \\
 a_{\underline{\lambda}}^{\dagger} &= \alpha_{\underline{\lambda}}^{\dagger} \cos h \theta_{\underline{\lambda}} + \beta_{\underline{\lambda}} \sin h \theta_{\underline{\lambda}},
 \end{aligned}$$

$$d_{\Delta} = \alpha_{\Delta}^{\dagger} \sin h \theta_{\Delta} + \beta_{\Delta} \cos h \theta_{\Delta},$$

$$d_{\Delta}^{\dagger} = \alpha_{\Delta} \sin h \theta_{\Delta} + \beta_{\Delta}^{\dagger} \cos h \theta_{\Delta}, \quad (2.16)$$

where  $\alpha_{\Delta}^{\dagger}$ ,  $\alpha_{\Delta}$  and  $\beta_{\Delta}^{\dagger}$ ,  $\beta_{\Delta}$  now represent the creation and annihilation operators for the two types of quasi-particles, the magnons. In order that the non-diagonal terms in these operators vanish, we must have

$$\tan h(2\theta_{\Delta}) = \frac{-4J_{AB}(S_A S_B)^{\frac{1}{2}} z_{\Delta}}{2J_{AB}z(S_A + S_B) + (H + H_A)g_A \mu_B - (H - H_B)g_B \mu_B} \quad (2.17)$$

The spin Hamiltonian now becomes diagonal and takes the form

$$H_{\Sigma} = \sum_{\Delta} \hbar \omega_{\Delta}^{\alpha} (\alpha_{\Delta}^{\dagger} \alpha_{\Delta} + \frac{1}{2}) + \sum_{\Delta} \hbar \omega_{\Delta}^{\beta} (\beta_{\Delta}^{\dagger} \beta_{\Delta} + \frac{1}{2}), \quad (2.18)$$

where it is evident that the spin wave spectrum splits up into two branches with energies  $\hbar \omega_{\Delta}^{\alpha, \beta}$  which are given, after neglecting the external and anisotropy fields and a constant term, by

$$E_{\Delta}^{\alpha} = \hbar \omega_{\Delta}^{\alpha} = \frac{4J_{AB} S_A S_B}{|S_A - S_B|} \lambda^2 a^2, \quad (2.19)$$

$$E_{\Delta}^{\beta} = \hbar \omega_{\Delta}^{\beta} = 2J_{AB}z(|S_A - S_B|) + \frac{4J_{AB} S_A S_B}{|S_A - S_B|} \lambda^2 a^2,$$

where we have considered the long wavelength region in which  $\lambda \cdot R_n \ll 1$  and 'a' is the lattice constant. Magnons belonging to the first branch  $E_{\Delta}^{\alpha}$  are said to be the 'acoustic' magnons and the dispersion relation, for our system, is parabolic in wave vector. The second branch  $E_{\Delta}^{\beta}$  represents the 'optical' magnons whose energy is much higher than those of the acoustic magnons.

### B. Magnon-Phonon Relaxation:

In order to obtain the interaction terms, the Hamiltonian  $H_{int}$  must be subjected to the same transformations which are applied to the magnon part  $H_m$ . After applying the spin wave transformations (2.13) and (2.14) to  $H_{int}$ , (2.12) becomes

$$\begin{aligned}
 H_{int} = & -i \frac{4}{\sqrt{N}} \left( \frac{\hbar}{2w_{QP} m} \right)^{\frac{1}{2}} \sum_{l,m} a_{QP} \cdot e^{iJ(\underline{R}_n)} (b_{QP}^+ - b_{-QP}) \\
 & \times \left[ (S_A S_B)^{\frac{1}{2}} (a_{1m} d_m + a_{1m}^+ d_m^+) + (S_A d_m^+ d_m + S_B a_{1m}^+ a_{1m}) \right] \\
 & \times ( e^{i\mathbf{q} \cdot \underline{R}_l^0} - e^{i\mathbf{q} \cdot \underline{R}_m^0} ) + \dots
 \end{aligned}$$

Now we apply the Fourier transformations (2.15) and the canonical transformation (2.16) to the above expression.

The summation over  $l$  and  $m$  is carried out by using the interference conditions such as

$$\sum_{l,m} \exp(i\lambda l - i\mu m) = \sum_{l,m} \exp(i\lambda l - i\mu m) \exp(i\lambda l - i\mu m)$$

Also since the expression is summed over  $\lambda$  and  $q$  which run through the first Brillouin zone, we are allowed to make such changes in the variables as

$$\lambda \rightarrow \lambda - q \quad \text{or} \quad q \rightarrow -q, \quad \text{etc.}$$

After some calculation we get,

$$H_{\text{int}} = \sum_{\lambda, q} A_{\lambda q} (\alpha_{\lambda} \alpha_{\lambda-q}^{\dagger} b_q^{\dagger} - \alpha_{\lambda}^{\dagger} \alpha_{\lambda-q} b_q) + B_{\lambda q} (\alpha_{\lambda} \beta_{\lambda-q} b_q^{\dagger} - \alpha_{\lambda}^{\dagger} \beta_{\lambda-q}^{\dagger} b_q) + C_{\lambda q} (\beta_{\lambda} \beta_{\lambda-q}^{\dagger} b_q - \beta_{\lambda}^{\dagger} \beta_{\lambda-q} b_q^{\dagger}) \quad (2.20)$$

with

$$A_{\lambda q} = D \left\{ (\gamma_{\lambda-q} - \gamma_{\lambda}) (S_A S_B)^{\frac{1}{2}} \sin h (\theta_{\lambda-q} - \theta_{\lambda}) + (1 - \gamma_{\lambda}) (S_B \cos h \theta_{\lambda} \cos h \theta_{\lambda-q} - S_A \sin h \theta_{\lambda} \sin \theta_{\lambda-q}) \right\},$$



$$\begin{aligned}
 B_{\underline{\lambda}q} &= D \left\{ (r_{\underline{\lambda}-q} - r_{\underline{\lambda}}) (S_A S_B)^{\frac{1}{2}} \cos h (\theta_{\underline{\lambda}-q} - \theta_{\underline{\lambda}}) \right. \\
 &\quad \left. + (1 - r_q) (S_B \cos h \theta_{\underline{\lambda}} \sin h \theta_{\underline{\lambda}-q} - S_A \sin h \theta_{\underline{\lambda}} \cos h \theta_{\underline{\lambda}-q}) \right\}, \\
 C_{\underline{\lambda}q} &= D \left\{ (r_{\underline{\lambda}-q} - r_{\underline{\lambda}}) (S_A S_B)^{\frac{1}{2}} \sin h (\theta_{\underline{\lambda}-q} - \theta_{\underline{\lambda}}) \right. \\
 &\quad \left. + (1 - r_q) (S_A \cos h \theta_{\underline{\lambda}} \cos h \theta_{\underline{\lambda}-q} - S_B \sin h \theta_{\underline{\lambda}} \sin h \theta_{\underline{\lambda}-q}) \right\}
 \end{aligned}
 \tag{2.21}$$

with  $D = -i \frac{4}{\sqrt{N}} \left( \frac{\hbar}{2w_q m} \right)^{\frac{1}{2}} \sum_{\underline{h}} \frac{e_{\underline{q}} \cdot e_{\underline{h}}}{\hbar} J(\underline{h})$  and we have dropped the polarization suffix  $p$ .

The Hamiltonian (2.20) shows explicitly the interaction of the acoustic and the optical magnons with the acoustic phonons. Thus the first term of (2.20) involves interactions of the acoustic phonons with the acoustic magnons and the last term expresses the interaction of acoustic phonons with optical magnons. These processes will conserve energy. However, since the energy of the optical magnon is much higher than that of the acoustic magnon, the strength of the last term will depend on the excitation of the higher frequency branch and at low temperatures, its population density is expected to be low. The processes connected with the second term of (2.20) which involves creation (annihilation) of two magnons, one in the lower branch and the other in the

higher branch, with the annihilation ( creation ) of an acoustic phonon do not seem to be possible in the long wavelength region in view of non-conservation of energy.

Thus in further calculations we drop the second term of (2.20). The phonon system is considered to be at a lower temperature  $T_1$  and the magnon system at  $T_m$ . Since the magnon-magnon relaxation time is known to be much smaller than the phonon-magnon relaxation time, the magnon-system can immediately attain equilibrium and we will have  $T_m = T$ , the temperature of the crystal. The excited phonons will come to the equilibrium by gaining energy from the spin system. According to time-dependent perturbation theory, the probability of transition from a state  $|\beta_i\rangle$  with energy  $E_i$  to a state  $|\beta_f\rangle$  with energy  $E_f$  is

$$W_{if} = \frac{2\pi}{\hbar} |(H_p)_{if}|^2 \delta(E_i - E_f),$$

where  $(H_p)_{if}$  is the matrix element of the perturbing potential  $H_p$  between  $|\beta_i\rangle$  and  $|\beta_f\rangle$ . Accordingly, the transition probabilities for the above processes are given by:

$$W_1(n_{\Delta}, n_{\Delta-q}, N_q \rightarrow (n_{\Delta}-1), (n_{\Delta-q}+1), (N_q+1)) \\ = \frac{2\pi}{\hbar} |A_{\Delta q}|^2 (n_{\Delta})(n_{\Delta-q}+1)(N_q+1) \delta(E_{\Delta-q} + E_q - E_{\Delta}),$$

$W_1(n_{\Delta}, n_{\Delta-q}, N_q \rightarrow (n_{\Delta}+1)(n_{\Delta-q}-1), (N_q-1))$  down the different body distribution functions with respective energy parameter  $= \frac{2\pi}{\hbar} |A_{\Delta q}|^2 (n_{\Delta}+1)(n_{\Delta-q})(N_q) \delta(E_{\Delta-q} + E_q - E_{\Delta})$ , the term in  $W_1$  corresponds to first order in  $\Delta T$ . The final expression is

$$W_3(n'_{\Delta}, n'_{\Delta-q}, N_q \rightarrow (n'_{\Delta}-1), (n'_{\Delta-q}+1), (N_q-1)) \\ = \frac{2\pi}{\hbar} |C_{\Delta q}|^2 (n'_{\Delta})(n'_{\Delta-q}+1)(N_q) \delta(E_{\Delta} + E_q - E_{\Delta-q}),$$

$$W_3(n'_{\Delta}, n'_{\Delta-q}, N_q \rightarrow (n'_{\Delta}+1), (n'_{\Delta-q}-1), (N_q+1)) \\ = \frac{2\pi}{\hbar} |C_{\Delta q}|^2 (n'_{\Delta}+1)(n'_{\Delta-q})(N_q+1) \delta(E_{\Delta} + E_q - E_{\Delta-q}).$$

Here  $N_q$  is the distribution function for phonons,  $n_{\Delta}$  and  $n'_{\Delta}$  the distribution functions for the magnons of lower and higher branches respectively. The rate of energy transfer from the magnon to the phonon system will be given by

$$\dot{Q} = \dot{Q}_{\alpha} + \dot{Q}_{\beta} = \sum_{\Delta q} \hbar \omega_q (\langle N_q \rangle_{\alpha} + \langle N_q \rangle_{\beta}) \\ = \frac{2\pi}{\hbar} \sum_{\Delta q} \hbar \omega_q \left\{ |A_{\Delta q}|^2 \left\{ (n_{\Delta})(n_{\Delta-q}+1)(N_q) \right. \right. \\ \left. \left. + |C_{\Delta q}|^2 \left\{ (n'_{\Delta}+1)(n'_{\Delta-q})(N_q+1) - (n'_{\Delta})(n'_{\Delta-q}+1)(N_q) \right\} \right\} \delta(E_{\Delta-q} + E_q - E_{\Delta}) \right. \\ \left. + |C_{\Delta q}|^2 \left\{ (n'_{\Delta}+1)(n'_{\Delta-q})(N_q+1) - (n'_{\Delta})(n'_{\Delta-q}+1)(N_q) \right\} \delta(E_{\Delta} + E_q - E_{\Delta-q}) \right\} \\ \dots (2.22)$$

