CHAPTER 2

THEORIES OF FERRO-, ANTIFERRO- AND FERRIMAGNETISM

2.1 The Effective Field Theory

2.1.1 The Weiss molecular field theory

The Weiss molecular field approximation was originally given by Pierre Weiss . This theory states that the ordering of the magnetic moment which induces ferro-, antiferro- and ferrimagnetism is due to the internal interaction between the spins of the magnetic atoms. Such an interaction between each magnetic atom and the remainder of the crystal is replaced by an effective field $\underline{H}_e$, called the exchange field, which is assumed to be proportional to the average magnetic moment of the crystal:

$$\underline{H}_e = q \, \underline{M}, \tag{2.1}$$

where q is a constant called the molecular field coefficient. Heisenberg[7] described this molecular field concept as a conse-

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[7] W. Heisenberg, *Zeitschrift fur Physik* 38, 411 (1926).

quence of a quantum mechanical exchange interaction which has no classical analog. The exchange interactions between spins of localized electrons can be taken into account by adding, to the Hamiltonian of the system, a term

$$\mathcal{H}_H = -2 \sum_{i,j} J_{ij} \underset{\sim}{S}_i \cdot \underset{\sim}{S}_j, \qquad (2.2)$$

called the Heisenberg Hamiltonian, where $\underset{\sim}{S}_i$, $\underset{\sim}{S}_j$ are the total spins of atom i, j, respectively and J_{ij} is the exchange integral which is negative for antiparallel spin interactions whereas it is positive for parallel spin interactions, therefore J_{ij} is positive for ferromagnetic interaction while it becomes negative for antiferro- and ferrimagnetic interaction. The derivation of the Heisenberg Hamiltonian together with the exchange integral are shown in Appendix A. Considering the case of identical interactions of nearest neighbour pairs only, we get the single atom Heisenberg Hamiltonian from eq. (2.2) of the form

$$\mathcal{H} = -2J \underset{\sim}{S}_i \cdot \sum_{j=1}^{z} \underset{\sim}{S}_j, \qquad (2.3)$$

where the sum is over the z nearest neighbours of the ith atom. In terms of the Weiss molecular field, the Hamiltonian for this ith atom is

$$\mathcal{H} = -\underset{\sim}{\mu}_i \cdot \underset{\sim}{H}_e = -g\mu_B \underset{\sim}{S}_i \cdot \underset{\sim}{H}_e, \qquad (2.4)$$

where g is the Lande g-factor and μ_B is the Bohr magneton. Equating eqs. (2.3) and (2.4) together with the definition of the exchange field $\mathbf{H}_e$ in eq. (2.1) give an approximate connection between the exchange integral J and the molecular field coefficient q by the relation

$$q = \frac{2zJ}{Ng^2\mu_B^2} , \qquad (2.5)$$

where N is the number of atoms in unit volume.

The molecular field treatment of a ferromagnet considers a system of atoms with total spins S. Each atom experiences the effective field $\mathbf{H}_e = q\mathbf{M}$ produced by its interaction with the other atoms. By the method of statistical mechanics[8], the susceptibility above the Curie temperature is given by the Curie-Weiss law:

$$\chi = \frac{C}{T - T_C} ; \quad T_C = qC, \qquad (2.6)$$

whereas experimentally, it is given accurately by

$$\chi = \frac{C}{T - \theta} , \qquad (2.7)$$

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[8] C. Kittel, *Introduction to Solid State Physics* (4th ed.; New York: John Wiley & Sons, Inc., 1971), p. 530.

where Θ is appreciably greater than the actual transition temperature T_C (see Fig. 1). Moreover, the molecular field theory predicts the relative magnetization at low temperatures according to the relation

$$\frac{M(T)}{M(0)} = 1 - 2e^{-2x}, \tag{2.8}$$

where $x = \frac{g\mu_B SH}{kT}$. Instead, it is found that the best fit with the experimental data for the magnetization near absolute zero is given by

$$\frac{M(T)}{M(0)} = (1 - AT^{3/2}), \tag{2.9}$$

the Block's $T^{3/2}$ temperature behavior.

In an antiferromagnet with two equivalent sublattices of antiparallel magnetic moments two different molecular fields $\underline{H}_{eA}$ and $\underline{H}_{eB}$ are expected to act on the A and B sublattices, respectively. The interactions giving rise to those fields are the interaction between the magnetic moments within each sublattice and the interaction between the magnetic moments of different sublattices. Generally, the latter is the strongest. Hence, we have

$$\underline{H}_{eA} = - q_{ii}\underline{M}_A - q_{AB}\underline{M}_B,$$

$$\underline{H}_{eB} = -q_{BA}\underline{M}_A - q_{ii}\underline{M}_B, \qquad (2.10)$$

where q_{ii} is the molecular field coefficient for the interaction between atoms within each sublattice and $q_{AB} = q_{BA}$ is the molecular field coefficient for the interaction between atoms in different sublattices. Following the same statistical treatment used to obtain eq. (2 .7), the susceptibility above the Néel point of an antiferromagnetic system turns out to be (see Fig. 1)

$$\chi = \frac{C}{T + \theta}, \qquad (2.11)$$

where
$$\theta = \frac{1}{2}C(q_{ii} + q_{AB}). \qquad (2.12)$$

The Néel temperature is given by

$$T_N = \frac{1}{2}C(q_{AB} - q_{ii}). \qquad (2.13)$$

Moreover, the susceptibility below the Néel point of an antiferromagnet is obtained by

