A THEORETICAL STUDY OF PHONON-MAGNON INTERACTION IN MAGNETICALLY ORDERED SYSTEMS

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Chapter 1

INTRODUCTION

A. Phonons: Matter in the solid state occurs usually in crystalline forms with the atoms, molecules or ions constituting the substance arranged in a three dimensional network of an appropriate symmetry. The ions* of a solid are never quiescent; they execute small oscillations around their equiblirium positions. Since the ions are strongly coupled with their neighbouring ones by elastic forces, the displacement of one ion from its equiblirium position will set forth a disturbance which will travel like a wave through the crystal. Thus, we can describe the vibrational state of the solid in terms of lattice waves having different wave vectors and velocities travelling through the crystal, their nature being governed by the elastic forces. Alternatively, the lattice vibrations in a crystal can be represented as a vibrational field where the field variables are the displacements of the ions from their equiblirium positions. In a quantum

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* From hereafter, by ions we shall mean the appropriate constituents of the solid under consideration, e.g. atoms, molecules etc.

field theoretic language this can be also described as a cloud of quantas of the vibrational field known as 'phonons'. Mathematically we can introduce the concept of phonons by considering the Hamiltonian for the lattice vibrations in the following form:

$$H_l = \frac{1}{2} \sum_{\underline{l},\underline{b}} \left(\frac{1}{m_b}\right) \underline{p}_{\underline{l},\underline{b}} \cdot \underline{p}_{\underline{l},\underline{b}} + V(\underline{h}_{\underline{l},\underline{b}}) \qquad (1)$$

Here the atoms are labelled by two symbols, $\underline{l},\underline{b}$ where $\underline{l}$ is the vector to the origin of the cell to which the atom belongs and $\underline{b}$ is the atom in the cell. The first term in eq.(1) represents the kinetic energy of the ions and the second term is the potential of the ions which is a function of $\underline{h}_{\underline{l},\underline{b}} \left(= \underline{x}_{\underline{l},\underline{b}} - \underline{l} + \underline{b}\right)$, the displacement of the ion at the site $(\underline{l},\underline{b})$ from its equiblirium position.

Now, if we expand the potential energy in Taylor's series in terms of the various displacements, we get

$$V(\underline{h}_{\underline{l},\underline{b}}) = V_o + \frac{1}{2} \sum_{\underline{l},\underline{b}} \sum_{\underline{l}',\underline{b}'} \underline{h}_{\underline{l},\underline{b}} \cdot \left[\frac{\partial^2 V}{\partial \underline{h}_{\underline{l},\underline{b}} \partial \underline{h}_{\underline{l}',\underline{b}'}}\right]_o \cdot \underline{h}_{\underline{l}',\underline{b}'} + \cdots$$

$$\equiv V_o + \frac{1}{2} \sum_{\underline{l},\underline{b}} \sum_{\underline{l}',\underline{b}'} \underline{h}_{\underline{l},\underline{b}} \cdot G_{\underline{l},\underline{b};\underline{l}',\underline{b}'} \cdot \underline{h}_{\underline{l}',\underline{b}'} + \cdots \qquad (2)$$

where we have retained terms only up to second order in the Taylor's expansion. Substituting eq.(2) in eq.(1) we get the Hamiltonian operator as

$$H_l = \frac{1}{2}\sum_{\underline{l},\underline{b}} \left(\frac{1}{m_b}\right) \underline{p}_{\underline{l},\underline{b}} \cdot \underline{p}_{\underline{l}',\underline{b}'} + \frac{1}{2}\sum_{\underline{l},\underline{b}}\sum_{\underline{l}',\underline{b}'} \underline{h}_{\underline{l},\underline{b}} \cdot G_{\underline{l},\underline{b};\underline{l}',\underline{b}'} \cdot \underline{h}_{\underline{l}',\underline{b}'} \tag{3}$$

The Hamiltonian H_1 can be written[1] in a diagonal form in terms of new operators $b^{\dagger}_{qp}$, b_{qp} defined by the following equations,

$$\underline{h}_{\underline{l},\underline{b}} = -i\left(\frac{\hbar}{2N m_b}\right)^{\frac{1}{2}} \sum_{\underline{q},p} \underline{e}_{\underline{q},\underline{b},p}\, \omega_{\underline{q},p}^{-\frac{1}{2}} \left(b^{\dagger}_{\underline{q},p} - b_{-\underline{q},p}\right) e^{-i\underline{q}\cdot\underline{l}}$$

$$\underline{p}_{\underline{l},\underline{b}} = \left(\frac{\hbar m_b}{2N}\right)^{\frac{1}{2}} \sum_{\underline{q},p} \underline{e}_{\underline{q},\underline{b},p}\, \omega_{\underline{q},p}^{\frac{1}{2}} \left(b_{\underline{q},p} + b^{\dagger}_{-\underline{q},p}\right) e^{i\underline{q}\cdot\underline{l}} \tag{4}$$

Here $\underline{q}, \underline{p}$ denote the wave vector and branch number[1] of the lattice wave respectively, $\underline{e}_{\underline{q},\underline{b},\underline{p}}$ the polarization vector and $\omega_{\underline{q},\underline{p}}$ the frequency of the lattice wave with wave vector $\underline{q}$, branch number p. The Hamiltonian (3) becomes in terms of these new operators as,

$$H_l = \sum_{\underline{q},p} \left(b^{\dagger}_{\underline{q},p}\, b_{\underline{q},p} + \frac{1}{2}\right) \hbar\omega_{\underline{q},p} \tag{5}$$

with $n_{qp} = b^{\dagger}_{qp}\, b_{qp}$ the occupation number operator which has got only positive integers as its eigenvalues. The eq.(5) suggests a simple physical interpretation of the lattice vibrational field, namely, that it consists of a system of noninteracting quantas known as 'phonons', each having an energy $\hbar\omega_{\underline{q},p}$. The distribution of phonons in the energy spectrum at any temperature will be gover-

ned by Bose statistics as the phonon creation and annihilation operators obey the commutation relations:

$$\left[b_{\underline{q},p}\ ,\ b^{\dagger}_{\underline{q}',p'}\right] = \delta_{\underline{q}\underline{q}'}\ \delta_{pp'} \qquad (6)$$

The higher order terms in the expansion of potential energy, eq.(2), so far neglected, if taken into account, will give rise to interactions betwen phonons and their motions will no longer be independent of each other.

B. Magnons: The first attempt to explain theoretically the magnetic properties of ferromagnets on an atomic model was made by Heisenberg[2] in 1928. According to his theory the magnetism in ferromagnets arises due to the spins of localized unpaired electrons arranged in a definite pattern and each spin being coupled strongly by exchange forces with its neighbours. He showed that the total Hamiltonian of a system consisting of atoms arranged in a regular three dimensional network with each atom having one unpaired electron can be written as,

$$H = C - 2\sum_{l<m} J_{lm}\ \underline{S}_l \cdot \underline{S}_m \qquad (7)$$

where C includes the contribution to the Hamiltonian independent of spin variables, $\underline{S}_l$ and $\underline{S}_m$ are the spin operators for the electrons belonging to atoms at $\underline{R}_l$ and $\underline{R}_m$. These operators can be written in the matrix form as,

$$S_x = \frac{1}{2}\begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix}, \quad S_y = \frac{1}{2}\begin{bmatrix} 0 & -i \\ i & 0 \end{bmatrix}, \quad S_z = \frac{1}{2}\begin{bmatrix} 1 & 0 \\ 0 & -1 \end{bmatrix} \tag{8}$$

These are known as 'Pauli matrices'. Further, J_{lm} is the exchange integral for the l^{th} and m^{th} electronic wave function with the explicit form,

$$J_{l,m} = \int \psi_l^*(1)\, \psi_m^*(2)\, V_{lm}\, \psi_l(2)\, \psi_m(1)\, d\tau_1 d\tau_2 \tag{9}$$

where V_{lm} is given as,

$$V_{lm} = \frac{e^2}{R_{lm}} + \frac{e^2}{r_{12}} - \frac{e^2}{|\underline{r}_1 - \underline{R}_m|} - \frac{e^2}{|\underline{r}_2 - \underline{R}_l|} \tag{10}$$

with small r denoting the electronic coordinates and capital R the nuclear coordinates. It is important to remember that the above interaction energy between spin variables is fundamentally of electrostatic origin. Spin enters primarily as a consequence of Pauli exclusion principle and indistinguiblity of electrons. Besides the exchange interaction considered in eq.(7), there are magnetic interactions of relativistic origin, namely the dipolar interaction with the following explicit form:

$$\sum_{j>i} g^2 \mu_B^2 \left[\frac{\underline{S}_i \cdot \underline{S}_j}{r_{ij}^3} - 3\, \frac{(\underline{S}_i \cdot \underline{r}_{ij})(\underline{S}_j \cdot \underline{r}_{ij})}{r_{ij}^5} \right] \tag{11}$$

However, as the strength of the dipolar interaction is considerably weaker than the exchange interaction, we shall not consider it in our present study.

Let us consider the number of states with different values of the z component of the total magnetic moment. With N electrons, the state with the magnetic moment $N\mu_B$ (μ_B being Bohr magneton) can be realized in only one way, namely, when all the spins are parallel. But the state with the total magnetic moment $M\mu_B (M < N)$ can occur in

$$n(M) = \frac{N!}{\left(\frac{N+M}{2}\right)! \left(\frac{N-M}{2}\right)!} \tag{12}$$

different ways. Assuming the distributions of states of a given M to be Gaussian, Heisenberg[2] deduced the temperature dependence of the magnetic moment as,

$$\frac{M}{M_\infty} = \tanh \eta \tag{13a}$$

where $$\eta = \frac{\mu_B H}{kT} + \frac{1}{2}\left(\gamma - \frac{\gamma^2}{2}\right)\frac{M}{M_\infty} + \frac{\gamma^2}{4z}\left(\frac{M}{M_\infty}\right)^3 \tag{13b}$$

and $$\gamma = \frac{z J_e}{kT} \tag{13c}$$

Here M is the magnetic moment at any temperature T, M_∞ the saturation magnetic moment and J_e the exchange integral for nearest neighbour interaction assumed to be the same for all the nearest neighbours. The eq.(13) is essentially the same as derived by Weiss[3] based on the macroscopic concept of molecular field.

In spite of its success to suggest an explanation of the origin of Weiss molecular field and the deduction of eq.(13), Heisenberg's simple theory

outlined above suffers from two serious defects. Firstly, it is based upon a simple Heitler-London description in which the periodicity of the lattice is not taken into account. Secondly, he arbitrarily assumes a Gaussian distribution of states of a given M. As the thermal properties strongly depend on this distribution a description derived from first principles is needed for quantitative work. Bloch[4] in an attempt to improve upon Heisenberg's work introduced the concept of spin wave. According to Bloch a spin wave is a single spin reversal moving over a large number of otherwise aligned spins in a crystal lattice. For the ground state, Bloch takes a determinantal wave function formed with atomic orbitals and with all the electrons in the same spin state as given below,

$$\psi_0 = \sum_P (-1)^P P\left[\phi_1(\underline{r}_1)\alpha_1 \phi_2(\underline{r}_2)\alpha_2 \cdots \phi_N(\underline{r}_N)\alpha_N\right] \qquad (14)$$

where P denotes all the possible permutation operations among all the electrons, ϕ denotes the orbital part of the wave function and α and β denote the two spin states of the electron. Bloch defines the eigen state with one spin wave of wave vector k as,

$$\psi_k = a_k \sum_n e^{2\pi i \underline{k}\cdot\underline{r}_N} \psi_n \qquad (15)$$

where ψ_n differs from ψ_o in that the spin of the electron on the n^{th} atom has been reversed. The eigenstate ψ_k has the eigenvalue as,

$$E_k = E_o + 2Je \sum_{\underline{\rho}} \left(1 - e^{2\pi i \underline{k}\cdot\underline{\rho}}\right) \tag{16}$$

where $\underline{\rho}$ is the nearest neighbour vector and the sum extends over all such vectors. Bloch showed that if the number of spin waves is small compared to the total number of all electrons, then the energy of the crystal in a state in which there are f spin waves of wave numbers $k_1, k_2, \ldots\ldots k_f$ is

$$E(k_1, \cdots k_f) = E_o + \sum_{\nu=1}^{f} \epsilon_s(k_\nu) \tag{17}$$

where

$$\epsilon_s(k_\nu) = E_k - E_o$$

Thus, the spin waves behave like particles which are so nearly independent of each other that their energies are additive. Further, Bloch deduced the following expression for M at low temperature keeping only the second order terms in the wave vector in the expansion of exponential in eq.(16):

$$\frac{M}{M_\infty} = 1 - \alpha \frac{2.612}{2\pi^2} \left(\frac{kT}{Je}\right)^{3/2} \tag{18}$$

where α is a constant characteristic of the lattice. Weiss has verified experimentally Bloch's $T^{3/2}$ law

for iron and nickel in the temperature range from 70° K to 20° K. Eq.(16) shows a very important qualitative difference betwen the Weiss model and the spin wave picture. According to the Weiss model, the energy required to reverse a spin from the completely saturated state is equal to the constant quantity $4k\Theta_c$ with Θ_c as the Curie temperature, where as in the present picture we find from eq.(16) a continuous distribution of energy values. In fact one finds that it is so easy to reverse a spin in the case of one dimensional array of spins that there is no ferromagnetism possible in this case.