These expressions are evaluated by writing down the different Bose distribution functions with respective energy parameters, writing  $\Delta T = T - T_1$  and expanding the terms in Taylor's series to first order in  $\Delta T$ . The final expression for  $\dot{Q}$  is

$$\dot{Q} = \frac{2\pi}{h} \frac{\Delta T}{T^2} \sum_{\Delta, q} \frac{(\hbar \omega q)^2}{k_B} \left\{ F^\alpha(\lambda q) |A_{\Delta q}|^2 e^{E_\Delta/k_B T} \delta(E_{\Delta-q} + E_q - E_\Delta) + F^\beta(\lambda q) |C_{\Delta q}|^2 e^{E_{\Delta-q}/k_B T} \delta(E_\Delta + E_q - E_{\Delta-q}) \right\} \quad (2.23)$$

with

$$F^{\alpha, \beta}(\lambda q) = \left[ (e^{E_\Delta^{\alpha, \beta}/k_B T} - 1) (e^{E_{\Delta-q}^{\alpha, \beta}/k_B T} - 1) (e^{E_q/k_B T} - 1) \right]^{-1}$$

Under the approximation  $\lambda q \ll 1$ , the constants  $A_{\Delta q}$  and  $C_{\Delta q}$  can be evaluated and to the lowest order in wavelength, they have the form

$$|A_{\Delta q}|^2 = |C_{\Delta q}|^2 = \frac{16}{h} \frac{\hbar}{w_q} (\epsilon J)^2 \frac{(S_A + S_B)^2}{2} q^4 a^4 \quad (2.24)$$

In the expression (2.23) for  $\dot{Q}$ , we change summation over  $\Delta$  and  $q$  to integration. To illustrate the procedure, we will consider the first part containing  $|A_{\Delta q}|^2$ , denoting it by  $\dot{Q}_1$ . Thus

$$\dot{Q}_1 = \frac{2\pi}{h} \frac{\Delta T}{T^2} \left[ \frac{\hbar v}{(2\pi)^3} \right]^2 \iint \frac{(\hbar \omega q)^2}{k_B} F^\alpha(\lambda q) |A_{\Delta q}|^2 e^{E_\Delta/k_B T} \delta(E_{\Delta-q} + E_q - E_\Delta) d\gamma_q d\gamma_\Delta$$

The result after the integration over the  $\gamma$  variables

with

$$d\gamma_{\underline{q}} = q^2 \sin \theta_{\underline{q}} dq d\theta_{\underline{q}} d\phi_{\underline{q}}$$

and  $d\gamma_{\underline{\lambda}} = \lambda^2 \sin \theta_{\underline{\lambda}} d\lambda d\theta_{\underline{\lambda}} d\phi_{\underline{\lambda}}$ .

$c$  is the velocity of sound.

The  $\delta$ -function is expanded in the following manner to facilitate integration over the angle variables:

$$\begin{aligned} \delta(E_{\underline{\lambda}-\underline{q}}^{\alpha} + E_{\underline{q}} - E_{\underline{\lambda}}^{\alpha}) &= \delta \left[ k_B \Theta_C a^2 ( (\underline{\lambda}-\underline{q})^2 - \underline{\lambda}^2 ) + k_B \Theta_D q a \right] \\ &= \frac{1}{2k_B \Theta_C a^2 \lambda q} \delta \left( \frac{q}{2\lambda} + \frac{\Theta_D}{2\Theta_C a} - \cos \theta_{\underline{\lambda}\underline{q}} \right), \end{aligned} \quad (2.25)$$

with  $k_B \Theta_C = 4 J_{AB} S_A S_B / (|S_A - S_B|)$  (2.25a)

and  $\Theta_D$  the Debye temperature of the crystal.

Here we write

$$\cos \theta_{\underline{\lambda}\underline{q}} = \cos \theta_{\underline{\lambda}} \cos \theta_{\underline{q}} + \sin \theta_{\underline{\lambda}} \sin \theta_{\underline{q}} \cos (\phi_{\underline{\lambda}} - \phi_{\underline{q}})$$

and with  $x = \cos (\phi_{\underline{\lambda}} - \phi_{\underline{q}})$  the  $\delta$ -function becomes

$$\frac{1}{2\lambda q \cdot k_B \Theta_C a^2 \sin \theta_{\underline{\lambda}} \sin \theta_{\underline{q}}} \cdot \delta(b - x),$$

where

$$b = \frac{1}{\sin \theta_{\underline{\lambda}} \sin \theta_{\underline{q}}} \left( \frac{q}{2\lambda} + \frac{\Theta_D}{2\Theta_C a} - \cos \theta_{\underline{\lambda}} \cos \theta_{\underline{q}} \right).$$

The result after the integration over the angular variables is

The quantity of  $P$  is given by the integral

$$P = \int \int q^6 \lambda F(\lambda q) e^{-E_\lambda / k_B T} d\lambda dq$$

with  $\frac{1}{\lambda^2} \leq \eta \leq \infty$

$$P = \frac{\pi^3 \Delta T}{T^2} \left[ \frac{Nv}{(2\pi)^3} \right]^2 \frac{\hbar}{k_B N M} \left[ e^{J(\frac{\hbar}{h})} \right]^2 (S_A + S_B)^2 \frac{\Theta_D}{\Theta_C} a^3.$$

where  $\eta = \alpha^2$

We introduce the following dimensionless variables at this stage

$$\eta = \frac{E^\alpha}{k_B T} = \frac{\Theta_C a^2}{T} \lambda^2$$

and

$$\xi = \frac{E_q}{k_B T} = \frac{\Theta_D a}{T} q.$$

The limits of integration are obtained from the  $\delta$ -function condition (2.25)

$$\frac{q}{2\lambda} + \frac{\Theta_D}{2\Theta_C a \lambda} = \cos \theta_{\lambda q}$$

or

$$\frac{1}{4\lambda^2} \left( q + \frac{\Theta_D}{\Theta_C a} \right)^2 \leq 1,$$

which gives  $\eta \geq \frac{(5 + \beta)^2}{4\beta}$

with  $\beta = \Theta_D^2 / \Theta_C T$ .

The ranges of integration are therefore chosen to be

$$\frac{(\xi + \beta)^2}{4\beta} \leq \eta \leq \infty, \\ 0 \leq \xi \leq \infty.$$

The integration is performed in the low temperature limit and including the second part of (2.23), we have

$$\dot{Q} = \frac{718.8}{\pi^2} \frac{\hbar N}{M} (S_A + S_B)^2 (\theta_J)^2 \frac{T^6 \Delta T}{k_B \theta_C^2 \theta_D^6} \exp(-\beta/4) \\ \left\{ 1 + \exp\left(-\frac{4(S_A - S_B)^2}{3 S_A S_B} \frac{\theta_C}{T}\right) \right\} \quad (2.26)$$

It can be seen that in the low temperature region ( T below 10°K ) the second term in the curly bracket of (2.26) which arises owing to the interaction of phonons with the higher magnon branch is negligible compared with the first. In the calculation of the relaxation time, we neglect this term. The relaxation time is related to  $\dot{Q}$  by<sup>25</sup>

$$\frac{1}{\tau_{mp}} = \frac{\dot{Q} \left( \frac{1}{C_s} + \frac{1}{C_l} \right)}{\Delta T} \quad (2.27)$$

where  $C_s$  and  $C_l$  are the spin and lattice specific heats of the crystal respectively. The lattice specific heat is known to be<sup>13</sup>

$$C_l = 234 N k_B \left( \frac{T}{\theta_D} \right)^3.$$

The spin specific heat can be easily calculated using the dispersion relation for the acoustic branch from (2.19), thus

$$C_s = \sum_{\Delta} \frac{dE_s}{dT}$$

and  $E_s = \sum_{\Delta} \frac{E_{\Delta}^{\alpha}}{e^{E_{\Delta}^{\alpha}/k_B T} - 1}$

After simple calculations we obtain for a b.c.c. crystal

$$C_s = \frac{Nk_B}{18.69} \left( \frac{T}{\theta_C} \right)^{3/2}$$

Using these values, the magnon-phonon relaxation time is given by

$$\frac{1}{\tau_{mp}} = \frac{718.8}{\pi^2} \left( \frac{\hbar}{Mk_B^2} \right) (S_A + S_B)^2 (\theta_J)^2 \frac{T^6}{\theta_C^2 \theta_D^6} e^{-\beta/4} (18.69) \left( \frac{\theta_C}{T} \right)^{3/2} + \frac{1}{234} \left( \frac{\theta_D}{T} \right)^3 \quad (2.28)$$

It is to be noted that, as in the case of a true ferromagnet, there is an exponential factor  $\exp(-\theta_D^2/4\theta_C T)$  in the expression for  $1/\tau_{mp}$ ; however, the other temperature factors are different in the present case.



and  
 C. Renormalization of the spin wave spectrum:

The spin wave spectrum derived in (2.19) for a ferrimagnet is valid only near the absolute zero of temperature. As the temperature rises, lattice vibrations come into play. Owing to their interaction with spin waves, the pure spin wave modes will no longer exist, and we must picture the elementary excitations as being coupled spin-phonon modes. The spin wave energy should therefore be appropriately renormalized after taking account of these interactions.

Similar calculations were first performed by Fröhlich<sup>26</sup> for electrons in the field of lattice vibrations and he obtained the renormalization of the velocity of sound and the interaction parameter. The procedure here is based on obtaining a canonical transformation and then choosing the transformation so that the first order terms vanish. We write the total Hamiltonian for our system as

$$H = H_0 + H_1$$

where  $H_0$  contains the diagonal spin and phonon parts,

$$H_0 = H_m + H_1 = \sum_{\Delta} \hbar \omega_{\Delta} \left( \alpha_{\Delta}^{\dagger} \alpha_{\Delta} + \frac{1}{2} \right) + \sum_{\underline{q}} \hbar \omega_{\underline{q}} \left( b_{\underline{q}}^{\dagger} b_{\underline{q}} + \frac{1}{2} \right)$$

and  $H_1$  is the interaction term involving acoustic branch of magnons ( first term of eq. (2.20) ) with frequency  $\omega_{\Delta q}$ . Thus

$$H_1 = \sum_{\Delta q} A_{\Delta q} ( \alpha_{\Delta} \alpha_{\Delta-q}^{\dagger} b_q^{\dagger} - \alpha_{\Delta}^{\dagger} \alpha_{\Delta-q} b_q ) .$$

Now we define a canonical transformation by

$$\begin{aligned} H_T &= e^{-iS} H e^{iS} \\ &= H_0 + (H_1 + i [H_0, S]) + \frac{1}{2} [H_1, S] + \dots, \end{aligned} \quad (2.29)$$

where  $S$  is suitably chosen as

$$S = \sum_{\Delta q} ( G_{\Delta q} \alpha_{\Delta} \alpha_{\Delta-q}^{\dagger} b_q^{\dagger} + G_{\Delta q}^* \alpha_{\Delta}^{\dagger} \alpha_{\Delta-q} b_q ) \quad (2.30)$$

with the constants  $G_{\Delta q}$  and  $G_{\Delta q}^*$  to be determined by the condition

$$H_1 + i [H_0, S] = 0 . \quad (2.31)$$

Using (2.30) for  $S$  and substituting in (2.31), we obtain for the constants

$$G_{\Delta q} = G_{\Delta q}^* = \frac{i A_{\Delta q}}{(E_{\Delta-q} + E_q - E_{\Delta})} . \quad (2.23)$$

Now using  $S$  along with the above constants, we calculate the remaining term of (2.29), i.e.,  $\frac{1}{2} [H_1, S]$ . It is seen

that this gives us some additional diagonal terms in the spin wave operators which will combine with  $H_0$  to give the renormalized frequency. Thus

$$H_T = \sum_{\underline{\lambda}} \hbar \omega_{\underline{\lambda}} (\alpha_{\underline{\lambda}}^+ \alpha_{\underline{\lambda}} + \frac{1}{2}) - 2 \sum_{\underline{\lambda}, \underline{q}} i A_{\underline{\lambda}, \underline{q}} G_{\underline{\lambda}, \underline{q}} (\alpha_{\underline{\lambda}}^+ \alpha_{\underline{\lambda}} + \frac{1}{2}) + 2 \sum_{\underline{\lambda}, \underline{q}} i (A_{\underline{\lambda}+\underline{q}, \underline{q}} G_{\underline{\lambda}+\underline{q}, \underline{q}} - A_{\underline{\lambda}, \underline{q}} G_{\underline{\lambda}, \underline{q}}) \langle N_{\underline{q}} \rangle (\alpha_{\underline{\lambda}}^+ \alpha_{\underline{\lambda}} + \frac{1}{2}) + \dots \quad (2.33)$$

where, as before,  $\langle N_{\underline{q}} \rangle = \langle b_{\underline{q}}^+ b_{\underline{q}} \rangle$  is the phonon distribution function. From this equation, the renormalized frequency becomes

$$\hbar \omega_{\underline{\lambda}}^r = \hbar \omega_{\underline{\lambda}} - 2i \sum_{\underline{q}} A_{\underline{\lambda}, \underline{q}} G_{\underline{\lambda}, \underline{q}} + 2i \sum_{\underline{q}} (A_{\underline{\lambda}+\underline{q}, \underline{q}} G_{\underline{\lambda}+\underline{q}, \underline{q}} - A_{\underline{\lambda}, \underline{q}} G_{\underline{\lambda}, \underline{q}}) \langle N_{\underline{q}} \rangle \quad (2.34)$$

This expression can be evaluated by changing summation into integration and noting the expressions for the parameters,

$$\log N_{\underline{q}} = \frac{1}{e^{\hbar \omega_{\underline{q}} / k_B T} - 1}$$

and (see eq. 2.16)

$$A_{\underline{\lambda}, \underline{q}} = -1 \sqrt{\frac{4}{V N}} \left( \frac{\hbar}{w_{\underline{q}} M} \right)^{\frac{1}{2}} (e^J) \frac{(S_A + S_B)}{z} q^2 a^2$$

The expression has been integrated in the low temperature approximations and for  $q \cdot a \ll 1$ . The second term of (2.34) only gives a temperature-independent energy shift. Thus, we have

$$\hbar\omega_{\Delta}^r = \hbar\omega_{\Delta} - \frac{(\theta_J)^2 (S_A + S_B)^2 a^6 k_B^4 T^5}{\pi^2 \hbar^4 c^6 M z^2 \theta_C} \cdot \frac{10^2}{a^{\lambda}} \log \left( 1 + \frac{8pa\lambda}{1-4pa\lambda} \right), \quad (2.36)$$

where we have assumed  $\hbar\omega_{\Delta} \gg k_B T$ , and  $p = \frac{\theta_C}{\theta_D}$ .

