Interactions of External Radiations with Magnons in Magnetic Crystals.

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Interactions between spin-waves and external radiations i.e. neutrons and photons have been investigated in antiferromagnetically ordered systems.

In Chapter I a survey of the present literature on neutron diffraction in magnetic solids and infrared spectra of solids is taken. The two theories proposed by Tanabe et al. and Halley & Silvera, to explain the two-magnon absorption in the infrared spectrum of antiferromagnetic materials are discussed.

In the second chapter of the dissertation the interaction between magnons and neutrons in antiferromagnetically coupled systems is derived by taking into account the mixing of ground and excited orbital states of localized electrons of magnetic atoms due to interactions with neutrons. This is relevent to magnetic crystals whose magnetic atoms allow orbital transitions owing to interaction with neutrons. The interaction terms are derived by formulating the Heisenberg type exchange interaction in terms of atomic states which are perturbed owing to the scattering with neutrons. For a two sub-lattice antiferromagnet the interaction terms have the form:

$$H_{m_n}(af) = \sum \Big[A_{\lambda q} \Big\{ \eta^{+}_{k'\sigma'} \eta_{k\sigma} \big(a^{\dagger}_{\lambda} a_{\lambda - q} + \beta^{+}_{\lambda - q} \beta_{\lambda} \big) \Big\}$$

$$+ B_{\lambda q} \Big\{ \eta^{+}_{k'\sigma'} \eta_{k\sigma} \big(a_{\lambda - q} \beta_{\lambda} + a^{+}_{\lambda - q} \beta^{+}_{\lambda} \big) \Big\} \Big]$$

where $A_{\lambda q}$ & $B_{\lambda q}$ are the coupling constants, $\eta^{+}_{k\sigma}(\eta_{k\sigma})$ the neutron creation (annihilation) operators for wave vector k and spin σ, $\alpha^{+}_{\lambda}(\alpha_{\lambda})$ and $\beta^{+}_{\lambda}(\beta_{\lambda})$ are the magnon creation (annihilation) operators respectively with wave vector λ and corresponding to the two branches. Scattering cross-sections for all the two-magnon Raman processes are calculated in terms of the matrix elements of the magnon-neutron interaction between the combined neutron-magnon states. The temperature dependence in the low temperature region of the scattering cross section ($d^2\sigma/dEd\Omega$) is given by

$$\frac{d^2\sigma}{dEd\Omega}(\text{total}) \propto \frac{k'}{k}\frac{T}{\theta_c}\left[\frac{5}{2}\, e^{\frac{\theta_c ab}{T}} + \frac{1}{2}\, e^{\frac{-\theta_c ab}{T}} + e^{\frac{2\theta_c ab}{T}}\right]$$

where , $b = \frac{\hbar^2(k^2 - k'^2)}{2m_n k_B \theta_c a}$, k and k' being the wave vectors of incident and scattered neutron. Order of magnitude estimate for two-magnon emission process has been done for NiF_2. This may perhaps explain the hump observed by Erickson before the (100) reflection in the neutron diffraction data.

The third chapter of the thesis deals with the magnon photon interaction in antiferromagnetic systems e.g. FeF_2, MnF_2 etc. Magnons and photons interact via the transverse optical lattice vibrations. The relative displacement $\delta u(r)$ between two ions carrying opposite charges, leads to a dielectric polarisation $P(r) = ne^* \delta u(r)$, e^* being the effective charge and n the number of ions pairs per unit volume. This polarisation gives rise to instantaneous 'local' electric field $E = \frac{4\pi P(r)}{3}$. The magnetic field associated with this electric field, from Maxwell's equation is

$$H(\underline{r}) = \sum_{\underline{q}} (C_{\underline{q}})(b^{+}_{\underline{q}} - b - \underline{q}) \times (e^{i.\underline{q}.\underline{r}} \underline{e}_{\underline{q}})$$

$C_{\underline{q}}$ being the coupling coefficient, $b^{+}_{\underline{q}}$, $b_{\underline{q}}$ the creation, annihilation operators for phonons and $\underline{e}_{\underline{q}}$ the unit polarisation vector. This magnetic field gives rise to a Zeeman term $H(\underline{r}).\underline{s}$. The transverse optical phonons also interact with the photons of the electro-magnetic radiation. The phonons are eliminated by a suitable canonical transformation giving the required magnon-photon interaction. The absorption coefficients for the two cases namely (a) the electric vector parallel and (2) perpendicular to the crystal axis are calculated giving a singularity in absorption at the frequency $w = 2(w_A + w_e)$, w_A and w_e being the anisotropy and exchange frequencies. The order of magnitude calculation for the ratio of the 2 absorption coefficients has been evaluated for FeF_2.

CHAPTER - 1

INTRODUCTION

The first attempt to explain theoretically the magnetic properties of ferromagnets on an atomic model was made by Heisenberg in 1928.[1] According to his theory the magnetism in ferromagnets arises due to the spins of localized unpaired electrons arranged in a definite pattern, each spin being coupled strongly by exchange forces with its neighbours. His description is based on a simple Heitler - London description in which periodicity of the lattice is not taken into account.

Bloch[2] in an attempt to improve upon Heisenberg's work introduced the concept of spin waves. According to Bloch, a spin wave is a single spin reversal moving over a large number of otherwise aligned spins in a crystal lattice. An alternative approach to introduce the concept of spin wave was made by Holstein and Primakoff (hereafter referred to as HP). They considered a three dimensional network of spins coupled ferromagnetically in an external magnetic field. They defined a set of co-ordinates which describe the quantum state of the system accurately. In terms of these coordinates the total Hamiltonian of the system consists of two parts; the first comprises terms quadratic in the new variables defined by HP[3] with the explicit form

$$H = \sum_{\underline{\lambda}} \hbar w_{\underline{\lambda}} \left(a^{+}_{\underline{\lambda}} a_{\underline{\lambda}} + \frac{1}{2} \right)$$

where $a^{+}_{\Delta} a_{\Delta}$ is the occupation number operator of the spin waves with positive integers as its eigenvalues. This measures the deviation of S_z from its maximum value S. This expression suggests that the spin system considered may be likened to a system of non-interacting particles with energy $\hbar\omega$. These are known as magnons which are Bosons. The other part of the Hamiltonian consists of higher order terms in the creation (a^{+}_{Δ}) and annihilation operators(a_{Δ}) and represents the mutual interaction between magnons. Dyson[4] has conclusively established that the linear Bloch theory with non-interacting spin waves is equivalent to HP theory neglecting higher than second order terms.

The extension of the concept of spin waves to the case of antiferromagnets is not straightforward. This was done by Hulthen,[5] Ziman,[6] Anderson,[7] Kubo[8] and others.[9,10] 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

$$H = 2J \sum_{n.n} \underline{S}_1 \cdot \underline{S}_m$$

l refers to an atom on sublattice 1 and m to one on sublattice 2, 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 cannot 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 spins in this plane would still be arbitrary. In an actual antiferromagnet this degeneracy is removed by the presence of anisotropy field.[10] This anisotropy field is such that the spins on sublattice 1 are preferentially 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

$$H = 2J \sum_{n.n} \underline{S}_l \cdot \underline{S}_m - H_A g \mu_B \left(\sum_l S_l^z - \sum_m S_m^z \right) - H_0 g \mu_B \left(\sum_j S_j^z \right).$$

g is the spectroscopic splitting factor, u_B is Bohr magneton being given by $\frac{e\hbar}{2m_e c}$ and H_0 is the external magnetic field. 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 $H_A \to \infty$ the ground state for the Hamiltonian operator 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[10] and others show that this model is only about 7 % wrong. Further confirmation of the validity of the model of interlocking sublatticeshas been provided by the experiments of Shull and Smart[11] 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 whereby the role of the 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 and the reverse to the other can be regarded as stable.

The energy spectrum and other properties of the elementary excitations described above can be studied by the magnetic resonance and neutron diffraction techniques.

Neutron diffraction furnishes a valuable approach in the study of magnetism on an atomic level. The basic characteristic of neutrons is the de Broglie wavelength

$\lambda = \hbar/mv$ associated with them. The most abundantly available thermal neutrons have the wavelength of the order of $\sim 1\ A^{\circ}$ which is of the same order as interatomic distances and hence neutrons are ideally suited for interatomic interference studies.

For neutrons interacting with solids there are two important interactions: (1) short range interaction with atomic nuclei, and (2) interaction of magnetic moment of the neutron with atomic moments. The cross-sections due to these two are of the same order of magnitude for magnetic crystals and hence the magnetic scattering can be measured with good accuracy. The magnetic scattering consists of (1) magnetic Bragg scattering which gives information about orientation and length of the individual magnetic moments in the ordered state and the shape of the magnetization density in a given ion; (2) diffuse scattering which is important especially above the order-disorder transition. Below transition temperature from the study of energy distribution of scattered neutrons, one can determine the spectrum of elementary excitations.

The basic concepts of magnetic scattering of neutrons were developed by Bloch[12] and by Schwinger.[13] Halpern and Johnson[14] derived the formulae for purely magnetic neutron scattering. They considered the two extreme cases of a set of uncoupled paramagnetic ions and of ferromagnetic systems with their electronic spins locked rigidly in an ordered

array. Van Hove[15] devised a very general time-dependent approach to both nuclear and magnetic scattering of slow neutrons. He considered pair correlation between spins at different times in a ferromagnetic crystal and used this to derive the general formulae for the angular and energy distribution of magnetically scattered neutrons.

A number of experimental and theoretical investigations have dealt with the effects of exchange coupling on the magnetic scattering of unpolarized neutrons in the paramagnetic and spin wave regions. In the paramagnetic region many investigations of purely magnetic scattering have been carried out by the method of moments. As early as 1939 Van Vleck[16] pointed out the relationship between the inelastic magnetic scattering in this region and the range of exchange constants of the pertinent lattices. For the cases of nearest neighbour exchange interaction Van Vleck calculated the second and fourth moments of neutrons energy transfer. However, he did not consider the interference effects due to short range magnetic order. Slotnick[17] took account of these effects and using stationary state methods, dealt with the purely magnetic scattering by paramagnetic exchange coupled lattices with interactions between nearest and next nearest neighbours.

De Gennes[18] studied the magnetic scattering by Van Hove's approach. He dealt mainly with very large values of T in the paramagnetic region so that the temperature dependence of the quantities of interest was disregarded. He treated the energy

transfer and the momentum transfer vectors of a neutron to be independent variables in contrast to the viewpoint taken by Slotnick and by Sa'enz.[19]

On the experimental side, Shull and is collaborators,[20] Erickson[21] and Brockhouse and others[22] observed the effects of short range order on the integrated cross-sections in the paramagnetic region. Bendt[23] studied inelastic magnetic scattering by indirect methods and Brockhouse and his collaborators studied it by direct energy analysis method. In a broad manner these studies confirm the theories of Van Vleck[16] and Slotnick.[17]

By combining the method of time correlation functions with the spin wave formulation of Holstein and Primakoff,[3] Van Hove[15] studied the purely magnetic one-magnon scattering of neutrons for ferromagnetic systems. Using this formulation and that of Ziman,[6] Elliot and Lowde[24] investigated the spin wave scattering for both ferromagnetic and antiferromagnetic systems. They also discussed the magneto-vibrational scattering and gave the rules of separating it experimentally from the purely magnetic scattering. Maleev[25] performed an improved calculation of the above type of ferromagnetic spin wave scattering. Kaplan[26] treated this problem for normal spinels with nearest neighbour exchange interactions. He based this treatment on his spin wave analysis of these spinels.[27]

On the experimental side of the investigation of spin wave scattering one may mention the work of Lowde[28] for iron

and of Riste, Blinowski and Janik[29] for magnetite involving no energy analysis of outgoing neutrons, and the studies of Brockhouse[30] on the energy spectrum of neutrons scattered by magnetite. These experimental studies support the main conclusions of Elliot and Lowde[24] and Kaplan.[27] Spin wave scattering of initially unpolarized neutrons for ferromagnets has also been investigated by Lowde and Umanatha[31] for iron and by R.N.Sinclair and Borckhouse[32] for a f.c.c. Co alloy.

A.W.Saenz[19] has considered the magnetic scattering of slow neutrons of arbitrary initial polarization by time-dependent operator approach for the case of complete orbital quenching of the magnetic ions. He divided it into purely magnetic and magneto-vibrational scattering involving zero phonon processes and nonzero phonon processes respectively. He applied the formulae to temperature regions which are sufficiently large or sufficiently small compared to the order-disorder transition temperature. He also extended the theory to the case of an arbitrary number of magnetic ions per magnetic unit cell. Nagai and Yashimori[33] gave detailed calculations on the spin wave spectrum of MnF_2 and on the scattering of neutrons by spin waves in this substance.

These theoretical investigations of the scattering of neutrons by magnons have mainly dealt with neutron scattering accompanying absorption or emission of a single magnon[19,24,34] i.e. one-magnon processes and the orbital effects have not been considered. Joshi and Sinha,[35] in their study of two-magnon

neutron scattering in ferrimagnets, have taken account of the orbital states which are admixed with the ground state of the atoms owing to their interaction with neutrons.

The light scattering by magnetic materials is another valuable tool for the study of magnetism. The photons of the electromagnetic field interact with the magnons in various orders giving one-magnon, two-magnon processes etc. These are analogous to ordinary Raman processes in which one or more phonons are emitted or absorbed by the photons in their interactions with matter.

The light scattering by the spin waves in antiferromagnets, especially the transition metal fluorides, has received considerable attention both experimentally and theoretically. The first observation of the magnon absorption was reported by Halley and Silvera[36] for FeF_2. Richards[37,38] reported a similar observation for antiferromagnetic MnF_2 and CoF_2. Various workers observed the magnon side-bands in magnetic systems.[39,42] Fleury et. al.[43] studied the light scattering (temperature dependence) of both one- and two-magnon processes for FeF_2. These might be Raman scattering of magnons.

First order or one-magnon scattering can be identified by the magnitude and temperature dependence of the frequency shift of the scattered light, by the polarization selection rules observed to govern the scattering and the disappearance of the scattered light as the sample temperature is raised above the Ne'el temperature. The identification of second

order or two-magnon scattering is similar though not so definite. The two-magnon absorption line found at 154.4 cm^{-1} in FeF_2 has the following properties:[44] (1) The absorption is greatest when the E vector of the incident radiation is parallel to the c-axis of the crystal, (2) The line broadens and disappears as the temperature is raised above the Neel temperature, (3) The absorption intensity is comparable to that of the antiferromagnetic resonance line observed at 52.7 cm^{-1}, (4) The frequency varies with temperature decreasing more slowly than the antiferromagnetic resonance frequency as $T \rightarrow T_N$, (5) The line does not disappear as $T \rightarrow 0$. (6) The line does not appear to broaden or split in the magnetic field.

Allen et al.[38] observed similar two-magnon absorption line at 100 cm^{-1} in MnF_2. The strength, temperature dependence and insensitivity to magnetic field are similar to those for FeF_2 mentioned above observed by Halley and to those for CoF_2 observed by Richards.[37] The ratio of integrated intensities with $E \parallel C$ - and $E \perp C$ - axis of the crystal is $\sim$ 7 for FeF_2 and $\sim$ 25 for MnF_2.

On the theoretical side Shen and Bloembergen[45] were the first to treat the interaction between light waves and spin waves in magnetic materials. They considered the interaction in a way analogous to the coupling of light with phonons and predicted magnon Raman effects in para, ferro- and antiferromagnetic solids. The total Hamiltonian can be written as

$$H = H_{mag} + H_{orb} + H_{int}$$

where the interaction Hamiltonian consists of spin-orbit, spin-radiation and orbit-radiation interactions. It has the form

$$H_{int} = \sum_{m,b} \lambda_{mb} \underline{L}_{mb} \cdot \underline{S}_{mb} - \sum_{m.b.} [e\underline{r}_{mb} \cdot \underline{E}_{mb} + \mu \underline{L}_{mb} \cdot \underline{H}_{mb}]$$

$$- \sum_{m.b} 2 \mu \underline{S}_{mb} \cdot \underline{H}_{mb} \quad .$$

$\underline{L}$ is the orbital angular momentum operator, λ_{mb} is the spin-orbit coupling constant, $\underline{E}_{mb}$ and $\underline{H}_{mb}$ the electric and magnetic fields of the radiation at the b^{th} atom in the m^{th} unit cell. $\mu \underline{L} \cdot \underline{H}$ and $2\mu \underline{S} \cdot \underline{H}$ correspond to magnetic dipole transitions. Density matrix formulation is used to derive the magnon wave equation. The coupling constant is found to be independent of temperature in the harmonic approximation. The gain effect for stimulated Raman effect has also been calculated. From this, they find that the spin Raman process appears to be two orders of magnitude smaller than the ordinary Raman effect in liquids. However, the interaction considered by Shen and Bloembergen[46] gives only Raman Processes, whereas, both one-magnon and two-magnon absor tion lines have been observed.