Slater[5] further extended Bloch's work by redifining the spin wave. Instead of ϕ_n he took the wave function $\phi_{n,R}$ which is constructed from ϕ_o by taking an electron from the atom at $\underline{r}_n$ to the atom at $\underline{r}_n+\underline{R}$ and reversing its spin. Thus, Bloch's spin waves are special case of that of Slater when R=0. Further, instead of using the ordinary atomic wave function for constructing the determinantal wave function as done by Bloch, Slater used an orthoganalized set of Wannier functions[6] χ_n constructed from Bloch type function $\chi_k(\underline{r})e^{2\pi i \underline{k}\cdot\underline{r}}$ by means of the equation,

$$\chi_n(\underline{r}-\underline{R}_n) = \sum_K \chi_K(\underline{r})\, e^{2\pi i \underline{k}\cdot(\underline{r}-\underline{R}_n)} \qquad (19)$$

Slater concludes that at low temperatures the Bloch type

spin waves are energetically more favourable than that considered by him.

An alternative approach to introduce the concept of spin wave was made by Holstein and Primakoff[7] leading to essentially the same results as obtained by Bloch. Following Bloch, they considered a three dimensional network of spins coupled ferromagnetically in an external magnetic field. They defined a set of coordinates which describe accurately the quantum state of the system. In terms of these coordinates they obtained the total Hamiltonian of the system consisting of two parts. The first part consists of terms quadratic in the new variables defined by HP[7] with the explicit form,

$$H = \sum_{\lambda} \hbar \omega_{\lambda} \left(a_{\lambda}^{\dagger} a_{\lambda} + \tfrac{1}{2} \right) \qquad (20)$$

where $a_{\lambda}^{\dagger} a_{\lambda}$ is the occupation number operator of spin waves with positive integers as its eigenvalues. This expression, as in the case of lattice waves, suggests that the spin system considered here may be likened to a system of non-interacting particles with energy $\hbar\omega_{\lambda}$ (known as magnons). Like phonons, magnons are Bosons. The other part of the Hamiltonian consists of higher order terms in $a_{\lambda}^{\dagger}$ and a_{λ} and represents the mutual interaction between magnons. Dyson[8] has conclusively established that the linear Bloch

theory with non-interacting spin waves, [redacted] is equivalent to HP theory neglecting higher than second order terms. Further, Oguchi[9] has shown the equivalence of HP method with that of Dyson which is the most rigorous treatment at low temperatures.

The extension of the concept of spin waves to the case of antiferromagnets is not straightforward. This was done by Hulthen[10], Ziman[11], Anderson[12], Kubo[13] and others[14,15]. The method essentially consists in writing the Hamiltonian of the spin system in terms of the spin deviation operators as in the case of ferromagnets. For the sake of simplicity we shall restrict ourselves to the simple and body centered cubic structures which can be divided into two interlocking sublattices 1 and 2 such that all the nearest neighbours of an ion on sublattice 1 are on sublattice 2 and vice versa. If we consider only the isotropic Heisenberg exchange interaction, the Hamiltonian of the spin system can be written as,

$$H = 2J \sum_{\substack{\text{nearest} \\ \text{neighbour}}} \underline{S}_l \cdot \underline{S}_m \qquad (21)$$

where l refers to an atom on sublattice 1 and m to one on sublattice2; J being positive. With this Hamiltonian the antiferromagnetic state with certain long range

order among the spin directions on sublattice 1 and a similar order among the spins on sublattice 2 in the opposite direction is highly degenerate as the common direction of the resulting antiparallel magnetic moment of the two sublattices is arbitrary. This degeneracy is such that it can not be removed by any external field, since for a not too large external field the spins would arrange themselves antiferromagnetically in a plane perpendicular to the external field ,but the directions of the spin in the plane would still be arbitrary. In an actual antiferromagnet this degeneracy is removed by the presence of the anisotropy field.[15] This anisotropy field must be of such a form that the spins on sublattice 1 are preferentialy aligned in the z direction, say, and those on sublattice 2 in the negative z direction. This will be the case when we assume an effective field H_A such that it points along the positive z direction at the sites of sublattice 1 and in the negative z direction at those of sublattice 2. Thus, the Hamiltonian becomes,

$$H = 2J \sum_{\substack{\text{nearest} \\ \text{neighbour}}} \underline{S}_l \cdot \underline{S}_m - H_A g\beta_B \left(\sum_l S_l^x - \sum_m S_m^z\right) - H g\beta_B \left(\sum_j S_j^z\right) \qquad (22)$$

So we find that the anisotropy field plays a very important role in the case of antiferromagnets which is not the case in ferromagnets. In the limiting case of $H_A \rightarrow \infty$, the ground state for the Hamiltonian operator (eq.(22)) is the state in which all the spins on sublattice 1 are pointing in the positive z direction and those on sublattice 2 in the negative z direction. This completely ordered state is used as the ground state in the spin wave theory of antiferromagnetism. Theoretical investigations of Van Vleck[15] and others show that this model is only about 7% wrong. Further confirmation of the validity of the model of interlocking sublattices has been provided by the experiments of Shull and Smart[16] on neutron diffraction. However, as the completely ordered state assumed in the usual spin wave theory as the ground state suffers from the ambiguity that the role of the two sublattices may be interchanged without affecting the ground state energy, one may expect a quantum mechanical resonance effect where by the roles of two sublattices are interchanged with each other. But this process is very slow and for all practical purposes the assignment of one type of spin to one sublattice the reverse to the other can be regarded as stable. The final form of the Hamilton-

ian is the same as eq.(20) except that here the dispersion relation is different from the case of ferromagnets.

C. Relaxation Process: Relaxation effects occur whenever the time a system needs to come to thermal equiblirium either within itself or with an external reservoir is comparable to the time in which changes in the external parameters are made. The following magnetic phenomena exhibit the relaxation effects:

1. Line width in ferromagnetic resonance or antiferromagnetic resonance absorption.[40]
2. Power saturation with intense radio frequency magnetic fields in resonance absorption phenomena.
3. Time delay in heat transfer betwen lattice and spin systems.
4. Velocity of propagation of the Bloch wall separating domains in ferromagnetic insulators.
5. Time delay in the establishment of a new equilibrium position for the magnetic moment of a specimen after a sudden change in the external magnetic field.

When a system of coupled spins is under the influence of a microwave electromagnetic field the temperature of the spin system, if entirely insulated from the surroundings, will rise. The population density

of the higher energy states will continue to rise till the absorption and stimulated emission become equal and in the stationary state no net absorption will take place. However, other processes which involve energy dissipation set in. The phonon-magnon interaction provides one of the required dissipation process in the case of ferro-,ferri- or antiferromagnetic systems. The physical process responsible for phonon-magnon relaxation is the modulation of spin interaction energies by the crystal field vibrations. Historically, the first theoretical study in this field was made by Akhiezer[17] for ferromagnets in 1946. He investigated the problem from a microscopic point of view by expanding the exchange and dipolar terms in power series of normal coordinates of the lattice vibration. However, Polder[18] has pointed out that the way in which Akhiezer replaces sums by integrals in his calculation probably tended to underestimate the effect of the modulation of short range exchange integrals and thus Akhiezer's calculations when properly revised do not yield with sufficiently short relaxation time to agree with experiment. In the last decade Kittel[19] and Abraham[20], Kaganov and Tsukernik[21] and others[14] have made phenomenological study of the problem using the macroscopic

concepts of the exchange energy density and magnetoelastic coupling energy. In what follows we shall discuss their method briefly. The study of phonon-magnon interaction by Kittel and Abraham is based on a macroscopic magnetoelastic interaction of the magnetisation or spin direction with the crystal lattice which accounts approximately for the observed magnetostriction in ferromagnets. This is expressed as,

$$H_{int} = B_1(\alpha_1^2 e_{xx} + \alpha_2^2 e_{yy} + \alpha_3^2 e_{zz}) + B_2(\alpha_1\alpha_2 e_{xy} + \alpha_2\alpha_3 e_{yz} + \alpha_3\alpha_1 e_{zx}) \quad (23)$$

Here α's are the direction cosines of the magnetisation and the e's are elastic strain tensor components. Values of the constants B_1 and B_2 are taken from the observed magnetostriction data. They find that the most important type of interaction is that involving scattering of a magnon with the emmision or absorption of a phonon. Calculated values of spin lattice relaxation time for nickle by Abraham and Kittel are of the right order of magnitude. Kaganov and Tsukernik start with the following interaction Hamiltonian,

$$H_{int} = H_{ms} + H_{ex} \quad (24)$$

where H_{ms} represents the magnetoelastic interaction

energy with the explicit form,

$$H_{ms} = \gamma \int M_i M_k e_{ik} \, dV \qquad (25)$$

Here γ is determined, as in the study by Kittel and Abraham, from magnetostriction experiments. Further, H_{ex} is given by,

$$H_{ex} = \lambda_1 \int \frac{\partial M_l}{\partial x_i} \cdot \frac{\partial M_l}{\partial x_k} u_{ik} \, dV + \lambda_2 \int \frac{\partial M_l}{\partial x_i} \cdot \frac{\partial M_l}{\partial x_i} u_{kk} \, dV \qquad (26)$$

The energy H_{ex} may be considered as arising due to the expansion of the exchange interaction energy $\int \alpha_{ik} \left(\frac{\partial M_l}{\partial x_i}\right)\left(\frac{\partial M_l}{\partial x_k}\right) dV$ in powers of u_{ik}. For the constants $\lambda_{1,2}$ Kaganov and Tsukernik chose the form $\frac{\beta_{1,2}}{2} \frac{\theta_c}{\mu M_0} a^2$ where $\beta_{1,2}$ are dimensionless constants of order unity. Proceeding in this way Kaganov and Tsukernik derived the expressions for spin-lattice relaxation time in ferromagnets and they found that in the temperature range $\mu M_0 \ll T \ll \theta_0^2/\theta_c$ and for $\theta_0^2/\theta_c \ll T < \theta_0/\theta_c$ the spinspin relaxation time T_{ss} is much shorter than the spin- lattice relaxation time T_{sp}. Further, they also considered the interaction processes involving two phonons, taking the interaction Hamiltonian as,

$$H_{int} = L^{ik}_{lmrs} \int \frac{\partial M_q}{\partial x_i} \cdot \frac{\partial M_q}{\partial x_k} u_{lm} u_{rs} \, dV \qquad (27)$$

$$\left(\text{with } L^{ik}_{lmrs} \sim \theta_c a^2/\mu M_0\right)$$

They find that these processes can be important only at temperatures below 1^{0} K where their treatment fails.

In the case of antiferromagnets, at least to author's knowledge, no theoretical or experimental study of phonon-magnon interaction has been made till now.

Although the phenomenological approach outlined above explains the experimental results in a fairly satisfactory way, any such approach in general suffers from two serious drawbacks. Firstly, such an approach does not provide a clear insight of the various physical processes involved. Secondly, the strength of the interaction can not be determined from first principles. Thus, it is desirable to study the interaction on an atomistic model from first principles.

The aim of the present study is to consider the phonon-magnon interaction from a microscopic point of view taking into account the mixing of excited and ground state atomic orbitals owing to crystal field oscillations. These perturbed states, when used for formulating the two electron interaction term in the second quantisation representation, furnish the relevent interaction term. This mechanism is applied to formulate the phonon-magnon interaction

terms for the case of ferro- and antiferromagnetic systems. The expressions of the relaxation time for the establishment of equilibrium between the spin and lattice systems are derived.[37,38,39]

Chapter 2

FORMULATION OF THE INTERACTION HAMILTONIAN[37]

A. Ferromagnets : For the study of phonon-magnon interaction first we consider the case of a simple ferromagnet consisting of magnetic atoms or ions regularly arranged in a crystal and each having in addition to closed shell ion core one localized d electron. The total Hamiltonian for such a system consists of the following parts:

$$H = H_{el} + H_l + H_c + H_z \quad , \tag{28}$$

where H_{el} is the Hamiltonian involving one electron coordinates, namely,

$$H_{el} = \sum_i p_i^2/2m + \sum_{i,n} U(\underline{r}_i - \underline{R}_n), \tag{29}$$

H_l is the lattice Hamiltonian expressed as in eq.(5)

$$H_l = \sum_{q\rho} \hbar\, \omega_{q\rho} \left(b^\dagger_{q\rho}\, b_{q\rho} + \tfrac{1}{2}\right) \quad ,$$

and H_c is the electron-elctron Coulomb interaction operator with the explicit form,

$$H_c = \sum_{i\neq j} \tfrac{1}{2}\, \frac{e^2}{r_{ij}} \equiv \tfrac{1}{2} \sum_{i\neq j} g_{ij} \tag{30}$$

and H_z is the Zeeman term,

$$H_z = -g\mu_B H_0 \sum_i S_{zi} \quad . \tag{31}$$

In the above, $p_i^2/2m$ is the kinetic energy operator of the i^{th} electron, $U(\underline{r}_i - R_n)$ is its potential energy at $\underline{r}_i$ due to the ion core at $\underline{R}_n$ and r_{ij} the distance between electrons i and j. For the present, the spin-orbit interaction, anharmonic terms in lattice vibrations and dipolar interactions between magnetic ions are not included. For the i^{th} electron the Hamiltonian(29) is rewritten as,

$$H_{el}^{(i)} = p_i^2/2m + U(\underline{r}_i - \underline{R}_i^\circ) + V_0 + H' \tag{32}$$

where $V_0 = \sum_{m \neq l} U(\underline{r}_i - \underline{R}_m^\circ)$;

$$H' = \sum_h \left(\frac{\partial V}{\partial \underline{R}_h}\right)_0 \cdot \delta\underline{R}_h + \frac{1}{2} \sum_{h,h'} \delta\underline{R}_h \cdot \left(\frac{\partial^2 V}{\partial \underline{R}_h \, \partial \underline{R}_h'}\right) \cdot \delta\underline{R}_h' + \cdots \tag{33}$$

and

$$\underline{R}_h = \underline{R}_l - \underline{R}_m$$

Here, $U(\underline{r}_i - R_i^\circ)$ represents the electrostatic potential acting on the electron when the ion-core to which the electron belongs is in its equilibrium position, V_0 is the static crystal field potential due to the nearest neighbour ion at R_i° and H' represents the first and higher order terms of the Taylor series development of the crystal field potential V due to neighbouring ions in terms of their relative displacements. We take the wave function $\phi_{a\sigma}$ as the solution of

$$\left[p^2/2m + U(\underline{r} - \underline{R}_\ell^\circ) + V_\circ\right] \phi_{a\sigma} = E_{a\sigma}\, \phi_{a\sigma} \qquad (34)$$

The eigenstates of the Hamiltonian (32), which includes H' as the perturbation, are represented as,

$$\psi_{a\sigma} = \phi_{a\sigma} + \sum_{\alpha} \frac{H'_{\alpha a}\, \phi_{a\sigma}}{E_\alpha - E_a} + \cdots\cdots \qquad (35)$$

where the Greek letters represent the excited states and Latin letters the ground state. With the above choice of one electron functions, we can write the total Hamiltonian in occupation number representation[22] as,

$$H = H_L + \sum_{a\sigma} E'_{a\sigma} A^\dagger_{a\sigma} A_{a\sigma} + \frac{1}{2} \sum_{abcd} \sum_{\sigma\sigma'} A^\dagger_{a\sigma} A^\dagger_{b\sigma'} \times \left\langle a\sigma, b\sigma' \middle| g_{12} \middle| c\sigma, d\sigma' \right\rangle A_{c\sigma} A_{d\sigma'} + H_{\text{ex}} \qquad (36)$$

where $A^\dagger_{a\sigma}$, $A_{a\sigma}$ represent the creation and annihilation operators respectively for the one electron states with eigenvalues $E'_{a\sigma}$ and obey the following relations:

$$\begin{aligned} A_i A^\dagger_\kappa + A^\dagger_\kappa A_i &= \delta_{i\kappa} \\ A_i A_\kappa + A_\kappa A_i &= 0 \\ A^\dagger_i A_i &= N_i \ , \end{aligned} \qquad (37)$$

where N_i is the occupation number operator for the state ψ_i. In the above equations subscripts κ, i denote both orbital and spin states. For the ground occupied states N_i is equal to I, unity operator, and for empty excited states N_i is equal to 0, null operator.