A few conclusions can be drawn from the above expression. The frequency of the spin waves decreases with the increase of temperature and the dependence is  $T^5$ . The second term of (2.36) is expected to approach the first term in magnitude before  $100^\circ\text{K}$ . Apparently, it shows that the spin wave energy of the acoustic mode diminishes rapidly. This only suggests that the acoustic magnon branch, which we have been considering, becomes unstable near this temperature and only the optical branch <sup>may</sup> play an important role at higher temperatures. The  $\lambda$ -dependent factor

$$\frac{1}{\lambda} \log \left( 1 + \frac{8pa\lambda}{1-4pa\lambda} \right)$$

seems to be almost constant over the range of  $\lambda = 10^4$  to  $10^6 \text{ cm}^{-1}$ . A similar treatment of the optical branch  $\omega_{\Delta}^{\beta}$  may improve the results at higher temperatures.

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### CHAPTER - 3

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## TRANSPORT DUE TO SPIN WAVES

### A. Thermal Conductivity of Ferrimagnetic Insulators:

The phonon-magnon interaction studied in the last chapter is useful for studying the transport properties of magnetic solids. For magnetically concentrated systems this interaction is found to be the overriding process.

Our object here is to study the thermal conductivity of simple magnetic ferrites. In the case of ferromagnetic metals, the conduction electron contribution to transport phenomena suppresses the spin-phonon effects. However, in the case of antiferromagnetic and ferrimagnetic insulators the conduction electron contribution is almost negligible and the spin-wave contribution can be studied in detail.

Thus, let us consider a simple b.c.c. ferrimagnet, as envisaged before. We consider a temperature gradient existing in the crystal. Since, at low temperatures, the magnon-magnon relaxation time is known to be much shorter than the phonon-magnon relaxation time, we consider the magnon system to be in equilibrium, and, as such the magnons do not take part in the transport of the heat current.<sup>27</sup> Thus only phonons, having a ~~gradient~~ <sup>gradient</sup> of the occupation number, are supposed to be the carriers of heat.

In the equilibration process, the phonon system will relax through different mechanisms such as the boundary scattering, mass defect and strain field scattering and spin wave scattering.

We thus start with the Hamiltonian

$$H = H_p + H_m + H_{p-p} + H_{p-m} + H_{p-d} \quad (3.1)$$

where the first two terms respectively stand for the diagonal parts of the phonon and the magnon Hamiltonians,  $H_{p-p}$  represents phonon-phonon interaction and  $H_{p-m}$  and  $H_{p-d}$  represent the phonon-magnon and phonon-defect terms respectively. However, in the following treatment, we have neglected the phonon-phonon or anharmonic terms, restricting ourselves to the low temperature region. We are considering only the acoustic branch of magnons and hence we take

$$H_{p-m} = \sum_{\Delta, q} A_{\Delta q} (\alpha_{\Delta} \alpha_{\Delta-q}^+ b_q^+ - \alpha_{\Delta}^+ \alpha_{\Delta-q} b_q) \quad (3.2)$$

Also

$$H_{p-d} = \sum_{q, q'} D_{qq'} (b_q^+, b_q + \text{h.c.}) \quad (3.3)$$

The coefficient  $D_{qq'}$ , arising from various sources has been given by Klemens<sup>28</sup> and is

$$D_{qq'}^2 = \frac{M^2}{N^2} w_q^2 w_{q'}^2 S^2$$

where  $M$  is the mass of the host atom,  $N$  the number of atoms and  $w_q$  the angular phonon frequency corresponding to  $q$ .  $S$  is given by

where  $S^2 = S_1^2 + (S_2 + S_3)^2$ , dimensions of the sample is a number. This is independent of the phonon where  $S_1$  is the contribution due to mass defect

$$S_1 = \frac{\Delta M}{M} \frac{1}{2\sqrt{3}} \quad (3.6)$$

$\Delta M$  being the mass difference;  $S_2$  arises from a difference in elastic constants,

$$S_2 = \frac{d(\epsilon^2)}{\epsilon^2} \frac{1}{\sqrt{6}}$$

where  $\epsilon = w/q$  is the velocity of waves;  $S_3$  is due to an elastic strain field around a point imperfection

our aim is to calculate the contribution due to the strain field  $S_3 = \frac{d(\epsilon^2)}{\epsilon^2} \frac{1}{\sqrt{6}}$

where  $Q$  is a constant of the order of 3-4. The number  $N_q$  is given by

The phonon relaxation time  $\tau$  is defined by the Boltzmann's equation

$$\frac{\partial N_q}{\partial t} = \frac{N_q^0 - N_q}{\tau} \quad (3.4)$$

where  $N_q$  is the phonon distribution number and  $N_q^0$  its equilibrium value. The contribution to  $\tau$  due to boundary and mass defect scattering is already known. Thus,



$$\frac{1}{\tau_{\text{(bound)}}} = \lambda = \frac{vc}{l}, \quad (3.5)$$

where  $c$  is the sound velocity,  $l$  the dimensions of the sample and  $v$  a number. This is independent of the phonon wave vector. Also

$$\frac{1}{\tau_{p-d}} = \beta \zeta^4, \quad (3.6)$$

where  $\beta$  is a constant and  $\zeta$  is conveniently defined in terms of the phonon wave vector by

$$\zeta = \frac{\hbar \omega_q}{k_B T} = \frac{\hbar c q}{k_B T}. \quad (3.7)$$

Thus our main aim is to calculate the contribution due to magnon interaction,  $\tau_{p-m}$ , from (3.4).

As before ( see 2.22 ) the rate of change of the occupation number  $N_q$  is given by

$$\begin{aligned} \left( \frac{\partial N_q}{\partial t} \right)_{p-m} = & \frac{2\pi}{\hbar} \sum_{\Delta} |A_{\Delta q}|^2 \left\{ (n_{\Delta})(n_{\Delta-q} + 1)(N_q + 1) \right. \\ & \left. - (n_{\Delta} + 1)(n_{\Delta-q})(N_q) \right\} \delta(E_{\Delta-q} + E_q - E_{\Delta}). \quad (3.8) \end{aligned}$$

Changing summation to integration and integrating out the angle variables to eliminate the  $\delta$ -function as before, we obtain

and

$$\left(\frac{\partial N_q}{\partial t}\right)_{p-m} = \frac{1}{h} \frac{\Delta T}{T} \frac{E_q}{k_B T} \frac{Nv}{8\pi k_B \Theta_C q a^2} \int |A_{\lambda q}|^2 F(\lambda q) \times e^{E_{\lambda}/k_B T} \lambda d\lambda, \quad (3.9)$$

in a Taylor series in powers of  $q$ .

where the various factors have been defined in chapter 2. Now integrating over the magnon wave vector  $\lambda$  the above expression gives

$$\left(\frac{\partial N_q}{\partial t}\right)_{p-m} = \frac{\Delta T}{T} \frac{E_q}{k_B T} \cdot \frac{v \cdot (eJ)^2 (S_A + S_B)^2 T}{\pi k_B \Theta_C c M z^2} \times \frac{e^{-\beta}}{(e^{-\beta} - 1)} q^2 \exp \left\{ - \frac{(\gamma + \beta)^2}{4\beta} \right\}. \quad (3.10)$$

The right hand side of the Boltzmann's equation (3.4) is also obtained by using the phonon distribution function and gives

$$\frac{N_q^0 - N_q}{T_{p-m}} = \frac{1}{T_{p-m}} \cdot \frac{\Delta T}{T} \frac{E_q}{k_B T} \cdot \frac{e^{-\beta}}{(e^{-\beta} - 1)^2}. \quad (3.11)$$

Therefore, equating (3.10) and (3.11) yields we get

$$\frac{1}{T_{p-m}} = R \cdot f(q), \quad (3.12)$$

where

$$R = \frac{v \cdot (eJ)^2 (S_A + S_B)^2 T}{\pi k_B c \Theta_C^2 M z^2}$$

$$\text{and } f(q) = q^2 (e^{\xi} - 1) \exp \left\{ - \frac{(\xi + \beta)^2}{4\beta} \right\},$$

To obtain the explicit  $q$ -dependence of  $f(q)$ , we expand it in a Taylor series in powers of  $q$ ,

$$f(q) = f(0) + qf'(0) + q^2 f''(0) + \dots$$

Retaining the lowest order term in this and changing over to  $\xi$ , we get

$$\frac{1}{\tau_{p-m}} = Q \xi^3 \quad (3.13)$$

with

$$Q = \frac{6k_B v(\theta_J)^2 (S_A + S_B)^2 T^3 \exp(-\beta/4)}{\pi \hbar^2 c^3 \theta_C^2 M z^2} \quad (3.14)$$

Thus, it is seen that the magnon contribution to the phonon relaxation time varies as the third power of the phonon wave vector for a ferrimagnet.

The total relaxation time is then given by combining (3.5, 3.6, 3.13) :

$$\frac{1}{\tau(\xi)} = A + Q \xi^3 + P \xi^4 \quad (3.15)$$

Manganese ferrite (MnFe<sub>2</sub>O<sub>4</sub>) and yttrium iron garnet (Y<sub>3</sub>Fe<sub>5</sub>O<sub>12</sub>) (YIG). The thermal conductivity of a substance is directly related to this phonon relaxation time by<sup>29</sup>

$$K = \frac{k_B^4 T^3}{2\pi^2 \hbar^3 c} \int_0^{\Theta_D/T} \xi^4 \gamma(\xi) \frac{e^{-\xi}}{(e^{\xi}-1)^2} d\xi,$$

which, with the help of (3.15), becomes

$$K = \frac{k_B^4 T^3}{2\pi^2 \hbar^3 c} \int_0^{\Theta_D/T} \frac{\xi^4}{(A + \frac{2}{3}\xi + \frac{1}{3}\xi^2)} \frac{e^{-\xi}}{(e^{\xi}-1)^2} d\xi. \quad (3.16)$$

This integral, in general, cannot be evaluated in a closed form. The integration is therefore performed graphically in the temperature range 5°K to 25°K. The discussion and comparison with experimental results is given in the following section.<sup>30</sup>

#### B. Comparison with experimental results:

Although our theory is based on a simple, two-sublattice b.c.c. structure, no such ferrima net is known to exist. The theory can, however, be applied to complicated systems while confining one's attention to the 'acoustic' branches of phonons and magnons and to low temperature regions. The insulating ferrites and garnets do indeed constitute the next ideal systems for comparing the results with the theory. For this purpose, we have chosen two systems, manganese ferrite ( $MnFe_2O_4$ ) and yttrium iron garnet ( $Y_3Fe_2Fe_3O_{12}$ ) (YIG).