For the explanation of two-magnon absorption line found in antiferromagnetic FeF_2, Halley and Silvera[36,44,46] proposed a mechanism which can be described as follows: The model includes magnon, phonon, photon and exciton fields in

zero order. The interaction Hamiltonian consists of spin-orbit, magnon-phonon, electric dipole and crystal field interactions and also the quadrupole dipole part of the Coulomb interaction between magnetic ions. The last one leads to a coupling between magnons and odd parity excitons. Physically it amounts to the removal of the centre of symmetry at an ion site, say j, by the excitation of a long wavelength magnon. The excitation of magnon leads to asymmetrical precession of spins at $j \pm \underline{\delta}$ as compared to the spin at j. The spins at $j \pm \underline{\delta}$ interact with their charge clouds via the spin-orbit interaction $\underline{L}.\underline{S}$. The charge clouds at $j \pm \underline{\delta}$ then interact with the charge cloud at j by the quadrupole dipole interaction. This together with the spin-orbit interaction $\underline{L}.\underline{S}$ at the ion site j leads to a coupling between two electronic levels of j which have even and odd parity respectively. Consider two electronic levels of an ion at j. The external electromagnetic field couples the two electronic states via the interaction $(-e/mc)\ \underline{A}.\underline{P}$. Here $\underline{A}$ is the vector potential, $\underline{P}$ the momentum operator, e and m the charge and mass of the electron, c is the velocity of light. The corresponding exciton state is coupled to a two-magnon state via the combined action of the coupling between magnons and excitons as discussed above and the spin-orbit interaction on j. Explicitly the spin-dependent quadrupole moment at $\underline{j} + \underline{\delta}$ is

$$Q_{j+\delta} = 2\lambda \sum_{m \neq 1} \langle \Gamma_1 | Q^{(QP)} | \Gamma_m \rangle \langle \Gamma_m | L_{j+\underline{\delta}} | \Gamma_1 \rangle \cdot \frac{S_{-i+\underline{\delta}}}{E_1 - E_m}$$

Here $Q^{(OP)}$ is the electron quadrupole-moment tensor operator at the site j, $\underline{L}$ the angular momentum operator and $|\Gamma_l\rangle$ and $|\Gamma_m\rangle$ the electronic ground and excited states. The potential at j arising from these spin-dependent moments at $\underline{j}+\underline{\delta}$ is

$$V_j = e \sum_{i=1}^{n} \sum_{\underline{\delta}} (\underline{X}_i - \underline{X}^{(o)}_{j+\underline{\delta}}) \cdot Q_{j+\underline{\delta}} \cdot (\underline{X}_i - \underline{X}^{(o)}_{j+\underline{\delta}}) / |\underline{X}_i - \underline{X}^{(o)}_{j+\underline{\delta}}|^5 .$$

Here $\underline{X}_i$ are the co-ordinates of n electrons of the j^{th} ion. This, in the second quantised formalism, in which electronic states are treated as excitons, leads to interaction between magnons and odd-parity excitons.

Using this mechanism Halley[44] and Silvera have calculated the two-magnon absorption frequency to be 154 cm^{-1} which is very close to the observed value 154.4 cm^{-1} for FeF_2. They observed this singularity in the absorption for the case when the electric field of the radiation is along the crystal axis. The ratio of the integrated intensities for $E \parallel C$ and $E \perp C$ estimated by them is found to be two orders of magnitude larger than the observed value.

According to Tanabe, Moriya and Sugano[47] the mechanism considered by Halley and Silvera seems to be defective for the reason that the quadrupole moment which is Hermitean and invariant with respect to time cannot be represented by a Hermitean operator proportional to $\underline{S}$ which changes it sign upon time reversal. This defect has been removed by Halley in the detailed derivation of the microscopic theory by not taking the average of Q in the ground state. The mechanism

discussed by Halley and Silvera is based on the use of spin-orbit interaction which couples the low lying orbital electronic levels to the ground state of the magnetic ion. This is not applicable for MnF_2 in which such states do not exist. For this case Halley[48] proposes alternative explanation in which the interaction between magnons and photons is via optical phonons.

The line shapes of the two-magnon absorption lines observed in FeF_2 and MnF_2 are very dissimilar. The total integrated intensities in the two cases differ by about a factor of 2. The polarization properties are also different. Due to these reasons, according to Halley, it is almost impossible to find a mechanism which can explain the two-magnon lines for both the materials.

Halley has proposed an alternative mechanism for the explanation of the two-magnon absorption line in MnF_2. The electric field couples to the optical phonon modes which in turn couple to the spin waves via the magnetoelastic interaction. The total Hamiltonian of the system is

$$H = H_{magnons} + H_{photons} + H_{phonons} + H_{phot.phon}$$

$$+ H^{(ex)}_{phon.spin} + H^{(dipole-di)}_{phon-spin}$$

The exchangestrictive interaction between spins and phonons is obtained by expanding the exchange integral in a Taylor series and keeping only the zero order and linear terms.

The exchange interaction should depend strongly on the phase differences between the displacements of fluorine and magnetic ions; whereas, the contribution of the dipole-dipole interaction to the spin-phonon interaction arises from the phase differences between the magnetic ions in a unit cell. These are expected to be small and important contribution to H_{int} should come from exchangestrictive part. Halley gives the effective spin Hamiltonian which leads to equivalent absorption. This is shown to include the terms of the form used by Allen et al.[38] in their phenomenological Hamiltonian.

In order to fit the line shape for MnF_2 it was necessary for Allen et al.[38] to include the coupling between distant pairs of ions falling off with the distance in an exponential manner of the form,

$$\pi(\underline{r}_i, \underline{r}_j) = \pi \exp\left(- |\underline{r}_j - \underline{r}_i| / r_o \right).$$

For a good fit they took the value of r_o to be 0.4 a_o where a_o is the Bohr radius. Similar dependence on the distance is obtained by Halley, r_o being $\sim 0.33\ a_o$.

The alternative mechanism proposed by Tanabe, Moriya and Sugano[47] can be explained as follows: Consider as a pair of ions in the ground state where the up and down spins are in the orbitals ϕ_a and ϕ_b. The transition electric dipole moment for the flip of both spins is given as:

$$\langle \phi_{a\uparrow}\phi_{b\downarrow} | \bar{P}_{eff} | \phi_{b\uparrow}\phi_{a\downarrow} \rangle = \sum_{\mu} \langle \phi_{a\uparrow}\phi_{b\downarrow} | \bar{P} | \phi_{\mu\uparrow}\phi_{b\downarrow} \rangle$$

$$\times \langle \phi_{\mu\uparrow}\phi_{b\downarrow} | V | \phi_{b\uparrow}\phi_{a\downarrow} \rangle / \Delta E(\phi_{\mu} \leftarrow \phi_{a})$$

$$+ \sum_{\nu} \langle \phi_{a\uparrow}\phi_{b\downarrow} | \bar{P} | \phi_{a\uparrow}\phi_{\nu\downarrow} \rangle \langle \phi_{a\uparrow}\phi_{\nu\downarrow} | V | \phi_{b\uparrow}\phi_{a\downarrow} \rangle / \Delta E(\phi_{\nu} \phi_{b})$$

+ Terms obtained by interchanging $\bar{P}$ and V.

Here $\bar{P}$ is the electric dipole moment operator, V is the Coulomb operator r_{12}^{-1} when ϕ_{ν} and ϕ_{μ} correspond to odd parity excited orbitals and V is the kinetic plus the crystalline field operator when $\phi_{\mu} = \phi_{b}$ and $\phi_{\nu} = \phi_{a}$. For $V = r_{12}^{-1}$ the matrix elements involved are nondiagonal exchange integrals; in other case they are the transfer integrals. In terms of the spin operators $\underline{S}_{a}$ and $\underline{S}_{b}$ the above equation can be put in its operator equivalent form as

$$\underline{P}_{eff} = \underline{\underline{x}}_{ab} \ (\underline{S}_{a} \cdot \underline{S}_{b}).$$

This vanishes if the pair of orbitals ϕ_{a} and ϕ_{b} has a centre of symmetry. For a periodic crystal, e.g., one-dimensional antiferromagnet it has the form

$$\underline{P}_{eff} = \underline{x} \sum_{j=even} (\underline{S}_{j} \cdot \underline{S}_{j+1} - \underline{S}_{j} \cdot \underline{S}_{j-1})$$

for nearest-neighbour interaction only. Since $\underline{x}$ transforms as a vector and j is at the centre of symmetry the negative sign appears.

This mechanism has been used by Moriya[49] to explain the far infrared absorption by two-magnon excitations in antiferromagnets. The absorption coefficients are calculated from the complex electric polarizability and give the two-magnon absorption frequencies for various fluorides as,

MnF_2 : $(E \parallel C) = 110\ cm^{-1}$ and $E \perp C = 101\ cm^{-1}$;

CoF_2 = 127 or 156 cm^{-1}

which may be compared with the value 120 cm^{-1} for CoF_2 reported by Richards.[37] Tanabe and Gondaira[50] have given the detailed derivation of the explicit expression for α. Assuming an effective spin-dependent moment

$$\underline{P} = \sum_{a,b} \sum_{i,i'} \underline{\alpha}(ai' \leftarrow i,b) \quad \underline{S}_a (i' \leftarrow i).\underline{S}_b) + (a \rightleftarrows b)$$

with

$$\underline{S}_a(i' \longrightarrow i') = \sum_{m'm} C^{+}_{ai'm'} C_{aim} \langle m' | \bar{S} | m \rangle$$

For the case of optical transition, Tanabe and Gondaira[51] have attempted an explanation of the optical sidebands in MnF_2.

Moriya[52] has studied the Raman scattering of light due to spin systems in magnetic materials by using the spin-dependent electric polarizability, with particular reference to iron group fluorides.

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As pointed out by Richards[38] both the mechanisms are modifications of the one proposed by Dexter [53] to explain the experimental results of Varsanyi and Dicke,[54] namely the optical absorption of a photon by a pair of atoms in co-operation. By considering the interaction between two ions to be the Coulomb interaction and retaining only the first order dipole-dipole term in the multipole expansion of the same, the transition probability for the double transition is calculated by using the first order perturbation theory.

In the present dissertation, we have proposed a new mechanism for explanation of the two-magnon absorption by antiferromagnetic systems. In a polar medium the relative displacement of oppositely charged ions $\delta\underline{u}(r)$, for a transverse optical vibrational mode, leads to a polarization $\underline{P}(r) = ne^* \delta\underline{u}(r)$ which produces an instantaneous local electric field. The corresponding magnetic field interacts with the spins giving the coupling of magnons and transverse optical phonons. The phonons in turn interact with the photons and photon-magnon interaction can be obtained by a suitable canonical transformation.

C H A P T E R - 2

TWO-MAGNON SCATTERING OF NEUTRONS

Theoretical studies of the scattering of neutrons by the collective modes of coupled spin systems (e.g. magnons) have so far been based on their interactions with spins of magnetic atoms. These calculations have mainly dealt with neutron scattering accompanying absorption or emission of a single magnon i.e. one-magnon processes.[24,34]

In the present chapter we consider neutron scattering processes involving orbital transitions in antiferromagnetic systems. This is done by taking into account the mixing of ground and excited orbital states of localized electrons of magnetic atoms due to interactions with neutrons. This mixing effect induced by neutrons is parallel to that caused by phonons in magnon-phonon interaction effects. The formulation of effective exchange Hamiltonian thus gives the relevant neutron-magnon interaction terms.

Inasmuch as in a simple two sub-lattice antiferromagnet the energies of the two magnon modes are comparable, there are many neutron - two-magnon interaction terms. Scattering cross-sections involving al these processes have been evaluated.

Formulation of the interaction terms:

We consider a two sub-lattice b.c.c. antiferromagnet where the magnetic atoms carry localised unpaired electrons in addition to the closed shell configuration. The atoms at different sublattices carry equal and opposite magnetic moments. Let us consider a beam of thermal neutrons incident on such an antiferromagnetic crystal. The Hamiltonian of the systems can be written as:[55]

$$H = \sum_{d} H_d + H_{neu} + \frac{1}{2} \sum_{l,m} H_{lm} + \sum_{d} H_{nd} \quad , \qquad (1)$$

where H_d is the Hamiltonism for a single atom, 'd' spanning both the sublattices designated by the running indices l and m ; H_{neu} is that of the neutrons, H_{lm} gives the exchange interaction between atoms l and m. H_{nd} gives the interaction between an incoming neutron and a magnetic atom as described by,

$$H_{nd} = + \quad H_{ns} + H_{no} \quad , \qquad (2)$$

where H_{ns} refers to interaction with the spin moment and H_{no} with the orbital moment of the magnetic atom. The individual eigenstates and eigenvalues of the neutrons and the atomic system are assumed to be known, namely,

$$H_{neu} \, |k\sigma\rangle = E_{k\sigma} \, |k\sigma\rangle \quad , \qquad (3)$$

$$\left.\begin{aligned} H_d \; |l_i\rangle &= E_{ai} \; |l_i\rangle \;, \\ H_d \; |m_i\rangle &= E_{bi} \; |m_i\rangle \;, \end{aligned}\right\} \qquad (4)$$

where $|k\,\sigma\rangle$ is the wave function of the neutron with wave vector k spin σ 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 state (here the ground state). The effect of H_{no} can be taken as a perturbation which causes transitions between different orbital states of an electron of the atom. Thus the perturbed orbital state $|l_{as}\rangle$ can be written as

$$|l_{as}\rangle = |l_{is}\rangle + \sum_f \frac{\langle l_f, k'\sigma' \,|\, H_{no} \,|\, l_i, \underline{k}\,\sigma\rangle \; a^+_{k'\sigma'} a_{\underline{k}\sigma} |l_{fs}\rangle}{(E_{ai} - E_{af})} \;, \qquad \ldots\ldots\ldots (5)$$

where k', σ' are the wave vector and spin of the scattered neutron and $|l_f\rangle$ the final state of the atom with energy E_{af}; s is the spin index of the electron.

In terms of the states described by (5) the effective exchange Hamiltonian between two magnetic atoms is formulated; namely:

$$\frac{1}{2}\sum_{l,m} H_{lm} = \frac{1}{2}\sum_{lm} \sum_{ss'} C^+_{as} \, C^+_{bs} \, \langle l_{as'}, m_{bs'} | H_{lm} | m_{bs'}, l_{as} \rangle C_{bs} C_{as} \;, \qquad \ldots\ldots\ldots (6)$$

where C^{+}_{as}, C_{as} represent the fermion creation and annihilation operators for an electron in the orbital state $|l_{as}\rangle$. For the orbital states which are singly occupied, the following relations hold between the above fermion operators and the conventional spin operators,[56]

$$
\begin{aligned}
& C^{+}_{a(+)} C_{a(+)} + C^{+}_{a(-)} C_{a(-)} \\
= \; & N_{a(+)} + N_{a(-)} = 1 \\
& N_{a(+)} - N_{a(-)} = 2S^{a}_{z} \qquad (7) \\
& C^{+}_{a(+)} C_{a(-)} = S^{a}_{+} = S^{a}_{x} + iS^{a}_{y} \\
& C^{+}_{a(-)} C_{a(+)} = S^{a}_{-} = S^{a}_{x} - iS^{a}_{y} \; .
\end{aligned}
$$

On making use of these relations as well as the states described by (5), the Hamiltonian in (6) can be written in terms of a pure exchange part and others involving exchange accompanied by neutron scattering

$$
H_{ex} = \frac{1}{2} \sum_{l,m} 2 J_{l,m} \underline{S}_{l} \cdot \underline{S}_{m} + \text{Constant} , \qquad (8)
$$

where

$$
J_{lm} = \langle l_{is}, m_{js}, | H_{lm} | m_{js}, l_{is'} \rangle ,
$$

and

$$H_{mn} = \sum_{l,m,f} \Big[\frac{\langle m_j, \underline{k}'\sigma' | H_{no} | m_f, \underline{k}\sigma \rangle \langle l_{is}, m_{fs} | H_{lm} | m_{js}, l_{is} \rangle}{\Delta E_b}$$

$$+ \frac{\langle l_i, \underline{k}'\sigma' | H_{no} | l_f \underline{k}\sigma \rangle \langle l_{fs} m_{js} | H_{lm} | m_{js}, l_{is} \rangle}{\Delta E_a}$$

$$+ \frac{\langle m_f, \underline{k}'\sigma' | H_{no} | m_j, \underline{k}\sigma \rangle \langle l_{is}, m_{js} | H_{lm} | m_{fs} l_{is} \rangle}{\Delta E_b}$$

$$+ \frac{\langle l_f, \underline{k}'\sigma' | H_{no} | l_f, \underline{k}\sigma \rangle \langle l_{is}, m_{js} | H_{lm} | m_{js} l_{fs} \rangle}{\Delta E_a} \Big] \times$$

$$\times \quad \eta^{+}_{\underline{k}'\sigma'} \eta_{\underline{k}\sigma} \underline{S}_l \cdot \underline{S}_m \quad . \qquad (9)$$

Here $\eta^{+}_{\underline{k}\sigma}$ $\eta_{\underline{k}\sigma}$ are the fermion creation and annihilation operators respectively for a neutron in the state $|\underline{k}\sigma\rangle$ and $\Delta E_a = E_{ai} - E_{af}$, $\Delta E_b = E_{bi} - E_{bf}$. $\underline{S}_l$ and $\underline{S}_m$ are the appropriate spin operators for the spins on the two sublattices. We use the neutron wave function

$$|\underline{k}\sigma\rangle = e^{i\underline{k}\cdot\underline{r}} |\sigma\rangle \qquad (10)$$

so that a typical matrix element, say, the one in the first term of (9) becomes,

$$\langle m_j, k'\sigma' \mid H_{no} \mid m_f, k\sigma \rangle = \langle m_j \sigma' \mid H_{no} \mid m_f \sigma \rangle \, e^{i\underline{q}\cdot\underline{R}m} \quad ,$$

where $\underline{q} = \underline{k}-\underline{k}'$. We now use the spin deviation operators of the two sublattices which are expressed below:[57]

$$\begin{aligned} S_l^+ &= S_l^x + iS_l^y = (2S)^{1/2} (1-n_l/2S)^{1/2} a_l \quad , \\ S_l^- &= S_l^x - iS_l^y = (2S)^{1/2} a_l^+ (1-n_l/2S)^{1/2} \qquad (11) \\ S - S_l^z &= a_l^+ a_l = n_l \quad (\text{the s in deviation}). \end{aligned}$$

Likewise, for the other sublattice,

$$\begin{aligned} S_m^+ &= (2S)^{1/2} d_m^+ (1 - n_m/2S)^{1/2} \\ S_m^- &= (2S)^{1/2} (1 - n_m/2S)^{1/2} d_m \qquad (12) \\ S &+ S_m^z = n_m \quad . \end{aligned}$$

These operators satisfy the commutation relations

$$aa^+ - a^+a = 1 \quad \text{and} \quad dd^+ - d^+d = 1 \, .$$

In terms of these operators, we can write

$$\begin{aligned} S_l \cdot S_m &= \tfrac{1}{2} (S_l S_m^- + S_l^- S_m^+) + S_{lz} S_{mz} \\ &= S (a_l d_m + a_l^+ d_m^+ + d_m^+ d_m + \text{e} + a_l^+ a_l) , \end{aligned}$$

where $a^{+}_{\underline{\lambda}}$, $a_{\underline{\lambda}}$ and $\beta^{+}_{\underline{\lambda}}$, $\beta_{\underline{\lambda}}$ represent the creation and annihilation operators for the two types of quasiparticles (magnons).