$$\chi_p = \frac{1}{3}\chi_{//} + \frac{2}{3}\chi_{\perp}, \qquad (2.14)$$

where $\chi_{//}$ is the susceptibility below the Néel point when the magnetic field is applied parallel to the direction of magnetization and is that for perpendicular applied field. It is easily shown[9] that

$$\chi_{//} = \frac{N u_B^2 g^2 S^2 B_s'(x_o)}{kT + \frac{1}{2}(q_{ii}+q_{AB})\, u_B^2 g^2 S^2 N B_s'(x_o)}, \qquad (2.15)$$

and

$$\chi_{\perp} = \frac{1}{q_{AB}}, \qquad (2.16)$$

where $B_s'(x_o)$ is the derivative of the Brillouin function with respect to its argument and $x_o = \frac{g\mu_B S}{kT}(q_{AB} - q_{ii})M_o$; $M_o = M_A = M_B$. The curves of $\chi_{//}$, $\chi_{\perp}$ and χ_p are shown in Fig. 2. Experimental data[10] shows that $\chi_{//}$ does not vanish at absolute zero.

The Weiss molecular field approach has also been applied to a ferrimagnet in which the two sublattices have different magnetization. In this case the molecular fields acting on the A and B sublattices are such as

$$\underline{H}_{eA} = - q_{AA}\underline{M}_A - q_{AB}\underline{M}_B,$$

$$\underline{H}_{eB} = - q_{BA} - q_{BB}\underline{M}_B. \qquad (2.17)$$

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[9] A.H. Morrish, op. cit., pp. 450-453.

[10] S. Forner, "High-Field Antiferromagnetic Resonance in Cr_2O_3," Physical Review 130, 183 (1963).

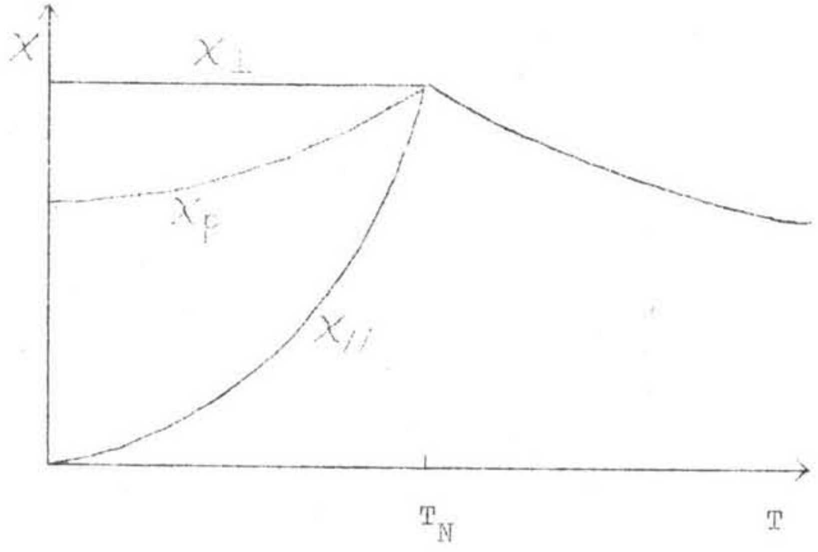

Fig. 2 Temperature dependence of the magnetic susceptibility of an antiferromagnetic substance.

However, now, $q_{AB} = q_{BA}$, $q_{AA} \neq q_{BB}$ and $\underline{M}_A \neq \underline{M}_B$. Appling a similar type of statistical analysis used for the previous two magnetic systems, it is found that the paramagnetic susceptibility of a ferrimagnetic system is

$$\frac{1}{\chi} = \frac{T}{C} - \frac{1}{\chi_o} - \frac{\sigma}{T - \theta'} , \qquad (2.18)$$

where χ_o, σ and θ' are constants characteristic of the material. The graphical representation of eq. (2.18) is a hyperbola shown in Fig. 1. Also, the Néel temperature of a ferrimagnet can be obtained as

$$T_N = -\frac{1}{2}(C_A q_{AA} + C_B q_{BB}) + \frac{1}{2}\left[(C_A q_{AA} - C_B q_{BB})^2 + 4C_A C_B q_{AB}^2\right]^{1/2}, \qquad (2.19)$$

where
$$C_A = \sum_i \frac{N_i g^2 \mu_B^2 S_i(S_i+1)}{3k} ,$$

and
$$C_B = \sum_j \frac{N_j g^2 \mu_B^2 S_j(S_j+1)}{3k} . \qquad (2.20)$$

The spontaneous magnetization of the system at any time is found from the equation

$$\underline{M}(T) = \underline{M}_A(T) - \underline{M}_B(T). \qquad (2.21)$$

At low temperatures, the resultant magnetization often varies with the temperature in a manner reminiscent of the ferromagnetic case, espectially, if the intrasublattice interactions q_{AA} and q_{BB} are of the same magnitude. However, some unusual variations are also possible.