In  $MnFe_2O_4$ , both the cations at tetrahedral ( $Mn^{2+}$ ) and octahedral ( $Fe^{3+}$ ) sites have the configuration  $d^5$  in  ${}^6S_{5/2}$  state. The system therefore will not present any charge exchange effects. The tetrahedral (A) and the octahedral (B) magnetic ion systems can be roughly approximated to a two sublattice ferrimagnet with a predominant A-B interaction. The various parameters entering into Q(3.14) have the following values;

$$\begin{aligned}
 v &= a^3 = (8.51 \times 10^{-8})^3 \text{ c.c.}, & S_A &= 5/2, \\
 S_B &= 2 \times 5/2 = 5, & \Theta_D &= 380^\circ K, \\
 \Theta_C &= 783^\circ K, & M &= 55 \times 10^{-24} \text{ gms.}, \\
 z &= 6, & c &= 10^5 \text{ cms./sec.}
 \end{aligned}$$

Here  $\Theta_D$  has been derived from the melting point data<sup>31</sup> in that the low temperature specific heat results for this material are not available. Also

$$\begin{aligned}
 (\Theta_J) &= 3.33 \times 10^{-6} \text{ dynes}, \\
 A &= 2 \times 10^{10} \text{ sec}^{-1}, & F &= 2 \times 10^8 \text{ sec}^{-1},
 \end{aligned}$$

so that

$$Q = 7.5 \times 10^6 T^3 \exp(-\beta/4) \text{ sec}^{-1}.$$

The theoretical curve drawn with these values of the parameters is shown along with the experimental points of Douthett and Friedberg<sup>32</sup> in fig. 1 in the temperature range

5°K to 25°K. As mentioned above, it is believed that beyond 15°K the three phonon processes take over.

Another type of ferrimagnets, to which the present calculations can be extended, are the magnetic garnets. Here we have chosen Yttrium iron garnet (YIG); the experimental measurements of thermal conductivity of YIG are reported by Douglass.<sup>33</sup> In this system also, the magnetic ions are distributed on two types of crystallographic sites. The cation sites are classified as a, c, d and accordingly the formula  $Y_3^{(c)} Fe_2^{(a)} Fe_3^{(d)} O_{12}$  expresses the distribution. Here also, the  $Fe^{3+}$  ions are in  $6S_{5/2}$  states ( $d^5$  configuration) and the ions at 'a' sites are coupled antiferromagnetically to those of 'd' sites. In view of the unequal number of ions in the sublattices, we get a ferrimagnetic system. For comparison with experiments, we have set out the following values of the parameters:

$$v = a^3 = (12.376 \times 10^{-8})^3 \text{ c.c.},$$

$$S_A = 5, \quad S_B = 7.5,$$

$$\theta_C = 550^\circ K, \quad \theta_D = 500^\circ K,$$

$$c = 3.843 \times 10^5 \text{ cms./sec.},$$

$$M = 92.27 \times 10^{-24} \text{ gms.},$$

$$(\theta_J/z) = 1.674 \times 10^{-7} \text{ dynes.}$$

The theoretical curve ranging from 5°K to 25°K is shown along with the experimental points in fig. 2. The agreement

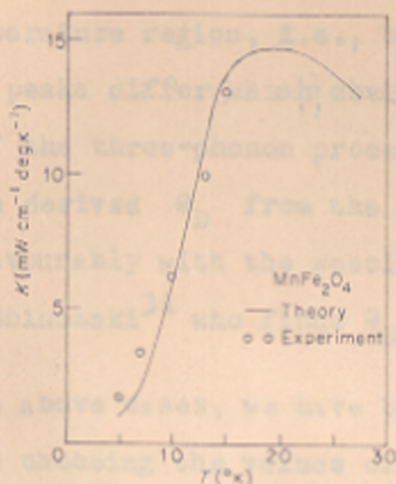


Figure 1. Thermal conductivity of  $MnFe_2O_4$ . Experimental points of Douthett and Friedberg are shown alongside method of estimation outlined by Blaha and Spachys.

After this choice of  $\tau$ ,  $\lambda$  and  $\nu$  are so chosen that they correspond to one point of the experimental curve. In each case the value  $\tau$  chosen falls in the range which would lead to the right order of relaxation time for the system. In both the figures the agreement with theory is very good in the region where spin wave contributions are expected to be important, i.e. around 10°K.

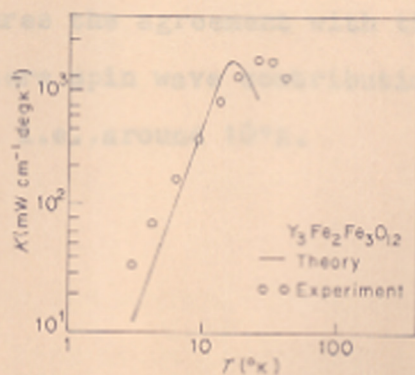


Figure 2. Thermal conductivity of  $Y_3Fe_2Fe_3O_{12}$ . Experimental points of Douglass are shown alongside

seems to be satisfactory although deviations appear in the very low temperature region, i.e., below 5°K. Also the conductivity peaks differ which obviously is owing to the appearance of the three-phonon processes at about 15°K. Here also we have derived  $\Theta_D$  from the melting point data which compares favourably with the specific heat measurements of Shinozaki<sup>34</sup> who finds  $\Theta_D$  of the same order.

In both the above cases, we have been guided by two considerations in choosing the values of the parameters  $\Theta_J$ , A and P. First we choose a suitable value of  $\Theta_J$  by a method of estimation outlined by Sinha and Upadhyaya.<sup>13,31</sup> After this choice of  $\Theta_J$ , A and P are so chosen that they correspond to one point of the experimental curve. In each case the value  $\Theta_J$  chosen falls in the range which would lead to the right order of relaxation time for the system. In both the figures the agreement with theory is very good in the region where spin wave contributions are expected to be important, i.e., around 10°K.



1. The function  $f(x) = x^2 + 3x - 4$  is a parabola opening upwards.

2. The vertex of the parabola is at  $(-1.5, -6.25)$ .

3. The x-intercepts are  $x = -4$  and  $x = 1$ .

4. The y-intercept is  $(0, -4)$ .

5. The graph is symmetric about the vertical line  $x = -1.5$ .

6. The maximum value of the function is  $-6.25$ .

7. The function is increasing on the interval  $(-1.5, \infty)$ .

8. The function is decreasing on the interval  $(-\infty, -1.5)$ .

**CHAPTER - 4**

9. The area of the triangle is  $10$  square units.

GREEN FUNCTION METHOD FOR FERRIMAGNETISM

The Green function theory for ferromagnetism has been developed by several authors explaining the temperature dependence of magnetization<sup>35-42</sup>, susceptibility and electrical resistivity<sup>43</sup> and has been extended to antiferromagnetism<sup>39</sup> and to more complicated structures such as simple spiral.<sup>38</sup> In the present work, we extend this theory to ferrimagnetism. Here we have evaluated the spin wave spectrum and the low temperature dependence of magnetization, which we consider in the following sections:

A: Spin Wave Spectrum:

We start with the spin dependent Hamiltonian (2.11)

$$H = 2 \sum_{l,m} J(l-m) \underline{S}_{lA} \cdot \underline{S}_{mB} - (H+H_A) g_A \mu_B \sum_l S_{lzA} - (H-H_B) g_B \mu_B \sum_m S_{mzB},$$

where we have used the exchange integral  $J(l-m)$  between the  $l$ -th and the  $m$ -th atoms instead of  $J_{AB}$ . In this chapter we have used units in which  $\hbar = 1$ . We define the two Green functions corresponding to the two sublattices<sup>44</sup>

$$G_t^a(l, l') = \langle\langle S_{lA}^+(t); e^{aS_{l'zA}} S_{l'A}^- \rangle\rangle = i \theta(t) \langle [S_{lA}^+(t), e^{aS_{l'zA}} S_{l'A}^-] \rangle,$$

$$F_t^a(m, l') = \langle\langle S_{mB}^+(t), e^{aS_{l'zA}} S_{l'A}^- \rangle\rangle$$

$$= i \Theta(t) \langle\langle [S_{mB}^+(t), e^{aS_{l'zA}} S_{l'A}^-] \rangle\rangle, \quad (4.1)$$

where 'a' is a real number. The equation of motion of these functions will involve such commutators as

$$[S_{lA}^+, H] \quad \text{and} \quad [S_{mB}^+, H], \quad \text{which are found to be}$$

$$[S_{lA}^+, H] = (H + H_A) g_A \mu_B S_{lA}^+ + 2 \sum_m J(1-m) (S_{lzA} S_{mB}^+ - S_{lA}^+ S_{mzB}),$$

$$[S_{mB}^+, H] = (H + H_B) g_B \mu_B S_{mB}^+$$

$$+ 2 \sum_l H(1-m) (S_{lzA} S_{mB}^+ - S_{lA}^+ S_{mzB}).$$

Using these in the equation of motion (1.35), we obtain

$$i \frac{d}{dt} G_t^a(l, l') = \Theta(t) \delta(t) \delta_{ll'} + (H + H_A) g_A \mu_B G_t^a(l, l') + 2 \sum_m J(1-m) \langle\langle (S_{lzA} S_{mB}^+ - S_{lA}^+ S_{mzB}); e^{aS_{l'zA}} S_{l'A}^- \rangle\rangle,$$

$$i \frac{d}{dt} F_c^a(m, l') = (H - H_B) \epsilon_{B, \mu\beta} F_c^a(m, l') - 2 \sum_I J(1-m) \langle\langle (S_{1zA} S_{mB}^+ - S_{1A}^+ S_{mzB}); e^{aS_{1'zA}} S_{1'A}^- \rangle\rangle,$$

where  $\Theta(a) = \langle\langle [S_{1A}^+, e^{aS_{1'zA}} S_{1'A}^-] \rangle\rangle$ . (4.3)

Equations (4.2) contain Green functions of higher order and we must have some criterion to decouple them. The usual decoupling approximation is due to Bogolyubov and Tyablikov<sup>20</sup>,

$$\langle\langle S_i^z S_j^+; B \rangle\rangle \xrightarrow{I \neq J} \langle S_i^z \rangle \langle\langle S_j^+; B \rangle\rangle,$$

where  $B$  is some operator. This means that we neglect the correlation between the  $S^z$  operator on the  $i$ -th site and the  $S^+$  operator on the  $j$ -th site. This decoupling procedure was improved by Callen<sup>40</sup> who uses

$$\langle\langle S_i^z S_j^+; B \rangle\rangle \xrightarrow{I \neq J} \langle S_i^z \rangle \langle\langle S_j^+; B \rangle\rangle - \alpha \langle S_i^- S_j^+ \rangle \langle\langle S_i^+; B \rangle\rangle, \quad (4.4)$$

where  $\alpha$  is a parameter. An argument based on a physical criterion that at any temperature  $\langle S^z \rangle - S^z$  should be self consistently small leads to the value

$$\alpha = \frac{\langle S^z \rangle}{2S^2}.$$

For our ferrimagnetic system with two sublattices, we use

$$\langle\langle S_i^z S_j^+; B \rangle\rangle \xrightarrow{i \neq j} \langle S_i^z \rangle \langle\langle S_j^+; B \rangle\rangle - \alpha_{A,B} \langle S_i^- S_j^+ \rangle \langle\langle S_i^+; B \rangle\rangle \quad (4.5)$$

with

$$\alpha_{A,B} \text{ being } = \frac{\langle S_{zA,B} \rangle}{2 S_A S_B}.$$

Using the decoupling

$\alpha_A$  or  $\alpha_B$  is to be used depending on whether the operator  $S^z$  which is split up into  $S^+$  and  $S^-$  is for the sublattice A or B. Recently, the decoupling procedure has been improved<sup>41</sup> by setting up the equation of motion of the higher order Green function instead of decoupling it when it occurs in the equation of motion of the lowest order Green function. This equation contains a still higher order Green function which is then approximated by an order of magnitude argument. Another improvement of Callen's approximation is due to Copeland and Qersch<sup>45</sup> who take  $\alpha = \langle S^z \rangle^x / 2 S^{x+1}$  and find that best agreement with experiments is obtained when  $x = 3$  giving  $\alpha = \langle S^z \rangle^3 / 2 S^4$ . However, as Tahir-Kheli and ter Haar<sup>35</sup> put it, the procedure of decoupling is at present far from being well understood, and here we have used Callen's approximation.