In order that the pure magnon terms be diagonal, we must have,

$$\tanh 2\theta_{\underline{\lambda}} = - \frac{w_e \gamma_{\underline{\lambda}}}{w_e + w_A} \qquad (15)$$

$$\approx - \gamma_{\underline{\lambda}} \qquad \text{for } w_e \gg w_A ,$$

with $w_e = 2z J^{AB} S / \hbar$ the exchange frequency, and $w_A = g\mu_B H_A / \hbar$, the frequency associated with the anisotropy energy, and

$$\gamma_{\underline{\lambda}} = \frac{1}{z} \sum_{h} e^{i\underline{\lambda}.\underline{r}^{o}_{h}} ,$$

$\underline{r}^{o}_{h}$ being the nearest-neighbour distances, z is the coordination number.

Using the relations (13) - (14) the magnon part and the interaction Hamiltonian takes the form

$$H_m + H_{mn}(s.f) = \sum [\hbar w^{\alpha}_{\underline{\lambda}} (a^{+}_{\underline{\lambda}} a_{\underline{\lambda}} + \frac{1}{2}) + \hbar w^{\beta}_{\underline{\lambda}} (\beta^{+}_{\underline{\lambda}} \beta_{\underline{\lambda}} + \frac{1}{2})]$$

$$+ \sum 8zS \langle l_i, \sigma' \mid H_{no} \mid l_f \sigma \rangle J / \Delta s_l$$

$$\times \Big\{ A_{\underline{\lambda}\underline{q}} (\eta^{+}_{\underline{k}'\sigma'} \eta_{\underline{k}\sigma} a^{+}_{\underline{\lambda}} a_{\underline{\lambda}-\underline{q}} + \eta^{+}_{\underline{k}'\sigma'} \eta_{\underline{k}\sigma} \beta^{+}_{\underline{\lambda}-\underline{q}} \beta_{\underline{\lambda}})$$

$$+ B_{\underline{\lambda}\underline{q}} (\eta^{+}_{\underline{k}'\sigma} \eta_{\underline{k}\sigma} a_{\underline{\lambda}-\underline{q}} \beta_{\underline{\lambda}} + \eta^{+}_{\underline{k}'\sigma'} \eta_{\underline{k}\sigma} a^{+}_{\underline{\lambda}-\underline{q}} \beta^{+}_{\underline{\lambda}}) \Big\} .$$

Here $\hbar w^{\alpha}_{\underline{\lambda}} = \hbar w^{\beta}_{\underline{\lambda}} = k_B \theta_C \, a\lambda$, neglecting the external magnetic field.

The coupling coefficients are given by

$$A_{\underline{\lambda q}} = (\gamma_{\underline{\lambda}} + \gamma_{\underline{\lambda}-\underline{q}}) \sinh (\theta_{\underline{\lambda}} + \theta_{\underline{\lambda}-\underline{q}}) + (1 + \gamma_{\underline{q}}) \cosh (\theta_{\underline{\lambda}} + \theta_{\underline{\lambda}-\underline{q}}) , \qquad (17)$$

$$B_{\underline{\lambda q}} = (\gamma_{\underline{\lambda}} + \gamma_{\underline{\lambda}-\underline{q}}) \cosh (\theta_{\underline{\lambda}} + \theta_{\underline{\lambda}-\underline{q}}) + (1 + \gamma_{\underline{q}}) \sinh (\theta_{\underline{\lambda}} + \theta_{\underline{\lambda}-\underline{q}}) ,$$

with $\theta_{\underline{\lambda}}$ as defined in (15).

In the neutron magnon interaction term (cf (16)) we have not included the hermitian conjugate processes since we are considering a particular incident neutron $|\underline{k}\sigma\rangle$ being scattered to the state $|\underline{k}'\sigma'\rangle$. The hermitian conjugate processes for magnon operators are already taken into account because of the summation over $\underline{\lambda}$. Equation (16) fully describes the interaction of neutrons with magnons involving orbital transitions of magnetic atoms.

Scattering cross section:

The scattering cross-section per unit energy, per unit solid angle is given in terms of neutron-magnon interaction H_{mn} by[57]

$$\frac{d^2\sigma}{dEd\Omega} = \frac{1}{h^4}\frac{k'}{k}\left(\frac{m_n}{2\pi}\right)^2 |\langle k'f \mid H_{mn} \mid ik\rangle|^2$$

$$\times\ \delta\left(E_k - E_{k'} + E_i - E_f\right) , \qquad (19)$$

where m_n is the mass of the neutron, $|i\rangle$ and $|f\rangle$ denote the initial and final states of the crystal.

For the antiferromagnetic system we consider scattering from processes involving magnons from both the magnon branches which differ only by Zeeman energy. The calculation of $d^2\sigma / dEd\Omega$ involves the matrix elements of the neutron-orbital moment interaction of the type $\langle l_i\sigma' \mid H_{no} \mid l_f\sigma\rangle$. These are evaluated as follows:

We can write

$$H_{no} = - \mu_l \cdot \underline{H}_n ,$$

where $\underline{H}_n$ is the magnetic field caused by the magnetic moment μ_n of the neutron, μ_l being the orbital moment of the atom. Then with $\underline{r} = \underline{R}_n - \underline{R}_l$

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

By standard vector algebra,

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

Now

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

so that

$$H_{no} = \mu_l \cdot \underline{\nabla} (\mu_n \cdot \underline{\nabla} \frac{1}{r}) - 4\pi \mu_l \cdot \mu_n \delta(\underline{r}) \cdot \qquad (20)$$

This expression can be written as

$$H_{no} = \left[\frac{\mu_l \cdot \mu_n}{r^3} - \frac{3(\mu_l \cdot \underline{r})(\mu_n \cdot \underline{r})}{r^5} \right] - \frac{8\pi}{3} \mu_l \cdot \mu_n \delta(\underline{r}) \cdot \qquad (21)$$

The first term denotes the dipolar interaction and is weaker than the last term and we neglect it in the following. The matrix element can then be written as

$$\langle l_i, \sigma' | H_{no} | l_f, \sigma \rangle = \langle l_i \sigma' | \int d^3 \underline{r} \cdot H_{no} | l_f \sigma \rangle$$

$$= - \frac{8\pi}{3} \langle l_i, \sigma' | \mu_l \cdot \mu_n | l_f \sigma \rangle \cdot \qquad (22)$$

The orbital and neutron magnetic moments are given by

$$\mu_l = \frac{-2|e|\hbar}{m_e c} \times \underline{Li}$$

$$\mu_n = g_n \frac{|e|\hbar}{m_n c} \underline{S_n} \qquad (23)$$

where e and m_e are electronic mass and charge respectively; $g_n = -1.91$, m_n, $\underline{S_n}$ neutron mass and spin operator; c velocity of light and $\underline{Li}$ the orbital angular momentum of atom l. Therefore

$$\mu_l \cdot \mu_n = -2 g_n \frac{e^2 \hbar^2}{m_e m_n c^2} \underline{L_i} \cdot \underline{S_n} ,$$

and

$$\langle l_i \sigma' \mid H_{no} \mid l_f \sigma \rangle = \frac{16}{3} \pi g_n \frac{e^2 \hbar^2}{m_e m_n c^2} \langle l_i, \sigma' \mid \underline{L}_i \cdot \underline{S}_n \mid l_f \sigma \rangle .$$

Here we can write

$$\underline{L} \cdot \underline{S}_n = L_z S_{nz} + \frac{1}{2} (L^+ S_n^- + L^- S_n^+) .$$

The first term $L_z S_{nz}$ does not give rise to any scattering and hence is neglected. The remaining part gives

$$\langle l_i \sigma' | H_{no} | l_f \sigma \rangle = \frac{8\pi}{3} g_n \frac{e^2 \hbar^2}{m_e m_n c^2} \langle l_i \sigma' \mid L^+ S_n^- + L^- S_n^+ \mid l_f \sigma \rangle$$

$$= \frac{8\pi}{3} g_n \frac{e^2 \hbar^2 F(q)}{m_e m_n c^2} \Big[\langle l_i(L') \mid L^+ \mid l_f(L) \rangle \langle \sigma' \mid S_n^- \mid \sigma \rangle$$

$$+ \langle l_i(L) \mid L^- \mid l_f(L) \rangle \langle \sigma' \mid S_n^+ \mid \sigma \rangle \Big] .$$

Here

$$F(\underline{q}) = \int d^3 \underline{r}_1 \, R_f^*(\underline{r}_1) R_i(\underline{r}_1) \, e^{i \underline{q} \cdot \underline{r}} ,$$

where $R_f(\underline{r}_1)$ and $R_i(\underline{r}_1)$ are the radial parts of the electronic function associated with the initial and final states. $|l_f(L)\rangle$ and $|l_i(L)\rangle$ thus denote the angular part of the corresponding state functions. Since we are considering the incident neutrons in a definite spin state, only one of the terms of the curly brackets will survive. For the incident neutron having spin $+\frac{1}{2}$, we get [39]

$$\langle l_i \sigma' | H_{no} | l_f \sigma \rangle = \frac{8\pi}{3} g_n \frac{e^2 \hbar^2 F(q)}{m_e m_n c^2} \left\{ (L+M)(L-M+1) \right\}^{1/2}$$

$$\times \left\{ \left(\frac{1}{2} + \frac{1}{2} \right) \left(\frac{1}{2} - \frac{1}{2} + 1 \right) \right\}^{1/2}$$

$$= \frac{8\pi}{3} g_n \frac{e^2 \hbar^2 F(q)}{m_e m_n c^2} \left\{ (L+M)(L-M+1) \right\}^{1/2}, \qquad (24).$$

where M is the orbital magnetic quantum number of the atom.

Using this relation, H_{mn} takes the form

$$H_{mn} = \sum_{f\,\underline{\lambda}} \frac{64\pi S}{3} \pi g_n \frac{e^2 \hbar^2 F(q)}{m_e m_n c^2} \left\{ (L+M)(L-M+1) \right\}^{1/2} \frac{J}{\Delta E_1}$$

$$\times \left\{ A_{\underline{\lambda q}} (\eta^+_{\underline{k}',\sigma'} \eta_{\underline{k}\sigma} \alpha^+_{\underline{\lambda}} \alpha_{\underline{\lambda}-\underline{q}} + \eta^+_{\underline{k}',\sigma'} \eta_{\underline{k}\sigma} \beta^+_{\underline{\lambda}-\underline{q}} \beta_{\underline{\lambda}}) \right.$$

$$\left. + B_{\underline{\lambda q}} (\eta^+_{\underline{k}',\sigma'} \eta_{\underline{k}\sigma} \alpha_{\underline{\lambda}-\underline{q}} \beta_{\underline{\lambda}} + \eta^+_{\underline{k}',\sigma'} \eta_{\underline{k}\sigma} \alpha^+_{\underline{\lambda}-\underline{q}} \beta^+_{\underline{\lambda}}) \right\}.$$

Now we evaluate the matrix element of the neutron magnon interaction H_{mn} which appears in the scattering cross-section. The combined states of the neutron and magnon can be written.

$$|i\underline{k}\rangle = e^{i.\underline{k}.\underline{r}_l} |i_{\underline{k}}, 0_{\underline{k}'}\rangle \, | \cdots \; n_{\underline{\lambda}_1}, \; n_{\underline{\lambda}_2} \; \cdots \rangle,$$

$$|f\underline{k}'\rangle = e^{i\underline{k}'.\underline{r}_m} |0_{\underline{k}}, 1_{\underline{k}'}\rangle \, | \cdots \; n'_{\underline{\lambda}_1}, \; n'_{\underline{\lambda}_2} \; \cdots \rangle.$$

then, say for the first process $\eta^+_{\underline{k}',\sigma'} \eta_{\underline{k}\sigma} \alpha^+_{\underline{\lambda}} \alpha_{\underline{\lambda}-\underline{q}}$

$$\langle \underline{k}'\, f \mid H_{mn}^{(1)} \mid i\ \underline{k} \rangle = \sum \phi A_{\underline{\lambda}\underline{q}}$$

$$\langle \ldots n_{\underline{\lambda}'_1} n_{\underline{\lambda}'_2} \ldots 1_{\underline{k}'}\ 0_{\underline{k}} \mid e^{-i\underline{k}'\cdot\underline{r}_m} n_{\underline{k}'}^{+} n_{\underline{k}} a_{\underline{\lambda}}^{+} a_{\underline{\lambda}-\underline{q}}\ e^{i\cdot\underline{k}\cdot\underline{r}_1} \mid$$

$$\ldots\ldots n_{\underline{\lambda}_1} n_{\underline{\lambda}_2} \ldots 0_{k'} , 1_{\underline{k}} \rangle$$

Here since the incident neutron state is $\mid i_{\underline{k}},\ 0'_{\underline{k}} \rangle$ and final neutron state is $\mid 0_{\underline{k}}\ i_{\underline{k}'} \rangle$ the neutron operators have unity as matrix element. The magnon operators yield

$$\langle \underline{k}' f \mid H_{mn}^{(1)} \mid i\underline{k} \rangle = \sum_{\underline{\lambda}} \phi A_{\underline{\lambda}\underline{q}} \times \left\{ (n_{\underline{\lambda}}+1)(n_{\underline{\lambda}}-\underline{q}) \right\}^{1/2} \times e^{i(\underline{k}\cdot\underline{r}_1 - \underline{k}'\cdot\underline{r}_m)} .$$

Therefore

$$\langle \underline{k}' f \mid H_{mn}^{(1)} \mid i\underline{k} \rangle \langle i\underline{k} \mid H_{mn}^{(1)} \mid f\underline{k}' \rangle = \sum_{\underline{\lambda}} \phi^2 A_{\underline{\lambda}\underline{q}}^2 (n_{\underline{\lambda}}+1)(n_{\underline{\lambda}}-\underline{q}) \quad \ldots \quad (25)$$

Here

$$\phi^2 = \left\{ \frac{64\pi z}{3} \mu_n \frac{e^2\hbar^2 F(q)}{m_e m_n c^2} \Delta \frac{J}{E_1} \left[(L+M)\ (L-M+1) \right]^{1/2} \right\}^2 \qquad (26)$$

The calculation of each process given in (16) is carried out separately. In the long wavelength approximation, retaining terms upto lowest order in the wave vectors, the coupling coefficients take the following simple forms.

$$A_{\underline{\lambda}\underline{q}}^2 = \frac{a^2}{2} \left\{ \frac{(\lambda^2 + 1\lambda - q|^2 - q^2)^2 + 4\lambda^2 |\lambda - q|^2}{2\lambda\, |\lambda - q|} \right.$$

$$\left. + 2\,(\lambda^2 + |\lambda - q|^2 - q^2) \right\} \qquad (27a)$$

The relevant expressions are

$$B^2_{\lambda q} = \frac{a^2}{z}\left\{\left(\frac{\lambda^2 + |\lambda - q|^2 - q^2)^2 + 4\lambda^2|\lambda - q|^2}{2\lambda|\lambda - q|}\right.\right.$$

$$\left.\left. - 2\ (\lambda^2 + |\lambda - q|^2 - q^2\ )\right\}\right. \qquad (27b)$$

The relevant expressions are

$$\frac{d^2\sigma}{dE d\Omega}(\alpha^+_{\lambda}, \alpha_{\lambda'}) = \frac{k'}{4\pi k}\ \frac{m_n^2}{\hbar^4} \sum_{\lambda} \phi^2\ A^2_{\lambda q}\ e^{-E^{\alpha}_{\lambda} - q\ /\ k_B T}$$

$$\times\ e^{-2W_q}\ \delta\left(E_k - E_{k'} - E^{\alpha}_{\lambda} + E^{\alpha}_{\lambda - q}\right) \qquad (28a)$$

$$\frac{d^2\sigma}{dE d\Omega}(\beta_{\lambda}\beta^+_{\lambda'}) = \frac{k'}{4\pi^2 k}\ \frac{m_n^2}{\hbar^4} \sum_{\lambda} \phi^2 A^2_{\lambda q}\ e^{-E^{\beta}_{\lambda}/k_B T}$$

$$\times\ e^{-2W_q}\ \delta\left(E_k - E_k + E^{\beta}_{\lambda} - E^{\beta}_{\lambda - q}\right) \qquad (28b)$$

$$\frac{d^2\sigma}{dE d\Omega}(\alpha_{\lambda}, \beta_{\lambda'}) = \frac{k'}{4\pi^2 k}\ \frac{m_n^2}{\hbar^4} \sum_{\lambda} \phi^2 B^2_{\lambda q}\ e^{-E^{\alpha}_{\lambda} - q\ /k_B T}$$

$$\times\ e^{-E^{\beta}_{\lambda}/k_B T}\ e^{-2W_q}\ \delta\left(E_k - E_{k'} + E^{\alpha}_{\lambda - q} + E^{\beta}_{\lambda}\right) \qquad (28c)$$

and

$$\frac{d^2\sigma}{dE d\Omega}(\alpha^+_{\lambda}, \beta^+_{\lambda}) = \frac{k'}{4\pi^2 k}\ \frac{m_n^2}{\hbar^4} \sum_{\lambda} \phi^2 B^2_{\lambda q}\ e^{-2W_q}$$

$$\times\ \left(e^{-(E^{\beta}_{\lambda} + E^{\alpha}_{\lambda - q}\ /k_B T)} + e^{-E^{\alpha}_{\lambda - q}/k_B T} + e^{-E^{\alpha}_{\lambda}\ /k_B T} + 1\right)$$

$$\times\ \delta\left(E_k - E_{k'} - E^{\alpha}_{\lambda - q} - E^{\beta}_{\lambda}\right)\ , \qquad (28d)$$

where, if we write $r_l = r^o_l + u_l$, then

$$e^{-2W_{\underline{q}}} = |\langle e^{i\underline{q}\cdot\underline{u}_l} \rangle|^2$$

We have used the equilibrium distribution function

$$n_{\underline{\lambda}} = (e^{E_{\underline{\lambda}}/k_BT} - 1)^{-1}$$

so that e.g.

$$(n^{\alpha}_{\underline{\lambda}}+1)(n^{\alpha}_{\underline{\lambda}-\underline{q}}) = \frac{e^{E^{\alpha}_{\underline{\lambda}}/k_BT}}{(e^{E^{\alpha}_{\underline{\lambda}}/k_BT}-1)(e^{+E^{\beta}_{\underline{\lambda}-\underline{q}}/k_BT}-1)}$$

$$\simeq e^{-E^{\alpha}_{\underline{\lambda}-\underline{q}}/k_BT}$$

in the low temperature region.