Further, for the orbital states which are singly occupied, the following relations hold between the above operators and the conventional spin operators:

$$\begin{aligned} A_a^\dagger(+)\,A_a(+) + A_a^\dagger(-)\,A_a(-) &\equiv 1 \\ N_a(+) + N_a(-) &\equiv 1 \\ N_a(+) - N_a(-) &= 2S_z^a \\ A_a^\dagger(+)\,A_a(-) &= S_+^a = S_x^a + i S_y^a \\ A_a^\dagger(-)\,A_a(+) &= S_-^a = S_x^a - i S_y^a \end{aligned} \tag{38}$$

where (+) and (-) indicate the two spin states of the electron. Now let us consider the third term in eq. (36) which with the help of the equation (35) can be written as:

$$\frac{1}{2}\sum_{abcd}\sum_{\sigma\sigma'} A_{a\sigma}^\dagger A_{b\sigma'}^\dagger \Big\langle \Big(\phi_a + \sum_\alpha \frac{H'_{\alpha a}\phi_\alpha}{\Delta E_\alpha}\Big)_\sigma \Big(\phi_b + \sum_\beta \frac{H'_{\beta b}\phi_b}{\Delta E_\beta}\Big)_{\sigma'} \Big| g_{12} \Big|$$

$$\Big(\phi_c + \sum_\gamma \frac{H'_{\gamma c}\phi_\gamma}{\Delta E_\gamma}\Big)_\sigma \Big(\phi_d + \sum_\delta \frac{H'_{\delta d}\phi_\delta}{\Delta E_\delta}\Big)_{\sigma'} \Big\rangle A_{c\sigma} A_{d\sigma'} \tag{39}$$

This expression gives terms of various orders in the perturbation Hamiltonian H' for the phonon-magnon interaction Hamiltonian. To illustrate the procedure for summing over the spin index we consider the term of the equation (39) containing the zeroth power of H' as given below:

$$\frac{1}{2}\sum_{abcd}\sum_{\sigma\sigma'} A_{a\sigma}^\dagger A_{b\sigma'}^\dagger \Big\langle \phi_{a\sigma}^{(1)}\,\phi_{b\sigma'}^{(2)} \Big| g_{12} \Big| \phi_{b\sigma}^{(1)}\,\phi_{a\sigma'}^{(2)} \Big\rangle A_{c\sigma} A_{d\sigma'} \tag{40}$$

Allowing σ, σ' to be in (+) and (−) state independently, the summation over indexes σ, σ' can be written as,

$$\sum_{\sigma\sigma'} A^{\dagger}_{a\sigma} A_{b\sigma'} A_{b\sigma} A_{a\sigma'} = A^{\dagger}_{a}(+) A^{\dagger}_{b}(+) A_{b}(+) A_{a}(+) + A^{\dagger}_{a}(+) A^{\dagger}_{b}(-) A_{b}(+) A_{a}(-) + A^{\dagger}_{a}(-) A^{\dagger}_{b}(+) A_{b}(-) A_{a}(+) + A^{\dagger}_{a}(-) A^{\dagger}_{b}(-) A_{b}(-) A_{a}(-) \tag{41}$$

Now, utilising the relations (37) and (38), eq.(41) can be written as,

$$\begin{aligned} &\sum_{\sigma\sigma'} A^{\dagger}_{a\sigma} A_{b\sigma'} A_{b\sigma} A_{a\sigma'} = \\ &= N_a(+) N_b(+) + S^{+}_{a} S^{-}_{b} + S^{-}_{a} S^{+}_{b} + N_a(-) N_b(-) \\ &= \frac{1}{4}\left(2S^{z}_{a}+1\right)\left(2S^{z}_{b}+1\right) + S^{+}_{a} S^{-}_{b} + S^{-}_{a} S^{+}_{b} + \frac{1}{4}\left(1-2S^{z}_{a}\right)\left(1-2S^{z}_{b}\right) \\ &= \frac{1}{2}\left(1 + 4\, \underline{S}_a \cdot \underline{S}_b\right) \end{aligned} \tag{42}$$

Similarly summing over spin states in other terms of eq.(36) we get the total Hamiltonian of the system as,

$$\begin{aligned} H = &\sum_{q\rho} \hbar\omega_{q\rho}\left(b^{\dagger}_{q\rho} b_{q\rho} + \tfrac{1}{2}\right) + \sum_{a} E'_a N_a + \frac{1}{2}\sum_{l\neq m} J(\underline{R}_{lm})\, P^{\sigma}_{lm} \\ &+ 2\sum_{h,\, l\neq m} {}^{\alpha}J(\underline{R}_{lm})\, P^{\sigma}_{lm} \cdot \delta\underline{R}_h + \sum_{l\neq m}\sum_{hh'}\left[{}^{\alpha}J'(\underline{R}_{lm}) + {}^{\alpha\beta}J(\underline{R}_{lm})\right. \\ &\left. + 2\,{}^{\alpha}J_{\beta}(\underline{R}_{lm})\right] P^{\sigma}_{lm}\, \delta\underline{R}_h \cdot \delta\underline{R}'_h + H_z \end{aligned} \tag{43}$$

where

$$P^{\sigma}_{lm} \equiv \frac{1}{2} + 2\,\underline{S}_l \cdot \underline{S}_m \;;$$

$$J(\underline{R}_{lm}) = \left\langle \phi^{(1)}_{l} \phi^{(2)}_{m} \middle| g_{12} \middle| \phi^{(1)}_{m} \phi^{(2)}_{l} \right\rangle$$

$$
\begin{aligned}
{}^{\alpha}J(\underline{R}_{\ell m}) &\equiv \sum_{\alpha} \langle \phi_\alpha \phi_m | g_{12} | \phi_m \phi_\ell \rangle \langle \phi_\alpha | \underline{V}^h | \phi_\ell \rangle \Big/ (E_\alpha - E_\ell) \\
{}^{\alpha\beta}J(\underline{R}_{\ell m}) &\equiv \sum_{\alpha\beta} \frac{\langle \phi_\alpha \phi_\beta | g_{12} | \phi_m \phi_\ell \rangle \langle \phi_\alpha | \underline{V}^h | \phi_\ell \rangle \langle \phi_\beta | \underline{V}^{h'} | \phi_m \rangle}{(E_\alpha - E_\ell)(E_\beta - E_m)} \\
{}^{\alpha}J_{\beta}(\underline{R}_{\ell m}) &= \sum_{\alpha\beta} \frac{\langle \phi_\alpha \phi_m | g_{12} | \phi_\beta \phi_\ell \rangle \langle \phi_\alpha | \underline{V}^h | \phi_\ell \rangle \langle \phi_\beta | \underline{V}^{h'} | \phi_m \rangle}{(E_\alpha - E_\ell)(E_\beta - E_m)} \\
{}^{\alpha}J'(\underline{R}_{\ell m}) &= \sum_{\alpha} \frac{\langle \phi_\ell \phi_m | g_{12} | \phi_m \phi_\ell \rangle \langle \phi_\alpha | \underline{V}^{hh'} | \phi_\ell \rangle}{(E_\alpha - E_\ell)} \\
\underline{V}^h &\equiv \left(\frac{\partial V}{\partial \underline{R}_h}\right)_o \quad ; \quad \underline{V}^{hh'} \equiv \left(\frac{\partial^2 V}{\partial \underline{R}_h \, \partial \underline{R}_{h'}}\right)_o \\
\underline{R}_{\ell m} &= \underline{R}^{\circ}_{\ell} - \underline{R}^{\circ}_{m}
\end{aligned}
\tag{44}
$$

The relative displacement $\delta \underline{R}_h$ between the ions at $\underline{R}_\ell$ and $\underline{R}_m$ can be expressed in terms of the creation and annihilation operators of phonons as[1]:

$$
\begin{aligned}
\delta \underline{R}_h &= \frac{1}{\sqrt{N}} \sum_{q\rho} (-i)\, \underline{e}_{q\rho} \left(\frac{\hbar}{2\omega_{q\rho} M}\right)^{1/2} \left(b^{\dagger}_{q\rho} - b_{-q\rho}\right)\left(e^{i \underline{q}\cdot \underline{R}^{\circ}_{\ell}} - e^{i \underline{q}\cdot \underline{R}^{\circ}_{m}}\right) \\
&\equiv \frac{1}{\sqrt{N}} \sum_{q\rho} \underline{g}_{q\rho} \left(b^{\dagger}_{q\rho} - b_{-q\rho}\right)\left(e^{i \underline{q}\cdot \underline{R}^{\circ}_{\ell}} - e^{i \underline{q}\cdot \underline{R}^{\circ}_{m}}\right)
\end{aligned}
\tag{45}
$$

We now use the Holstein and Primakoff[7] spin deviation and other operators:

$$
\begin{aligned}
S^{+}_{\ell} &= (2S)^{1/2} \left(1 - \frac{a^{\dagger}_{\ell} a_{\ell}}{2S}\right)^{1/2} a_{\ell} \\
S^{-}_{\ell} &= (2S)^{1/2}\, a^{\dagger}_{\ell} \left(1 - \frac{a^{\dagger}_{\ell} a_{\ell}}{2S}\right)^{1/2} \\
S - S^{z}_{\ell} &= a^{\dagger}_{\ell} a_{\ell} = n_{\ell} \text{ (the spin deviation operator)}
\end{aligned}
\tag{46}
$$

where $a_\ell^\dagger$ and a_ℓ are the well known creation and annihilation operators defined by Holstein and Primakoff. Following them we shall neglect quantities of the type $\langle n_\ell / 2S \rangle$ and higher order terms. In view of the usual commutation relations satisfied by the spin operators, namely,

$$[S_x, S_y] = i S_z \tag{47}$$

$a_\ell^\dagger, a_\ell$ must satisfy the following commutation relations:

$$[a_\ell, a_m^\dagger] = \delta_{\ell m}$$

and $$[a_\ell, a_m] = [a_\ell^\dagger, a_m^\dagger] = 0 \tag{48}$$

However, in order to satisfy the above commutation relations it is necessary for $a_\ell^\dagger$, a_ℓ etc. to be infinite matrices. Therefore, n_ℓ can take on integer values ranging from 0 to ∞. However, only those values of n, for which $0 \leqslant n \leqslant 2S$, have any physical meaning. The discrepancy is only apparent since the transitions from states with $n_\ell > 2S$ to states with $n_\ell < 2S$ will never occur; e.g.

$$S_\ell^- \psi_{2S} = (2S)^{1/2} (2S+1)^{1/2} \left(1 - \frac{2S}{2S}\right) \psi_{2S+1} = 0 \tag{49}$$

Using eq. (45) and eq. (46) the part of the Hamiltonian involving spin deviation and phonon creation and annihilation operators can be written as,

$$H_{int} = \frac{8}{\sqrt{N}} \sum_{\ell,m} \sum_{qp}{}^{\alpha} J(\underline{R}_{\ell m}) \cdot \underline{g}_{qp} (a_\ell^\dagger a_m - a_m^\dagger a_m)(b_{qp}^\dagger - b_{-qp}) \times$$

$$\times\left(e^{i\underline{q}\cdot\underline{R}^{\circ}_{l}} - e^{i\underline{q}\cdot\underline{R}^{\circ}_{m}}\right) + 4/N \cdot \sum_{l,m}\sum_{h h'}\sum_{q\beta q'\beta'}\left[{}^{\alpha}J'(\underline{R}_{lm}) + {}^{\alpha\beta}J(\underline{R}_{lm})\right.$$

$$\left. + 2^{\alpha}J_{\beta}(\underline{R}_{lm})\right]\underline{g}_{q\beta}\,\underline{g}_{q'\beta'}\left(a^{\dagger}_{l}a_{m} - a^{\dagger}_{m}a_{m}\right)\left(b^{\dagger}_{q\beta} - \underline{b}_{q\beta}\right)\left(b^{\dagger}_{q'\beta'} - \underline{b}_{q'\beta'}\right)\times$$

$$\times\left(e^{i\underline{q}\cdot\underline{R}^{\circ}_{l}} - e^{i\underline{q}\cdot\underline{R}^{\circ}_{m}}\right)\left(e^{i\underline{q}'\cdot\underline{R}^{\circ}_{l}} - e^{i\underline{q}'\cdot\underline{R}^{\circ}_{m}}\right) + \ldots\ldots \qquad (50)$$