In the second stage, we Fourier transform the Green functions. Now we Fourier transform the Green functions, in the first stage, from time to energy variables by

$$G_{\mathbf{t}}^{\mathbf{a}}(1,1') = \int_{-\infty}^{\infty} G_{\mathbf{E}}^{\mathbf{a}}(1,1') e^{-i\mathbf{E}\mathbf{t}} d\mathbf{E}$$

and

$$F_{\mathbf{t}}^{\mathbf{a}}(m,1') = \int_{-\infty}^{\infty} F_{\mathbf{E}}^{\mathbf{a}}(m,1') e^{-i\mathbf{E}\mathbf{t}} d\mathbf{E}.$$

Using the decoupling equation (4.5) along with these transforms, equations (4.2) give

$$\begin{aligned} G_{\mathbf{E}}^{\mathbf{a}}(1,1') & \left[ \mathbf{E} - (\mathbf{H} + \mathbf{H}_A) \mathbf{g}_{A, \mu\beta} + \sum_m J(1-m) \left\{ 2 \langle S_{zB} \rangle - 2\alpha_B \langle S_{1A}^- S_{mB}^+ \rangle \right\} \right] \\ & - \sum_m F_{\mathbf{E}}^{\mathbf{a}}(m,1') \left[ J(1-m) \left\{ 2 \langle S_{zA} \rangle - 2\alpha_A \langle S_{1A}^+ S_{mB}^- \rangle \right\} \right] \\ & - \frac{\Theta(\mathbf{g})}{2\pi} \delta_{\mathbf{t}\mathbf{t}'} = 0, \end{aligned} \quad (4.6a)$$

$$\begin{aligned} \sum_I G_{\mathbf{E}}^{\mathbf{a}}(1,1') & \left[ J(1-m) \left\{ 2 \langle S_{zB} \rangle - 2\alpha_A \langle S_{1A}^- S_{mB}^+ \rangle \right\} - F_{\mathbf{E}}^{\mathbf{a}}(m,1') \right] \\ & \left[ \mathbf{E} - (\mathbf{H} + \mathbf{H}_B) \mathbf{g}_{B, \mu\beta} + \sum_I J(1-m) \left\{ 2 \langle S_{zA} \rangle - 2\alpha_B \langle S_{1A}^+ S_{mB}^- \rangle \right\} \right] = 0. \end{aligned} \quad (4.6b)$$

In the second stage, we Fourier transform the different functions to the reciprocal lattice space by

$$\begin{aligned}
 G_{\mathbb{E}}^a(l, l') &= \frac{2}{N} \sum_{\Delta} e^{i(l-l') \cdot \Delta} G_{\mathbb{E}}^a(\Delta), \\
 F_{\mathbb{E}}^a(m, l') &= \frac{2}{N} \sum_{\Delta} e^{i(m-l') \cdot \Delta} F_{\mathbb{E}}^a(\Delta), \\
 \langle e^{i a S_{1'A} S_{1'A}^+ S_{mB}^-} \rangle &= \frac{2}{N} \sum_{\Delta} e^{i(m-l') \cdot \Delta} \Psi_{\mathbb{E}}(\Delta, a), \\
 J(m-l') &= \frac{2}{N} \sum_{\Delta} e^{i(m-l') \cdot \Delta} J(\Delta).
 \end{aligned}
 \tag{4.7}$$

With the use of these transforms, equations (6) now give us two simultaneous equations in the two Green functions

$$\begin{aligned}
 (E-d_1) G_{\mathbb{E}}^a(\Delta) - f_1(\Delta) F_{\mathbb{E}}^a(\Delta) - \frac{\alpha A}{2\pi} &= 0, \\
 f_2(\Delta) G_{\mathbb{E}}^a(\Delta) - (E-d_2) F_{\mathbb{E}}^a(\Delta) &= 0
 \end{aligned}
 \tag{4.8}$$

with

$$\begin{aligned}
 d_1 &= (H+H_A) \mathcal{E}_A \mu_B - 2 \langle S_{zB} \rangle J(0) - \frac{4\alpha A}{N} \sum_{\Delta'} J(\Delta') \mathcal{N}_F(\Delta', 0), \\
 f_1(\Delta) &= 2 \langle S_{zA} \rangle J(\Delta) + \frac{4\alpha B}{N} \sum_{\Delta'} J(\Delta - \Delta') \mathcal{N}_F(\Delta', 0), \\
 d_2 &= (H-H_B) \mathcal{E}_B \mu_A - 2 \langle S_{zA} \rangle J(0) - \frac{4\alpha B}{N} \sum_{\Delta'} J(\Delta') \mathcal{N}_F(\Delta', 0), \\
 f_2(\Delta) &= 2 \langle S_{zB} \rangle J(\Delta) + \frac{4\alpha A}{N} \sum_{\Delta'} J(\Delta - \Delta') \mathcal{N}_F(\Delta', 0).
 \end{aligned}
 \tag{4.9}$$

These simultaneous equations are similar to those obtained for the antiferromagnetic case<sup>39</sup> but there is a significant

difference in the energy parameters of (4.9). Solving these equations, we obtain

$$\left. \begin{aligned} G_E^a(\lambda) &= \frac{\Theta(a)}{2\pi(\Delta_+(\lambda) - \Delta_-(\lambda))} \left\{ \frac{\Delta_+(\lambda) - d_2}{E - \Delta_+(\lambda)} - \frac{\Delta_-(\lambda) - d_2}{E - \Delta_-(\lambda)} \right\}, \\ F_E^a(\lambda) &= \frac{\Theta(a)}{4\pi} \frac{f_2(\lambda)}{(\Delta_+(\lambda) - \Delta_-(\lambda))} \left\{ \frac{1}{E - \Delta_+(\lambda)} - \frac{1}{E - \Delta_-(\lambda)} \right\}, \end{aligned} \right\} \quad (4.10)$$

where the energy expressions are

$$\Delta_{\pm}(\lambda) = \frac{1}{2} \left[ d_1 + d_2 \pm \left\{ (d_1 - d_2)^2 + 4f_1(\lambda)f_2(\lambda) \right\}^{\frac{1}{2}} \right]. \quad (4.11)$$

The above Green functions possess singularities and it is well known that the poles of the Green function give the energies of the elementary excitations<sup>46</sup> of the system considered. Thus for our spin wave system there are two branches of the quasi-particle energy,  $\Delta_{\pm}(\lambda)$ . We evaluate these in the low temperature limit. To make any further progress one is forced to make certain approximations owing to the complexity of the parameters in equation (4.9). Firstly we neglect all exchange interactions except between nearest neighbours, so that, for the system considered

$$J(\lambda) = J_{AB} \sum_{\delta} \exp(i\lambda \cdot \delta) = J_{AB}^2 f_{\lambda}, \quad (4.12)$$

where  $\delta$  is the vector to the nearest neighbour. Secondly, at low temperatures, we assume that the z-components of spins are not much different from the total spin magnitude.

Thus



$$\langle S_{zA} \rangle \rightarrow S_A ; \text{ and } \langle S_{zB} \rangle \rightarrow -S_B.$$

The function  $\Psi_F(\underline{\lambda}, 0)$  is the Fourier transform of  $\langle S_{1A}^- S_{mB}^+ \rangle$  which, in effect, is the propagator of spin deviation. As the temperature tends to zero, the spins will become more and more localized giving rise to negligible cross-correlation.<sup>47</sup> Thus, for our purpose, we can approximate  $\Psi_F$  to zero. Lastly, we neglect the anisotropy fields and the external field. These approximations lead to a good deal of simplification of the parameters in (4.9) and we have

$$\left. \begin{aligned} d_1 &= 2S_B J_{AB} z, & d_2 &= -2S_A J_{AB} z, \\ f_1(\underline{\lambda}) &= 2S_A J_{AB} z \gamma_{\underline{\lambda}}, & f_2(\underline{\lambda}) &= -2S_B J_{AB} z \gamma_{\underline{\lambda}}. \end{aligned} \right\} (4.13)$$

Using these expressions in (4.11) we obtain

$$\left. \begin{aligned} \Delta_+(\underline{\lambda}) &= \frac{4J_{AB} S_A S_B}{|S_A - S_B|} \lambda^2 a^2, \\ \Delta_-(\underline{\lambda}) &= 2J_{AB} z (|S_A - S_B|) + \frac{4J_{AB} S_A S_B}{|S_A - S_B|} \lambda^2 a^2. \end{aligned} \right\} (4.14)$$

These equations for the quasi-particle energies are identical with those obtained earlier by the spin wave method<sup>23</sup> (see (2.19)). Thus the spectrum of spin waves at low temperatures consists of two distinct branches, the first corresponds to the 'acoustic' mode and the second to the 'optic'<sub>al</sub> mode.

### B. Magnetization of the ferrimagnet:

Now we are in a position to calculate the magnetization of the system. As discussed in the introduction and enumerated in many other articles,<sup>22,48,49</sup> the Fourier transforms  $\Psi(\Delta, a)$  of the correlation functions, which are called the spectral density functions, are related to the Green functions by

$$\left. \begin{aligned} \Psi_G(\Delta, a) &= i \int_{-\infty}^{\infty} \frac{G_{E+i\alpha}^a(\Delta) - G_{E-i\alpha}^a(\Delta)}{e^{E/k_B T} - 1} e^{-iEt} dE, \\ \Psi_F(\Delta, a) &= i \int_{-\infty}^{\infty} \frac{F_{E+i\alpha}^a(\Delta) - F_{E-i\alpha}^a(\Delta)}{e^{E/k_B T} - 1} e^{-iEt} dE, \end{aligned} \right\} (4.15)$$

where  $\alpha$  is an infinitesimal factor. By using (4.10) for the Green functions the above expressions can be integrated. The procedure will be shown for  $\Psi_G(\Delta, a)$ .

By substituting (4.10) in (4.15) and putting  $t = 0$  we obtain

$$\Psi_G(\Delta, a) = \frac{i \Theta(a)}{2\pi(\Delta_+(\Delta) - \Delta_-(\Delta))} \int_{-\infty}^{\infty} (e^{E/k_B T} - 1)^{-1} \left\{ \frac{\Delta_+(\Delta) - d_2}{E - \Delta_+(\Delta) + i\alpha} - \frac{\Delta_-(\Delta) - d_2}{E - \Delta_-(\Delta) - i\alpha} \right\} dE.$$

The integration is easily performed by a standard theorem

$$\int \frac{F(x)}{x-a+i0} dx = P \int \frac{F(x)}{x-a} dx + i\pi F(a),$$

where  $P$  indicates the principal value of the integral.

Carrying out the integration for  $\Psi_F$  in a similar way, we obtain

$$\left. \begin{aligned} \Psi_G(\lambda, a) &= \Theta(a) \phi_G(\lambda), \\ \Psi_F(\lambda, a) &= \Theta(a) \phi_F(\lambda) \end{aligned} \right\} \quad (4.16)$$

with

$$\left. \begin{aligned} \phi_G(\lambda) &= \frac{1}{\Delta_+(\lambda) - \Delta_-(\lambda)} \left\{ \frac{\Delta_+(\lambda) - d_2}{e^{\Delta_+(\lambda)/k_B T} - 1} - \frac{\Delta_-(\lambda) - d_2}{e^{\Delta_-(\lambda)/k_B T} - 1} \right\}, \\ \phi_F(\lambda) &= \frac{f_2(\lambda)}{\Delta_+(\lambda) - \Delta_-(\lambda)} \left\{ \frac{1}{e^{\Delta_+(\lambda)/k_B T} - 1} - \frac{1}{e^{\Delta_-(\lambda)/k_B T} - 1} \right\}. \end{aligned} \right\} \quad (4.17)$$

We define\*

$$\left. \begin{aligned} \Psi(a) &= \frac{2}{N} \sum_{\lambda} \Psi(\lambda, a), \\ \Phi &= \frac{2}{N} \sum_{\lambda} \phi(\lambda), \end{aligned} \right\} \quad (4.18)$$

so that (4.16) takes the form

$$\Psi(a) = \Theta(a) \cdot \Phi. \quad (4.19)$$

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\*Wherever the subscript is dropped, it can be either G or F.

The quantity  $\bar{\Psi}(0)$  is directly related to magnetization and the magnetization, in turn, is related to  $\bar{\Psi}$  by the equation ( see equation (52) of Callen<sup>40</sup> )

$$\langle S_z \rangle = \frac{(S-\bar{\Psi})(1+\bar{\Psi})^{2S+1} + (S+1+\bar{\Psi})(\bar{\Psi})^{2S+1}}{(1+\bar{\Psi})^{2S+1} - (\bar{\Psi})^{2S+1}},$$

which at low temperatures, can be reduced by expanding in powers of  $\bar{\Psi}$  to

$$\langle S_z \rangle = S - \bar{\Psi} + O(\bar{\Psi}^{2S+1}). \quad (4.20)$$

Thus our object is to evaluate  $\bar{\Psi}_G$  and  $\bar{\Psi}_F$  which will straightaway give the magnetization.