For actual calculation we ignore the Zeeman and anisotropy energies for the two magnon branches. Under this approximation the relevant δ-function can be eliminated by integration over the angle variables with the help of the following deduction, e.g.

$$\delta(E_{\underline{k}}-E_{\underline{k}'} - E^{\alpha}_{\underline{\lambda}} + E^{\beta}_{\underline{\lambda}-\underline{q}})$$

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

$$= \frac{(b-\lambda)}{qk_B\theta_c a}\,\delta\left(\cos\theta_{\underline{\lambda}\underline{q}} - \frac{q^2-b^2+2b}{2q}\right), \qquad (29)$$

where

$$b = \frac{\hbar^2(k^2-k'^2)}{2m_n k_B\theta_c a} \qquad (30)$$

and we have used

$$E^{\alpha,\beta}_{\underline{\lambda}} = \hbar w_{\underline{\lambda}} = k_B \theta_c a \lambda$$

Choosing the z axis in the direction of $\underline{q}$ the integration over $\underline{\lambda}$ gives

$$\frac{d^2\sigma}{dEd\Omega}(a^+_{\underline{\lambda}} a_{\underline{\lambda}'}) = \frac{k'}{16\pi^4 k} \frac{m_n^2 N a^4 \phi^2}{\hbar^4 z k_B \theta_c q} e^{\theta_c ab/T} e^{-2W_{\underline{q}}}$$

$$\times \left\{ \frac{(q^2-b^2)^2}{2a} \left(\frac{T}{\theta_c}\right) + \frac{4b(q^2-b^2)}{a^2} \left(\frac{T}{\theta_c}\right)^2 \right.$$

$$\left. + \frac{8(3b^2-q^2)}{a^2} \left(\frac{T}{\theta_c}\right)^3 - \frac{96b}{a^4} \left(\frac{T}{\theta_c}\right)^4 + \frac{192}{a^5} \left(\frac{T}{\theta_c}\right)^5 \right\}$$

..... (31a)

$$\frac{d^2\sigma}{dEd\Omega}(a^+_{\underline{\lambda}}, \beta^+_{\underline{\lambda}'}) = \frac{k}{16\pi^4 k} \frac{m_n^2 N a^4 \phi^2}{\hbar^4 z k_B \theta_c q} e^{-2W_{\underline{q}}}$$

$$\times \left(2 + e^{\theta_c ab/T} + e^{2\theta_c ab/T}\right) \times \left(\frac{(q^2-b^2)^2}{4a} \frac{T}{\theta_c}\right) \quad (31b)$$

$$\frac{d^2\sigma}{dEd\Omega}(\alpha_{\Delta}, \beta_{\Delta'}) = \frac{k'}{4\pi^4 k} \frac{m_n^2 N a^4 \phi^2}{\hbar^4 z k_B \theta_c q} e^{-\theta_c ab/T} \times e^{-2W_{\underline{q}}} \times \frac{(q^2-b^2)^2}{4a} \frac{T}{\theta_c}$$

..... (31c)

and

$$\frac{d^2\sigma}{dEd\Omega}(\beta_\Delta,\beta_\Delta^+,) = \frac{k'}{4\pi^4 k} \frac{m_n^2 Na^4 \gamma^2}{\hbar^4 z k_B \theta_c q} e^{-2W_q}$$

$$x \left\{ \frac{(q^2-b^2)^2}{2a} \frac{T}{\theta_c} + \frac{4b(b^2-q^2)}{a^2} \left(\frac{T}{\theta_c}\right)^2 + \frac{8(3b^2-q^2)}{a^3} \left(\frac{T}{\theta_c}\right)^3 \right.$$

$$\left. + \frac{96b}{a^4} \left(\frac{T}{\theta_c}\right)^4 + \frac{192}{a^5} \left(\frac{T}{\theta_c}\right)^5 \right\} . \qquad (31d)$$

Here N is the number of unit cells per unit volume, a the lattice parameter, k_B - Boltzmann constant and θ_c Neel temperature of the system.

In the low temperature region the above expression can be written as,

$$\frac{d^2\sigma}{dEd\Omega}(\text{total}) = \frac{k'}{16\pi^4 k} \frac{m_n^2 Na^3 \gamma^2}{\hbar^4 z k_B \theta_c, q} (q^2-b^2)^2 e^{-2W_q} \; x \; \frac{T}{\theta_c}$$

$$x \left\{ e^{\theta_c ab/T} + \frac{1}{4}(2+3e^{\theta_c ab/T} + e^{2\theta_c ab/T}) + e^{-\theta_c ab/T} + 2 \right\}.$$

These results have been published in J. Phys. C. (Proc. Phys. Soc.). Sr.2, Vol.1. 1968.

Discussion

In the preceeding sections, we have given a formal treatment of the two-magnon scattering of neutrons involving orbital transitions of the magnetic atoms in a simple

antiferromagnet. In the absence of experimental results where the effects discussed above will show up, it is not possible to consider any quantitative comparison between experiment and theory. It is expected that the orbital effects visualized in the calculations will be present in magnetic systems having rare earth or actinide elements. Among the possible specific cases suitable for the above studies, we may mention the antiferromagnetic systems UO_2. Recent studies of the form factor of U^{4+} ion in this system show the presence of large orbital effects. (Frazer et al. 1965).[60] A careful experimental investigation of the diffuse inelastic scattering around the Bragg peaks could perhaps reveal interesting features. Two-magnon absorption or emission cross-sections, namely

$$\frac{d^2\sigma}{d\omega d\Omega}(\alpha_{\underline{\lambda}}, \beta_{\underline{\lambda}'}) \quad \text{or} \quad \frac{d^2\sigma}{d\omega d\Omega}(\alpha^+_{\underline{\lambda}'} \beta^+_{\underline{\lambda}'})$$

may manifest as side effects.

We see from the expressions of the scattering cross-sections (cf. equations (31a) to (31d) that they depend on temperature in a complicated way. The expressions can, however, be simplified somewhat for the case of very low temperatures. The total scattering cross-section for this situation is given in (32) where the first two terms arise from processes ($\alpha^+_{\underline{\lambda}}, \alpha_{\underline{\lambda}'}$) and ($\alpha^+_{\underline{\lambda}}, \beta^+_{\underline{\lambda}'}$) and the last two terms ~~cross-sections~~ from ($\alpha_{\underline{\lambda}}, \beta_{\underline{\lambda}'}$) and ($\beta_{\underline{\lambda}}, \beta^+_{\underline{\lambda}'}$).

In the low temperature region and for $b > 0$, the former processes will dominate. Two-magnon emission processes ($\alpha^{+}_{\underline{\lambda}}$, $\beta^{+}_{\underline{\lambda}}$) will lead to considerable slowingdown of neutrons. In fact, two-magnon excitations have been observed in the infrared absorption by some antiferromagnetic substances (Halley and Silvera, 1965).[36] These we will study in the next chapter.

A careful examination of the neutron diffraction results of Erickson[21](1953) on NiF_2 at $25^{o}K$ shows a side hump before (100) reflections. This may perhaps, among other effects be attributed to two-magnon absorption processes involved in the scattering of neutrons. Although NiF_2 may not be an ideal system for test, we shall nevertheless consider an order of magnitude estimate of the scattering cross-sections for such processes. The values of the parameters involved are taken as:

$k' = 3.891 \times 10^{8}\ cm^{-1}$ $\qquad k = 5.184 \times 10^{8}\ cm^{-1}$

$q = k = k - k' = 1.293 \times 10^{8}\ cm^{-1}$

$\theta_c = 73.22^{o}K.$ $\qquad \theta_D = 450^{o}K$ $\qquad T = 25^{o}K$

$a = 4.6505\ A^{o}$, $\qquad C = 3.0837\ A^{o}$, $\qquad z = 8$

$S = 1$, $\qquad (L+M)(L-M+1) \quad 10$, assuming that Ni^{2+} ions are in 3F states.

$F(q) = 0.91$ for Ni^{2+} (taken from the calculated form factors for various transition metal ions as given by Nathan, 1964)[61]

$M_{ni} = 58.69$ atomic mass units, $\qquad m_n = 1.6747 \times 10^{-27}$ gms

$I_n = -1.91$; $J = -11.2 \times 10^{-16}$ ergs. $\Delta E_L \sim 1$ eV

Substituting these values in (26) we get

$$\phi^2 = 14.3 \times 10^{-88}$$

and

$$b = \hbar^2(k^2 - k'^2) / 2m_n k_B \theta_c a$$

$$= 0.833 \times 10^8$$

$$2W_{\underline{q}} = 3\hbar^2 q^2 / 4M_{Ni} K_B \theta_D = 2.92 \times 10^{-3}$$

Thus

$$e^{-2W_q} \sim 1$$

and

$$e^{-\theta_c e^{ab/T}} \simeq 0.122 \times 10^{-4}$$

Making use of these values, we finally get the differential scattering cross-section for two-magnon absorption (cf.equation 33a) for the system NiF_2 at $25^\circ K$ as 0.074 barns / ev per unit solid angle.

It has been emphasised earlier that it is difficult to draw a comparison with experimental situations in quantitative details. The figures are given to indicate an order of magnitude estimate of the scattering cross-section which would ensure from one of the processes discussed in the present paper. Perhaps some future experimental work in this direction might lead to a detailed assessment of the effects predicted.

CHAPTER - 3

Two-magnon absorption in antiferromagnets

We consider an insulator compound of transition element in which the electronic states of magnetic ions are described by localized 'd' orbitals. In crystals, the fivefold degeneracy of the atomic d-levels is lifted by the crystalline electric field. Assuming that the ground orbital state of an ion is non-degenerate and that the level separation between the ground state and the lowest excited state is sufficiently larger then the spin-orbit coupling and the interatomic exchange interaction energies, the spin system can be described by the usual spin Hamiltonian, which for a simple two sub-lattice antiferromagnet can be written as,

$$H_m = 2J_1 \sum_{l,m} \underline{S}_l \cdot \underline{S}_m - Hg\mu_B \left(\sum_l S_l^z + \sum_m S_m^z \right)$$

$$- H_A g\mu_B \left(\sum_l S_l^z - \sum_m S_m^z \right) \qquad (1)$$

Here J_1 is the exchange interaction energy between a corner and a neighbouring body center ion, H and H_A are the external magnetic field and the single ion anisotropy field respectively. l and m run over the N ions of the two sublattices. We use the equations (11 to 14) from chapter 2, to go over to the spin wave representation. The Hamiltonian thus assumes the following form

$$H_m = \sum_{\underline{\lambda}} [\, \hbar w^{\alpha}_{\underline{\lambda}}\, \alpha^{+}_{\underline{\lambda}}\, \alpha_{\underline{\lambda}} + \hbar w^{\beta}_{\underline{\lambda}}\, \beta^{+}_{\underline{\lambda}}\, \beta_{\underline{\lambda}} \,] \qquad (2)$$

where

$$\hbar w^{\alpha}_{\underline{\lambda}} = \hbar w^{\beta}_{\underline{\lambda}}$$

$$= \hbar \left[w_{BZ}^2 - w_e^2 \cos^2 \frac{a\lambda_x}{2} \cos^2 \frac{a\lambda_y}{2} \cos^2 \frac{a\lambda_z}{2} \right]^{1/2}$$

neglecting the external magnetic field. The angle $\theta_{\underline{\lambda}}$ is defined in the present case as

$$\tanh 2\theta_{\underline{\lambda}} = \frac{-w_e \gamma_{\underline{\lambda}}}{w_e + w_A} \quad , \qquad (3)$$

with

$$\gamma_{\underline{\lambda}} = \cos \frac{a\lambda_x}{2} \cos \frac{a\lambda_y}{2} \cos \frac{c\lambda_z}{2} \quad , \qquad (4)$$

a and c being the lattice parameters.

$$w_{BZ} = w_A + w_e \quad . \qquad (5)$$

Next we consider the magnetic field associated with the transverse optical vibrations of the lattice. Consider the simple case of two ions per unit cell of the host ionic crystal, the ions carrying the charges $+e^*$ and $-e^*$ and having masses M^+ and M^- respectively. The relative displacement $\delta u(\underline{r})$ of the ions from their equilibrium positions ($\underline{r}+$, $\underline{r}-$) can be described as

$$\delta \underline{u}(\underline{r}) = \delta \underline{u}(\underline{r}+) - \delta \underline{u}(\underline{r}-) \qquad (6)$$

In general the displacement of the k^{th} ion in the i^{th} unit cell of the crystal is given, under harmonic approximation, by,[62]

$$\delta u(\begin{smallmatrix} l \\ k \end{smallmatrix}) = (-i) \sum_{q,j} \left(\frac{\hbar}{2NM_k \Omega_{jq}} \right)^{1/2} (b^{+}_{jq} - b_{j-q}) \times e^{iq.r_l} \; e(k \mid \tfrac{q}{j}) \;, \tag{7}$$

where b^{+}_{jq}, b_{jq} are the phonon creation, annihilation operators, M_k is the mass of the k^{th} atom, N the number of unit cell comprising the lattice, Ω_{jq} the mode branch frequency; q the wave vector and j the mode branch index, $e(k \mid \frac{q}{j})$ is the normalised eigenvector of the dynamical matrix satisfying the normality and closure conditions

$$\sum_{\alpha,k} e^{+}_{\alpha} (k \mid \tfrac{q}{j}) e_{\alpha} (k \mid \tfrac{q}{j'}) = \delta_{jj'}, \tag{8}$$

and

$$\sum_{j} e^{*}_{\alpha} (k' \mid \tfrac{q}{j}) \; e_{\beta} (k \mid \tfrac{q}{j}) = \delta_{\alpha\beta} \delta_{kk'} .$$

For the case of a diatomic ionic crystal in the transverse optical mode of vibration, we have for $q \longrightarrow 0$

$$\sqrt{M_+} \; e(+ \mid \tfrac{q}{t}) = \sqrt{M_-} \; e(- \mid \tfrac{q}{t}) , \tag{9}$$

where 't' stands for transverse optical mode branch.

In the long wavelength approximation, the relative displacement of the two ions in the l^{th} unit cell can be written down as

$$\delta\underline{u}(\underline{r}_l) = (-i) \sum_{\underline{q}+} \left(\frac{\hbar}{2M\omega_{t\underline{q}}} \right)^{1/2} (b^+_{t\underline{q}} - b_{t-\underline{q}}) \, e^{i\underline{q}.\underline{r}_l}$$

$$\times \left(\frac{1}{\sqrt{M_+}} \, e\left(+ \middle| \frac{q}{t} \right) - \frac{1}{\sqrt{M}} \, e\left(- \middle| \frac{q}{t} \right) \right) \qquad \ldots\ldots\ldots\ldots\ldots\ldots (10)$$

On combining this with (9), the dipolar displacement reduces to

$$\delta\underline{u}(\underline{r}_l) = (-i) \sum_{\underline{q}t} \left(\frac{\hbar}{2M\omega_{t\underline{q}}} \right)^{1/2} (b^+_{t\underline{q}} - b_{t-\underline{q}}) \, e^{i\underline{q}.\underline{r}_l} \underline{e}_{\underline{q}t} \qquad \ldots\ldots\ldots (11)$$

where M is the reduced mass of the ion pair. $\underline{e}_{\underline{q}t}$ is the unit polarisation vector associated with the transverse optical mode t and wave vector $\underline{q}$ i.e.

$$|\underline{e}_{\underline{q}t}| = 1 \qquad \text{and} \qquad \underline{q} . \underline{e}_{\underline{q}t} = 0$$

The dielectric polarisation associated with the optical mode is given by

$$\underline{P}(\underline{r}) = ne^* \, \delta \underline{u}(\underline{r}) , \qquad (12)$$

where 'n' is the number of ion-pairs per unit volume.