2.1.2 The Oguchi's method (The constant coupling approximation)

In the Weiss molecular field theory all the exchange interactions in the crystal are replaced by an effective field and the spin-spin interaction has not been taken into account. Oguchi[11] has treated the exchange interaction between a pais of nearest neighbour atoms selected at random: the coupling with the other atoms is represented by an effective field. The Oguchi's method has been improved[12] by allowing the effective field to be not only a function of magnetization, but also a function of temperature. Using the usual statistical analysis, this method yields the results similar to those obtained from the Weiss molecular field theory at temperatures well above the transition temperature whereas in the vicinity of the transition temperature, it gives more reliable results. Moreover,

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[11] T. Oguchi, Progress of Theoretical Physics 13, 148 (1955).

[12] P.W. Kasteleijn and J. Van Kranendonk, "Constant Coupling Approximation for Heisenberg Ferromagnetism," Physica XXII, 317 (1956).

the transition temperature predicted by this theory is appreciably different from that obtained from the Weiss molecular field theory. However, for temperatures appreciably less than the transition temperature, the constant coupling method fails badly.

2.1.3 The Bethe-Peierls-Weiss method

The principle of the Bethe[13]-Peierls[14]-Weiss[15] (B-P-W) approximation considers a cluster of atoms that consists of an arbitrary central atom and its z nearest neighbours. The interaction of the neighbour atoms with each other and with the central atom is treated by the exchange interaction. The effect of the atoms outside the cluster is represented by an effective field $\underset{\sim}{H}_e$ which acts on the z neighbour atoms of the cluster. The effective field is determined by a consistency condition. The transition temperature is recognized as the temperature below which $\underset{\sim}{H}_e$ is nonzero.

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[13] H.A. Bethe, "Statistical Theory of Superlattices," Proceeding of the Royal Society of London A150, 552 (1935).

[14] R.E. Peierls, "Statistical Theory of Superlattices with Unequal Concentrations of the Components," Proceeding of the Royal Society of London A154, 207 (1936).

[15] P.R. Weiss, "The Application of the Bethe-Peierls Method to Ferromagnetism," Physical Review 74, 1493 (1948).

This method has also been applied to antiferro-[16] and ferrimagnetism[17]. The results obtained are in good agreement with those of the Weiss molecular field approach at temperatures well above the transition temperature. However, at temperatures near the transition temperature, this method yields the paramagnetic susceptibility which does not follow the Curie-Weiss law: the $1/\chi$ versus T curve is concave in a ferromagnet while it is convex in an antiferromagnet, as observed experimentally. For the case of ferrimagnetism, the $1/\chi$ versus T curve calculated by this method and the Weiss molecular field approach differ appreciably for some restricted temperatures above the Neel point; at higher temperatures, however, they approach one another. Moreover, this method yields the results for the transition temperatures that are almost the same as obtained by the constant coupling approximation.

Finally, it should be noted that the Bethe-Peierls-Weiss method is concerned only with the local ordering within a cluster and neglects long-range order and spin-wave effects that are

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[16] Y.Y. Li, "Magnetic Moment Arrangements and Magnetocrystalline Deformations in Antiferromagnetic Compounds," Physical Review 100, 627 (1955).

[17] J.S. Smart, "Application of the Bethe-Weiss Method to Ferrimagnetism," Physical Review 101, 585 (1956).

important at low temperatures. Hence, the validity of this method is therefore confined to temperatures in the vicinity of the transition temperature.

2.2 The Spin Wave Theory

Bloch[18] invented the concept of a spin wave, from which Herring and Kittel[19] developed a simple phenomenological spin wave theory. At absolute zero, a spin system is completely ordered and is in its ground state. The first excited state may presumably be obtained by raising the temperature slightly so that one spin is reversed. However, an excitation of much lower energy can be obtained by allowing all spins to share the reversal. These low-lying elementary excitations of the spin system have a wavelike form and are called spin waves or, when quantized, magnons. The formation of a spin wave is illustrated by Fig. 3 for a one-dimensional ferromagnetic system. The usual approximation should be made by assuming that spin waves are independent and the superposition is possible only as long as the number of the spin waves is small, that is, for temperatures

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[18] F. Bloch, Zeitschrift fur Physik 61, 206 (1930); 74, 295 (1932).

[19] C. Herring and C. Kittel, "On the Theory of Spin Waves in Ferromagnetic Media," Physical Review 81, 869 (1951).