From equations (4.14) it is seen that  $\Delta_-(\lambda)$  contains the large term  $2J_{AB}z(\phi_A - S_B)$  ('optical' branch). Since this appears in the exponential in the denominator of (4.17) it will give a negligible contribution<sup>4,23</sup> as compared to that due to the first terms of (4.17) at low temperatures. Now we use (4.17) in (4.18) to evaluate  $\bar{\Psi}$  and change summation over  $\Delta$  to integration to obtain:

$$\bar{\Psi}_G = \frac{2}{N} \frac{Na^3}{2\pi^2} \int \lambda^2 d\lambda \cdot \frac{\Delta_+(\lambda) - d_z}{\Delta_+(\lambda) - \Delta_-(\lambda)} \cdot \frac{1}{e^{\Delta_+(\lambda)/k_B T} - 1}.$$

At low temperatures, the exponential term can be expanded as

$$\frac{1}{e^{\Delta_+/k_B T} - 1} = \frac{e^{-\Delta_+/k_B T}}{1 - e^{-\Delta_+/k_B T}} = \sum_{\lambda=1}^{\infty} e^{-\lambda \Delta_+/k_B T}$$

Using expression for  $\Delta_+$ , the integral simply reduces to Gaussian integral and yields

$$\begin{aligned} \bar{\Psi}_G &= \frac{S_A}{4\pi^{3/2} |S_A - S_B|} \zeta\left(\frac{3}{2}\right) \left(\frac{T}{\Theta_C}\right)^{3/2} + O(T^{5/2}), \\ \bar{\Psi}_F &= \frac{S_B}{4\pi^{3/2} |S_A - S_B|} \zeta\left(\frac{3}{2}\right) \left(\frac{T}{\Theta_C}\right)^{3/2} + O(T^{5/2}), \end{aligned} \quad (4.21)$$

where  $\Theta_C$  is defined in (2.25a).

Writing the equation (4.20) for the two sublattices in the form

$$\begin{aligned} \langle S_{zA} \rangle &= S_A - \bar{\Psi}_G, \\ \langle S_{zB} \rangle &= -S_B - \bar{\Psi}_F, \end{aligned}$$

we obtain

$$\begin{aligned}
 \langle S_z(\text{Total}) \rangle &= \langle S_{zA} \rangle + \langle S_{zB} \rangle \\
 &= (S_A - S_B) - \frac{\zeta(\frac{3}{2})}{32\pi^{3/2}} \left\{ \frac{k_B T (|S_A - S_B|)}{J_{AB} S_A S_B} \right\}^{3/2} + \dots
 \end{aligned}
 \tag{4.22}$$

This gives us the low temperature dependence of magnetization. It can be noted that this dependence agrees well with that obtained<sup>4</sup> by the spin wave method for a b.c.c. ferrimagnet and exhibits the  $T^{3/2}$  dependence of magnetization.

Thus we have evaluated expressions for the spin wave frequencies (4.14) and the magnetization (4.22). In both the cases we have restricted the calculations to the low temperature region. As the temperature rises the correlation functions such as  $\langle S_{1A}^- S_{mB}^+ \rangle$  can no longer be neglected. In this case the equations involve such sums as

$$\sum_{\underline{\lambda}'} J(\underline{\lambda} - \underline{\lambda}') \chi_{\underline{\lambda}'}(\underline{\lambda}, 0)$$

which are, in general, difficult to solve without some specific approximations. Nevertheless, the exact agreement of the expressions for the quasi-particle energies and magnetization at low temperatures obtained here with those of the spin wave theory is very encouraging and suggests a strong possibility of extending this theory to high temperatures.

## CHAPTER - 5

1. Introduction

2. Objectives

3. Conclusion

### MAGNETIC SCATTERING OF NEUTRONS

The present chapter is devoted to a topic somewhat different from those studied in the first few chapters. However, there exists a great similarity between phonon-magnon interaction and neutron-magnon interaction. In the phonon-magnon interaction (chapter 2) we considered the energy flow from the spin system to the lattice or vice-versa when one or the other is excited. In the present chapter, we consider the magnetic system to be in an equilibrium state at a certain temperature and the neutrons as responsible for perturbing this system.

In recent years, the neutron scattering technique has been exploited to furnish valuable information about the spin dependent properties of magnetically ordered systems. The scattering mechanisms of slow neutrons involving nuclear and magnetic interactions with the atoms have been studied by several authors<sup>50,51</sup>. In the magnetic scattering two types of effects can arise owing to the spin and the orbital states of the atoms. Most of the theoretical work seems to be confined to the study of interaction with the spins alone and various authors<sup>52,53</sup> have discussed neutron scattering with spin waves in exchange coupled magnetic lattices, in particular, involving one magnon processes.



However, the states of some paramagnetic atoms may involve orbital transitions also in the course of such interactions.<sup>50,54,55</sup> In the present work we have taken into account such orbital states which are admixed with ground state of the atoms owing to their interaction with neutrons in the formulation of the effective exchange interaction. We will now consider the formulation of the interaction Hamiltonian and the neutron scattering cross-sections for ferrimagnetics.

A. Formulation of the interaction terms:

We consider a system of magnetic atoms having localized unpaired electrons in addition to the closed shell configuration. The sublattices of the crystal, assumed to be of the b.c.c. type, are designated by the running indices  $l$  and  $m$ , the corresponding atoms having unequal and opposite spin magnetic moments. Let us consider a beam of slow neutrons incident on such a ferrimagnetic crystal. The Hamiltonian of the system can be written as

$$H = \sum_d H_d + H_{\text{neu}} + \sum_{l,m} H_{lm} + \sum_d H_{nd}, \quad (5.1)$$

where  $H_d$  is the Hamiltonian for a single atom, 'd' spans both  $l$  and  $m$ ,  $H_{\text{neu}}$  is that of the neutrons,  $H_{lm}$  describes the exchange interaction between atoms  $l$  and  $m$ .  $H_{nd}$  is the

Hamiltonian for the neutron-atom magnetic interaction and includes both spin and orbital moments:<sup>50</sup>

$$H_{nd} = H_{ns} + H_{no}, \quad (5.2)$$

where  $H_{ns}$  pertains to interaction with the spins of the atoms and  $H_{no}$  with the orbital moments. The individual eigenstates and eigenvalues of the neutrons and the atomic system are assumed to be known, namely,

$$H_{neu} |k\sigma\rangle = E_{k\sigma} |k\sigma\rangle, \quad (5.3)$$

$$\begin{aligned} H_d |l_i\rangle &= E_{ai} |l_i\rangle, \\ H_d |m_i\rangle &= E_{bi} |m_i\rangle, \end{aligned} \quad (5.4)$$

where  $|k\sigma\rangle$  is the wave function of the neutron with wave vector  $\underline{k}$ , spin  $\sigma$  and energy  $E_{k\sigma}$ ;  $|l_i\rangle$  and  $|m_i\rangle$  are the eigenstates of the atoms  $l$  and  $m$  respectively with energies  $E_{ai}$  and  $E_{bi}$ , 'i' indicating the initial (here ground) state. We represent the orbital states of the atoms which are perturbed owing to interaction with neutrons as

$$|l_{as}\rangle = |l_{is}\rangle + \sum_f \frac{\langle l_f, \underline{k}'\sigma' | H_{no} | l_i, \underline{k}\sigma \rangle}{(E_{ai} - E_{af})} |l_f\rangle, \quad (5.5)$$

where we are considering only the orbital interaction  $H_{no}$ ;  $\underline{k}'$ ,  $\sigma'$  are the wave vector and spin of the scattered

neutron and  $|l_f\rangle$  the final state of the atom with energy  $E_{af}$ . Also  $s$  denotes the electron spin. Now we formulate the exchange Hamiltonian of the magnetic lattice in the presence of neutrons by making use of the states

$|l_{as}\rangle$  and  $|m_{bs}\rangle$  etc. This can be written as

$$\frac{1}{2} \sum_{l,m} H_{lm} = \frac{1}{2} \sum_{lm} \sum_{ss'} c_{as}^+ c_{bs}^+ \langle l_{as}, m_{bs} | H_{lm} / m_{bs}, l_{as} \rangle c_{bs} c_{as} \quad (5.6)$$

where  $c_{as}^+$ ,  $c_{as}$  represent the fermion creation and annihilation operators for an electron in the orbital state  $|l_{as}\rangle$ . On making use of the perturbed states (5.5), the Hamiltonian can be written in terms of a pure exchange part and others involving exchange accompanied by neutron scattering:

$$H_m = \sum_{l,m} \sum_{s,s'} c_{ls}^+ c_{ms}^+ J_{AB} c_{ms} c_{ls} = 2J_{AB} \cdot S_{lA} \cdot S_{mB} + \text{const.} \quad (5.7)$$

and

$$H_{mn} = \sum_{lmf} 2 \left[ \frac{\langle m_j, k' \sigma' | H_{no} / m_f, k \sigma \rangle \langle l_{is}, m_{fs} | H_{lm} / m_{js}, l_{is} \rangle}{\Delta E_b} \right. \\ + \frac{\langle l_i, k' \sigma' | H_{no} / l_f, k \sigma \rangle \langle l_{fs}, m_{js} | H_{lm} / m_{js}, l_{is} \rangle}{\Delta E_a} \\ + \frac{\langle m_f, k' \sigma' | H_{nl} / m_j, k \sigma \rangle \langle l_{is}, m_{js} | H_{lm} / m_{fs}, l_{is} \rangle}{\Delta E_b} \\ \left. + \frac{\langle l_f, k' \sigma' | H_{no} / l_i, k \sigma \rangle \langle l_{is}, m_{js} | H_{lm} / m_{js}, l_{fs} \rangle}{\Delta E_a} \right] \\ \times \eta_{k' \sigma'}^+ \eta_{k \sigma} S_{lA} \cdot S_{mB} \quad (5.8)$$

where  $\Delta E_a = E_{ai} - E_{af}$ ,  $\Delta E_b = E_{bi} - E_{bf}$  and  $\eta_{\underline{k}\sigma}^+$ ,  $\eta_{\underline{k}\sigma}$  are respectively the creation and annihilation operators for the neutron in the state  $|\underline{k}\sigma\rangle$ . Other terms of (5.7) have been defined in previous chapters. In writing the magnon-neutron Hamiltonian,  $H_{mn}$ , we have already carried out summations over spin states  $s$  and  $s'$ . We use the neutron wave function

$$|\underline{k}\sigma\rangle = e^{i\underline{k}\cdot\underline{r}} |\sigma\rangle \quad (5.9)$$

and a typical matrix element, say, that in the first term of (5.8) becomes

$$\langle m_j, \underline{k}'\sigma' / H_{no} / m_f, \underline{k}\sigma \rangle = \langle m_j, \sigma' / H_{no} / m_f, \sigma \rangle e^{-i\underline{q}\cdot\underline{r}_m},$$

where  $\underline{q} = \underline{k} - \underline{k}'$ . Now we use the H-P<sup>10</sup> operators which lead to

$$\begin{aligned} \underline{S}_{1A} \cdot \underline{S}_{mB} &= \frac{1}{2} (S_{1A}^+ S_{mB}^- + S_{1A}^- S_{mB}^+) + S_{1zA} S_{mzB} \\ &= (S_A S_B)^{\frac{1}{2}} (a_{1m} d_m + a_{1m}^+ d_m^+) + S_A d_m^+ d_m + S_B a_{1m}^+ a_{1m}. \end{aligned} \quad (5.10)$$