The instantaneous 'local' electric field E as seen by an ion embedded in the polarised medium can be arrived at by using the cavity model of Lorentz[63] i.e. we consider

a 'physically small' spherical cavity around the site of interest; large compared to the lattice constant, but small compared to some characteristic length - the wavelength of the polarisation wave in the present case. This imposes a long wavelength approximation on the derivation. The local field is given by

$$\underline{E}(\underline{r}) = \frac{4\pi \underline{P}(\underline{r})}{3} \qquad (13)$$

(Gaussian units)

This relation remains valid, even if the polarisation varies linearly across the dimension of the cavity. This, in the present case implies that the long wavelength approximation mentioned above is not so restrictive. One can reasonably put the small wavelength limit to be of the order of 10 times the lattice - constant. Next we derive the magnetic field associated with this electric field. From Maxwell's equations we have

$$\frac{1}{C} \frac{\partial \underline{H}(\underline{r})}{\partial t} = -\underline{\nabla} \times \underline{E}(\underline{r}) \qquad (14)$$

where capital C is the velocity of light in vacuum. Substituting from (10) and (12) for $\underline{E}(\underline{r})$ and integrating the resulting equation, we get

$$\underline{H}(\underline{r}) = \sum_{t\underline{q}} \left(\frac{4\pi mec}{3\Omega_{t\underline{q}}} \right) \left(\frac{\hbar}{MN\Omega_{t\underline{q}}} \right)^{1/2} (b^{+}_{t\underline{q}} - b_{t-\underline{q}})$$

$$\times \underline{\nabla} \times \left(e^{i\underline{q}\cdot\underline{r}} \underline{e}_{t\underline{q}} \right) \qquad (15)$$

taking account of the fact that $\underline{H}(\underline{r})$ involves the phonon operators $b^{+}_{t\underline{q}}$, $b_{t\underline{q}}$ having implicit time dependence of the type of $e^{i\Omega_{t\underline{q}}}$. The magnetic field thus obtained is a 'near zone' field acting locally at the site in question. It follows the variation of polarisation adiabatically. It is thus distinct from the radiation field which propagates as an independent mode - the photons of the electromagnetic field. The latter couples strongly with the transverse optical modes near the cross over point of the two dispersion characteristics, modifying the dispersion relations condiderably at and below that point. The diagonalisation of this interaction has been considered by Hopfield.[64] In the present case, the photons considered have $w_{k\lambda} > \Omega_{k\mu}$ for optical phonons of interest.

Making use of the vector identity

$$\underline{\nabla} \times (\phi \underline{A}) = \phi \,\mathrm{curl}\, \underline{A} - \underline{A} \times \mathrm{grad}\, \phi \qquad (16)$$

where ϕ is a scalar field ($= e^{i\cdot\underline{q}\cdot\underline{r}}$ in the present case) and $\underline{A}$ as the vector field ($= \underline{e}_{\underline{q}t}$ in the above relation) we have

$$\underline{H}(\underline{r}) = \sum_{\underline{q}t} \left(\frac{4\pi n e^{*} c}{3\Omega_{t\underline{q}}} \right) \left(\frac{\hbar}{2MN\Omega_{t\underline{q}}} \right)^{1/2} (b^{+}_{t\underline{q}} - b_{t-\underline{q}})$$

$$\times i e^{i\cdot\underline{q}\cdot\underline{r}} \, (\underline{q} \times \underline{e}_{t\underline{q}}) \qquad (17)$$

It is clear from the form of (16) that there will be a non-vanishing magnetic field at a point $\underline{r}$ only for the transverse optical mode.

Neglecting the wavevector ($\underline{q}$) dependence of the transverse optical mode frequency Ω_{tq}, we have

$$\underline{H}(\underline{r}) = \sum_{t\underline{q}} \underline{I}_{t\underline{q}} \left(b^{+}_{t\underline{q}} - b_{t-\underline{q}} \right) e^{i.\underline{q}.\underline{r}} \tag{18}$$

with

$$\underline{I}_{t\underline{q}} = \left(\frac{4\pi ne^{*}C}{3\Omega_{ot}} \right) \left(\frac{\hbar}{2MN\Omega_{ot}} \right)^{1/2} \left(\underline{q} \times \underline{e}_{t\underline{q}} \right) . \tag{19}$$

The interaction of this field with the spin magnetic moment can be written as

$$H_{sp} = - g \mu_B \sum_{l} \underline{H}(\underline{r}_l) . \underline{S}_l \tag{20}$$

The transition metal fluorides FeF_2, MnF_2 etc. appear in rutile structure. There are six atoms per unit cell, there being consequently 12 phonon modes, nine being optical modes. The magnetic field associated with any one infrared active transverse optical mode will contain more effective mass M* and effective charge e*. We consider those infrared active optical phonon modes which have point group symmetry A_{2u} (one mode) and E_u (three doubly degenerate modes). Thus we have

$$\begin{aligned} H_{sp} = &- i \sum_{\underline{q},lt} \left(b^{+}_{\underline{q}t} - b_{-\underline{q}t} \right) \left(\underline{k}_{t\underline{q}} . \underline{S}_l \right) e^{i.\underline{q}.\underline{r}_l} \\ &- i \sum_{\underline{q},mt} \left(b^{+}_{\underline{q}t} - b_{-\underline{q}t} \right) \left(\underline{k}_{t\underline{q}} . \underline{S}_m \right) e^{i.\underline{q}.\underline{r}_m} \end{aligned} \tag{21}$$

where t stands for A_{2u} and $\sum_{i=1}^{6} E_u^i$,

$$\underline{k}_{t\underline{q}} = g \mu_B \underline{I}_{t\underline{q}} \tag{22}$$

g being the spectroscopic splitting factor.

Using the relations (11 - 14) from chapter 2, to go over to the spin wave representation, we can write the magnon-phonon interaction explicitly as,

$$H_{mp} = \sum_{\underset{\sim}{q}} (b^{+}_{\underset{\sim}{q}A_{2u}} - b_{-\underset{\sim}{q}A_{2u}}) [A^{-}_{\underset{\sim}{q}A_{2u}} (\alpha_{\underset{\sim}{q}} + \beta^{+}_{\underset{\sim}{q}}) + A^{+}_{\underset{\sim}{q}A_{2u}} (\alpha^{+}_{-\underset{\sim}{q}} + \beta_{-\underset{\sim}{q}})] + \sum_{\underset{\sim}{q}} \sum_{i=1}^{6} (b^{+}_{\underset{\sim}{q}E^{i}_{u}} - b_{-\underset{\sim}{q}E^{i}_{u}}) \times B^{z}_{\underset{\sim}{\lambda q}E^{i}_{u}} (\alpha^{+}_{\underset{\sim}{\lambda}}\alpha_{\underset{\sim}{q}+\underset{\sim}{\lambda}} - \beta_{\underset{\sim}{\lambda}}\beta^{+}_{\underset{\sim}{\lambda}+\underset{\sim}{q}}) + C^{z}_{\underset{\sim}{\lambda q}E^{i}_{u}} (\alpha^{+}_{\underset{\sim}{\lambda}}\beta^{+}_{\underset{\sim}{\lambda}+\underset{\sim}{q}} - \alpha_{\underset{\sim}{q}+\underset{\sim}{\lambda}}\beta_{\underset{\sim}{\lambda}}) . \quad \ldots\ldots\ldots\ldots (23)$$

The coupling coefficients are given by

$$A^{\pm}_{\underset{\sim}{q}t} = \frac{-i}{2} (2SN)^{1/2} (\sin h\, \theta_{\underset{\sim}{q}} + \cos h\, \theta_{\underset{\sim}{q}}) k^{\pm}_{t\underset{\sim}{q}}$$

$$B^{z}_{\underset{\sim}{\lambda q}t} = i k^{z}_{t\underset{\sim}{q}} \cos h (\theta_{\underset{\sim}{\lambda}} + \theta_{\underset{\sim}{\lambda}+\underset{\sim}{q}}) \qquad (24)$$

$$C^{z}_{\underset{\sim}{\lambda q}t} = i k^{z}_{t\underset{\sim}{q}} \sin h (\theta_{\underset{\sim}{\lambda}+\underset{\sim}{q}} - \theta_{\underset{\sim}{\lambda}})$$

Now we consider the interaction between the lattice vibrations and electromagnetic field. Referring to a perfect crystal as a system composed of periodically ordered lattice centres and electromagnetic radiation, the Hamiltonian describing the motion of lattice centres in the electromagnetic field is given by

$$H = \sum_{l,k} \frac{1}{2m_k^{(i)}} \left[\underline{p}_{lk}^{(i)} - \frac{e_k^{(i)}}{c} \underline{A}(\underline{x}_{lk}) \right]^2 + \sum_{kk'} e_k^{(i)} \phi(\underline{x}_{lk}) \qquad (25)$$

The electromagnetic field is defined by the four vector

$$A_\mu(\underline{x},t) = [\underline{A}(\underline{x},t),\ i\phi(x,t)] \qquad \mu = 1,2,3,4 \ .$$

where $\underline{A}$ is the vector potential and ϕ the scaler potential which we take to be zero by a suitable gauge transformation. Here $p_{lk}^{(i)}$ is the momentum of the i^{th} particle, of the (lk) centre. $m_k^{(i)}$ and $e_k^{(i)}$ and mass and charge of the i^{th} particle. The phonon-photon interaction Hamiltonian obtained from (25) is given by [66]

$$H_{ph-photon} = -\sum_{l} e\hbar \frac{\pi}{NV} \sum_{k\ \underline{q}t} \frac{z}{\sqrt{M}} \left(\underline{p}_{lk}^{\underline{q}t} \cdot \underline{A}_{lk} \right)$$

$$- \frac{e\hbar}{2N} \sqrt{\frac{\pi k}{2V}} \sum_{l} \sum_{k'} \sum_{\substack{\underline{q}t \\ \underline{q}'t'}} \frac{z}{M_k}$$

$$\left[(\underline{p}_{lk}^{\underline{q}t} \cdot \underline{A}_{lk})(\underline{k} \cdot \underline{n}_{lk}^{\underline{q}'t'}) + (\underline{k} \cdot \underline{n}_{lk}^{\underline{q}'t'})(\underline{p}_{lk}^{\underline{q}t} \cdot \underline{A}_{lk}) \right] .$$

..... (26)

Introducing the second quantisation representation by

$$\left.\begin{aligned} \underline{n}_{lk}^{\underline{q}t} &= \frac{i}{\sqrt{\Omega_{\underline{q}t}}} \left[b_{\underline{q}t}^{+} - b_{-\underline{q}t} \right] e^{i\underline{q}\cdot\underline{r}} \\ \underline{p}_{lk}^{\underline{q}t} &= \sqrt{\Omega_{\underline{q}t}} \left[b_{\underline{q}t}^{+} + b_{-\underline{q}t} \right] e^{-i\underline{q}\cdot\underline{r}} \end{aligned}\right\} \qquad (27)$$

and

$$A_{lk} = \frac{1}{\sqrt{w_{\underline{k}}}} \left[a_{\underline{k}r}^{+} + a_{-\underline{k}r} \right] e^{-i\underline{k}\cdot\underline{l}}$$

the interaction Hamiltonian takes the form,

$$H_{ph-phonon} =$$

$$= \sum_{\underline{k}} D_{\underline{k}A_{2u}} [b^{+}_{-\underline{k}A_{2u}} a^{+}_{\underline{k}z} + b^{+}_{\underline{k}A_{2u}} a_{-\underline{k}z} + b_{\underline{k}A_{2u}} a^{+}_{\underline{k}z} + b_{\underline{k}A_{2u}} a_{-\underline{k}z}]$$

$$+ \sum_{\underline{k}} \sum_{i=1}^{6} D_{\underline{k}E^{i}_{u}} [b^{+}_{-\underline{k}E^{i}_{u}} a^{+}_{\underline{k}y} + b^{+}_{\underline{k}E^{i}_{u}} a_{-\underline{k}y} + b_{\underline{k}E^{i}_{u}} a^{+}_{-\underline{k}y} + b_{\underline{k}E^{i}_{u}} a_{-\underline{k}y}]$$

$$+ \sum_{\underline{q}\underline{q}'} F_{\underline{q}\underline{q}'A_{2u}} \{ - [b^{+}_{\underline{q}A_{2u}} b_{-\underline{q}'A_{2u}} a_{-\underline{q}-\underline{q}'z} + b^{+}_{\underline{q}A_{2u}} b^{+}_{-\underline{q}'A_{2u}} a_{\underline{q}+\underline{q}'z}$$

$$+ b_{-\underline{q}A_{2u}} b_{-\underline{q}'A_{2u}} a^{+}_{-\underline{q}-\underline{q}'z} + b_{-\underline{q}A_{2u}} b_{-\underline{q}A_{2u}} a_{\underline{q}+\underline{q}'z}]$$

$$+ [b^{+}_{\underline{q}A_{2u}} b^{+}_{\underline{q}'A_{2u}} a^{+}_{-\underline{q}-\underline{q}'z} + b^{+}_{\underline{q}A_{2u}} b^{+}_{\underline{q}'A_{2u}} a_{\underline{q}+\underline{q}'z}$$

$$+ b_{-\underline{q}A_{2u}} b^{+}_{\underline{q}'A_{2u}} a_{-\underline{q}-\underline{q}'z} + b_{-\underline{q}A_{2u}} b_{\underline{q}'A^{+}_{2u}} a_{\underline{q}+\underline{q}'z}] \} + \cdots$$

+ similar terms containing both phonons from $\sum_{i=1}^{6} E^{i}_{u}$ modes and photons polarised along x or y direction. ..... (28)

Here

$$D_{\underline{k}t} = e\hbar \sqrt{\frac{\pi N \Omega_{\underline{q}t}}{w_k}} \frac{z}{m} (\underline{e}_{-\underline{k}t} \cdot \underline{\epsilon}_{\underline{k}r}) \qquad (29)$$

and

$$F_{\underline{q}\underline{k}} = - e\hbar \sqrt{\frac{\pi\hbar}{2vw_k}} \frac{z}{M^{*}} (\underline{e}_{\underline{q}t} \cdot \underline{\epsilon}_{\underline{q}r}) (\underline{k} \cdot \underline{e}_{-\underline{q}-\underline{k}t}) \qquad (39)$$

$\underline{\epsilon}_{\underline{k}r}$ being the unit polarisation vector for the photons

of wave vector $\underline{k}$. t refers to A_{2u} or $\sum_{i=1}^{6} E_u^i$.

Thus the total Hamiltonian is,

$$H = H_m + H_{ph} + H_{phot} + H_{m-ph} + H_{ph-phot}$$

$$= \sum_{\underline{\lambda}} [\hbar w^{\alpha}_{\underline{\lambda}} (\alpha^+_{\underline{\lambda}} \alpha_{\underline{\lambda}} + \tfrac{1}{2}) + \hbar w^{\beta}_{\underline{\lambda}} (\beta^+_{\underline{\lambda}} \beta_{\underline{\lambda}} + \tfrac{1}{2})]$$

$$+ \sum_{\underline{q}t} \hbar \Omega_{ot} (b^+_{\underline{q}t} b_{\underline{q}t} + \tfrac{1}{2}) + \sum_{\underline{k}} \hbar c |k| (a^+_{\underline{k}r} a_{\underline{k}r} + \tfrac{1}{2})$$

$$+ \sum_{\underline{q}} (b^+_{\underline{q}A_{2u}} - b_{-\underline{q}A_{2u}}) [A^-_{\underline{q}} (\alpha_{\underline{q}} + \beta^+_{\underline{q}}) + A^+_{\underline{q}} (\alpha^+_{-\underline{q}} + \beta_{-\underline{q}})]$$

$$+ \sum_{\underline{q}} \sum_{i=1}^{6} (b^+_{\underline{q}E_u^i} - b_{-\underline{q}E_u^i}) [B^z_{\underline{\lambda}\underline{q}E_u^i} (\alpha^+_{\underline{\lambda}} \alpha_{\underline{\lambda}+\underline{q}} - \beta_{\underline{\lambda}} \beta^+_{\underline{\lambda}+\underline{q}})$$

$$+ C^z_{\underline{\lambda}\underline{q}E_u^i} (\alpha^+_{\underline{\lambda}} \beta^+_{\underline{\lambda}+\underline{q}} - \alpha_{\underline{q}+\underline{\lambda}} \beta_{\underline{\lambda}})]$$

$$+ \sum_{\underline{k}} D_{\underline{k}A_{2u}} (b_{-\underline{k}A_{2u}} a^+_{\underline{k}z} + b^+_{\underline{k}A_{2u}} a_{-\underline{k}z} + b_{\underline{k}A_{2u}} a^+_{\underline{k}z} + b_{\underline{k}A_{2u}} a_{-\underline{k}z})$$

+ Similar terms with phonons from $\sum_{i=1}^{6} E_u^i$ and photons polarised along x or y

$$+ \sum_{\underline{q}\underline{q}'i=1}^{6} F_{\underline{q}\underline{q}'E_u^i} \{ - [b^+_{\underline{q}E_u^i} b^+_{-\underline{q}'E_u^i} a^+_{-\underline{q}-\underline{q}'x} + b^+_{\underline{q}E_u^i} b^+_{-\underline{q}'E_u^i} a_{\underline{q}+\underline{q}'y}$$

$$+ b_{-\underline{q}E_u^i} b_{-\underline{q}E_u^i} a^+_{-\underline{q}-\underline{q}'x} + b_{-\underline{q}'E_u^i} b_{-\underline{q}E_u^i} a_{\underline{q}+\underline{q}'y}] +$$

$$+ \Big[b^{+}_{\underline{q}E^{i}_{u}} \, b^{+}_{\underline{q}E^{i}_{u}} \, a^{+}_{-\underline{q}-\underline{q}',x} + b^{+}_{\underline{q}E^{i}_{u}} \, b_{\underline{q}'E^{i}_{u}} \, a_{\underline{q}+\underline{q}',x}$$

$$+ b_{-\underline{q}E^{i}_{u}} \, b^{+}_{\underline{q}'E^{i}_{u}} \, a^{+}_{-\underline{q}-\underline{q}'E^{i}_{u}} + b_{-\underline{q}E^{i}_{u}} \, b^{+}_{\underline{q}'E^{i}_{u}} \, a_{\underline{q}+\underline{q}',x} \Big] \Big\}$$

+ Similar terms containing both the phonons of A_{2u} mode and photons polarised along z.