We now make use of the spin wave Fourier transformation of the spin deviation operators, defined by,

$$\begin{aligned} a_{\lambda} &= \left(1/\sqrt{N}\right)\sum_{l}\exp\left(i\underline{K}_{\lambda}\cdot\underline{R}^{\circ}_{l}\right)\,a_{l} \\ a^{\dagger}_{\lambda} &= \left(1/\sqrt{N}\right)\sum_{l}\exp\left(-i\underline{K}_{\lambda}\cdot\underline{R}^{\circ}_{l}\right)\,a^{\dagger}_{l} \\ a_{l} &= \left(1/\sqrt{N}\right)\sum_{\lambda}\exp\left(-i\underline{K}_{\lambda}\cdot\underline{R}^{\circ}_{l}\right)\,a_{\lambda} \\ a^{\dagger}_{l} &= \left(1/\sqrt{N}\right)\sum_{\lambda}\exp\left(i\underline{K}_{\lambda}\cdot R^{\circ}_{l}\right)\,a^{\dagger}_{\lambda} \end{aligned} \qquad (51)$$

where $\underline{K}$ is the reduced wave vector for quantised spin waves (magnons). In terms of $a^{\dagger}_{\lambda}$ and a_{λ}, which are respectively the creation and annihilation operators for magnons, the interaction Hamiltonian (50) takes the elegant form,

$$H_{int} = \frac{4}{\sqrt{N}}\sum_{h\lambda q\beta} 2s^{\alpha}J(R^{\circ}_{h})\left[e^{i\underline{K}_{\lambda}\cdot\underline{R}^{\circ}_{h}} - e^{i(\underline{K}_{\lambda}-\underline{q}_{\nu})\cdot R^{\circ}_{h}} + e^{-i\underline{q}_{\nu}\cdot\underline{R}^{\circ}_{l}} - 1\right]\underline{g}_{q\nu\beta}\times$$

$$\times\, a^{\dagger}_{\lambda-q_{\nu}}\,a_{\lambda}\left(b^{\dagger}_{q\nu\beta} - b_{-q\nu\beta}\right) + \frac{4s}{N}\sum_{hh'}\sum_{q\beta\lambda}\sum_{q'\beta'}\left[{}^{\alpha}J'(\underline{R}^{\circ}_{l}) + {}^{\alpha\beta}J(R^{\circ}_{l}) + 2^{\alpha}J_{\beta}(\underline{R}^{\circ}_{l})\right]\times$$

$$\underline{g}_{q\nu\beta}\,\underline{g}_{q'\nu'\beta'}\left[e^{i\underline{K}_{\lambda}\cdot\underline{R}^{\circ}_{h}} - e^{i(\underline{K}_{\lambda}-q_{\nu})\cdot\underline{R}^{\circ}_{h}} + e^{-i\underline{q}_{\nu}\cdot\underline{R}^{\circ}_{h}} + e^{-iq'_{\nu'}\cdot\underline{R}^{\circ}_{h'}} - 1 + e^{i\left\{(\underline{K}_{\lambda}-q_{\nu})\cdot\underline{R}^{\circ}_{h} - q'_{\nu'}\cdot\underline{R}^{\circ}_{l}\right\}}\right.$$

$$\left. - e^{i(\underline{K}_{\lambda}\cdot\underline{R}^{\circ}_{h} - \underline{q}'_{\nu'}\cdot\underline{R}^{\circ}_{h})} - e^{-i(\underline{q}_{\nu}\cdot\underline{R}^{\circ}_{h} + \underline{q}'_{\nu'}\cdot\underline{R}^{\circ}_{h})}\right]a^{\dagger}_{\lambda-q_{\nu}-q'_{\nu'}}\,a_{\lambda}\left(b^{\dagger}_{q\nu\beta} - b_{-q\nu\beta}\right)\times$$

$$\times\left(b^{\dagger}_{q'\nu'\beta'} - b_{-q'\nu'\beta'}\right) + \ldots\ldots \qquad (52)$$

In arriving at (52) the summation over l has been carried out utilising the relation[1]

$$\sum_{l} e^{i\underline{S}\cdot\underline{R}_l^0} = 0 \quad \text{for} \quad \underline{S} \neq \underline{K}$$
$$= N \quad \text{for} \quad \underline{S} = \underline{K} \qquad (53)$$

where $\underline{K}$ is the vector of the reciprocal lattice including zero and $\underline{S}$ stands for the phonon or magnon wave vectors. It may be noted that only normal processes i.e. $K = 0$ are under consideration here. The various exchange integrals occurring in (52) are taken for the nearest neighbours and hence $\underline{R}_\lambda^0 = \underline{R}_l^0 - \underline{R}_m^0$, where l and m are suffixes for the nearest neighbours. Finally, the following interference conditions, which follows from the relation (53) between the magnons and phonons wave vectors, may be noted which hold respectively for the one and two phonon processes and have been already utilised in deriving (52) :

$$\underline{K}'_\lambda + \underline{q}_r - \underline{K}_\lambda = 0 \qquad (54)$$

$$\underline{K}'_\lambda + \underline{q}_r + \underline{q}'_{r'} - \underline{K}_\lambda = 0 \qquad (55)$$

The various interaction terms in eq. (52) can be pictorially represented as in fig. 1 and 2. The solid line indicates magnon and the broken curve phonon. The first term in (52) represents the one phonon direct process which involves the creation or annihilation of a phonon of wave vector $\underline{q}_r$ accompanied with the scatte-

Fig. 1

One phonon process

$\underline{b} - \underline{\gamma}$
$\underline{q}$
$\underline{\gamma}$
(a)
$\underline{b} + \underline{\gamma}$
$\underline{\gamma}$
$\underline{q}$
(b)

Fig. 2

Two phonon process

$\bar{b}$
$\bar{b}$
γ
$\bar{b} - \bar{b} - \bar{\gamma}$

(a)

$\bar{b}$
$\bar{b}$
$\bar{\gamma}$
$\bar{b} + \bar{b} - \bar{\gamma}$

(b)

$\bar{b}$
$\bar{b}$
$\bar{\gamma}$
$\bar{b} + \bar{b} + \bar{\gamma}$

(c)

ring of two magnons (with wave vectors $\underset{\sim}{K}_\lambda$ and $\underset{\sim}{K}'_\lambda$). This is pictorially represented in fig. 1(a) and 1(b) respectively. The second term contains Raman process involving the creation and annihilation of two phonons (with wave vectors $\underset{\sim}{q}$ and $\underset{\sim}{q}'$) accompanied with the scattering of two magnons. Fig. 2(b) represents the Raman process involving phonons and magnons and 2(a) and 2(c) represent the processes involving the simultaneous creation and annihilation of two phonons. In the next chapter we consider the equilibration between magnon and phonon system when the former is at a slightly higher temperature compared to the latter.

B. Antiferromagnets:[38] As discussed in the introduction we restrict ourselves to a case of two interpenetrating simple cubic lattices of magnetic ions, each having in addition to the closed electron shell ion core one localised d electron. The spins on sublattice 1 point up and those on sublattice 2 point down. The two together form a body centered cubic structure of the magnetic system. With the help of the above model and following the procedure outlined in section A we get the total Hamiltonian of the system including anisotropy terms as,

$$H = H_l + H_{el} + H_{ex} + H_z + H_{an} + H_{int}. \qquad (56)$$

where the symbols respectively stand for contributions to the total Hamiltonian due to lattice, one electron terms, isotropic exchange, Zeeman, anisotropy and interaction terms. H_l and H_{el} are as before given by eq. (5) and eq. (29) respectively. H_{ex} can be written as,

$$H_{ex} = \frac{1}{2} \sum_{lm} J(\underline{R}^{o}_{lm}) P^{\sigma}_{l,m} \qquad (57)$$

In eq.(57) l runs over ions on sublattice 1 and m over sublattice 2. Further, the explicit forms of H_z, H_{an} and H_{int} are given below.

$$H_z = - g \mu_B H \sum_{j=l,m} S^z_j \qquad (58)$$

where $\underline{H}$ is the external magnetic field pointing in the z direction and the other symbols have their usual significance.

$$H_{an} = - H_a g \mu_B \left(\sum_l S^z_l - \sum_m S^z_m \right) \qquad (59)$$

H_a being the anisotropy field.

$$H_{int} = \sum_{l,m} 2 J(\underline{R}_{lm}) P^{\sigma}_{lm} \cdot \delta \underline{R}_l + \text{higher order terms.} \qquad (60)$$

Here we restrict ourselves to interactions involving one phonon only[23].

We now transform the spin dependent parts of

the Hamiltonian (56) in terms of the spin deviation operators of the two sublattices which are expressed below:

$$S_l^+ = S_l^x + iS_l^y = (2S)^{1/2}(1 - n_l/2S)^{\frac{1}{2}}\, a_l$$
$$S_l^- = S_l^x - iS_l^y = (2S)^{1/2} a_l^\dagger (1 - n_l/2S)^{1/2} \qquad (61)$$
$$S - S_l^z = a_l^\dagger a_l = n_l \text{ (the spin deviation operator)}$$

Likewise, for the other sublattice,

$$S_m^+ = (2S)^{1/2}\, d_m^\dagger (1 - n_m/2S)^{1/2}$$
$$S_m^- = (2S)^{1/2} (1 - n_m/2S)^{1/2} d_m \qquad (62)$$
$$S + S_m^z = n_m$$

These operators satisfy the commutation relations

$$\left[a_m , a_l^\dagger\right] = \delta_{ml} \text{ and } \left[d_l , d_m^\dagger\right] = \delta_{lm} \qquad (63)$$

and rest of the commutation brackets being zero. Using eqs.(61) and (62) the spin dependent part of the Hamiltonian (56) can be written as,

$$\begin{aligned} H_s &= H_{ex} + H_z + H_{an} + H_{int} \\ &= \text{constant} + \sum_{l,m} sJ(R_{lm}^\circ)(a_l d_m + a_l^\dagger d_m^\dagger + a_l^\dagger a_l + d_m^\dagger d_m) + \\ &\quad g\mu_B\Big\{H\Big(\sum_l a_l^\dagger a_l - \sum_m d_m^\dagger d_m\Big) + H_A\Big(\sum_l a_l^\dagger a_l + \sum_m d_m^\dagger d_m\Big)\Big\} + \\ &\quad \frac{4S}{\sqrt{N}} \sum_{l,m} \sum_{q\rho} \underline{g}_{q\rho}^{\alpha} J(R_{lm}^\circ)\left[a_l d_m + a_l^\dagger d_m^\dagger + a_l^\dagger a_l + d_m^\dagger d_m\right] \times \\ &\quad \times\left[b_{q\rho}^\dagger - b_{q\rho}\right]\left(e^{i\underline{q}\cdot\underline{R}_l^\circ} - e^{i\underline{q}\cdot\underline{R}_m^\circ}\right) + \ldots\ldots \end{aligned} \qquad (64)$$

In writing eq.(64) we have neglected the terms of the type $n_{l\text{ or }m}/2S$ and higher order terms in the expansion of $\left(1 - \frac{n}{2S}\right)^{1/2}$ and constant includes the terms independent

of spin deviation operators. The Hamiltonian H_s can be written in the spin wave representation by making use of the Fourier transforms of the spin deviation operators given by,

$$
\begin{aligned}
a_\ell &= (2/N)^{1/2} \sum_\lambda \exp(-i \underline{K}_\lambda \cdot \underline{R}^\circ_\ell)\, a_\lambda \\
a^\dagger_\ell &= (2/N)^{1/2} \sum_\lambda \exp(i \underline{K}_\lambda \cdot \underline{R}^\circ_\ell)\, a^\dagger_\lambda \\
d_m &= (2/N)^{1/2} \sum_\lambda \exp(i \underline{K}_\lambda \cdot \underline{R}^\circ_m)\, d_\lambda \\
d^\dagger_m &= (2/N)^{1/2} \sum_\lambda \exp(-i \underline{K}_\lambda \cdot \underline{R}^\circ_m)\, d^\dagger_\lambda
\end{aligned}
\qquad (65)
$$

where the propagation vector $\underline{K}_\lambda$ runs over $N/2$ points of the first Brillouin zone of the reciprocal space of the sublattice. With the help of (65), we can express (64) as,

$$
\begin{aligned}
H_s = \text{constant} &+ \sum_\lambda z J(\underline{R}^\circ_h) S\{\gamma_\lambda (a_\lambda d_\lambda + a^\dagger_\lambda d^\dagger_\lambda) + (a^\dagger_\lambda a_\lambda + d^\dagger_\lambda d_\lambda)\} + \\
&+ g\mu_B \{(H + H_A) \sum_\lambda a^\dagger_\lambda a_\lambda + (H_A - H) \sum_\lambda d^\dagger_\lambda d_\lambda\} + \frac{4Sz}{\sqrt{N}} \sum_\lambda \sum_{q\nu} g_{q\nu} \cdot \\
&{}^{\alpha}\underline{J}(\underline{R}^\circ_h) \Big[(\gamma_{\lambda-q} - \gamma_\lambda)\, a_\lambda d_{\lambda-q} + (\gamma_{\lambda+q} - \gamma_\lambda)\, a^\dagger_\lambda d^\dagger_{\lambda+q} + (1 - \gamma_q)\, a^\dagger_\lambda a_{\lambda+q} \\
&+ (\gamma_q - 1)\, d^\dagger_\lambda d_{\lambda-q} \Big] \big[b^\dagger_{q\nu} - b_{-q\nu} \big]
\end{aligned}
\qquad (66)
$$

where

$$
\gamma_\lambda \equiv \Big(\sum_h e^{\pm i \underline{K}_\lambda \cdot \underline{R}^\circ_h} \Big) \Big/ z \; ; \quad \underline{R}^\circ_h = \underline{R}^\circ_\ell - \underline{R}^\circ_m \qquad (67)
$$