Then we Fourier transform the operators  $a_{1m}$  and  $d_m$  to the reciprocal space and use the relations (5.9) and (5.10) in (5.8). Summations over  $l$  or  $m$  give us certain momentum conservation conditions such as

$$\underline{k} - \underline{k}' \pm (\underline{\lambda} - \underline{\lambda}') = \underline{k} - \underline{k}' \mp \underline{q} = 0$$

for different terms.  $\underline{q}$  is the difference between wave vectors of the incident and the outgoing neutrons. Invoking the canonical transformation as given in (2.16'), (5.8) becomes

$$\begin{aligned}
H_{mn} = & \sum_{\Delta} 2\pi \left\{ \left( \frac{J_1}{\Delta E_b} P_{\Delta q} \langle m_j, \sigma' / H_{no} / m_f, \sigma \rangle \right. \right. \\
& + \frac{J_2}{\Delta E_a} X_{\Delta q} \langle l_1, \sigma' / H_{no} / l_f, \sigma \rangle \left. \right) \eta_{k\sigma'}^* \eta_{k\sigma} \alpha_{\Delta}^+ \alpha_{\Delta-q} \\
& + \left( \frac{J_1}{\Delta E_b} Q_{\Delta q} \langle m_j, \sigma' / H_{no} / m_f, \sigma \rangle \right. \\
& + \frac{J_2}{\Delta E_b} Y_{\Delta q} \langle l_1, \sigma' / H_{no} / l_f, \sigma \rangle \left. \right) \eta_{k\sigma'}^* \eta_{k\sigma} \alpha_{\Delta}^+ \beta_{\Delta} \\
& + \left( \frac{J_1}{\Delta E_b} R_{\Delta q} \langle m_j, \sigma' / H_{no} / m_f, \sigma \rangle \right. \\
& \left. + \frac{J_2}{\Delta E_a} Z_{\Delta q} \langle l_1, \sigma' / H_{no} / l_f, \sigma \rangle \right) \eta_{k\sigma'}^* \eta_{k\sigma} \beta_{\Delta}^+ \beta_{\Delta} \left. \right\} \quad (5.11)
\end{aligned}$$

with

$$\begin{aligned}
J_1 &= \langle l_{1s}, m_{fs}, / H_{lm} / m_{js}, l_{1s}' \rangle, \\
J_2 &= \langle l_{fs}, m_{js}, / H_{lm} / m_{js}, l_{1s}' \rangle.
\end{aligned} \quad (5.12)$$

The coupling constants are given by

$$\begin{aligned}
P_{\Delta q} = & (S_A S_B)^{\frac{1}{2}} (r_{\Delta-q} \sin h \theta_{\Delta} \cos h \theta_{\Delta-q} + r_{\Delta} \cos h \theta_{\Delta} \sin h \theta_{\Delta-q}) \\
& \text{where } \theta_{\Delta} \text{ is defined in (3.17).} \\
& + (S_A \sin h \theta_{\Delta} \sin h \theta_{\Delta-q} + S_B r_{\Delta} \cos h \theta_{\Delta} \cos h \theta_{\Delta-q}),
\end{aligned}$$

$$\begin{aligned}
 Q_{\Delta q} &= 2(S_A S_B)^{\frac{1}{2}} (r_{\Delta-q} \cos h \theta_{\Delta} \cos h \theta_{\Delta-q} + r_{\Delta} \sin h \theta_{\Delta} \sin h \theta_{\Delta-q}) \\
 &\quad + 2(S_A \cos h \theta_{\Delta} \sin h \theta_{\Delta-q} + S_B r_q \sin h \theta_{\Delta} \cos h \theta_{\Delta-q}), \\
 R_{\Delta q} &= (S_A S_B)^{\frac{1}{2}} (r_{\Delta-q} \cos h \theta_{\Delta} \sin h \theta_{\Delta-q} + r_{\Delta} \sin h \theta_{\Delta} \cos h \theta_{\Delta-q}) \\
 &\quad + (S_A \cos h \theta_{\Delta} \cos h \theta_{\Delta-q} + S_B r_q \sin h \theta_{\Delta} \sin h \theta_{\Delta-q}), \\
 X_{\Delta q} &= (S_A S_B)^{\frac{1}{2}} (r_{\Delta-q} \cos h \theta_{\Delta} \sin h \theta_{\Delta-q} + r_{\Delta} \sin h \theta_{\Delta} \cos h \theta_{\Delta-q}) \\
 &\quad + (S_A r_q \cos h \theta_{\Delta} \cos h \theta_{\Delta-q} + S_B \sin h \theta_{\Delta} \sin h \theta_{\Delta-q}), \\
 Y_{\Delta q} &= 2(S_A S_B)^{\frac{1}{2}} (r_{\Delta-q} \sin h \theta_{\Delta} \sin h \theta_{\Delta-q} + r_{\Delta} \cos h \theta_{\Delta} \cos h \theta_{\Delta-q}) \\
 &\quad + 2(S_A r_q \sin h \theta_{\Delta} \cos h \theta_{\Delta-q} + S_B \cos h \theta_{\Delta} \sin h \theta_{\Delta-q}), \\
 Z_{\Delta q} &= (S_A S_B)^{\frac{1}{2}} (r_{\Delta-q} \sin h \theta_{\Delta} \cos h \theta_{\Delta-q} + r_{\Delta} \cos h \theta_{\Delta} \sin h \theta_{\Delta-q}) \\
 &\quad + (S_A r_q \sin \theta_{\Delta} \sin h \theta_{\Delta-q} + S_B \cos h \theta_{\Delta} \cos h \theta_{\Delta-q}),
 \end{aligned}
 \tag{5.13}$$

where  $\theta_{\Delta}$  is defined in (2.17).

In the neutron-magnon interaction term, i.e., (5.11) we have not included the hermitian conjugate processes since we are considering a particular incident neutron  $|\underline{k}\rangle$  being scattered to the state  $|\underline{k}'\rangle$ . The hermitian conjugate processes of the magnon operators are already taken into account because of the sum over  $\underline{\lambda}$ . Equation (5.11) fully describes the interaction of neutrons with magnons involving orbital transition of the magnetic atoms. The exchange integrals  $J_1$  and  $J_2$  defined in (5.12) are different since these exchange integrals involve one excited orbital state of one or the other atom.

#### B. Scattering cross section:

The scattering cross-section per unit range of energy, per unit solid angle, is given in terms of the neutron-magnon interaction  $H_{mn}$  by <sup>56</sup>

$$\frac{d^2\sigma}{d\Omega d\omega} = \frac{1}{k} \frac{k'}{k} \left(\frac{m_n}{2k}\right)^2 |\langle \underline{k}' f | H_{mn} | \underline{k} \rangle|^2 \delta(E_{\underline{k}} - E_{\underline{k}'}, -\hbar\omega_{\underline{\lambda}} + \hbar\omega_{\underline{\lambda}-\underline{q}}), \quad (5.14)$$

where  $m_n$  is the mass of the neutron,  $i$  and  $f$  denote the initial and the final states of the crystal,  $E_{\underline{k}}$  is the kinetic energy of the neutron of wave vector  $\underline{k}$ . ~~and we have used units in which  $\hbar = 1$ .~~

Once again, restricting ourselves to low temperatures, we consider only the 'acoustic' branch of magnons and, thus,

We take for  $H_{mn}$  only the first term of (5.11). This involves matrix elements of the neutron-orbital moment interaction of the type  $\langle l_1, m_1 / H_{no} / l_2, m_2 \rangle$ . These are evaluated as follows.

We can write

$$H_{no} = -\mu_1 \cdot H_n,$$

where  $H_n$  is the magnetic field caused by the magnetic moment  $\mu_n$  of the neutron,  $\mu_1$  being the orbital moment of the atom. Then with  $\underline{r} = \underline{x}_n - \underline{x}_1$

$$H_n = \text{curl} \frac{\mu_n \times \underline{r}}{r^3} = -\text{curl} \left( \mu_n \times \nabla \frac{1}{r} \right).$$

By standard vector algebra

$$\text{curl} \left( \mu_n \times \nabla \frac{1}{r} \right) = -(\mu_n \cdot \nabla) \nabla \frac{1}{r} + \mu_n \nabla^2 \frac{1}{r}.$$

Now  $\nabla^2 \frac{1}{r} = -4\pi \delta(\underline{r}),$

so that

$$H_{no} = \mu_1 \cdot \nabla \left( \mu_n \cdot \nabla \frac{1}{r} \right) - 4\pi \mu_1 \cdot \mu_n \delta(\underline{r}). \quad (5.15)$$

This expression can be written as

$$H_{no} = \left\{ \frac{\mu_1 \cdot \mu_n}{r^3} - \frac{3(\mu_1 \cdot \underline{r})(\mu_n \cdot \underline{r})}{r^5} \right\} - \frac{8\pi}{3} \mu_1 \cdot \mu_n \delta(\underline{r}). \quad (5.16)$$



The curly bracket above denotes the dipolar interaction and for our purpose we neglect it here. The matrix element can then be written as

$$\begin{aligned} \langle l_1, \sigma' / H_{no} / l_f, \sigma \rangle &= \langle l_1, \sigma' / \int d^3r \cdot H_{no} / l_f, \sigma \rangle \\ &= - \frac{8\pi}{3} \langle l_1, \sigma' / \mu_l \cdot \mu_n / l_f, \sigma \rangle. \end{aligned} \quad (5.16)$$

The orbital and neutron magnetic moments are given by

$$\begin{aligned} \mu_l &= \frac{-2 e \hbar k}{m_e c} \underline{l}_1, \\ \mu_n &= g_n \frac{e \hbar k}{m_n c} \underline{s}_n, \end{aligned} \quad (5.17)$$

where  $m_e$  is the electronic mass,  $\underline{l}_1$  the orbital angular momentum of atom 1 and  $\underline{s}_n$  the neutron angular momentum,  $g_n = -1.91$ . Therefore

$$\mu_l \cdot \mu_n = -2 g_n \frac{e^2 \hbar^2}{m_e m_n c^2} \underline{l}_1 \cdot \underline{s}_n$$

and

$$\langle l_1, \sigma' / H_{no} / l_f, \sigma \rangle = \frac{16}{3} \pi g_n \frac{e^2 \hbar^2}{m_e m_n c^2} \langle l_1, \sigma' / \underline{l}_1 \cdot \underline{s}_n / l_f, \sigma \rangle.$$

Here we can write

$$\underline{l}_1 \cdot \underline{s}_n = L_{1z} S_{nz} + \frac{1}{2} (L_1^+ S_n^- + L_1^- S_n^+).$$

The first term,  $L_{1z} S_{nz}$ , does not give rise to any scattering involving orbital transition and hence we drop it here. The remaining part gives

$$\begin{aligned} \langle l_1, \sigma' / H_{no} / l_f, \sigma \rangle &= \frac{8\pi}{3} \epsilon_n \frac{e^2 \hbar^2}{m_e m_n c^2} \times \langle l_1, \sigma' / L_1^+ S_n^- + L_1^- S_n^+ / l_f, \sigma \rangle \\ &= \frac{8\pi}{3} \epsilon_n \frac{e^2 \hbar^2}{m_e m_n c^2} \left\{ \langle l_1 / L_1^+ / l_f \rangle \langle \sigma' / S_n^- / \sigma \rangle + \langle l_1 / L_1^- / l_f \rangle \langle \sigma' / S_n^+ / \sigma \rangle \right\}. \end{aligned}$$

Since we are considering the incident neutrons with a particular spin, one or the other term of the curly bracket will survive. If the incident neutron is supposed to have spin  $+\frac{1}{2}$ , we get<sup>57</sup>

$$\begin{aligned} \langle l_1, \sigma' / H_{no} / l_f, \sigma \rangle &= \frac{8\pi}{3} \epsilon_n \frac{e^2 \hbar^2}{m_e m_n c^2} \left\{ (L_1 + M_1)(L_1 - M_1 + 1) \right\}^{\frac{1}{2}} \\ &\quad \times \left\{ \left( \frac{1}{2} + \frac{1}{2} \right) \left( \frac{1}{2} - \frac{1}{2} + 1 \right) \right\}^{\frac{1}{2}} \\ &= \frac{8\pi}{3} \epsilon_n \frac{e^2 \hbar^2}{m_e m_n c^2} \left\{ (L_1 + M_1)(L_1 - M_1 + 1) \right\}^{\frac{1}{2}}, \quad (5.18a) \end{aligned}$$

where  $M_1$  is the orbital magnetic quantum number of the atom 1. Similarly

$$\langle m_j, \sigma' / H_{no} / m_f, \sigma \rangle = \frac{8\pi}{3} \epsilon_n \frac{e^2 \hbar^2}{m_e m_n c^2} \left\{ (L_2 + M_2)(L_2 - M_2 + 1) \right\}^{\frac{1}{2}}. \quad (5.18b)$$

Using these relations, the first term of  $H_{mn}$  of (5.11) becomes

$$H_{mn} = \sum_{\Delta} (A_1 P_{\Delta q} + A_2 X_{\Delta q}) \eta_{\underline{k}'}^+ \eta_{\underline{k}} \alpha_{\Delta}^+ \alpha_{\Delta-q}$$

where

$$A_1 = \frac{16\pi}{3} \epsilon_n \frac{e^2 k^2}{m_e m_n c^2} \cdot \frac{J_1}{\Delta E_b} \left\{ (L_1 + M_1)(L_1 - M_1 + 1) \right\}^{\frac{1}{2}}$$

and

$$A_2 = \frac{16\pi}{3} \epsilon_n \frac{e^2 k^2}{m_e m_n c^2} \cdot \frac{J_2}{\Delta E_a} \left\{ (L_2 + M_2)(L_2 - M_2 + 1) \right\}^{\frac{1}{2}}$$