..... (31)

In order to get the magnon-photon interaction by summing over the optical phonons, we apply a canonical transformation defined by

$$H_T = e^{-iS} H e^{iS} .$$

Assuming S to be small and expanding the exponential we get

$$H_T = \left(1 - iS - \frac{S^2}{2} + \ldots \right) H \left(1 + iS - \frac{S^2}{2} + \ldots \right)$$

$$= H_o + H_{int} + \frac{i}{2}[H, S] - \frac{1}{2}[(H_{int}, S), S] ,$$

where

$$H = H_o + H_{int}$$

The form of S assumed in the present case is:

$$S = \sum_{\underline{q}\ t=A_{2u}} \Big\{ \gamma_{1\underline{q}t} b^{+}_{-\underline{q}t} \alpha_{\underline{q}} + \gamma_{2\underline{q}t} b^{+}_{\underline{q}t} \beta^{+}_{\underline{q}} + \gamma_{3\underline{q}t} b^{+}_{\underline{q}t} \alpha^{+}_{-\underline{q}}$$

$$+ \gamma_{4\underline{q}t} b^{+}_{\underline{q}t} \beta_{-\underline{q}} - \gamma_{5\underline{q}t} b_{-\underline{q}t} \alpha_{\underline{q}} - \gamma_{6\underline{q}t} b_{-\underline{q}t} \beta^{+}_{\underline{q}} - \gamma_{7\underline{q}t} b_{-\underline{q}t} \alpha^{+}_{-\underline{q}}$$

$$- \gamma_{8\underline{q}t} b_{-\underline{q}} + \beta_{-\underline{q}} \Big\} +$$

$$+ \sum_{\underline{q}\underline{\lambda}} \sum_{t=\sum_u^i=1}^{6} \Big\{ \Theta_{1\underline{\lambda}\underline{q}t} b^{+}_{\underline{q}t} \alpha^{+}_{\underline{\lambda}} \alpha_{\underline{q}+\underline{\lambda}} - \Theta_{2\underline{\lambda}\underline{q}t} b^{+}_{\underline{q}t} \beta_{\underline{\lambda}} \beta^{+}_{\underline{q}+\underline{\lambda}}$$

$$+ \Theta_{3\underline{\lambda}\underline{q}t} b^{+}_{\underline{q}t} \alpha^{+}_{\underline{\lambda}} \alpha^{+}_{\underline{q}+\underline{\lambda}} - \Theta_{4\underline{\lambda}\underline{q}t} b^{+}_{\underline{q}t} \alpha_{\underline{q}+\underline{\lambda}} \beta_{\underline{\lambda}} - \Theta_{5\underline{\lambda}\underline{q}t} b_{-\underline{q}t} \alpha^{+}_{\underline{\lambda}} \alpha_{\underline{q}+\underline{\lambda}} +$$

$$+ \Theta_{6\underline{\lambda}\underline{q}t} b_{-\underline{q}t} \beta_{\underline{\lambda}} \beta^{+}_{\underline{q}+\underline{\lambda}} - \Theta_{7\underline{\lambda}\underline{q}t} b_{-\underline{q}t} \alpha^{+}_{\underline{\lambda}} \beta^{+}_{\underline{q}+\underline{\lambda}} + \Theta_{8\underline{\lambda}\underline{q}t} b_{-\underline{q}t} \alpha_{\underline{q}+\underline{\lambda}} \beta_{\underline{\lambda}} \Big\}$$

$$+ \sum_{\underline{q}tr} \Big\{ \delta_{1\underline{q}tr} b^{+}_{\underline{q}t} a^{+}_{-\underline{q}r} + \delta_{2\underline{q}tr} b^{+}_{\underline{q}t} a_{\underline{q}r} + \delta_{3\underline{q}tr} b_{-\underline{q}t} a^{+}_{\underline{q}r} +$$

$$+ \delta_{4\underline{q}tr} b_{-\underline{q}t} a_{\underline{q}r} \quad (t=A_{2u},\ r=Z;\ t = \sum_{i=1}^{6} s_u^i ,\quad r = x \text{ or } y) \Big\}$$

$$+ \sum_{\underline{q}\underline{q}'tr} \Big\{ -[\, \psi_{1\underline{q}\underline{q}'rt} b^{+}_{\underline{q}t} b^{+}_{-\underline{q}'t} a^{+}_{-\underline{q}-\underline{q}'r} + \psi_{2\underline{q}\underline{q}'tr} b^{+}_{\underline{q}t} b^{+}_{-\underline{q}'t} a_{\underline{q}+\underline{q}'r}$$

$$+ \psi_{3\underline{q}\underline{q}'rt} b_{-\underline{q}t} b_{-\underline{q}'t} a^{+}_{-\underline{q}-\underline{q}'r} + \psi_{4\underline{q}\underline{q}'tr} b_{-\underline{q}t} b_{-\underline{q}'t} a_{\underline{q}+\underline{q}'r} \,]$$

$$+ [\, \psi_{5\underline{q}\underline{q}'t} b^{+}_{\underline{q}t} a^{+}_{-\underline{q}-\underline{q}'r} + 4_{\underline{q}\underline{q}'rt} b^{+}_{\underline{q}t} b^{+}_{\underline{q}'t} a_{\underline{q}+\underline{q}'r} +$$

$$+ \Psi_{7\underline{q}\underline{q}'rt}{}^{b}{}_{-\underline{q}t}{}^{a}{}_{b}{}^{+}_{\underline{q}'t}{}^{a}{}^{+}_{-\underline{q}-\underline{q}'r} + \Psi_{8\underline{q}\underline{q}'rt}{}^{b}{}_{-\underline{q}t}{}^{b}{}^{+}_{\underline{q}'t}\, a_{\underline{q}+\underline{q}'r} \Big]$$

$$\left(t = A_{2u},\ r = z;\quad t = \sum_{i=1}^{6} E_u^i,\quad r = x \text{ or } y \right) \Big\}$$

..... (32)

The coefficients of S are determined from the condition

$$H_{int} + \frac{i}{2}\left[H_o, S \right] = 0 \qquad (33)$$

They are given by

$$\gamma_{1\underline{q}A_{2u}} = \frac{iA^{+}_{\underline{q}A_{2u}}}{2\left(- E^{+}_{\underline{q}} + \hbar\Omega_{oA_{2u}} \right)} \qquad (34a)$$

$$\gamma_{2\underline{q}A_{2u}} = \frac{iA^{-}_{\underline{q}A_{2u}}}{2\left(E^{\beta}_{\underline{q}} + \hbar\Omega_{oA_{2u}} \right)} \qquad (34b)$$

$$\gamma_{3\underline{q}A_{2u}} = \frac{iA^{+}_{\underline{q}A_{2u}}}{2\left(E^{a}_{\underline{q}} + \hbar\Omega_{oA_{2u}} \right)} \qquad (34c)$$

$$\gamma_{4\underline{q}A_{2u}} = \frac{iA^{-}_{\underline{q}A_{2u}}}{2\left(- E^{\beta}_{\underline{q}} + \hbar\Omega_{oA_{2u}} \right)} \qquad (34d)$$

$$\gamma_{5\underline{q}A_{2u}} = \frac{-iA^{-}_{\underline{q}A_{2u}}}{2\left(E^{a}_{\underline{q}} + \hbar\Omega_{oA_{2u}} \right)} \qquad (34e)$$

$$Y_{6\underline{q}A_{2u}} = \frac{-iA^{-}_{\underline{q}A_{2u}}}{2\left(-E^{\beta}_{\underline{q}} + \hbar\Omega_{oA_{2u}}\right)} \tag{34f}$$

$$Y_{7\underline{q}A_{2u}} = \frac{-iA^{+}_{\underline{q}A_{2u}}}{2\left(-E^{\alpha}_{\underline{q}} + \hbar\Omega_{oA_{2u}}\right)} \tag{34g}$$

$$Y_{8\underline{q}A_{2u}} = \frac{-iA^{-}_{\underline{q}A_{2u}}}{2\left(E^{\beta}_{\underline{q}} + \hbar\Omega_{oA_{2u}}\right)} \tag{34h}$$

$$\Theta_{1\underline{\lambda}\underline{q}E^{i}_{u}} = \frac{-iB^{z}_{\underline{\lambda}\underline{q}E^{i}_{u}}}{2\left(\hbar\Omega_{oE^{i}_{u}} + E^{\beta}_{\underline{\lambda}} - E^{\beta}_{\underline{q}+\underline{\lambda}}\right)} \tag{35a}$$

$$\Theta_{2\underline{\lambda}\underline{q}E^{i}_{u}} = \frac{-iB^{z}_{\underline{\lambda}\underline{q}E^{i}_{u}}}{2\left(\hbar\Omega_{oE^{i}_{u}} + E^{\alpha}_{\underline{\lambda}+\underline{q}} + E^{\beta}_{\underline{\lambda}}\right)} \tag{35b}$$

$$\Theta_{3\underline{\lambda}\underline{q}E^{i}_{u}} = \frac{iC^{z}_{\underline{\lambda}\underline{q}E^{i}_{u}}}{2\left(\hbar\Omega_{oE^{i}_{u}} + E^{\alpha}_{\underline{\lambda}} + E^{\beta}_{\underline{q}+\underline{\lambda}}\right)} \tag{35c}$$

$$\Theta_{4\underline{\lambda}\underline{q}E^{i}_{u}} = \frac{iC^{z}_{\underline{q}E^{i}_{u}}}{2\left(-2\hbar\Omega_{oE^{i}_{u}} + E^{\alpha}_{\underline{\lambda}+\underline{q}} + E^{\beta}_{\underline{\lambda}}\right)} \tag{35d}$$

$$\theta_{5\underline{\lambda}\underline{q}E_u^i} = \frac{iB^z_{\underline{\lambda}\underline{q}E_u^i}}{2(\hbar\Omega_{oE_u^i} + E^{\alpha}_{\underline{\lambda}+\underline{q}} - E^{\alpha}_{\underline{\lambda}})} \tag{35e}$$

$$\theta_{6\underline{\lambda}\underline{q}E_u^i} = \frac{iB^z_{\underline{\lambda}\underline{q}E_u^i}}{2(-2\hbar\Omega_{oE_u^i} + E^{\beta}_{\underline{\lambda}+\underline{q}} - E^{\beta}_{\underline{\lambda}})} \tag{35f}$$

$$\theta_{7\underline{\lambda}\underline{q}E_u^i} = \frac{iC^z_{\underline{\lambda}\underline{q}E_u^i}}{2(\hbar\Omega_{oE_u^i} - E^{\alpha}_{\underline{\lambda}} - E^{\beta}_{\underline{\lambda}+\underline{q}})} \tag{35g}$$

$$\theta_{8\underline{\lambda}\underline{q}E_u^i} = \frac{iC^z_{\underline{\lambda}\underline{q}E_u^i}}{2(-\hbar\Omega_{oE_u^i} - E^{\beta}_{\underline{q}+\underline{\lambda}} - E^{\beta}_{\underline{\lambda}})} \tag{35h}$$

$$\phi_{1\underline{q}tr} = \frac{iD_{\underline{q}tr}}{2(\hbar\Omega_{ot} + \hbar w_c)} = -\phi_{4\underline{q}tr} \tag{36a}$$

$$\phi_{2\underline{q}tr} = \frac{iD_{\underline{q}tr}}{2(\hbar\Omega_{ot} - \hbar w_c)} = -\phi_{3\underline{q}tr} \tag{36b}$$

$$\psi_{1\underline{q}\underline{q}'rt} = \frac{iF_{\underline{q}\underline{q}'rt}}{2\hbar w_c} = \psi_{2\underline{q}\underline{q}'rt} \tag{37a}$$

$$\psi_{3\underline{q}\underline{q}'rt} = \frac{-iF_{\underline{q}\underline{q}'rt}}{2(2\hbar\Omega_{ot} - \hbar w_c)} = -\psi_{6\underline{q}\underline{q}'rt} \tag{37b}$$

$$\Psi_{4\underline{q}\underline{q}'rt} = \frac{-iF_{\underline{q}\underline{q}'rt}}{2(\hbar w_c + 2\hbar\Omega_{ot})} = -\Psi_{5\underline{q}\underline{q}'rt} \qquad (37c)$$

$$\Psi_{7\underline{q}\underline{q}'rt} = \frac{iF_{\underline{q}\underline{q}'rt}}{2\hbar w_c} = -\Psi_{8\underline{q}\underline{q}'rt} \qquad (37d)$$

Calculation of absorption coefficient

Consider $\underline{E} \perp \underline{C}$, say along x direction. The photons are hence polarised along x and the phonons of $\sum_{i=1}^{6} E_u^i$ modes are involved. The two-magnon creation terms come from $\frac{i}{2}[H_{int}, S]$. We consider only those processes where magnons from 2 different branches are involved. The external magnetic field has no effect on the line since the extra energies of the magnons from the two branches because of the external magnetic field are $\pm g\mu_B H$ the sum of the being zero. The corresponding interaction terms are,

$$H_{m-1} = \frac{i}{2}[H_{int}, S]$$

$$= \frac{i}{2} \sum_{\underline{\lambda}\underline{q}i=1}^{6} [-D_{\underline{q}E_u^i x} (\theta_{8\underline{\lambda}\underline{q}E_u^i} + \theta_{4\underline{\lambda}\underline{q}E_u^i})$$

$$+ C^z_{\underline{\lambda},-\underline{q}E_u^i} (\delta_{3\underline{q}\,E_u^i x} + \delta_{1\underline{q}E_u^i x})] a^+_{\underline{q}x} \alpha_{\underline{q}+\underline{\lambda}} \beta_{\underline{\lambda}}$$

$$+ [D_{-\underline{q}E_u^i x} (\theta_{7\,\underline{q}E_u^i} + \theta_{3\,\underline{q}E_u^i}) - C^z_{\underline{\lambda},-\underline{q}E_u^i}$$

$$(\delta_{4\underline{q}E_u^i x} + \delta_{2\underline{q}E_u^i x})] a_{-\underline{q}x}\ \alpha^+_{\underline{q}+\underline{\lambda}}\ \beta^+_{\underline{\lambda}} \qquad (38)$$

For the two-magnon creation process using equations (34-37), the coefficient of $a_{-\underline{q}} x\, a^{\alpha +}_{\underline{q}+\underline{\lambda}}\, \beta^{+}_{\underline{\lambda}}$ is given by,

$$\left[\frac{E_{\underline{\lambda}} + E_{\underline{\lambda}+\underline{q}}}{2\left[(\hbar \Omega_{oE_u^i})^2 - (E^{\alpha}_{\underline{\lambda}} + E^{\alpha}_{\underline{\lambda}+\underline{q}})^2 \right]} - \frac{E_c}{\left[(\hbar \Omega_{oE_u^i})^2 - E_c^2 \right]} \right]^2 \cdot \qquad (39)$$

The transition probability from the initial state $|i\rangle$ to the final state $|f\rangle$ is given by

$$W(i \rightarrow f) = \frac{2\pi}{\hbar} \left| \langle H_{int} \rangle \right|^2 \delta (\hbar w_i - \hbar w_f) \cdot$$

In the present case W assumes the form,

W(1 photon, no magnon $\rightarrow$ zero photon, two magnons)

$$= \frac{2\pi}{\hbar} \left(\frac{e^2 \hbar^3 \pi}{2VM^{*2} w_e} \right) \left(\frac{4\pi n e^{*} C g \mu_B}{3\Omega_{oE_u^i}} \right)^2 \frac{z}{M^{*}}$$

$$x \sum_{q} \frac{q^2}{3} \left[\frac{E^{\alpha}_{\underline{\lambda}} + E^{\alpha}_{\underline{\lambda}+\underline{q}}}{2\left[(\hbar \Omega_{oE_u^i})^2 - (E^{\alpha}_{\underline{\lambda}} + E^{\alpha}_{\underline{\lambda}+\underline{q}})^2 \right]} - \frac{E_c}{\left[(\hbar \Omega_{oE_u^i})^2 - E_c^2 \right]} \right]^2 n_{E_c}$$

$$x \left(1 + e^{-E^{\alpha}_{\underline{\lambda}}/k_B T} \right) \left(1 + e^{-E^{\beta}_{\underline{\lambda}+\underline{q}}/k_B T} \right)$$

$$x \sin h^2 \left(\theta_{\underline{\lambda}+\underline{q}} - \theta_{\underline{\lambda}} \right) \delta \left(E_c - E^{\alpha}_{\underline{\lambda}} - E^{\beta}_{\underline{\lambda}+\underline{q}} \right) . \qquad (40)$$

We consider the approximate dispersion relation,

$$E^{\alpha,\beta}_{\underline{\lambda}} = \hbar w^{\alpha\beta}_{\underline{\lambda}} = \hbar \left[w_{Bz}^2 - w_e^2 \cos^2 \frac{a\lambda_x}{2} \cos^2 \frac{a\lambda_y}{2} \cos^2 \frac{c\lambda_z}{2} \right]^{1/2}$$

..... (41)

We have from (3) $\tanh 2\theta_{\underline{\lambda}} = -A\gamma_{\underline{\lambda}}$.