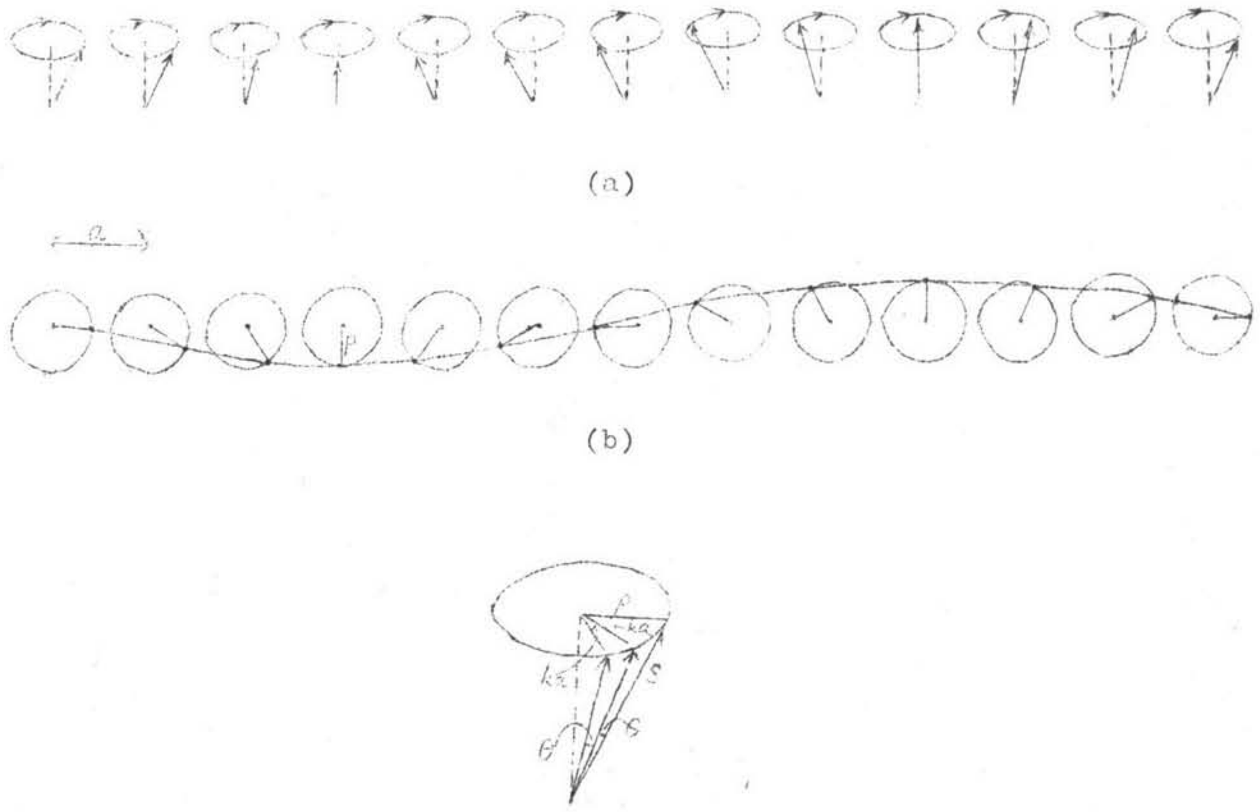

Fig. 3 Illustrating a spin wave with wave vector k.

(a) The precessing spins viewed in perspective.

(b) The spins viewed from above, showing one wavelength.

(c) The angular relationships between the spins of three successive atoms.

well below the transition temperature.

Consider a spin system of the localized spin model in a ferromagnet which has the Heisenberg interaction given by eq. (2.2):

$$\mathcal{H} = -2 \sum_{i,j} J_{ij} \underset{\sim}{S}_i \cdot \underset{\sim}{S}_j .$$

The equation of motion for each spin is

$$\hbar \frac{d}{dt} \underset{\sim}{S}_i = \frac{1}{i} \left[\underset{\sim}{S}_i , \mathcal{H} \right] . \qquad (2.22)$$

Using the commutation relation

$$\left[\underset{\sim}{S}_i , (\underset{\sim}{S}_i \cdot \underset{\sim}{A}) \right] = i \underset{\sim}{A} \times \underset{\sim}{S}_i ,$$

we obtain

$$\hbar \frac{d}{dt} \underset{\sim}{S}_i = \underset{\sim}{S}_i \times \sum_j 2J_{ij} \underset{\sim}{S}_j . \qquad (2.23)$$

Suppose that the spins are slightly disturbed, and set

$$\underset{\sim}{S}_i = \underset{\sim}{S}_o + \delta \underset{\sim}{S}_i , \quad \underset{\sim}{S}_j = \underset{\sim}{S}_o + \delta \underset{\sim}{S}_j , \qquad (2.24)$$

where $\underset{\sim}{S}_o$ is the spin value in the ground state.

Then eq. (2.23) becomes

$$\hbar \frac{d}{dt} \delta \underline{S}_i = \sum_j 2J_{ij}(\delta \underline{S}_i - \delta \underline{S}_j) \times \underline{S}_o \qquad (2.25)$$

by ignoring terms of degree higher than 1.

The solution of eq. (2.25) can be written as

$$\delta \underline{S}_i = \underline{A}_k e^{i\omega_k t - i\underline{k}\cdot\underline{r}_i} \qquad (2.26)$$

which is in the form of travelling waves.

Therefore eq. (2.25) becomes

$$i\hbar \omega_k \underline{A}_k = \left[\sum_j 2J_{ij} \left(1 - e^{i\underline{k}\cdot(\underline{r}_i - \underline{r}_j)} \right) \right] \underline{A}_k \times \underline{S}_o. \qquad (2.27)$$

Assuming that the considering spin is at the origin of the coordinate system and the exchange integrals between the given atom and its z nearest neighbouring atoms are all equal to J, then the frequency ω_k of the spin wave having wave vector $\underline{k}$ is immediately obtained from eq. (2.27) as

$$\omega_k = \frac{2J}{\hbar} S \sum_{j=1}^{z} \left(1 - e^{-i\underline{k}\cdot\underline{r}_j} \right). \qquad (2.28)$$

With this solution it can be seen from eq. (2.26) that

$$\delta S_{ix} = A_k \cos(\omega_k t - \mathbf{k}.\mathbf{r}_i),$$

$$\delta S_{iy} = -A_k \sin(\omega_k t - \mathbf{k}.\mathbf{r}_i) \qquad (2.29)$$

which expresses the propagation of the classical spin wave in the direction of $\mathbf{k}$ (see Fig. 3).