Now we have to evaluate the matrix element of the neutron-magnon interaction  $H_{mn}$  which appears in the scattering cross-section (5.14). The combined states of neutron and magnons can be written as

$$|i\underline{k}\rangle = e^{i\underline{k} \cdot \underline{r}_1} |1_{\underline{k}}\rangle |\dots n_{\Delta_1}, n_{\Delta_2} \dots\rangle,$$

$$|f\underline{k}'\rangle = e^{i\underline{k}' \cdot \underline{r}_m} |0_{\underline{k}'}\rangle \dots n_{\Delta_1}, n_{\Delta_2} \dots$$

Then

$$\langle \underline{k}' f | H_{mn} | i \underline{k} \rangle = \sum_{\Delta} (A_1 P_{\Delta q} + A_2 X_{\Delta q}) \langle \dots n_{\Delta_1}, n_{\Delta_2} \dots, 0_{\underline{k}'} | e^{-i\underline{k}' \cdot \underline{r}_m} \eta_{\underline{k}'}^+ \eta_{\underline{k}} \alpha_{\Delta}^+ \alpha_{\Delta-q} e^{i\underline{k} \cdot \underline{r}_1} | 1_{\underline{k}}, n_{\Delta_1}, n_{\Delta_2} \dots \rangle$$

Here again, since the incident neutron state  $|1_{\underline{k}}\rangle$  is occupied and the final neutron state  $|0_{\underline{k}'}\rangle$  is empty, the neutron operators will give unity. The magnon operators yield

In evaluating the sum (5.17), we use

$$\langle \underline{k}'f/H_{mn}/ik \rangle = \sum_{\lambda} (A_1 P_{\lambda q} + A_2 X_{\lambda q})$$

$$\times \left\{ (n_{\lambda} + 1)(n_{\lambda - q}) \right\}^{\frac{1}{2}} e^{i(\underline{k} \cdot \underline{r}_1 - \underline{k}' \cdot \underline{r}_m)}$$

Therefore

$$\langle \underline{k}'f/H_{mn}/ik \rangle \langle ik/H_{mn}/\underline{k}'f \rangle$$

where we conveniently choose the direction of  $\underline{k}$  to be in the  $\lambda$  direction.

$$= \sum_{\lambda} (A_1 P_{\lambda q} + A_2 X_{\lambda q})^2 (n_{\lambda} + 1)(n_{\lambda - q}) \quad (5.19)$$

The expression  $(A_1 P_{\lambda q} + A_2 X_{\lambda q})^2$  is evaluated by using (5.13) retaining terms upto lowest order in the wave vectors. This approximation is of interest for the rare earth ions in which case, the 4f shell has a small radius. We obtain

$$(A_1 P_{\lambda q} + A_2 X_{\lambda q})^2 = R_1 \cdot \frac{q^2 a^2}{z} - R_2 (\lambda^2 + \lambda'^2) \frac{a^2}{z}$$

where

$$R_1 = \frac{1}{2} A_1^2 z^2 \frac{S_A S_B (S_A^2 - 6S_A S_B + S_B^2)}{(S_A - S_B)^2} + 2A_1 A_2 z^2 S_A S_B$$

$$R_2 = 2A_2^2 z^2 S_A S_B + 2A_1 A_2 z^2 \frac{(3S_A^5 S_B + 4S_A^4 S_B^2 + 22S_A^3 S_B^3 + 4S_A^2 S_B^4 + 3S_A S_B^5)}{(S_A - S_B)^4}$$

and Using the above results and changing the limits of integration, (5.19) becomes

$$\lambda' = \lambda - q$$

In evaluating the sum (5.19), we must utilize the  $\delta$ -function of (5.14), i.e.,

$$\delta(E_{\underline{k}} - E_{\underline{k}'} - \hbar\omega_{\underline{\lambda}} + \hbar\omega_{\underline{\lambda}'}) = \frac{1}{2\lambda q k_B \theta_c a^2} \delta \left\{ \frac{\hbar^2 (\underline{k}^2 - \underline{k}'^2)}{4m_n \lambda q k_B \theta_c a^2} + \frac{q}{2\lambda} - \cos \theta_{\underline{\lambda}} \right\},$$

where we conveniently choose the difference vector  $\underline{q}$  to be in the  $z$  direction without loss of generality and  $\theta_{\underline{\lambda}}$  is the angle between the magnon vector  $\underline{\lambda}$  and the  $z$  axis.  $\hbar\omega_{\underline{\lambda}}$  is the ferrimagnetic acoustic mode branch energy given by the first equation of (2.19). Also, we use the equilibrium distribution function

$$n_{\underline{\lambda}} = \left( e^{\hbar\omega_{\underline{\lambda}}/k_B T} - 1 \right)^{-1}$$

so that

$$\begin{aligned} (n_{\underline{\lambda}+\underline{q}}+1)(n_{\underline{\lambda}-\underline{q}}) &= \frac{e^{\hbar\omega_{\underline{\lambda}}/k_B T}}{(e^{\hbar\omega_{\underline{\lambda}}/k_B T} - 1)(e^{\hbar\omega_{\underline{\lambda}-\underline{q}}/k_B T} - 1)} \\ &= e^{-\hbar\omega_{\underline{\lambda}-\underline{q}}/k_B T} \end{aligned}$$

in the low temperature region.

Using the above results and changing summation to integration, (5.19) becomes:

$$\begin{aligned}
 & \langle \underline{k}'f/H_{mn}/i\underline{k} \rangle \langle i\underline{k}/H_{mn}/\underline{k}'f \rangle \\
 & \frac{Nv}{4\pi^2} \int \lambda^2 \sin \theta_{\underline{\lambda}} d\theta_{\underline{\lambda}} d\lambda \left\{ R_1 \frac{q^2 a^2}{z} - R_2 (2\lambda^2 + q^2 - 2q \cos \theta_{\underline{\lambda}}) \frac{a^2}{z} \right\} \\
 & \times e^{-\pi \omega_{\underline{\lambda}} - q/k_B T} \frac{1}{2\lambda q k_B \theta_c a^2} \delta \left\{ \frac{-\hbar^2 (\underline{k}^2 - \underline{k}'^2)}{4m_n \lambda q k_B \theta_c a^2} + \frac{q}{2\lambda} - \cos \theta_{\underline{\lambda}} \right\}, \quad (5.21)
 \end{aligned}$$

This gives us the neutron scattering cross-section where  $v$  is the volume of the unit cell ( $v = a^3$ ) and  $N$  is their number in the crystal.

The integration over  $\theta_{\underline{\lambda}}$  is easily carried out by substituting  $x = \cos \theta_{\underline{\lambda}}$  which eliminates the  $\delta$ -function and gives

$$\begin{aligned}
 & \frac{Nv}{4\pi^2} \int_0^{\infty} \lambda d\lambda \left\{ R_1 \frac{q^2 a^2}{z} - R_2 (2\lambda^2 - p^2) \frac{a^2}{z} \right\} \\
 & \times \frac{1}{2q k_B \theta_c a^2} \exp \left( -k_B \theta_c \lambda^2 a^2 + \frac{\theta_c}{T} p^2 a^2 \right), \quad (5.20)
 \end{aligned}$$

where<sup>o</sup>

$$p^2 = \frac{-\hbar^2 (\underline{k}^2 - \underline{k}'^2)}{2m k_B \theta_c a^2}.$$

Integrating this expression and substituting in the equation (5.14) for the scattering cross-section, we obtain

<sup>o</sup>It is hoped that there would be no confusion between this  $p$  and the  $p$  defined after (2.36) which are quite different.

$$\frac{d^2\sigma}{dE d\Omega} = \frac{k'}{k} \left( \frac{m_p}{2\pi} \right)^2 \frac{N a}{8\pi^2 k_B \theta_c q^4 k^4} \left[ (R_1 q^2 + R_2 p^2) \frac{T}{\theta_c^2} - R_2 \frac{T^2}{\theta_c^2 z a^2} \right] \exp \left( \frac{\theta_c}{T} p^2 a^2 \right). \quad (5.21)$$

This gives us the neutron scattering cross-section <sup>for</sup> from a ferrimagnetic crystal at low temperatures taking into account the orbital magnetic moments of the atoms.

CHAPTER - 6



CONCLUDING REMARKS

In the foregoing chapters, we have developed a theory of interaction of magnons with different agencies such as phonons and neutrons from the microscopic point of view. In particular we have considered ferrimagnetic crystals. The magnon dispersion relations obtained in chapter 2(2.19) show a quadratic dependence on the wave vector for a simple b.c.c. ferrimagnet. For more complicated lattices such as magnetic spinels and garnets, the phonon-magnon interaction decreases rapidly with increasing wave frequency. Terms get very involved. Differences in the acoustic spin wave frequency arise only in the coefficients of  $\lambda^2 a^2$  in (2.19), namely<sup>7</sup>,

$$\tau_{w\Delta}(\text{magnetite}) = \frac{11}{10} \frac{J_{AB} S_A (S_{B1} + S_{B2})}{S_{B1} + S_{B2} - S_A} \lambda^2 a^2$$

and<sup>58</sup>

$$\tau_{w\Delta}(\text{YIG}) = \frac{5}{10} (5 J_{ad} - 8 J_{aa} - 3 J_{dd}) \lambda^2 a^2.$$

For a rough comparison of the relaxation time with available experimental results, we choose the following values of the parameters in (2.28) for the relaxation time. Consider a system with  $M = 55$ ,  $S_A = 2$ ,  $S_B = 2.5$ ,  $\theta_c = 200^\circ\text{K}$ ,  $\theta_D = 200^\circ\text{K}$  and  $J$  ranging between  $10^{-7}$  and  $10^{-6}$  dyne. Then (2.38) gives  $\tau_{mp}$  at  $10^\circ\text{K}$  in the range  $10^{-5}$  to  $10^{-7}$  sec. Although the above is a tentative range of values, this is fairly

close to the observed range in some systems.<sup>59,60</sup> The range of values chosen for  $\Theta_J$  seems to be quite reasonable for magnetic spinels and garnets, as seen by the estimates made for other substances.<sup>13,31</sup> It is to be noted that the expression for  $\gamma_{mp}$  is quite sensitive to the magnitudes of  $\Theta_c$  and  $\Theta_D$  and for a comparison with a given system one must use the appropriate values.

The renormalization of the spin wave frequency shows a very striking feature, i.e., the spin wave frequency decreases rapidly with temperature owing to the interaction with lattice vibrations. A similar investigation of the renormalization of the phonon energy in magnetically ordered systems ( i.e., owing to the interaction with magnons ) can be done and may furnish some valuable information about the temperature dependence of the Debye temperature in such systems.

The application of these calculations to thermal conductivity is straightforward and the agreement with experimental results quite conclusive. Although deviations appear in the conductivity peaks, one must bear in mind that the original phonon-magnon interaction theory is meant for two interpenetrating cubic sublattices of the magnetic ions. It was hoped that the interaction mechanism involving 'acoustic' modes of phonons and magnons will hold in low temperature regions even in the complicated lattices such as ferrites and garnets. Although this hope has been largely fulfilled one

could still desire a better agreement. The discrepancies may be connected with the complicated ion sublattices as well as the imperfections and defects of various kinds. Perhaps the simple expression of defect scattering,  $P \propto \omega^4$ , is not capable of fully explaining the results. However, the agreement in the region where spin wave contribution is important strongly supports the mechanism considered here.

In chapter 4, the Green's function method is applied to ferrimagnetism, which gives the spinwave spectrum and the low temperature dependence of magnetization. Since we have restricted ourselves to the low temperature region, a good deal of simplification has been possible. The theory can be extended to complicated ferrimagnets with many sublattices and to high temperatures.

In the neutron-magnon interaction, we have considered only the contribution from the orbital magnetic moments of the ions. The interaction of neutrons with spin magnetic moments has been a subject of considerable interest in the last decade and in almost all of these calculations the orbital moment was supposed to be quenched. However, as mentioned earlier, this approximation can not be made in the case of rare earth ions. We believe that the additional mechanism involving the orbital moment will improve the agreement between experiment and theoretical prediction including only spin moment. Unfortunately, neutron scattering

cross-section data for rare earth ferrites or garnets does not seem to be available although such experiments on rare earth metals have been done. A similar theory could be developed for ferro- and antiferro-magnetic systems for which the neutron scattering data already exist and may provide a check to the above statement.

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