Therefore,

$$\sinh^2(\theta_{\underline{\lambda}+\underline{q}} - \theta_{\underline{\lambda}}) = \frac{1}{2}\left[\frac{1-A^2\gamma_{\underline{\lambda}}\,\gamma_{\underline{\lambda}+\underline{q}}}{\sqrt{1-A^2\gamma_{\underline{\lambda}}^2}\,\sqrt{1-A^2\gamma_{\underline{\lambda}+\underline{q}}^2}} - 1\right]$$

$$= \frac{1}{2}\left[\left(1-A^2\cos^2\frac{a\lambda_x}{2}\cos^2\frac{a\lambda_y}{2}\cos\frac{c\lambda_z}{2}\cos\frac{c(\lambda_z+q)}{2}\right)\right.$$

$$\times\left(1+\frac{1}{2}A^2\cos^2\frac{a\lambda_x}{2}\cos^2\frac{a\lambda_y}{2}\cos\frac{c\lambda_z}{2}\right)$$

$$\left.\times\left(1+\frac{1}{2}A^2\cos^2\frac{a\lambda_x}{2}\times\cos^2\frac{a\lambda_y}{2}\cos^2\frac{c(\lambda_z+q)}{2}\right) - 1\right]$$

(assuming q to be along z.)

$$\simeq \frac{1}{2}\,\frac{A^2}{4}\cos^2\frac{a\lambda_x}{2}\cos^2\frac{a\lambda_y}{2}\left(\cos\frac{c(\lambda_z+q)}{2} - \cos\frac{c\lambda_z}{2}\right)^2.$$

Now

$$\cos c\,\frac{\lambda_z+q}{2} = \cos\frac{c\lambda_z}{2}\cos\frac{cq}{2} - \sin\frac{c\lambda_z}{2}\sin\frac{cq}{2}$$

$$\simeq \cos\frac{c\lambda_z}{2} - \frac{cq}{2}\sin\frac{c\lambda_z}{2} \text{ for small } q.$$

Therefore,

$$\left(\cos c\,\frac{\lambda_z+q}{2} - \cos c\,\frac{\lambda_z}{2}\right)^2 = \frac{c^2q^2}{4}\sin^2\frac{c\lambda_z}{2}$$

Therefore

$$\sinh^2(\theta_{\underline{\lambda}+\underline{q}} - \theta_{\underline{\lambda}}) = \frac{A^2c^2q^2}{16}\cos^2\frac{a\lambda_x}{2}\cos^2\frac{a\lambda_y}{2}\sin^2\frac{c\lambda_z}{2} \qquad (42)$$

The absorption coefficient for the two magnon process is proportional to w_c times the transition probability (where w_c is the radiation frequency), hence

$$\alpha_{\perp}(w_c) = \emptyset \left(\frac{e^2\hbar^2\pi^2}{VM^2}\right)\left(\frac{4\pi n e^{*} C g \mu_B}{3\Omega_{oE_u^i}}\right)^2$$

$$x \sum_{q} \frac{q^2}{3} \frac{A^2 c^2 q^2}{16} \sum_{\underline{\lambda}} \left[\frac{E^{\alpha}_{\underline{\lambda}} + E^{\alpha}_{\underline{\lambda}+q}}{2\left[\left(\hbar\Omega_{oE_u^i}\right)^2 - \left(E^{\alpha}_{\underline{\lambda}} + E^{\alpha}_{\underline{\lambda}+\underline{q}}\right)^2\right]} - \frac{E_c}{\left[\left(\hbar\Omega_{oE_u^i}\right)^2 - E_c^2\right]}\right]^2 n_{E_c} \left(1+e^{-E^{\alpha}_{\underline{\lambda}}/k_B T}\right)\left(1+e^{-E^{\beta}_{\underline{\lambda}+q}/k_B T}\right)$$

$$x \cos^2 \frac{a\lambda_x}{2} \cos^2 \frac{a\lambda_y}{2} \sin^2 \frac{c\lambda_z}{2} \delta\left(E_c - E^{\alpha}_{\underline{\lambda}} - E^{\beta}_{\underline{\lambda}+\underline{q}}\right)$$

..... (43)

Changing from summation to integration, $\alpha_{\perp}(w_c)$ can be written as

$$\alpha_{\perp}(w_c) = \emptyset \left(\frac{V}{8\pi^2}\right)\left(\frac{e^2\hbar^2}{M^{*2}}\right)\left(\frac{4\pi n e^{*} C g \mu_B}{3\Omega_{oE_u^i}}\right)^2$$

$$\frac{C^2 A^2}{48} \int_0^{6m} q^5 dq \int_0^{\pi/a} \left(\frac{E_c}{2\left[\left(\hbar\Omega_{oE_u^i}\right)^2 - E_c^2\right]}\right)^2$$

$$x \; d\lambda_x \, d\lambda_y \, d\lambda_z \, n_{E_c} \left(1+e^{-Ec/k_B T}\right)^2 \cos^2 \frac{a\lambda_x}{z} \cos^2 \frac{a\lambda_y}{z}$$

$$x \sin^2 \frac{c\lambda_z}{z} \frac{1}{\hbar} \delta\left(w_c - 2w_{\underline{\lambda}}\right) \qquad (44)$$

Consider the δ-function appearing in the above integration; we use the following property of δ-function,

$$\delta(x^2-a^2) = \frac{1}{2a} [\delta(x-a) + \delta(x+a)] \qquad a \gtrless 0$$

However if the coefficient of this δ-function in the integral is even with respect to change of sign in x, one obtains

$$\delta(x^2-a^2) = \frac{2}{2a} \delta(x-a).$$

Therefore

$$\delta(w_c - 2w_\lambda) = \frac{1}{2} w_c \quad \delta(w_c^2 - 4w_\lambda^2)$$

$$w_c^2 = 4w_\lambda^2 = 4[w_{Bz}^2 - w_e^2 \cos^2 \frac{a\lambda_x}{2} \cos \frac{a\lambda_y}{z} \cos^2 \frac{c\lambda_z}{2}].$$

Therefore

$$\frac{4w_{Bz}^2 - w_c^2}{4w_c^2} = \cos^2 \frac{a\lambda_x}{2} \cos^2 \frac{a\lambda_y}{2} \cos^2 \frac{c\lambda_z}{2}$$

and $\delta(w_c - 2w_\lambda) = w_c \delta[F(w_c) - \cos^2 \frac{a\lambda_y}{2} \cos^2 \frac{c\lambda_z}{2} \cos^2 \frac{a\lambda_x}{2}]$,

..... (45)

where

$$F(w_c) = \frac{4w_{Bz}^2 - w_c^2}{4w_c^2} \qquad (46)$$

$$\alpha_{\perp}(w_c) = \left(\frac{\emptyset V e^2 \hbar^2 c^2 A^2}{48 \times 8M^{*2}} \right) \left(\frac{4ne^* C g \mu_B}{3\Omega_{oE_u^i}} \right)^2 \times \frac{6m^7}{7}$$

$$\times \left[\frac{E_c}{2[(\hbar\Omega_{oE_u^i})^2 - E_c^2]} \right]^2 n_{E_c} \left(1 + e^{-E_c/2k_B T} \right)^2$$

$$\times \int d\lambda_x d\lambda_y d\lambda_z \cos^2 \frac{a\lambda_x}{2} \cos^2 \frac{a\lambda_y}{2} \sin^2 \frac{c\lambda_z}{2}$$

$$\times \frac{w_c}{\cos^2 \frac{a\lambda_x}{2} \cos^2 \frac{a\lambda_y}{2}} \delta \left(\frac{F(w_c)}{\cos^2 \frac{a\lambda_x}{2} \cos^2 \frac{a\lambda_y}{2}} - \cos^2 \frac{c\lambda_z}{2} \right)$$

Consider ,

$$I = \int d\lambda_x d\lambda_y d\lambda_z \sin^2 \frac{c\lambda_z}{2} \delta \left(\frac{F(w_c)}{\cos^2 \frac{a\lambda_x}{2} \cos^2 \frac{a\lambda_y}{2}} - \cos^2 \frac{c\lambda_z}{2} \right)$$

Substituting ,

$$\cos^2 \frac{c\lambda_z}{2} = x \, , d\lambda_z = - x^{-1/2} (1 - x)^{-1/2} dx ,$$

I becomes

$$I = - \frac{w_c}{c} \int d\lambda_x d\lambda_y x^{-1/2} (1-x)^{1/2} \delta \left(\frac{F(w_c)}{\cos^2 \frac{a\lambda_x}{2} \cos^2 \frac{a\lambda_y}{2}} - x \right) dx$$

$$= - \frac{w_c}{c} \frac{1}{F(w_c)} \int d\lambda_x d\lambda_y \cos \frac{a\lambda_x}{2} \cos \frac{a\lambda_y}{2}$$

$$\times \left(1 - \frac{F(w_c)}{\cos^2 \frac{a\lambda_x}{2} \cos^2 \frac{a\lambda_y}{2}} \right)^{1/2}$$

Let

$$\frac{a\lambda_x}{2} = x, \quad \frac{a\lambda_y}{2} = y, \quad d\lambda_x d\lambda_y = 4a^2 dxdy .$$

Therefore

$$I = \frac{-4w_c}{a^2 c\sqrt{F(w_c)}} \int dxdy \cos x \cos y \left(1 - \frac{F(w_c)}{\cos^2 x \cos^2 y}\right)^{1/2} .$$

The limits are determined from the condition

$1 - \frac{F(w_c)}{\cos^2 x \cos^2 y} > 0$ in order that I be real.

$$I > \frac{F(w_c)}{\cos^2 x \cos^2 y} \qquad \text{therefore } \cos y > \frac{F^{1/2}}{\cos x}$$

Therefore

$$y < \cos^{-1} \left(\frac{F(w_c)^{1/2}}{\cos x} \right) ,$$

and

$$\cos^2 x > F \qquad \text{therefore} \qquad x < \cos^{-1} (F^{1/2}) .$$

Hence

$$I = - \frac{4w_c}{a^2 c\sqrt{F(w_c)}} \int_0^{\cos^{-1}(F^{1/2})} dx \int_0^{\cos^{-1}(F^{1/2}/\cos x)} \cos x \cos y \, dy \; x \left(1 - \frac{F(w_c)}{\cos x \cos y}\right)^{1/2}$$

This in the limit $F(w_c) \longrightarrow 0$ i.e. $w_c \longrightarrow 2w_{Bz}$ assumes the form

$$I \rightarrow \frac{4w_c}{a^2 c \sqrt{F(w_c)}} \left[\int_0^{\pi/2} \cos x \, dx \right]^2 ,$$

therefore ,

$$\alpha_1(w_c) = \emptyset \left(\frac{VA^2 e^2 \hbar 6 m^7 w_c}{3 \times 14 \, a^2 M^{*2}}\right) \left(\frac{ne^* C g \mu_B}{3 \Omega_{oE_u^i}}\right)^2$$

$$\times \left[\frac{E_c}{2[(\hbar\Omega_{oE_u^i})^2 - E_c^2]}\right]^2 e^{-E_c/k_B T} (1+e^{-E_c/2k_B T})^2 \times \frac{1}{\sqrt{F(w_c)}}$$

..... (47)

II. $E \| C$: In this case the photons are polarised along the z axis and hence phonons of A_{2u} made are involved. The required two-magnon term comes from the $[(H_{int}, S); S]$ giving the coefficient of the two-magnon process as,

$$\frac{S^2 e^2 \hbar^3 \pi}{72 M^{*2} w_c V} \frac{q^6}{3} \left(\frac{4\pi n e^* C g \mu_B}{3\Omega_{oA_{2u}}}\right) \left(\frac{\hbar}{2M\Omega_{oA_{2u}}}\right)^2$$

$$\times (\sinh 2\theta_{\underline{q}} + \cosh 2\theta_{\underline{q}})(\sinh 2\theta_{-\underline{\lambda}-\underline{q}} + \cosh 2\theta_{-\underline{\lambda}-\underline{q}})$$

$$\times \left[\frac{2\hbar\Omega_{oA_{2u}} (E_{\underline{\lambda}} + E_{\underline{\lambda}+\underline{q}} + E_c)}{[4(\hbar\Omega_{oA_{2u}})^2 - E_c^2][(\hbar\Omega_{oA_{2u}})^2 - E_{\underline{\lambda}}^2]} + \frac{\hbar\Omega_{oA_{2u}} (E_{\underline{\lambda}} + E_{\underline{\lambda}+\underline{q}})}{2[(\hbar\Omega_{oA_{2u}})^2 - E_{\underline{\lambda}}^2]^2} \right]$$

Defining the transition probability and absorption coefficient in the same manner as above and carrying out the integration we get ,

$$\alpha_{\parallel}(w_c) = \left(\frac{\emptyset V s^2 e^2 \hbar^2 6m^9}{16x72x27\,\pi^3 M^{*2}}\right)\left(\frac{4\pi n e^* C g \mu_B}{3\Omega_{0A_{2u}}}\right)^4$$

$$x \quad \left(\frac{\hbar}{2M^*\Omega_{0A_{2u}}}\right)^2 \frac{4w_c}{a^2 c\,/F(w_c)}$$

$$x \int_0^{\cos^{-1}(F(w_c)^{1/2}} dx \int_0^{\cos^{-1}(F^{1/2}/\cos x)} dy$$

$$\left[\sec x \sec y + \frac{A^2 c^2 q^2}{B} \cos^3 x \cos^3 y \; x \left(1 - \frac{F}{\cos^2 x \cos^2 y}\right)\right]$$

$$x \left(1 - \frac{F}{\cos^2 x \cos^2 y}\right)^{-1/2} ,$$

which in the limit $F \rightarrow 0$ assumes the form

$$\alpha_{\parallel}(w_c) = \left(\frac{2\emptyset V w_c s^2 e^2 \hbar m^9}{27\pi^2 M^{*2} a^2 c}\right)\left(\frac{4\pi n e^* C g \mu_B}{3\Omega_{0A_{2u}}}\right)^4$$

$$x \left(\frac{\hbar}{2M\Omega_{0A_{2u}}}\right)^2 x \left[\frac{\hbar\Omega_{0A_{2u}} E_c}{[4(\hbar\Omega_{0A_{2u}})^2 - E_c^2]^2}\right]^2 e^{-E_c/k_B T}$$

$$x \left(1+e^{-E_c/2k_B T}\right)^2 x \frac{1}{/F(w_c)}$$

$$x \left[\left[\int_0^{\pi/2} \sec x \, dx\right]^2 + \frac{A^2 c^1 q^2}{8}\left[\int_0^{\pi/2} \cos^3 x \, dx\right]^2\right]$$

..... (48)

This shows that there is a singularity at $w_c = 2w_{BZ}$ for both $\alpha_{\parallel}$ and $\alpha_{\perp}$ and hence the present mechanism predicts absorption at $w_c = 2w_{BZ}$ for both. Taking more exact relation for $w_{\underline{km}}$ (i.e. including the exchange interaction energy between ions neighbouring along z axis, as well as both the anisotropy constants of tetragonal and hexagonal nature, the singularity will occur at,

$$w_c = 2w_e \left[(1+d)^2 - e^2 - \frac{4(1+d)}{w_e^2} - \frac{Z_2 J_2}{Z_1 J_1} \right]^{1/2}$$

with $d = 2D/8J$, $e = 2E/8J$, $\hbar w_e = 8J$, $Z_1 = 8$, $Z_2 = 2$.

The ratio of the two absorption coefficients is

$$\frac{\alpha_{\parallel}}{\alpha_{\perp}} = \frac{7S^2 6m^2}{9\pi^2 c^2 A^2} \left(\frac{4\pi n e^* C g \mu_B}{3\Omega_{oA_{2u}}} \right)^2 \left(\frac{\Omega_{oE_u^i}}{\Omega_{oA_{2u}}} \right)^2 \left(\frac{\hbar}{2M^* \Omega_{oA_{2u}}} \right)^2$$

$$\times \frac{(\hbar \Omega_{oE_u^i})^4}{(\hbar \Omega_{oA_{2u}})^6} . \qquad (49)$$

DISCUSSION

In the present chapter we have considered the magnon-phonon interaction via the transverse optical phonons. A new mechanism for the coupling between magnons and transverse-phonons has been used. The conventional mechanism of coupling between spins and the lattice is through the spin-orbit and the orbit-lattice couplings. The former is essentially an intra-atomic coupling while the latter is quite generally given by an expression of the type

$$H_{OL} = C \int \rho(\underline{r}) \text{ div } \underline{u}(\underline{r}) \, d^3r \quad , \tag{50}$$

where $\rho(\underline{r})$ is the electronic charge density for the orbit in question and $\underline{u}(\underline{r})$ the lattice displacement at the point $\underline{r}$ of the host continuum. This expression is identically zero for the transverse mode since div $\underline{u}(\underline{r}) = 0$ for transverse mode. Thus the conventional mechanism fails to couple the magnons to the transverse optical phonons.

We have considered the magnetic field associated with the transverse optical mode. This field gives rise to the Zeeman type coupling with the spins leading in turn to the magnon phonon interaction. There is a strong evidence in favour of the magnetic field considered. Kumar and Sinha have been successful in explaining the spin-phonon relaxation due to transverse optical mode by the mechanism considered above.

Halley and Silver[a] have ascribed the two-ion coupling to the interaction between a spin-orbit induced electronic quandrupole moment of atom j and the electronic dipole moment of atom i. The even parity excited state energies of the magnetic ions appear as energy denominators in their theoretical expressions for π_i. For Fe^{2+} these energies are in the region 1000 to 2000 cm^{-1} above the ground state; for Mn^{2+} about 20,000 cm^{-1} above the ground state. The energy denominator appears in the 4th power and hence their theory predicts the magnon absorption in MnF_2 to be about four orders of magnitude smaller than in FeF_2. Experimentally the two intensities are found to be comparable.

Recently Halley has proposed another mechanism for phonon magnon interaction. According to his throy, the photon absorption arises from indirect coupling of the spins to the field through the infrared-active optical phonons via the exchange-strictive part of the spin-phonon interaction. Inasmuch as the exchange integrals involve purely orbital functions of the electrons, ~~its~~ their modulation by transverse optical phonons seem unlikely. This follows from the arguments following equation, (50).

By the present mechanism we obtain an absorption at $2w_{BZ}$ which is equal to 154 cm^{-1} for FeF_2. The result is in agreement with that obtained by Halley and Silver[a] for the same. The absorption coefficient shows a temperature dependence of the form

$$e^{-E_c/k_BT}\left(1+e^{-E_c/2k_BT}\right)^2.$$

Due to the lack of sufficient experimental results, this cannot be compared with the observed temperature dependence.