Equation (2.28) is the dispersion relation $\omega(\mathbf{k})$ for spin waves with nearest neighbour interactions. If each lattice point is at the center of inversion, i.e., all nearest neighbours are at a distance $\pm\,\mathbf{r}_j$ from the given lattice point, eq. (2.28) simplifies to

$$\omega_k = \frac{2J}{\hbar} S \sum_{j=1} (1 - \cos \mathbf{k}.\mathbf{r}_j). \qquad (2.30)$$

For the one dimensional system with lattice constant a, eq. (2.30) is reduced to

$$\omega_k = \frac{4J}{\hbar} S(1 - \cos ka). \qquad (2.31)$$

At long wavelength $ka \ll 1$, this is reduced to

$$\hbar\omega_k \cong (2JSa^2)k^2. \qquad (2.32)$$

The dispersion relation for spin waves in a one dimensional ferromagnet with nearest neighbour interactions as given by

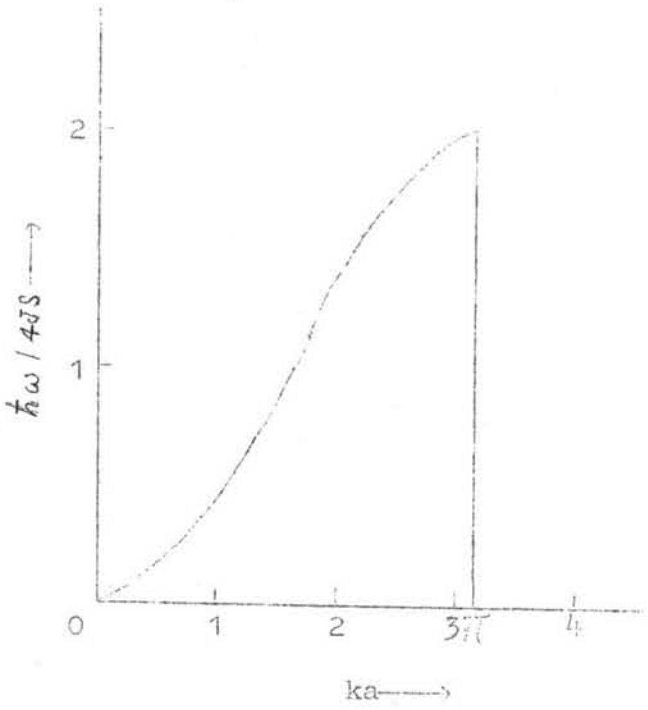

Fig. 4 Dispersion relation for spin waves in a ferromagnet in one dimension with nearest neighbour interactions.

eqs. (2.31) and (2.32) is shown in Fig. 4.

By virtue of the fact that quantum theory allows only integral values for the z component of the total spin quantum number, only a number of spin waves of wave vector $\underline{k}$ is possible, that is, the spin waves are quantized and the energy of quantization is

$$\epsilon_k = n_k \hbar \omega_k, \qquad (2.33)$$

where n_k is an integer equal to the number of magnons of wave vector $\underline{k}$. These are analogous to lattice vibrations or phonons therefore the magnons may be thought of as particles subject to Bose statistics. The spin wave theory predicts a decrease in spontaneous magnetization when the magnons are thermally excited, according to the relation

$$\frac{M(T)}{M(0)} = 1 - \frac{A}{2S}\left(\frac{T}{\Theta}\right)^{3/2}, \qquad (2.34)$$

where $\Theta = 2JS/k_B$, and A is a constant depending on the crystal structure.

The spin waves in an antiferromagnet are calculated by the analogous method to that used in the case of ferromagnetism. The results obtained are quite different from those for ferromagnetism. The dispersion relationship for an antiferromagnetic material is

$$\omega_k = \omega_e \left(1 - \gamma_k^2\right)^{1/2}, \qquad (2.35)$$

where

$$\omega_e = \omega_0 S = \left(\sum_j 2 J_{ij} \hbar\right) S \qquad (2.36a)$$

and

$$\gamma_k = \sum_j 2 \left|J_{ij}\right| e^{i\underset{\sim}{k}\cdot(\underset{\sim}{r}_j - \underset{\sim}{r}_i)} \Big/ \sum_j 2 \left|J_{ij}\right|. \qquad (2.36b)$$

For small $\underset{\sim}{k}$, the following relation holds:

$$\omega_k \propto \omega_e |\underset{\sim}{k}|. \qquad (2.37)$$

Kubo[20] showed by the spin wave theory that the parallel susceptibility of an antiferromagnet is given by

$$\chi_{//} = \frac{l N g^2 \mu_B^2 k_B^2}{z^3 J^3 S^3} T^2, \qquad (2.38)$$

where l is a constant depending on the type of the lattice, and that the perpendicular susceptibility is given by

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[20] R. Kubo, "The Spin-Wave Theory of Antiferromagnetics," Physical Review 87, 568 (1952).

$$\chi_{\perp} = \frac{N g^2 \mu_B^2}{4|J|z} \tag{2.39}$$

which is T independent as predicted by the molecular field approach.