In the above $J(\underline{R}^\circ_h)$ and ${}^{\alpha}J(\underline{R}^\circ_h)$ are assumed to be the same for all nearest neighbour interactions; the summation over h extends to nearest neighbours. Further, in deriving (66) and (64) we have carried out

the summation over l or m utilising the following interference conditions, for the terms in the square brackets,

$$\underline{K}'_\lambda - \underline{K}_\lambda = \pm \underline{q}_\nu \tag{68}$$

with the plus sign before $\underline{q}_\nu$ being used for the first and the last terms and minus sign for the second and the third terms in the first square bracket of (66). It can be seen from (66) that the pure spin part of H_s, i.e. the terms in the curly brackets, is not diagonal. To diagonalise the pure spin part as well as to write the interaction terms in the same representation we make use of the following cannonical transformation:

$$\begin{aligned} a_\lambda &= \alpha_\lambda \operatorname{ch}\theta_\lambda + \beta^\dagger_\lambda \operatorname{sh}\theta_\lambda \\ a^\dagger_\lambda &= \alpha^\dagger_\lambda \operatorname{ch}\theta_\lambda + \beta_\lambda \operatorname{sh}\theta_\lambda \\ d_\lambda &= \alpha^\dagger_\lambda \operatorname{sh}\theta_\lambda + \beta_\lambda \operatorname{ch}\theta_\lambda \\ d^\dagger_\lambda &= \alpha_\lambda \operatorname{sh}\theta_\lambda + \beta^\dagger_\lambda \operatorname{ch}\theta_\lambda \end{aligned} \tag{69}$$

with $\operatorname{ch}\theta = \cosh\theta$, $\operatorname{sh}\theta = \sinh\theta$, $\tanh 2\theta_\lambda = -\frac{\omega_e \gamma_\lambda}{\omega_e + \omega_A}$ (70)

and the symbols

$$\omega_e = 2z\,S\,J(R^\circ_\lambda)/\hbar$$

and $\omega_A = g\,\mu_B H_A/\hbar$.

Using the magnon creation and annihilation operators $\alpha^\dagger, \alpha$ and $\beta^\dagger, \beta$ the pure magnon ~~and interaction~~ Hamiltonian in (66) takes the forms given below:

$$H_m = \sum_\lambda \hbar\omega^+_\lambda \left(\alpha^\dagger_\lambda \alpha_\lambda + \tfrac{1}{2}\right) + \sum_\lambda \hbar\omega^-_\lambda \left(\beta^\dagger_\lambda \beta_\lambda + \tfrac{1}{2}\right) \tag{71}$$

where $\omega_\lambda^{\pm} = \left\{ (\omega_A + \omega_e)^2 - \omega_e^2 \gamma_\lambda^2 \right\}^{1/2} \pm \omega_H$

and $\omega_H = g \mu_B H / \hbar$

The above equation for $\underline{K}_\lambda = 0$ gives the well known relation for AFMR frequency. Thus, we see that in the case of antiferromagnets there are two distinctly different type of magnons described by the creation and annihilation operators $\alpha_\lambda^+, \alpha_\lambda$ and $\beta_\lambda^+, \beta_\lambda$ with energies $\hbar \omega_\lambda^+$ and $\hbar \omega_\lambda^-$ respectively.

Likewise, the phonon-magnon interaction terms reduce to the following form:

$$H_{int} = \sum_{\lambda q p} \Big[A_{\lambda q p} \left(\alpha_\lambda \alpha_{\lambda-q}^\dagger b_{qp}^\dagger - \alpha_\lambda^\dagger \alpha_{\lambda-q} b_{qp} \right) + B_{\lambda q p} \left(\alpha_\lambda \beta_{\lambda-q} b_{qp}^\dagger - \alpha_\lambda^\dagger \beta_{\lambda-q}^\dagger b_{qp} \right) + A_{\lambda q p} \left(\beta_\lambda \beta_{\lambda-q}^\dagger b_{qp} - \beta_\lambda^\dagger \beta_{\lambda-q} b_{qp}^\dagger \right) \Big] \qquad (72)$$

where

$$A_{\lambda q p} = \frac{4 S z \alpha \underline{J}(R_n)}{\sqrt{N}} \cdot \underline{g}_{qp} \Big[(\gamma_{\lambda-q} - \gamma_\lambda) \, sh(\theta_{\lambda-q} - \theta_\lambda) + (1 - \gamma_q) \, ch(\theta_{\lambda-q} - \theta_\lambda) \Big]$$

$$B_{\lambda q p} = \frac{4 S z \alpha \underline{J}(R_n)}{\sqrt{N}} \cdot \underline{g}_{qp} \Big[(\gamma_{\lambda-q} - \gamma_\lambda) \, ch(\theta_{\lambda-q} - \theta_\lambda) + (1 - \gamma_q) \, sh(\theta_{\lambda-q} - \theta_\lambda) \Big] \qquad (73)$$

In deriving (72) from the interaction part of (66) we have omitted the terms which represent processes involving simultaneous creation or annihilation of two magnons and one phonon since they will not conserve

energy. The diagramatic representation of the interactions described by the Hamiltonian (72) will be similar to fig. 1 as in the case of ferromagnets. It may be noted that in the processes involving β and $\beta^{\dagger}$ (cf. eq. (72)) the momentum conservation law for particles is not apparently satisfied. However, as we are dealing with quasiparticles, the momentum of the particle should not be taken identically equal to $\hbar k$.

In the next chapter we shall evaluate the charecteristic time of the magnon-phonon relaxation in antiferromagnets.

Chapter 3

MAGNON - PHONON RELAXATION PROCESSES [37,38]

A. Ferromagnets: We now study the relaxation processes due to various types of interactions discussed in the previous chapter.

(i) One phonon process:

First, we calculate the transition probabilities of the various processes connecting the initial and final states using the well known time dependent perturbation expression,[22]

$$W_{if} = (2\pi/\hbar) \left| H_p \right|^2_{if} \delta(E_i - E_f) \qquad (74)$$

It is expedient to note the properties of the creation and annihilation operators pertaining to the magnon and phonon systems.

$$\begin{aligned}
a_\lambda^\dagger \left| \cdots n_\lambda \cdots \right\rangle &= (n_\lambda + 1)^{1/2} \left| \cdots n_{\lambda+1} \cdots \right\rangle \\
a_\lambda \left| \cdots n_\lambda \cdots \right\rangle &= (n_\lambda)^{1/2} \left| \cdots n_{\lambda-1} \cdots \right\rangle \\
b_{q\nu}^\dagger \left| \cdots N_{q\nu} \cdots \right\rangle &= (N_{q\nu}+1)^{1/2} \left| \cdots N_{q\nu}+1 \cdots \right\rangle \\
b_{q\nu} \left| \cdots N_{q\nu} \cdots \right\rangle &= N_{q\nu}^{1/2} \left| \cdots N_{q\nu}-1 \cdots \right\rangle
\end{aligned} \qquad (75)$$

Here, $\left| \cdots n_\lambda \cdots \right\rangle, \left| \cdots N_{q\nu} \cdots \right\rangle$ represent the eigenkets of the unperturbed magnon and phonon Hamiltonians respecively. The explicit form of the former is given below:

$$H_{mag} = \sum_\lambda \hbar \omega_\lambda (a_\lambda^\dagger a_\lambda + 1/2) \qquad (76)$$

where $\hbar\omega_\lambda = \sum_h 2J(R_h^\circ)S\left(e^{i\underline{K}_\lambda\cdot\underline{R}_h^\circ}-1\right) + g\mu_B H$

and μ_B and H are the Bohr magneton and the external magnetic field: the last term is the Zeeman term. The eigenvalues of the magnon Hamiltonian operator, for $|\underline{K}_\lambda\cdot\underline{R}_h^\circ| \ll 1$ and for cubic crystals, can be written as

$$E_m = \sum_\lambda \left(2JSK_\lambda^2 a^2 + g\mu_B H\right)\left(n_\lambda + \tfrac{1}{2}\right) \tag{77}$$

which gives us the dispersion relation, apart from a constant term involving H,

$$\hbar\omega_\lambda = k\theta_c a^2 K_\lambda^2 \tag{78}$$

with $\theta_c = 2JS/k$, k the Boltzman constant and 'a' the nearest neighbour distance. For eigenkets of the combined ground state Hamiltonians (76) and (5), we use the notation

$$|\cdots n_\lambda \cdots ; \cdots N_{q\nu p}\cdots\rangle \tag{79}$$

Using (74), (75) and (79) the transition probability for the emission and absorption of a phonon can be expressed as

$$W\left(n_{\lambda-q_\nu}, n_\lambda, N_{q\nu p} \longrightarrow (n_{\lambda-q_\nu}+1), (n_\lambda - 1), (N_{q\nu p}+1)\right)$$
$$= \frac{2\pi}{\hbar}\left|\phi_{\lambda q\nu p}\right|^2 (n_{\lambda-q_\nu}+1)(n_\lambda)(N_{q\nu p}+1)\,\delta\left(E_{\lambda-q_\nu} + E_{q_\nu} - E_\lambda\right) \tag{80}$$

$$W\left(n_{\lambda-q_\nu}, n_\lambda, N_{-q\nu p} \longrightarrow (n_{\lambda-q_\nu}+1), (n_\lambda - 1), (N_{-q\nu p}-1)\right)$$
$$= \frac{2\pi}{\hbar}\left|\phi_{\lambda q\nu p}\right|^2 (n_{\lambda-q_\nu}+1)(n_\lambda) N_{-q\nu p}\,\delta\left(E_{\lambda-q_\nu} - E_{-q_\nu} - E_\lambda\right) \tag{81}$$

where $\phi_{\lambda q\nu p} = \frac{4}{\sqrt{N}}\sum_h 2S^\alpha \underline{J}(R_h^\circ)\cdot\underline{\mathcal{G}}_{q\nu p}\cdot$
$$\cdot\left[e^{i\underline{K}_\lambda\cdot\underline{R}_h^\circ} - e^{i(\underline{K}_\lambda - \underline{q}_\nu)\cdot\underline{R}_h^\circ} - 1 + e^{-i\underline{q}_\nu\cdot\underline{R}_h^\circ}\right] \tag{82}$$

The rate of transfer of energy between the magnon and phonon systems is expressed as

$$\dot{Q} = \sum_{q\rho} \hbar\omega_{q\rho} \langle \dot{N}_{q\rho} \rangle$$

$$= \frac{2\pi}{\hbar} \sum_{q\rho\lambda} |\phi_{\lambda q\rho}|^2 \hbar\omega_{q\rho} \delta(E_{\lambda-q} + E_q - E_\lambda) \cdot$$

$$\cdot [(n_{\lambda-q}+1)(n_\lambda)(N_{q\rho}+1) - (n_{\lambda-q})(n_\lambda+1)(N_{q\rho})] \qquad (83)$$

The δ function in (80) to (83) ensures the energy conservation. Let T_s and T_ℓ be the spin and lattice temperatures respectively which govern the equilibrium Bose distribution of magnons and phonons. Thus with $T_s = T$ and $\Delta T = T_s - T_\ell$ we can write (83) after developing the terms containing $T - \Delta T$ in terms of Taylor series in powers of ΔT. Keeping only first order terms, we get:

$$\dot{Q} = \frac{2\pi}{\hbar} \frac{\Delta T}{T^2} \sum_{q\rho\lambda} |\phi_{\lambda q\rho}|^2 \frac{(\hbar\omega_{q\rho})^2}{k} \cdot$$

$$\cdot \frac{e^{E_\lambda/kT}}{(e^{E_{\lambda-q}/kT}-1)(e^{E_\lambda/kT}-1)(e^{E_q/kT}-1)} \delta(E_{\lambda-q} + E_q - E_\lambda) \qquad (84)$$

We now change the summation into integration, and use the Debye approximation, treating the solid as an elastic homogeneous and isotropic medium. Further, we assume that all normal modes of vibration travel with the same velocity v_o, the velocity of sound. Further, $\phi_{\lambda q\rho}$ can be simplified using the approximation[24]

$$\left(\frac{\partial V}{\partial \underline{R}_h} \cdot \delta \underline{R}_h\right) \approx \left|\frac{\partial V}{\partial \underline{R}_h}\right| \left|\delta \underline{R}_h\right| ; \quad |e_{q\rho}| \approx 1$$

and hence

$$|g_{q\rho}| \approx (\hbar/2\omega_{q\rho} M)^{1/2}$$

$$\dot{Q} = \frac{2\pi}{\hbar}\,\frac{2^8}{N}\,\frac{\Delta T}{T^2}\,S^2\left[\alpha J(R_\lambda^\circ)\right]^2\left(\frac{\hbar^2 v_s^2 a^4}{2Mk}\right)a^4\left(\frac{Na^3}{(2\pi)^3}\right)^2 \cdot$$
$$\cdot\int q_\nu\left(K_\lambda^2 q_\nu^2 \cos^2\theta_{\lambda q_\nu}\right)\frac{e^{E_\lambda/kT}}{\left(e^{E_{\lambda-q_\nu}/kT}-1\right)\left(e^{E_\lambda/kT}-1\right)\left(e^{E_{q_\nu}/kT}-1\right)} \cdot$$
$$\cdot\,\delta\left(E_{\lambda-q_\nu}+E_{q_\nu}-E_\lambda\right)d\tau_\lambda\,d\tau_{q_\nu} \qquad (85)$$

where
$$d\tau_\lambda = K_\lambda^2\,dK_\lambda\,\sin\theta_\lambda\,d\theta_\lambda\,d\phi_\lambda$$
$$d\tau_{q_\nu} = q_\nu^2\,dq_\nu\,\sin\theta_{q_\nu}\,d\theta_{q_\nu}\,d\phi_{q_\nu}$$