The order of magnitude of the ratio of the absorption coefficients has been calculated for FeF_2. The various quantities involved have the following values:

S -	5/2	$\hbar$ ~	10^{-27} erg. sec.
M* -	10^{-23} gms	c -	3×10^{-8} cms.
n -	10^{23} per unit volume	e* -	4.8×10^{-18} esu.
C -	3×10^{10} cm/sec.	μ_B -	10^{-20} erg guass^{-1}.
A -	w_e/w_e+w_A 0.7298	σ_n -	10^{6} cm^{-1}

The following values for Ω have been considered

(1) $\Omega_{oE_u^i} = 4.8 \times 3 \times 10^{12}$ sec^{-1}

(2) $= 3.2 \times 10^{12}$ sec^{-1}

(3) $= 2.0 \times 3 \times 10^{12}$ sec^{-1}

and

$$\Omega_{oA_{2u}} = 4.4 \times 3 \times 10^{12} \text{ sec}^{-1}$$

With these values the ratio comes to be of the order of 10, in excellent agreement with experimental observation which is found to be between 7 and 13.

A plot of a_{11} against radiation frequency is given in fig.1 according to the formula obtained here. For comparison experimental results of Silvera and Hadley[44] are also given. It appears that ~~these are in~~ there is an excellent agreement between the theoretical formula obtained here and the experimental results, particularly below the resonance frequency. The slight discrepancy above the resonance frequency may be due to the magnon-magnon interactions which are not considered in the present analysis.

It is interesting to compare the expression for intensity as a function of frequency as $T \longrightarrow 0$, as given by Moriya, with that obtained in the present case. Moriya's expression is given by

$$a_{TSM}(w_{\underline{k}\lambda}) = \frac{C_{TSM}\, w_{\underline{k}\lambda}^3}{F^{1/2}} \int_0^{\cos^{-1}(F^{1/2})} dx \times \int_0^{\cos^{-1}(F^{1/2}/\cos x)} dy \cos x \cos y \left(1 - \frac{F}{\cos^2 x \cos^2 y}\right)^2$$

In the limit $T \longrightarrow 0$, our expression assumes the form

$$a(w_{\underline{k}\lambda}) = \frac{\phi\, w_{\underline{k}\lambda}^3}{[(\hbar\Omega_0)^2 - \hbar w_{\underline{k}}^2]^2} \times \frac{1}{F^{1/2}} \times \int_0^{\cos^{-1}(F^{1/2})} dx \int_0^{\cos(F^{1/2}/\cos x)} dy \cos x \cos y \left(1 - \frac{F}{\cos x \cos y}\right)^{\frac{1}{2}}$$

Thus the variation of α with frequency is found to be similar except for the term $1/[\,\Omega_o^2 - w_{\underline{k}\lambda}^2\,]^2$ occurring in our expression. For the photon frequency of interest, namely 154 cm^{-1} for FeF_2, the photon frequencies are far removed and hence the factor $1 / [\,\Omega_o^2 - w_{\underline{k}\lambda}^2\,]^2$ introduces a modification in the magnitude of α, without introducing an additional singularity.

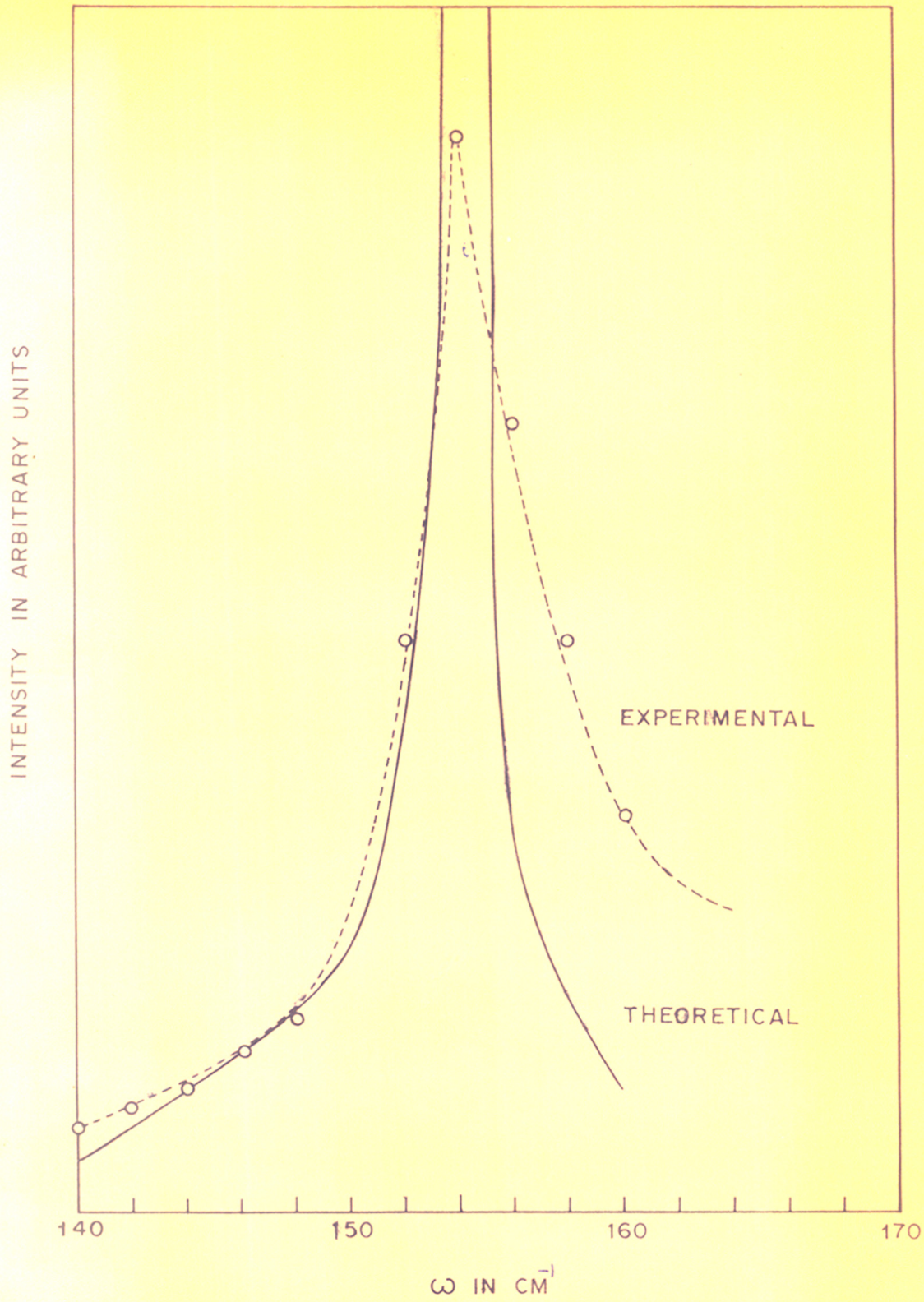

FIG. 1. GRAPH OF INTENSITY OF ABSORPTION v/s THE ABSORPTION FREQUENCY FOR FeF_2.

APPENDIX - 1

Here we give the pertinent details of the integration involved in the calculation of the scattering cross-section: From (28a)

$$\frac{d^2\sigma}{dEd\Omega}(a^+_{\underline{\lambda}}, a_{\underline{\lambda}'}) = \frac{k'}{4\pi 2k}\,\frac{m_2^2}{\hbar^4}\sum_{\underline{\lambda}} \phi^2 A^2_{\underline{\lambda}\underline{q}}\, e^{-E_{\underline{\lambda}}-\underline{q}/k_BT}$$

$$e^{-2W_{\underline{q}}}\,\delta\,(E_{\underline{k}} - E_{\underline{k}'} - E^a_{\underline{\lambda}} + E^a_{\underline{\lambda}-\underline{q}})$$

Taking the z axis along the direction of $\underline{q}$ and changing from summation to integration

$$\frac{d^2\sigma}{dEd\Omega} = \frac{k'}{4\pi^2 k}\,\frac{a^2 m_n^2}{2\hbar^4}\,q^2\,\frac{Nv}{4\pi^2}\int \lambda^2 d\lambda \sin\theta\, d\theta$$

$$\times\left[\frac{(\lambda^2 + |\lambda-q|^2 - q^2)^2 + 4\lambda^2|\lambda-q|^2}{2\lambda|\lambda-q|} + 2(\lambda^2+|\lambda-q|^2 - q^2)\right]$$

$$\times\; e^{-\theta_c a/T}\times(\lambda^2+q^2-2\lambda q\cos\theta)^{1/2}\times\delta(E_{\underline{k}}-E_{\underline{k}'}-E^a_{\underline{\lambda}}+E^a_{\underline{\lambda}-\underline{q}})$$

Now consider the δ-function.

Using the property of δ-functions, namely $\delta(ax) = 1/|a|\,\delta(x)$ where a is a constant, we can write the above δ-function as

$$\frac{1}{k_B\theta_c a}\,\delta\left(\frac{\hbar^2(k^2-k'^2)}{2m_n k_B\theta_c a} - \lambda + |\lambda - q|\right)$$

Next we use the property

$$\delta(f(x)) = \sum_{\alpha_i} \frac{\delta(x-\alpha_i)}{\left.\frac{df}{dx}\right|_{x=\alpha_i}}$$

where α_i is the i^{th} root of $f(x) = 0$.

In the present case we consider the argument as a function of $\cos\theta$. We write

$$f(\cos\theta) = b-\lambda+(\lambda^2+q^2-2\lambda q\cos\theta)^{1/2},$$

so that the root is given by

$$\cos\theta = \frac{-b^2+2\lambda b+q^2}{2\lambda q} = \alpha,$$

and

$$\left.\frac{df}{d\cos\theta}\right)_{\cos\theta=\alpha} = -\frac{\lambda q}{\lambda-b}.$$

Therefore the integral becomes

$$I = \int \lambda^2 d\lambda \sin\theta\, d\theta\, x \left[\frac{(2\lambda^2-2q\lambda\cos\theta)^2+4\lambda^2(\lambda^2+q^2-2\lambda q\cos\theta)}{2(\lambda^2+q^2-2\lambda q\cos\theta)^{1/2}} + 2(2\lambda^2-2\lambda q\cos\theta)\right] x\ e^{-\theta_c a/T(\lambda^2+q^2-2\lambda q\cos\theta)^{1/2}}$$

$$+ \quad -\lambda+b/\lambda q\ \delta\left(\frac{q^2+2b-b^2}{2\lambda q}-\cos\theta\right)$$

Carrying out the integration over the angle variable with the help of δ-function, I assumes a very simple form ,

$$I = \int \lambda(b-\lambda)\,d\lambda\, e^{-\theta_c a/T(\lambda-b)} \; x\left[\frac{(2\lambda^2+b^2-2b\lambda-q^2)^2+4\lambda^2(\lambda^2+b^2-2b\lambda)}{2\ (\lambda-b)}\right.$$

$$\left. + 2(2\lambda^2+b^2-2b\lambda-q^2)\right]$$

which after straightforward integration gives the differential scattering cross-section given in (31a).

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REFERENCES

1. W. Heisenberg, Z. Physik, 49, 619 (1928)
2. F.Bloch, Z. Physik, 74, 295 (1932).
3. T.Holstein and H.Primakoff, Phys. Rev., 58, 1098 (1940).
4. F.J.Dyson, Phys. Rev., 102, 1217 and 1230 (1956).
5. L.Hulthen, Proc. Acad. Sci. Amsterdam, 39, 190 (1936).
6. J.M.Ziman, Proc. Phys. Soc. (London) A65, 540, and 548 (1952).
7. P.W.Anderson, Phys. Rev., 86, 694 (1952).
8. R.Kubo, Phys. Rev., 87, 568 (1952).
9. For a more comprehensive bibliography see C.Kittel, International Conference on Magnetism and Crystallography, Kyoto, Japan, 2, 161 (1961).
10. J.Van Kranendonk and J.H.Van Vleck, Revs. Modern Phys., 30, 1 (1958).
11. C.G.Shull and J.S.Smart, Phys. Rev., 76, 1256 (1949).
12. F.Bloch, Phys. Rev., 50, 259 (1936); ibid 51, 994 (1937)
13. J.Schwinger, Phys. Rev. 51, 544 (1937).
14. O.Halpern and M.H.Johnson, Phys. Rev. 55, 848 (1939).
15. L.Van Hove, Phys. Rev., 95, 249 and 1375 (1954).
16. J.H.Van Vleck, Phys.Rev., 55, 924 (1939).
17. M.Slotnick, Phys, Rev. 83, 1226 (1965).
18. P.G. de Gennes, J.Phys. Chem. Solids, 4, 1223 (1958).
19. A.W.Sa'enz, Phys. Rev. 119, 1542 (1960).
20. C.G.Shull, W.A.Strauser, E.O.Wollan, Phys. Rev. 83,333 (1951).
21. R.A.Erickson, Phys. Rev. 90, 779 (1953).
22. B.N.Brockhouse, L.M.Corliss and J.M.Hastings, Phys. Rev. 98, 1721 (1955).

23. P.J.Bendt, Phys. Rev. 98, 1721 (1955).

24. R.J.Elliot and R.D.Lowde, Proc. Roy. Soc. (London) A230, 46 (1955).

25. S.V.Maleev, J.Exptl. Theoret. Phys. U.S.S.R., 33, 1010 (1957); Soviet Phys. - J.E.T.P. 6 (33),776 (1958)

26. T.A.Kaplan, Bull. Am. Phys. Soc. 4, 178 (1959).

27. T.A.Kaplan, Phys. Rev., 109, 782 (1958).

28. R.D.Lowde , Proc. Roy. Soc. (London) A235, 305 (1956).

29. T.Riste, K.Blinowski and J.Janik, J.Phys. Chem. Solids 9, 153 (1959).

30. B.N.Brockhouse, Phys. Rev. 106, 859 (1957); 111, 1273 (1958).

31. R.D.Lowde and N.Umanatha, Phys. Rev. Letters, 4,452 (1960).

32. R.N.Sinclair and B.N.Brockhouse, Phys. Rev., 120,1638 (1960).

33. O.Nagai and A.Yashimori, Progr. Theoret. Phys. (Kyoto), 25, 595 (1961).

34. A.W.Sa'enz, Phys.Rev., 125, 1940 (1962).

35. A.W.Joshi and K.P.Sinha, Proc. Phys. Soc. (G.B.) 91, 97 (1967).

36. J.W.Halley and I.Silvera, Phys. Rev. Letters, 15,654 (1965).

37. P.L.Richards, Bull. Am. Phys. Soc., 10, 33 (1965).

38. S.J.Allen Jr., R.Loudon, and P.L.Richards, Phys. Rev. Letters, 16, 463 (1966).

39. R.L.Greene, D.D.Sell, W.M.Yen, A.L.Shawlow and R.M.White, J.Appl. Phys. 37, 1229 (1966).

40. P.G.Rusell, D.S.McClure and J.W.Stout, Phys. Rev. Letters 16, 176 (1966).

41. L.F.Johnson, R.E.Dietz and H.J.Guggenheim, Phys. Rev. Letters 16, 841 (1966); J.Tylicki and W.M.Yen, Phys. Rev. 166, 488 (1968).

42. R.Stevenson, Phys. Rev. 152, 531 (1966)
G.Zanmarchi and P.F.Bongers, Solid State Communications 6, 27 (1968).
J.P.Van der Ziel and H.J.Guggenheim, Phys. Rev. 166, 479 (1968).

43. P.A.Fleury, S.P.S.Porto, L.E.Cheesman and H.J.Guggenheim, Phys. Rev. Letters, 17, 84 (1966);
P.A.Fleury and R.Loudon, Phys. Rev. 166, 514 (1968).

44. I.Silvera and J.W.Halley, Phys. Rev. 149, 415 (1966).

45. Y.R.Shen and N.Bloembergen, Phys. Rev., 143, 406 (1963);
R.Loudon, Adv. in Phys. 17, No.66, 243 (1968);
T.Moriya and Michiko Inoue, Technical Report of ISSP, Sr.A, No.302, 1968.

46. J.W.Halley, Phys. Rev. 149, 423 (1966).

47. Y.Tanabe, T.Moriya and S.Sugano, Phys. Rev. Letters, 15, 1023 (1965).

48. J.W.Halley, Phys. Rev. 154, 458 (1967).

49. T.Moriya, J.Phys. Soc. Japan, 21, 926 (1966).

50. Y.Tanabe and K.Gondaira, J.Phys.Soc. Japan, 21, 1527 (1966).

51. Y.Tanabe and K.Gondaira, J. Phys.Soc.Japan, 22, 573 (1967).

52. T.Moriya, J. Phys. Soc. Japan,23, 490 (1967).

53. D.L.Dexter, Phys.Rev. 126, 1962 (1962).

54. F.Varsanyi and G.J.Dieke, Phys. Rev. Letters, 7, 442 (1961).

55. P.G. de Gennes, Magnetism, Vol.III, Edited by G.T.Rado and H.Suhl (Academic Press, New York 1963).

56. U.N.Upadhyaya, Ph.D.Thesis, University of Poona, 1964 (Unpublished).

57. C.Kittel, 'Quantum Theory of Solids', (John Wiley and Sons, New York, 1963).

58. U.N.Upadhyaya and K.P.Sinha, Phys. Rev. 130, 939 (1963).

59. L.D.Landau and E.M.Liftshitz, 'Quantum Mechanics' (Pergamon Press, London, 1958).

60. B.C.Frazer, G.Shorane, D.E.Cox and C.E.Olsen, Phys.Rev. 140, A1148, (1965).

61. R.Nathans, Kjeller Report of the Advanced Course on Magnetic Exchange Interaction in Transition Metal Compounds, 153, (1964).

62. A.A.Maradudin, E.W.Montroll and G.H.Weiss, Solid state Physics, 4, 1963. (Academic Press, New York).

63. A.A.Lorentz, 'The Theory of Electrons' Dover Publications.

64. J.J.Hopfield, Phys. Rev. 112, 1555 (1958).

65. M.Balkanski, P.Moch and G.Parisot, J.Chem. Phys., 44, 940 (1966).

66. V. Gallina and M. Omini, Nuovo Cimento, 33, 1031 (1964).