For the case of ferrimagnets, a number of calculations has been made, both semiclassical and quantum mechanical, concerning the spin waves. Since ferrimagnetic materials posses a spontaneous magnetization, the dispersion relationship would be quadratic. The theories confirm that for the lowest branch, which is important at low temperatures, $\omega \propto k^2$. Moreover, Kaplan[21] developed the approximate theory of ferrimagnetic spin waves by taken into account only the exchange interactions between different sublattices and carried out the calculation to second order in the magnitude of wave vector $\underline{k}$. This theory shows four branches of the dispersion relation all of which are quadratic and two are identical in the classical limit to the ones obtained by H. Kaplan[22]. It then follows that at low temperatures the temperature dependence of the spontaneous magnetization has the same form of the $T^{3/2}$ law for the ferromagnetic

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[21] T.A. Kaplan, "Approximate Theory of Ferrimagnetic Spin Waves," Physical Review 109, 782 (1958).

[22] H. Kaplan, "A Spin-Wave Treatment of the Saturation Magnetization of Ferrites," Physical Review 86, 121 (1952).

case, namely,

$$\frac{M(T)}{M(0)} = 1 - A_{exp}T^{3/2}. \qquad (2.40)$$

Here A_{exp} depends on the exchange interaction between and within the sublattices and the number of nearest neighbours. It has been found experimentally[23] that $MnFe_2O_4$ does indeed follow the $T^{3/2}$ law up to $T/T_N \approx 0.7$ whereas for the spinels $NiFe_2O_4$, $CoFe_2O_4$ and Fe_3O_4, a better fit with a T^2 law is found.

However, the spin wave description is a good approximation only at low temperatures, but it is not valid for high temperatures where the interactions between magnons should be taken into account.

2.3 The Holstein-Primakoff Method

Holstein and Primakoff[24] have attacked on the problem of spin-wave interactions by considering the effect of applying large magnetic fields to a ferromagnetic system. The ground state Hamiltonian of the system is then

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[23] R. Pauthenet, Annals of Physics (Paris) 7, 710 (1952).

[24] T. Holstein and H. Primakoff, "Field Dependence of the Intrinsic Domain Magnetization of a Ferromagnet," Physical Review 58, 1098 (1940).

$$\mathcal{H} = -2\sum_{i,j} J_{ij}\, \underline{S}_i\cdot\underline{S}_j - g\mu_B H\sum_i S_i^z, \quad J_{ij} > 0. \quad (2.41)$$

In this method, the spin operators are written in the form of Boson operators as

$$S_i^+ = S_{ix}+iS_{iy} = a_i^*(2S)^{1/2}\left(1-\frac{n_i}{2S}\right)^{1/2}, \qquad (2.42a)$$

$$S_i^- = S_{ix}-iS_{iy} = (2S)^{1/2}\left(1-\frac{n_i}{2S}\right)^{1/2} a_i, \qquad (2.42b)$$

$$S_i^z = n_i - S, \qquad (2.42c)$$

where a_i and a_i^* are the lowering and raising operators subject to Bose statistics:

$$a_i^*|n\rangle = (n+1)^{1/2}|n+1\rangle, \quad a_i|n\rangle = n^{1/2}|n-1\rangle \qquad (2.43)$$

with the commutation relation

$$\left[a_i, a_j^*\right] = \delta_{ij}, \quad \left[a_i, a_j\right] = \left[a_i^*, a_j^*\right] = 0 \qquad (2.44)$$

and n_i is the Boson number operator:

$$n_i = a_i^* a_i \qquad (2.45)$$

having the eigenvalue n_i indicated in the state $|n_i\rangle$.

Using eqs. (2.42a)-(2.42c), the Hamiltonian (2.41) can be written as

$$\mathcal{H} = E_o - S \sum_{i,j} J_{ij}(a_i^* - a_j^*)\left(1 - \frac{n_i}{2S}\right)a_j + g\mu_B H \sum n_i, \quad (2.46)$$

where the constant E_o is the ground state energy defined by

$$E_o = - NzJS^2 - Ng\mu_B HS. \quad (2.47)$$

For travelling waves of wave vector $\underline{k}$, setting the canonical transformation:

$$a_i = \frac{1}{\sqrt{N}} \sum_{\underline{k}} e^{i\underline{k}.\underline{r}_i} a_{\underline{k}}, \quad a_i^* = \frac{1}{\sqrt{N}} \sum_{\underline{k}} e^{-i\underline{k}.\underline{r}_i} a_{\underline{k}}^* \quad (2.48)$$

with its inverse transformation

$$a_{\underline{k}} = \frac{1}{\sqrt{N}} \sum_{i} e^{-i\underline{k}.\underline{r}_i} a_i, \quad a_{\underline{k}}^* = \frac{1}{\sqrt{N}} \sum_{i} e^{i\underline{k}.\underline{r}_i} a_i^*, \quad (2.49)$$

where
$$a_{\underline{k}}^* a_{\underline{k}} = n_{\underline{k}}. \quad (2.50)$$

hence, the Hamiltonian in terms of $a_{\underline{k}}$'s is

$$\mathcal{H} = E_o - S \sum_{i,j} J_{ij} \sum_{\underline{k}_1\underline{k}_2} \frac{a_{\underline{k}_1}}{N} \left(e^{i\underline{k}_1\cdot\underline{r}_i} - e^{i\underline{k}_1\cdot\underline{r}_j} \right)$$

$$\times \left(1 - \frac{1}{2S} \sum_{\underline{k}_3\underline{k}_4} \frac{a^*_{\underline{k}_3}}{N} e^{i\underline{k}_3\cdot\underline{r}_i} \frac{a_{\underline{k}_4}}{N} e^{-i\underline{k}_4\cdot\underline{r}_i} \right) \frac{a_{\underline{k}_2}}{N} e^{-i\underline{k}_2\cdot\underline{r}_j}$$

$$+ g\mu_B H \sum_i \sum_{\underline{k},\underline{k}'} \frac{a^*_{\underline{k}}}{N} e^{i\underline{k}\cdot\underline{r}_i} \frac{a_{\underline{k}'}}{N} e^{-i\underline{k}'\cdot\underline{r}_i}. \qquad (2.51)$$