In proceeding further we make use of the δ function relation

$$E_{\lambda-q_\nu} + E_{q_\nu} - E_\lambda = 0$$

$$\text{i.e. } 2K_\lambda q_\nu\theta_c k a^2\left(\frac{q_\nu}{2K_\lambda} - \cos\theta_{\lambda q_\nu} + \frac{\theta_D}{2\theta_c K_\lambda a}\right) = 0 \qquad (86)$$

with $\theta_D = \hbar v_s/ka$

Making use of the condition

$$\cos^2\theta_\lambda + \cos^2\theta_{q_\nu} + f^2 - 2f\cos\theta_\lambda\cos\theta_{q_\nu} \leqslant 1,$$

where $f = \frac{1}{2K_\lambda}\left(q_\nu + \frac{\theta_D}{\theta_c a}\right)$ which follows simply from (86), we integrate over the angle variables utilising the δ function property

The final result is written in terms of the variables

$$\eta = E_\lambda/kT = \theta_c a^2 K_\lambda^2/T$$
$$\xi = E_{q_\nu}/kT = a\theta_D q_\nu/T$$

$$\text{Thus } \dot{Q} = D\int_0^\infty d\xi \int_{\frac{(\xi+\beta)^2}{4\beta}}^\infty d\eta\;\xi^4\,\frac{(\xi+\beta)^2}{4}\,\frac{e^\eta}{(e^{\eta-\xi}-1)(e^\eta-1)(e^\xi-1)} \qquad (87)$$

where
$$D = \frac{4N}{\pi^3}\left(\frac{\hbar}{Mk}\right)\frac{\Delta T}{T^2}\,\frac{T^8}{\theta_D^6\,\theta_c^2}\,S^2\left[\alpha J(R_\lambda^\circ)\right]^2 \qquad (88)$$
$$\beta \equiv \theta_D^2/\theta_c T$$

For integrating (88) we follow closely the method suggested earlier[17] and find out the values for the limiting cases of low and high temperatures.

Low temperature limit: ($\beta \gg 1$)

$$\dot{Q} = \frac{2^9 \times 3N}{\pi^3} \left(\frac{\hbar}{Mk}\right) \frac{\Delta T}{T^2} \frac{T^6}{\theta_D^2 \theta_c^4} \exp(-\beta/4) S^2 \left[\alpha J(R_k^0)\right]^2 \qquad (89)$$

The exponential decrease of rate of energy transfer with decreasing temperature in (89) is the same as that obtained by Akhiezer[17] and others[20,21]; however, there are important differences in other factors.

High temperature limit: ($\beta \to 0$)

In integrating (87) for the above case the upper limit for ξ is taken as θ_D/T and in the final expression the terms in θ_D/T with power higher than two are neglected. Thus, we get

$$\dot{Q} = \frac{2N}{3\pi^3} \left(\frac{\hbar}{Mk}\right) \Delta T \left(\frac{\theta_D}{\theta_c^2}\right)^2 S^2 \left[\alpha J(R_k^0)\right]^2 \qquad (90)$$

It may be noted that $\dot{Q}$ in the high temperature limit becomes independent of temperature in our mechanism.

(ii) Two phonon process:

The second term in (52) constitutes the two phonon interaction Hamiltonian. In this, however, the processes represented by fig.2(a) and 2(b) will be relatively unimportant in that it is highly improbable

that a very high energy magnon exists which is capable of breaking into two phonons and a magnon or vice versa. Accordingly these processes are neglected. We consider the other two terms which represent the Raman processes involving phonons. The rate of energy transfer owing to two phonon Raman processes can be written as,

$$\dot{Q}_R = \sum_{q\nu\beta} \hbar\,\omega_{q\nu\beta} \langle \dot{N}_{q\nu\beta} \rangle$$

$$= \frac{2\pi}{\hbar} \sum_{q\nu\beta} \sum_{q'\beta'} \hbar\,\omega_{q\nu\beta} \Big| \sum_{\lambda\lambda'} \Phi^{\lambda\lambda'}_{\lambda q q'} \Big|^2 \Big[(n_{\lambda-q-q'}+1)(n_\lambda)(N_{q\nu\beta}+1) \times (N_{-q'\beta'}) - (n_{\lambda-q-q'})(N_{q\nu\beta})(n_\lambda+1)(N_{-q'\beta'}+1) \Big] \delta\left(E_{\lambda-q-q'} + E_{q\nu} - E_\lambda - E_{-q'\beta'}\right) \tag{91}$$

As before we define $\Delta T = T_s - T_\ell = T - T_\ell$ and making use of the Taylor's series expansion we get for $\lambda = \lambda'$

$$\dot{Q}_R = \frac{2\pi}{\hbar} \sum_{\lambda q q'} \left(\frac{\hbar^2}{k}\right) \frac{\Delta T}{T^2} \Big| \sum_{\lambda} \Phi^{\lambda\lambda}_{\lambda q q'} \Big|^2 \omega_{q\nu\beta} \left(\omega_{q\nu\beta} - \omega_{-q'\beta'}\right) \times$$

$$\times \frac{e^{(E_\lambda + E_{-q'\beta'})/kT}}{\left(e^{E_{\lambda-q-q'}/kT} - 1\right)\left(e^{E_{\lambda}/kT} - 1\right)\left(e^{E_{q\nu\beta}/kT} - 1\right)\left(e^{E_{-q'\beta'}/kT} - 1\right)}$$

$$\times\ \delta\left(E_{\lambda-q-q'} + E_{q\nu\beta} - E_\lambda - E_{-q'\beta'}\right) \tag{92}$$

The explicit form for $\sum_{\lambda} \Phi^{\lambda\lambda}_{\lambda q q'}$ can be obtained by expanding the second term in eq.(52) up to fourth order in wave vectors (the second order terms become identically equal to zero) i.e.,

$$\sum_{h} \phi^{hh}_{\lambda q q'} = \frac{2}{3N}\left[{}^{\alpha}J'(R_h^\circ) + {}^{\alpha\beta}J(R_h^\circ) + 2^{\alpha}J_\beta(R_h^\circ)\right]\frac{\hbar}{M}\cdot$$

$$\cdot\frac{s\,\dot{a}^4}{(\omega_{qp}\,\omega_{-q'p'})^{1/2}}\Big[K_\lambda^2 q_h q_h' \cos\theta_{qq'} - K_\lambda q_h^2 q_h' \cos\theta_{\lambda q'} - K_\lambda q_h q_h'^2 \cos\theta_{\lambda q}$$

$$+ 2K_\lambda^2 q_h q_h' \cos\theta_{\lambda q}\cos\theta_{\lambda q'} - 2K_\lambda q_h^2 q_h' \cos\theta_{\lambda q}\cos\theta_{qq'} -$$

$$2K_\lambda q_h q_h'^2 \cos\theta_{\lambda q'}\cos\theta_{qq'}\Big] \qquad (93)$$

We now change from summation to integration; however, a rigorous integration is hopelessly complicated owing to the occurence of several angle variables in the δ function as well as in $\left|\sum_h \phi^{hh}_{\lambda q q'}\right|^2$. One can at best attempt to give an order of magnitude estimate for certain specific conditions. We estimate (93) for the case when a high energy magnon is almost completely annihilated creating a high energy phonon and a very low energy magnon as a result of collision with a very low energy phonon. The most important term in the square bracket in (93) is then of the type $K_\lambda q_h q_h'^2 \cos\theta_{\lambda q} \sim K_\lambda q_h q_h'^2$. Considering this we get, apart from numerical factors,

$$\dot{Q}_R \approx \frac{1}{\pi^5}\,\frac{Nk^3}{M^2k^2}\,\frac{\Delta T}{T^2}\left(\frac{T}{\theta_D}\right)^{12}\left(\frac{T}{\theta_c}\right)^{5/2}\frac{1}{\theta_D}\,s^2\,J_R^2 \qquad (94)$$

where

$$J_R \equiv \left[{}^{\alpha}J'(\underline{R}_h^\circ) + {}^{\alpha\beta}J(\underline{R}_h^\circ) + 2^{\alpha}J_\beta(\underline{R}_h^\circ)\right] \qquad (95)$$

Likewise, when we consider the interaction which involves a small transfer of energy betwen the phonon and magnon system i.e. for the term $K_\lambda^2 q_\lambda q_\lambda' \cos\theta_{q_\lambda q_\lambda'} \sim K_\lambda^2 q_\lambda q_\lambda'$ we get

$$\dot{Q}_R \approx \frac{1}{\pi^5} \frac{N\hbar^3}{M^2 k^2} \frac{\Delta T}{T^2} \left(\frac{T}{\theta_D}\right)^{10} \left(\frac{T}{\theta_c}\right)^{7/2} \frac{1}{\theta_D} S^2 J_R^2 \tag{96}$$

(iii) Relaxation time for equilibration:

Now we shall calculate the characteristic time for the phonon-magnon relaxation for the establishment of equilibrium betwen the magnon and phonon systems owing to the one phonon and two phonon processes discussed in the preceding sections. The relevent expression is given by[20]

$$\frac{1}{\tau_{sl}} = \frac{\dot{Q}\,(1/C_s + 1/C_l)}{\Delta T} \tag{97}$$

where τ_{sl} is the relaxation time, C_s and C_l the specific heats of the spin and lattice systems respectively. The following explicit forms for these are taken for the low temperature limit:

$$C_s = \frac{15}{32} \frac{1}{\pi^{3/2}} kN \left(\frac{T}{\theta_c}\right)^{3/2} \tag{98}$$

$$C_l = \frac{12\pi^4}{5} kN \left(\frac{T}{\theta_D}\right)^3 \tag{99}$$

Using the expressions for $\dot{Q}_s$ in the preceding sections the various relaxation times are given below.

One phonon process:

(a) Low temperature limit: ($\beta \gg 1$)

$$\frac{1}{\tau_{sp}} = \frac{3 \times 2^9}{\pi^3} \left(\frac{\hbar}{Mk^2}\right) \frac{T^4}{\theta_D^2 \theta_c^4} \left(\frac{\theta_c}{T}\right)^{3/2} \left(11 + \frac{\beta^{3/2}}{234}\right) e^{-\beta/4} s^2 \left[\alpha J(R_L^0)\right]^2 \tag{99}$$

(b) High temperature limit: ($\beta \to 0$)

In the high temperature limit the specific heat of the lattice reduces to the well known expression $3Nk$. For the spin system, however, analogous expression has not been suggested so far. The expression (98) for C_s is valid only at low temperature. As in the case of crystal lattice vibration, if we take the upper limit of magnon wave vectors $\frac{1}{a}$, then for the high temperature limit one arrives at the expression for the specific heat of the spin system as $C_s = Nk/6\pi^2$, which is much smaller than the lattice specific heat at high temperatures. It is interesting to note that the characteristic temperature for the spin system implicit in the above assumption is the same as the temperature θ_c. Thus, (97) becomes, neglecting $1/C_\ell$ compared to $1/C_s$,

$$\frac{1}{\tau_{sp}} = 4 \left(\frac{\hbar}{Mk^2}\right) \left(\frac{\theta_D^2}{\theta_c^4}\right) s^2 \left[\alpha J(R_L^0)\right]^2 \tag{100}$$

which is independent of temperature. This point has

not been noted before; however, one must bear in mind that the spin wave approximation breaks down at high temperature regions and there may not be any physical significance in calculations pertaining to high temperature limits.

Two phonon process:

Corresponding to the two cases implied in equations (94) and (96), the relaxation times for the two phonon processes at low temperatures are given by the following expressions:

$$\left(\frac{1}{\tau_{sp}}\right)_{R_1} \approx \frac{1}{\pi^5}\left(\frac{\hbar^3}{M^2k^3}\right)\left(\frac{T}{\theta_D}\right)^{10}\left(\frac{T}{\theta_D^3\theta_c}\right)\left(11+\frac{\beta^{3/2}}{234}\right)S^2J_R^2 \qquad (101)$$

and

$$\left(\frac{1}{\tau_{sp}}\right)_{R_2} \approx \frac{1}{\pi^5}\left(\frac{\hbar^3}{M^2k^3}\right)\left(\frac{T}{\theta_D}\right)^{10}\left(\frac{1}{\theta_c^2\theta_D}\right)\left(11+\frac{\beta^{3/2}}{234}\right)S^2J_R^2 \qquad (102)$$

The corresponding expressions for the high temperature limit can be obtained from (94) and (96) by dividing them by $\Delta T N k/6\pi^2$.