The Hamiltonian (2.51) can be written in terms of the occupation number in the form[25]

$$\mathcal{H} = E_o + \sum_{\underline{k}} A_{\underline{k}} a^*_{\underline{k}} a_{\underline{k}} + \sum_{\underline{k},\underline{k}'} \left(\frac{1}{2} B_{\underline{k}} a_{\underline{k}} a_{\underline{k}'} + \frac{1}{2} B^*_{\underline{k}} a^*_{\underline{k}} a^*_{\underline{k}'} \right), \qquad (2.52)$$

where $A_{\underline{k}}$ and $B_{\underline{k}}$ are some constants in terms of $\underline{k}$.

The above Hamiltonian can be diagonalized by introducing the quasi-particle operators defined by the Bogolubov transformation:

$$a_{\underline{k}} = (\cosh u_{\underline{k}}) a_{\underline{k}} + (\sinh u_{\underline{k}}) a^*_{\underline{k}'}, \qquad (2.53a)$$

$$a^*_{\underline{k}} = (\cosh u_{\underline{k}}) a^*_{\underline{k}} + (\sinh u_{\underline{k}}) a_{\underline{k}'} \qquad (2.53b)$$

with $u_{\underline{k}} = u_{\underline{k}'} = u^*_{\underline{k}}$.

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[25] Reference 24, p. 1103.

Using the transformation in eqs. (2.53a) and (2.53b), the Hamiltonian which is diagonal in the occupation number representation can be obtained and is called $\mathcal{H}_D$. It is convenient to denote the Fourier transform of J_{ij} by $J(\underline{k})$, defined as

$$J(\underline{k}) = \frac{1}{2N} \sum_{i,j} e^{i\underline{k}\cdot(\underline{r}_i - \underline{r}_j)} J_{ij}. \qquad (2.54)$$

And in this manner one obtains

$$\mathcal{H}_D = E_o + \sum_{\underline{k}} \left[2SJ(0) - 2SJ(\underline{k}) + g\mu_B H \, n_{\underline{k}} \right]$$

$$- \frac{1}{N} \sum_{\underline{k},\underline{k}'} \left[J(0) + J(\underline{k}-\underline{k}') - J(\underline{k}) - J(\underline{k}') \right] n_{\underline{k}} n_{\underline{k}'}. \quad (2.55)$$

The study of $\mathcal{H}_D$ gives an important result of the nonlinear magnon energies $\epsilon(\underline{k})$, such as

$$\epsilon(\underline{k}) = \hbar\omega(\underline{k}) - \frac{1}{NS} \sum_{\underline{k}'} \left[\hbar\omega(\underline{k}) + \hbar\omega(\underline{k}') - \hbar\omega(\underline{k}-\underline{k}') - \hbar\omega(\underline{0}) \right] n_{\underline{k}'}.$$

$$(2.56)$$

For nearest neighbour interactions,

$$\overline{\epsilon(\underline{k})} \leq \overline{h\omega(\underline{k})}. \qquad (2.57)$$

So it is clear that all magnon energies can be reduced by increasing the occupation number $n_{\underset{\sim}{k}}$, as in a discussion of thermal effects. However, the inequality of eq. (2.57) is reversed in nearest-neighbour ferrimagnets which have long-range interactions.

The diagonal part of the Hamiltonian gives a theory of noninteracting spin waves, identical with the linear approximation of Bloch. The nondiagonal part represents the effect of spin wave interactions and adds to the diagonal part by perturbation theory. However, the first successful analysis of the spin wave interactions was made by Dyson[26] who showed that the spontaneous magnetization is approximated by

$$\frac{M(T)}{M(0)} = 1 - \frac{A}{2S}\left(\frac{T}{\theta}\right)^{3/2} - \frac{A_1}{S}\left(\frac{T}{\theta}\right)^{7/2} + \frac{A_3}{S}\left(\frac{T}{\theta}\right)^{4} . \qquad (2.58)$$

The Holstein-Primakoff formalism has also been applied to antiferro- and ferrimagnetism[27]. The results obtained yield two branches of magnon energies with the lower branch, at long wavelength, quadratic for ferrimagnets and becoming approximately linear for antiferromagnets.

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[26] F.J. Dyson, "General Theory of Spin-Wave Interaction," Physical Review 102, 1217 (1956).

[27] T. Nagamura and M. Bloch, "Temperature Dependence of the Exchange Stiffness in Ferrimagnets," Physical Review 132, 2528 (1963).

2.4 The Green's Function Method

The double-time temperature dependent Green's function has been used to study various spin systems. The definition and some properties of the Green's function are mentioned in Appendix B according to Tyablikov[28]. The Fourier transform of the Green's function involving the operators A and B is a function of E, and is denoted by $\langle\langle A;B\rangle\rangle_E$. It satisfies the equation of motion:

$$E\langle\langle A;B\rangle\rangle_E = (1/2\pi)\langle[A,B]\rangle + \langle\langle[A,\mathcal{H}];B\rangle\rangle_E, \qquad (2.59)$$

where the square bracket denotes a commutator and the single angular bracket $\langle\ldots.\rangle$ indicates an average with respect to the canonical ensemble. From the analytical properties of the Green's function, the correlation function $\langle B(t')A(t)\rangle$ can be obtained from the relation[29]

$$\langle B(t')A(t)\rangle = \lim_{\epsilon\to 0^+} i\int_{-\infty}^{+\infty} \frac{\langle\langle A;B\rangle\rangle_{E=\hbar\omega+i\epsilon} - \langle\langle A;B\rangle\rangle_{E=\hbar\omega-i\epsilon}}{\exp(\hbar\omega/kt)-1} \times e^{-i\omega(t-t')}\,d\omega. \qquad (2.60)$$

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[28] S.V. Tyablikov, *Methods in Quantum Theory of Magnetism* (New York: Plenum Press, Inc., 1967), Ch. VII, pp. 205-240.

[29] *Ibid.*, pp. 210-215.

Due to the appearance of higher order Green's functions of eq. (2.59), various decoupling approximations must be employed to obtain the solution of the chain of equations for the Green's functions.

The singularities of the Green's functions determine the energy of elementary excitations of the system and the correlation functions give the values of critical temperatures. Other macroscopic as well as microscopic quantities can also be obtained. These results are applicable to a wide range of temperatures.

The technique of double-time temperature dependent Green's function was used by Bogolyubov and Tyablikov[30] to analyze the Heisenberg ferromagnet with spin 1/2. Tahir-Kheli and ter Haar[31] have made the extension of the theory to higher spin. Here, A and B are spin operators associated with different lattice sites. The essential approximation in the methods of Tyablikov and of Tahir-Kheli and ter Haar consists of the decoupling approximation such as,

$$\langle\langle A_l^z A_{l'}^+;B\rangle\rangle_E \xrightarrow[l\neq l']{} \langle A^z\rangle\langle\langle A_l^+;B\rangle\rangle_E \qquad (2.61)$$

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[30] N.N. Bogolyubov and S.V. Tyablikov, <u>Soviet Physics-Doklady</u> <u>4</u>, 604 (1959).

[31] R. Tahir-Kheli and D. ter Haar, "Use of Green's Functions in the Theory of Ferromagnetism. I. General Discussion of the Spin-S Case," <u>Physical Review</u> <u>127</u>, 88 (1962).

in which the fluctuations of A_1^z are ignored and replaced this operator by its average value. The replacement (2.61) is called the Tyablikov decoupling approximation or the random phase approximation (RPA).

The results of this approximation can almost explain the behaviors in the high-temperature region as well as in the range of temperatures near the transition temperature, but they are not reliable at low temperatures. Callen[32] developed a self-consistency decoupling approximation which approximately valids through the entire temperature range. This will be described in Chapter 3. Other decoupling schemes[33,34] have also been made for many purposes with successful results. In addition, this

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[32] H.B. Callen, "Green Function Theory of Ferromagnetism," *Physical Review* 130, 890 (1963).

[33] M.E. Lines, "Sensitivity of Curie Temperature to Crystal-Field Anisotropy. I. Theory," *Physical Review* 156, 534 (1967).

[34] I.P. Fittipaldi and M.D. Coutinho, Jr., "Spin-1/2 modified Callen decoupling procedures in the Green's-function theory of ferromagnetism," *Physical Review B* 10, 4808 (1974).

method has been applied to antiferromagnetism[35-39] as well as ferrimagnetism[40] using different decoupling schemes.

2.5 Comparison of the Different Aproaches

We shall now briefly compare the theories. All effective field methods are high temperature approximations and are unreliable at low temperatures. The Oguchi's method predicts the transition temperature lower than that given by the Weiss molecular field in good agreement with the results obtained by the B-P-W method. The B-P-W method as well as the Oguchi's method

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[35] F.B. Anderson and H.B. Callen, "Statistical Mechanics and Field Induced Phase Transitions of the Heisenberg Antiferromagnet," Physical Review 136, A1068 (1964).

[36] J.H. Barry, "Statistical-Mechanical Theory of Resonance Susceptibility in Heisenberg Antiferromagnetism," Physical Review 174, 531 (1968).

[37] D.K. Ghosh, "Green's-Function Theory of an Antiferromagnet," Physical Review B 8, 392 (1973).

[38] K. Fujii, "Heisenberg Antiferromagnet Using the Green's Function Theory," Progress of Theoretical Physics 51, 1283 (1974).

[39] R.H. Swendsen, "Modified Callen decoupling in the Green's-function theory of Heisenberg antiferromagnets," Physical Review B 11, 1935 (1975).

[40] D.A. Yablonskii, "Quantum Theory of an Isotropic Two-Sublattice Ferrimagnet in Applied Magnetic Fields," Soviet Physics-Solid State 14, 2468 (1973).

can account for the behaviors experimentally observed in the vicinity of the transition temperature. The spin wave theory and the nonlinear theory of Holstein-Primakoff account for the physical facts at temperatures well below the transition temperature but fail badly for higher temperature regions due to the appearance of spin wave interactions. The only method which is valid throughout the entire temperature range is the Green's function method. It predicts the transition temperature lower than that given by the Weiss molecular field approach and give the results below this temperature in accordance with the spin wave theory, whereas at high temperatures, the results obtained are similar to those of the effective field theories.