B. Antiferromagnets:

As in the case of ferromagnets first we calculate the transition probabilities for the various types of interactions described by the interaction Hamiltonian (72). These are given below:

$$W\left[n_\lambda, n_{\lambda-q}, N_{qp} \longrightarrow (n_\lambda-1)(n_{\lambda-q}+1)(N_{qp}+1)\right]$$
$$= \frac{2\pi}{\hbar}\left|A_{\lambda qp}\right|^2 (n_\lambda)(n_{\lambda-q}+1)(N_{qp}+1)\,\delta\left(E_{\lambda-q}+E_q-E_\lambda\right) \qquad (103a)$$

$$W\left[n_\lambda, n_{\lambda-q_\nu}, N_{q_\nu p} \longrightarrow (n_\lambda+1)(n_{\lambda-q_\nu}-1)(N_{q_\nu p}-1)\right]$$
$$= \frac{2\pi}{\hbar}\left|A_{\lambda q_\nu p}\right|^2 (n_\lambda+1)(n_{\lambda-q_\nu})(N_{q_\nu p})\,\delta\left(E_{\lambda-q_\nu}+E_{q_\nu}-E_\lambda\right) \quad (103b)$$

$$W\left[n_\lambda, n'_{\lambda-q_\nu}, N_{q_\nu p} \longrightarrow (n_\lambda-1)(n'_{\lambda-q_\nu}-1)(N_{q_\nu p}+1)\right]$$
$$= \frac{2\pi}{\hbar}\left|B_{\lambda q_\nu p}\right|^2 (n_\lambda)(n'_{\lambda-q_\nu})(N_{q_\nu p}+1)\,\delta\left(E_\lambda+E'_{\lambda-q_\nu}-E_{q_\nu}\right) \quad (104a)$$

$$W\left[n_\lambda, n'_{\lambda-q_\nu}, N_{q_\nu p} \longrightarrow (n_\lambda+1)(n'_{\lambda-q_\nu}+1)(N_{q_\nu p}-1)\right]$$
$$= \frac{2\pi}{\hbar}\left|B_{\lambda q_\nu p}\right|^2 (n_\lambda+1)(n'_{\lambda-q_\nu}+1)(N_{q_\nu p})\,\delta\left(E_\lambda+E'_{\lambda-q_\nu}-E_{q_\nu}\right) \quad (104b)$$

$$W\left[n'_\lambda, n'_{\lambda-q_\nu}, N_{q_\nu p} \longrightarrow (n'_\lambda-1)(n'_{\lambda-q_\nu}+1)(N_{q_\nu p}-1)\right]$$
$$= \frac{2\pi}{\hbar}\left|A_{\lambda q_\nu p}\right|^2 (n'_\lambda)(n'_{\lambda-q_\nu}+1)(N_{q_\nu p})\,\delta\left(E'_\lambda+E_{q_\nu}-E'_{\lambda-q_\nu}\right) \quad (105a)$$

$$W\left[n'_\lambda, n'_{\lambda-q_\nu}, N_{q_\nu p} \longrightarrow (n'_\lambda+1)(n'_{\lambda-q_\nu}-1)(N_{q_\nu p}+1)\right]$$
$$= \frac{2\pi}{\hbar}\left|A_{\lambda q_\nu p}\right|^2 (n'_\lambda+1)(n'_{\lambda-q_\nu})(N_{q_\nu p}+1)\,\delta\left(E'_\lambda+E_{q_\nu}-E'_{\lambda-q_\nu}\right) \quad (105b)$$

where $n_\lambda, n'_\lambda, N_{q_\nu p}$ respectively represent the occupation numbers of magnons associated with energies $\hbar\omega^+_\lambda$, $\hbar\omega^-_\lambda$ and phonons of energy $\hbar\omega_{q_\nu p}$. The δ functions ensure the conservation of energy. The rate of transfer of energy between the magnon and phonon systems is given as,

$$\dot{Q} = \dot{Q}_\alpha + \dot{Q}_{\alpha\beta} + \dot{Q}_\beta$$
$$= \sum_{q_\nu p}\left[\left\langle\dot{N}_{q_\nu p}\right\rangle_\alpha + \left\langle\dot{N}_{q_\nu p}\right\rangle_{\alpha\beta} + \left\langle\dot{N}_{q_\nu p}\right\rangle_\beta\right]\hbar\omega_{q_\nu p}$$
$$= \frac{2\pi}{\hbar}\sum_{\lambda q_\nu p}\hbar\omega_{q_\nu p}\Big[\left|A_{\lambda q_\nu p}\right|^2\left\{(n_\lambda)(n_{\lambda-q_\nu}+1)(N_{q_\nu p}+1)-\right.$$

$$-(n_\lambda+1)(n_{\lambda-q_r})(N_{q_r p})\}\,\delta(E_{\lambda-q_r}+E_{q_r}-E_\lambda)$$

$$+|B_{\lambda q_r p}|^2\{(n_\lambda)(n'_{\lambda-q_r})(N_{q_r p}+1)-(n_\lambda+1)(n'_{\lambda-q_r}+1)(N_{q_r p})\}\times$$

$$\delta(E_{q_r}-E_\lambda-E'_{\lambda-q_r})+|A_{\lambda q_r p}|^2\{(n'_\lambda+1)(n'_{\lambda-q_r})(N_{q_r p}+1)$$

$$-(n'_\lambda)(n'_{\lambda-q_r}+1)(N_{q_r p})\}\,\delta(E'_\lambda+E_{q_r}-E'_{\lambda-q_r})\Big] \qquad (106)$$

In proceeding further, we neglect the Zeeman energy contribution to the magnon energy i.e. $\omega_\lambda^+=\omega_\lambda^-=\omega_\lambda$. Hence, n_λ and n'_λ may be expressed by the same Bose distribution function $1/[\exp(E_\lambda/kT)-1]$. As before, we define $\Delta T = T_s - T_l = T - T_l$ and making use of the Taylor's series expansion of terms containing $(T-\Delta T)$ in powers of ΔT and keeping only the first order terms, we get

$$\dot{Q} = \frac{2\pi}{\hbar}\frac{\Delta T}{T^2}\sum_{\lambda q_r p}\frac{(\hbar\omega_{q_r p})^2}{k}F(\lambda,q_r,p)\Big[|A_{\lambda q_r p}|^2 e^{E_\lambda/kT}\delta(E_\lambda\pm E_{q_r}-E_\lambda)$$

$$+e^{E_{q_r}/kT}|B_{\lambda q_r p}|^2\delta(E_{\lambda-q_r}+E_\lambda-E_{q_r})+|A_{\lambda q_r p}|^2 e^{E_{\lambda-q_r}/kT}\delta(E_\lambda+E_{q_r}-E_{\lambda-q_r})$$

(107)

where

$$F(\lambda,q_r,p)\equiv\frac{1}{(e^{E_{\lambda-q_r}/kT}-1)(e^{E_\lambda/kT}-1)(e^{E_{q_r}/kT}-1)} \qquad (108)$$

We now change from summation to integration and use the following dispersion relation for magnons which is obtained by neglecting ω_A compared to ω_e.

$$\hbar\omega_\lambda = \hbar\omega_e\left(1-\gamma_\lambda^2\right)^{1/2} \approx 2JSk_\lambda a(2z)^{1/2} \equiv k\theta_c K_\lambda a \tag{109}$$

where we have used the approximation $\left(\underline{K}_\lambda \cdot \underline{R}_h\right) \ll 1$.
The above equation defines the parameter θ_c .

Let us now consider the forms of the coefficients $|A_{\lambda q p}|^2$ and $|B_{\lambda q p}|^2$ under the approximation $\left(\underline{K}_\lambda \cdot \underline{R}_h\right) \ll 1$. We get

$$\begin{aligned}|A_{\lambda q p}|^2 = \frac{16S^2z^2}{N}\left(\frac{\hbar}{2\omega_{q p}M}\right)\left[\alpha J(\underline{R}_b^0)\right]^2\Big[\tfrac{1}{2}\Big\{&\left(\gamma_{\lambda-q}-\gamma_\lambda\right)^2+\left(1-\gamma_q\right)^2\Big\}\,ch\,2\left(\theta_{\lambda-q}-\theta_\lambda\right)+\left(\gamma_{\lambda-q}-\gamma_q\right)\left(1-\gamma_q\right)sh\,2\left(\theta_{\lambda-q}-\theta_\lambda\right)\\&+\tfrac{1}{2}\Big\{\left(1-\gamma_q\right)^2-\left(\gamma_{\lambda-q}-\gamma_\lambda\right)^2\Big\}\Big]\end{aligned} \tag{110}$$

Here, if we use $\tanh 2\theta_\lambda = \dfrac{-\omega_e\gamma_\lambda}{\omega_e+\omega_A} \approx -\gamma_\lambda$

$$\left.\begin{aligned}ch\,2\left(\theta_{\lambda-q}-\theta_\lambda\right) &= \left(1-\gamma_{\lambda-q}\gamma_\lambda\right)\Big/\sqrt{\left(1-\gamma_{\lambda-q}^2\right)\left(1-\gamma_\lambda^2\right)}\\ \text{and}\quad sh\,2\left(\theta_{\lambda-q}-\theta_\lambda\right) &= \left(\gamma_\lambda-\gamma_{\lambda-q}\right)\Big/\sqrt{\left(1-\gamma_{\lambda-q}^2\right)\left(1-\gamma_\lambda^2\right)}\end{aligned}\right\} \tag{111}$$

eq. (110) becomes

$$\begin{aligned}|A_{\lambda q p}|^2 = \frac{16S^2z^2}{N}\left(\frac{\hbar}{2\omega_{q p}M}\right)\left[\alpha J(\underline{R}_b^0)\right]^2 \times\Bigg[&\frac{\tfrac{1}{2}\Big\{\left(\gamma_{\lambda-q}-\gamma_\lambda\right)^2+\left(1-\gamma_q\right)^2\Big\}\left(1-\gamma_{\lambda-q}\gamma_\lambda\right)-\left(\gamma_{\lambda-q}-\gamma_\lambda\right)^2\left(1-\gamma_q\right)}{\sqrt{\left(1-\gamma_{\lambda-q}^2\right)\left(1-\gamma_\lambda^2\right)}}+\\&\tfrac{1}{2}\Big\{\left(1-\gamma_q\right)^2-\left(\gamma_{\lambda-q}-\gamma_\lambda\right)^2\Big\}\Bigg]\end{aligned} \tag{112}$$

This can be further simplified if we restrict our

study to low wave vector phonons and magnons . Thus, for a cubic crystal we shall have,

$$\gamma_\lambda = \frac{1}{z}\sum_h e^{\pm i \underline{K}_\lambda \cdot \underline{R}_h^\circ}$$
$$= \frac{1}{z}\left(z - K_\lambda^2 a^2 + \cdots\right)$$
$$\approx 1 - \frac{K_\lambda^2 a^2}{z} \qquad (113)$$

Using the above approximation eq.(112) becomes

$$|A_{\lambda q_\nu b}|^2 = \frac{16 S^2}{N}\left(\frac{\hbar}{2\omega_{q_\nu} M}\right)\left[\alpha J(R_b^\circ)\right]^2 \frac{1}{k^4}\left\{\left[\frac{1}{\theta_c^4}\left(E_\lambda^6 - E_\lambda^4 E_{\lambda-q_\nu}^2 - E_\lambda^2 E_{\lambda-q_\nu}^4 + E_{\lambda-q_\nu}^6\right) + \frac{1}{\theta_D^4}\left(E_{q_\nu}^4 E_\lambda^2 + E_{q_\nu}^4 E_{\lambda-q_\nu}^2\right) - \frac{2}{\theta_D^2\theta_c^2}\left(E_{q_\nu}^2 E_\lambda^4 + E_{q_\nu}^2 E_{\lambda-q_\nu}^4 - 2E_{q_\nu}^2 E_\lambda^2 E_{\lambda-q_\nu}^2\right)\right]\Big/\left(4E_{\lambda-q_\nu}E_\lambda\right) + \frac{1}{2}\left[\frac{E_{q_\nu}^4}{\theta_D^4} - \frac{1}{\theta_c^4}\left(E_\lambda^4 + E_{\lambda-q_\nu}^4 - 2E_\lambda^2 E_{\lambda-q_\nu}^2\right)\right]\right\} \qquad (114)$$

where we have used the relations given below.

$$E_\lambda = k\theta_c a k_\lambda \quad , \quad E_{q_\nu} = k\theta_D a q_\nu$$

We get a similar expression for $|B_{\lambda q_\nu b}|^2$ except that the sign before the second square bracket in eq.(114) is minus. It is interesting to recall that as in the case of ferromagnets the dependence of $|A_{\lambda q_\nu b}|^2$ is of the fourth order in propagation vectors. However, the present expression is more involved than the corresponding expression for ferromagnets. We discuss the integration of three terms in eq.(106) i.e. $\dot{Q}_\alpha$, $\dot{Q}_{\alpha\beta}$

and $\dot{Q}_\beta$ separately.

Thus, we have

$$\dot{Q}_\lambda = \frac{2\pi}{\hbar}\frac{\Delta T}{T^2}\frac{1}{k}\left[\frac{Na^3}{8\pi^3}\right]^2 \times \int E_{q'}^2 |A_{\lambda q' p}|^2 F(\lambda q' p)\, e^{E_\lambda/kT}\, \delta\left(E_{\lambda-q'} + E_{q'} - E_\lambda\right) d\tau_\lambda\, d\tau_{q'} \tag{115}$$

where

$$d\tau_\lambda = K_\lambda^2\, dK_\lambda \sin\theta_\lambda\, d\theta_\lambda\, d\phi_\lambda$$

$$d\tau_{q'} = q'^2\, dq'\, \sin\theta_{q'}\, d\theta_{q'}\, d\phi_{q'}$$

Integrating over angle variables with the help of δ function which gives a factor proportional to $E_{\lambda-q'}$, the above expression reduces to the following form.

$$\dot{Q}_\lambda = \frac{8\pi^3}{\hbar}\frac{\Delta T}{T^2 k^3 \theta_c^2 a^2}\left[\frac{Na^3}{8\pi^3}\right]^2 \times \int E_{q'}^2 |A_{\lambda q' p}|^2 E_{\lambda-q'} F(\lambda q' p)\, K_\lambda q'\, dK_\lambda\, dq' \tag{116}$$

This expression with the help of the dimensionless variables $\eta \equiv E_\lambda/kT$ and $\xi \equiv E_{q'}/kT$ can be written as

$$\dot{Q} = G\int_0^\infty \int_{\xi r}^\infty \phi^+(\eta,\xi)\, \frac{e^\eta\, \xi^2\, \eta(\eta-\xi)}{(e^{\eta-\xi}-1)(e^\eta-1)(e^\xi-1)}\, d\xi\, d\eta \tag{117}$$

where $G = \frac{N}{\pi^3}\left(\frac{\hbar}{Mk}\right)\frac{\Delta T}{T^2}\frac{T^{10}}{\theta_D^2\theta_c^4} S^2\left[{}^\alpha J\left(\underline{R}_\lambda^\circ\right)\right]^2$,

$$\phi^\pm(\eta,\xi) = \left\{\left[\frac{1}{\theta_c^4}\left(8\eta^4\xi^2 - 16\eta^3\xi^3 + 14\eta^2\xi^4 - 6\eta\xi^5 + \xi^6\right)\right.\right.$$

$$\left.\left. + \frac{1}{\theta_D^4}\left(2\eta^2\xi^4 - 2\eta\xi^5 + \xi^6\right) + \frac{2}{\theta_D^2\theta_c^2}\left(4\eta\xi^5 - 4\eta^2\xi^4 - \xi^6\right)\right]\right/ 4\eta(\eta-\xi)$$

$$\pm \frac{1}{2}\left[\frac{\xi^4}{\theta_D^4} + \frac{1}{\theta_c^4}\left(4\eta\xi^3 - 4\eta^2\xi^2 - \xi^4\right)\right]\Bigg\} \tag{118}$$

and $\gamma = (\theta_c + \theta_D)/2\theta_D$. For $\theta_c > \theta_D$, $\gamma > 1$; however, for $\theta_c < \theta_D$ we have to use $\gamma = 1$ to satisfy the δ function condition. In the case of low temperature limit the integral in eq.(118) is easily evaluated following the procedure used in the case of ferromagnets. We get $\dot{Q}_\alpha$ (low temperature limit)

$$= 10^4\, G\left[\frac{1\cdot7}{\theta_c^4} + \frac{1\cdot7}{\theta_D^4} - \frac{3\cdot3}{\theta_D^2\,\theta_c^2}\right] \tag{119}$$

Following the above procedure, we get, after making use of the δ function while integrating, the values of $\dot{\theta}_{\alpha\beta}$ and $\dot{Q}_\beta$. Summing all the three expressions, we obtain

$\dot{Q}$ (low temperature limit)

$$= \frac{10^4}{N}\left(\frac{\hbar}{MR}\right)\frac{\Delta T}{T^2}\,\frac{T^{10}}{\theta_D^2\,\theta_c^4}\,S^2\left[\alpha\, J(\underline{R}_\lambda^\circ)\right]^2\left[\frac{4\cdot7}{\theta_c^4} + \frac{5\cdot3}{\theta_D^4} - \frac{9\cdot6}{\theta_D^2\,\theta_c^4}\right] \tag{120}$$

Relaxation time for equilibration:

As in the case of ferromagnets, the relaxation time for phonon-magnon interaction τ_{sp} is expressed as,

$$\frac{1}{\tau_{sp}} = \frac{\dot{Q}\left(\frac{1}{c_s} + \frac{1}{c_l}\right)}{\Delta T} \tag{121}$$

The spin system specific heat C_s for a b.c.c. antiferromagnet will be given by[15]

$$C_s = 4Nk\left(\frac{T}{\theta_c}\right)^3 \tag{122}$$

Substituting eqs. (99), (120) and (122) into (121) we obtain

$$\frac{1}{\tau_{sp}} = \frac{10^4}{\pi^3}\left(\frac{\hbar'}{MR^2}\right)\frac{T^5}{\theta_D^2\theta_c^4}\left(\frac{\theta_c^3}{4}+\frac{\theta_D^3}{234}\right)\left(\frac{4.7}{\theta_c^4}+\frac{5.3}{\theta_D^4}-\frac{9.6}{\theta_B^3\theta_c^2}\right)\times \times S^2\left[\alpha J\left(\underline{R}_k^\circ\right)\right]^2 \tag{123}$$

It is interesting to note that the above expression gives a simple law of the temperature dependence of the spin-lattice relaxation time in antiferromagnets, namely,

$$\tau_{sp} \propto \frac{1}{T^5} \tag{124}$$

Chapter 4

Estimates and Discussion[37,38]

A. Ferromagnets: In the expressions for the various relaxation times derived in the previous chapter most of the factors are easily determined except those involving $^{\alpha}J(\underline{R}_{\ell}^{\circ})$ and J_R. A knowledge of these will require the calculation of the exchange integrals involving excited orbitals as well as the matrix elements of the various derivatives of the crystal field potential with respect to excited and ground state orbitals. We consider an order of magnitude estimate of these quantities for a body centered cubic system such as iron. The excited orbitals for iron are taken to be the 4p empty orbitals. Thus, the transitions involved are from 3d to 4p, which for the cubic symmetry of the system can be achieved owing to the odd vibrations of the surrounding ions with respect to the ion in question. For the actual estimate we consider[24] a collinear system of three ions lying on the body diagonal of the unit cell. The potential energy of an electron at the central ion in the field of two diagonally opposite nearest neighbours at the equilibrium position can be taken as,

$$V = -\left(\frac{ze^2}{|\underline{R}_o - \underline{r}|} + \frac{ze^2}{|\underline{R}_o + \underline{r}|}\right) \qquad (125)$$

where R_o is the distance of the neighbouring ion from the central ion, r the coordinate of the electron referred to the central ion and ze is the charge of the ion. For an odd vibration of the unit chosen, the first and second derivatives of the potential are:

$$|\underline{V}^h| = |(\partial V/\partial \underline{R}_h)_o| = 4ze^2 r/R_o^3$$

$$|\underline{V}^{hh}| = |(\partial^2 V/\partial \underline{R}_h^2)_o| = 6ze^2 r/R_o^4$$

Thus, the matrix elements required have the following form,

$$\langle \psi_{3dz^2} | \underline{V}^h | \psi_{4p_z} \rangle \sim 4ze^2 r_o/R_o^3 \qquad (126)$$

$$\langle \psi_{3dz^2} | \underline{V}^{hh} | \psi_{4p_z} \rangle \sim 6ze^2 r_o/R_o^4 \qquad (127)$$

with r_o standing for the radius of the ion. For iron the charge of the ion core is taken for that of the ion without the 4s electrons, i.e. $+2e$ and the radius $r_o \sim 1\ A^\circ$. Thus, with $R_o = 2.5\ A^\circ$, the value of $\langle \psi_{3dz^2} | \underline{V}^h | \psi_{4p_z} \rangle \sim 1.25 \times 10^{-3}$ dynes. Likewise the value of (127) is 0.75×10^5 dynes/cm.

The energy denominator $\Delta E_\alpha = E_\alpha - E_a$ is taken to be the difference in 4p and 3d bands energy levels

for iron series metals. This is estimated from X-ray spectroscopy data[25] as $\Delta E_{s} \sim 5 ev$. A reasonable range will be 5 to 10 ev.; however, to be on the safe side we shall use the value $\Delta E_{s-p} \sim$ 10 ev. For an estimate of the exchange integrals of the type $\langle \psi_{4p}^{(1)} \psi_{3d} \mid g_{12} \mid \psi_{3d}^{(1)} \psi_{3d}^{(2)} \rangle$ we make use of the calculations of similar integrals in certain other studies[26]. Its value is of the order of 0.01 a.u.(0.27 ev.). We use the value of 0.1 ev. in our estimate. Thus, for iron at $T = 10^{\circ} K$,and with $\theta_c = 1000^{\circ} K$ and $\theta_D = 500^{\circ} K$, we get the estimate of $(1/\tau_{sp}) \approx 2.5 \times 10^{6}\ sec^{-1}$ i.e. $\tau_{sp} \sim 10^{-6}$ sec. for the one phonon process taking $S = 1$. This estimate seems to be of the right order of magnitude as can be seen by comparing with the values suggested earlier[20]. For the high temperature limit we get the value of $(1/\tau_{sp})$ as $5 \times 10^{10} sec^{-1}$. Thus, the relaxation time τ_{sp} is of the order of 10^{-11} sec. This value may be taken to be the limiting value since $(1/\tau_{sp})$ in this limit becomes independent of temperature. From this it is reasonable to expect that at room temperature τ_{sp} will be around 10^{-8} to 10^{-9} sec. in agreement with the value suggested by Bloembergen and Damon[27].

B. Antiferromagnets: We shall now apply the theory of phonon-magnon relaxation developed earlier to some specific systems. Unfortunately we can not compare the theoretical estimates with any experimental value in that none is available for any system. The system which can be taken as fairly closely resembling the model chosen i.e. , a body centered cubic distribution of magnetic ions with each interpenetrating simple cubic lattice representing one of the two sublattices, is perhaps MnF_2 (body centered tetragonal structure) on which some AFMR experiments have been carried out. The estimated exchange field for this system is of the order of 10^5 and $H_A \sim 10^3$ Oersteds. Following Weiss approximation, θ_c is estimated to be 30° K for $T_N \sim 70^\circ$ K(the Néel temperature for MnF_2). A rough measure of θ_D can be obtained by making use of the Lindeman relation[28]:

$$\theta_D = B\left(T_M / M V^{2/3}\right)^{1/2}$$

where M is the mean atomic weight and V is the mean atomic volume. Using a value of the constant B equal to 115 we obtain $\theta_D \sim 50^\circ$ K. One gets nearly the same value by using the formula[28]

$$\theta_D \approx (\hbar/k)\left(10\alpha/9MX\right)^{1/2}$$

where χ is the compressibility. The value of χ is taken from the data of Benedek and Kushida[29]. In estimating

$$^{\alpha}J(\underline{R}_k^0) = \sum_{\alpha} \langle \phi_\alpha | \underline{V}^k | \phi_i \rangle \langle \phi_\alpha \phi_m | V_{12} | \phi_m \phi_i \rangle / \Delta E_\alpha$$

the procedure is the same as discussed for the case of ferromagnets. $\langle \phi_\alpha | \underline{V}^k | \phi_i \rangle$ is expected to be of the order of 1×10^{-3} dynes as shown from our earlier estimate for ferromagnets as well as the ligand field calculations on inorganic complexes.[30] The exchange integral of the type $\langle \phi_\alpha \phi_m | V_{12} | \phi_m \phi_i \rangle$ may be estimated from the latest calculations of Freeman and Watson[31]. On the basis of their calculation a minimum value of the above integral can be taken to be of the order of 10^{-3} ev. Actually, for the present integral one may expect a larger value because of the extended nature of the excited orbitals. Further, the superexchange effects are mainly responsible for the spin coupling in magnetic compounds such as MnF_2 and the excited orbitals are to be chosen on the magnetic centers or linear combinations of atomic orbitals involving magnetic as well as nonmagnetic ions. Thus, using a value of $\Delta E_\alpha \equiv E_\alpha - E_0 \sim 10$ ev. , we get $^{\alpha}J(\underline{R}_k^0)$ of the order of 1×10^{-7} dynes. If one identifies $^{\alpha}J(\underline{R}_k^0)$

with $(dJ/dR_{\lambda}^{\circ})$, another estimate can be made from the data of Benedek and Kushida[29]. Using their values, namely,

$$(1/T_N)(\partial T_N/\partial P) = (4.4 \times 10^{-6})/(kg/cm^2)$$

$$(1/a)(\partial a/\partial P) = -(0.45 \times 10^{-6})/(kg/cm^2)$$

$$(1/c)(\partial c/\partial P) = -(0.31 \times 10^{-6})/(kg/cm^2)$$

where P is the hydrostatic pressure, a and c being the lattice constants of MnF_2, we get $\propto J(R_{\lambda}^{\circ}) \sim dJ/dR_{\lambda}^{\circ} \sim 6 \times 10^{-8}$ dynes. This is in rough agreement with the value noted above; however, to be on the safe side, we shall use a value of $\propto J(R_{\lambda}^{\circ})$ to be of the order of 10^{-8} dyn. Thus, with S = 5/2 and at T = 10° k, we obtain

$$1/\tau_{sp} \approx 10^{8} \text{ sec}^{-1}$$

i.e., τ_{sp} at 10° k is of the order of 10^{-8} sec. However, keeping in view the uncertainties in the values taken for the various parameters involved, the above may be considered to be a tentative estimate.

In contrast to the ferromagnets an important difference is the absence of an exponential temperature factor in the expression for τ_{sp} in antiferromagnets. This arises owing to the linear dispersion relation for antiferromagnets, namely, $E_{\lambda} \propto |K_{\lambda}|$. From the

experimental results on the line width of AFMR on MnF_2 we expect the magnon-magnon relaxation time to be of the order of 10^{-10} sec. at such low temperatures. Thus, the assumption implicit in our theoretical analysis that the magnons are in statistical equilibrium with each other seems to be justified. It is desirable to have more experimental results on the AFMR of various antiferromagnets before a more quantitative estimate is attempted.

Recently Nettelton[32] has used the matrix elements calculated here for one-phonon interactions with spin waves for the case of antiferromagnets to calculate the phonon relaxation time. The resulting analytical expression has been fitted to measurements by Slack on single crystals of MnF_2, CoF_2 and MnO. Nettelton finds very good fit for MnF_2, for which experiment and theory agree from 0° k to the maximum at 14.7° k where three phonon processes take over.

The foregoing analysis shows that the mechanism suggested here is of impotance in the relaxation processes involving the ordered spin and lattice systems. Although, the estimates have been made for cubic systems, the process may have wider validity for other systems such as the ferrites and garnets.

That such mixing of excited states with ground state doesoccur is supported by the absorption spectra study for systems containing the transition metal ions[33].

Although, we have restricted ourselves in the present study to the consideration of direct exchange interaction as the mechanism responsible for magnetic ordering, it is not difficult to take into account the other possible mechanisms such as the indirect exchange interaction through nonmagnetic ions studied by Kramers[34], Anderson[35], Sinha[26] and others[36] and Zener's theory of magnetic ordering. In fact, some recent calculations such as by Watson and Freeman[31], Carr and others show that the direct exchange mechanism is not the dominant mechanism of magnetic ordering for transition metals. However, it seems that whatever be the dominant mechanism of magnetic ordering the mixing of ground and excited states of the magnetic ions due to crystal field oscillations will play an important role in the interactions betwen phonons and magnons.

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