

**Spin Waves in Ferromagnetic and Antiferromagnetic
Metals**

A Thesis

**presented for the degree of Doctor of Philosophy of the
University of London and the Diploma of Imperial College**

by

Bernard Eric Armin Fischer

Mathematics Department,

Imperial College,

London S.W.7

Abstract

A general method of calculating spin wave energies to the lowest power in the wave vector q is described, which is applicable to both metallic and non-metallic magnets in the absence of anisotropy, though particular attention is directed towards the metals of the 3d transition series. Though the approach uses Green's functions, neither a diagram technique nor the decoupling of equations of motion is used. Instead of these approximations, the method assumes a form for the generalised frequency dependent susceptibility $\chi(q, \omega)$, expands in powers of q/ω , and compares the resulting equation with the equations of motion of $\chi(q, \omega)$ expanded in powers of q/ω . In this way an exact formula for D , where Dq^2 is the energy of ferromagnetic spin waves, and a reliable formula for c , where cq is the energy of antiferromagnetic spin waves, are calculated.

Earlier rigorous results on the contribution of spin wave interactions to D in the Heisenberg model are obtained more quickly and parallel work on the one band, short-range interaction, itinerant electron model of a strong ferromagnet is for the first time put on a rigorous footing. An estimate is made of the strength of spin wave interactions in nickel.

Similarly the linear theory of spin waves in the Heisenberg antiferromagnet and first order corrections are

easily derived. A new formula for c in the itinerant electron model is obtained, which disagrees with and goes beyond, previous calculations. The Heisenberg result is found as a limiting case. The shortcomings of other calculations and the problem of the interaction of spin waves with single particle excitations are described. c is then calculated for chromium, the only weakly antiferromagnetic pure metal, and for antiferromagnetic alloys containing local moments. In the latter case our formula for c is shown to be an alternative to using the Ruderman-Kittel interaction. In the last chapter a description of strongly antiferromagnetic -manganese is outlined within the itinerant electron model.

Acknowledgements

I am indebted to my supervisor, Dr D M Edwards, for his help and encouragement, and for initiating the joint work on spin wave interactions in ferromagnets. Useful discussions were had with Dr R D Lowde and his colleagues at A.E.R.E. Harwell in connection with measurements of spin wave energies in metallic antiferromagnets.

Contents

Chapter 1	The Itinerant Electron Model	
I	Exchange in Metals	page 1
II	Green's Functions, Susceptibility, Neutron Scattering	10
III	Magnons	16
Chapter 2	Spin Wave Interactions	
I	Introduction	20
II	General Formula for the Spin Wave Energy	22
III	The Heisenberg Ferromagnet	27
IV	The Itinerant Electron Model	38
Chapter 3	The Heisenberg Antiferromagnet	
I	Exact Results	47
II	Linear Theory	49
Chapter 4	Antiferromagnetism in the Band Model	
I	Oscillatory Spin Polarisation in the Free Electron Gas	58
II	The Short-range One-band Model	61
III	Single Particle States and the Ground State	65
IV	Susceptibility and Spin Waves	70

V Spin Waves with Fermi Level not in the Gap	page 76
VI Appendix	84
Chapter 5 Spin Waves in Chromium	
I The Two Band Approximation	87
II The Spin Density State of Chromium	88
III The Ground State	94
IV Spin Waves	103
Chapter 6 Spin Waves in a System of Itinerant Electrons and Local Moments	
I Local Moments in Alloys and Rare Earths	108
II The Ground State	111
III Spin Wave Energy	120
Chapter 7 Spin Correlations in Gamma Manganese	
I The Magnetisation in Gamma Manganese	128
II The Strong Correlation Model	130
References	i-ix

Chapter 1

The Itinerant Electron Model

1 Exchange in Metals

The energy of a group of electrons depends on their spin orientations, even in the absence of any spin dependent terms in the Hamiltonian of the electrons. This purely quantum mechanical effect, arising from the exclusion principle, gives rise to exchange energies which determine the properties of most magnetic materials. Various sorts of exchange coupling are known e.g. direct exchange, Ruderman-Kittel, itinerant exchange and superexchange. The simplest to handle mathematically and the earliest one investigated is direct exchange (Herring 1966a) which will now be described.

For a system with one available orbital per site, N sites, and one electron per site, the electron states can be put into correspondence with the states of a spin space defined by spin operators $\underline{S}_i$ on every site $\underline{R}_i$. Then the Hamiltonian H , approximating the low energy eigenstates of the system, has the following simple form in terms of the spin space operators (Dirac 1929)

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

where J_{ij} is the difference in energy between the configuration in which the spins on R_i and R_j are antiparallel and that in which they are parallel. (1.1) is known as the Heisenberg Hamiltonian and is the simplest expression involving the spin operators which has rotational symmetry. (1.1) does not only apply to direct exchange; J_{ij} could be defined to correspond to superexchange or Ruderman-Kittel exchange coupling.

This Hamiltonian does not apply to the itinerant electron model of a metal in which electrons are in Bloch states labelled by a wave vector $\underline{k}$ (Bloch 1929). The electrons do not remain on one atom long enough for $\underline{R}_i$ to be defined. How the exchange coupling arises for itinerant electrons is discussed in this section. A much more complete and detailed account is given in the review by Herring (1966b).

In order to determine the energy $\epsilon(\underline{k})$ of electrons with wave vector $\underline{k}$, in a metal, a band structure calculation is done (Callaway 1964). The elements of the second half of the 3d transition series, chromium, manganese, iron, cobalt and nickel form the iron group of magnetic metals. It is with these metals that we shall be mainly concerned. For them, one has as the basis for the band structure, the

wave functions corresponding to the five degenerate 3d atomic wave functions of different symmetry, plus atomic s and p functions.

In general it is found (Fletcher 1952, Yamashita et al 1963, Hubbard and Dalton 1968 for nickel) that the density of states has a narrow peaky portion of high density, corresponding to predominantly atomic d functions, superimposed on a wide flat portion of low density, corresponding to mainly s and p functions. When one includes the Coulomb interaction between the electrons in the Hamiltonian it is simplest to assume that hybridisation between the d, s, and p bands is small so that the Coulomb interaction has distinguishable terms corresponding to d-d, s-d and s-s interactions. The s bands are free electron-like and the interacting free electron gas is unlikely to be magnetic (Herring 1966b). So the s bands are usually neglected when looking for the cause of the magnetism. In the d band the electron charge density is concentrated near the nuclei of the solid, so the Coulomb interaction is strong. This localisation in the d band is also the reason for the band being narrow.

The easiest way of including interactions is to apply the Hartree-Fock approximation so that one remains with a one electron problem. The Hartree-Fock approximation

introduces a self-consistent exchange field into the one electron Hamiltonian. This field is the molecular field of Stoner Theory (Slater 1947, Wohlfarth 1953) and splits the up and down spin bands by an energy ΔE , called the exchange splitting.

The interaction, in a basis of Bloch functions of tight-binding form, in the Hartree-Fock approximation, will include integrals between states with parallel spin of the type

$$-J_{\underline{m}\underline{k},\underline{n}\underline{k}'} = - \iint \psi_{\underline{m}\underline{k}}^*(\underline{r}_1) \psi_{\underline{n}\underline{k}'}^*(\underline{r}_2) \frac{e^2}{r_{12}} \psi_{\underline{m}\underline{k}}(\underline{r}_2) \psi_{\underline{n}\underline{k}'}(\underline{r}_1) d\underline{r}_1 d\underline{r}_2 \quad (1.2)$$

$$\text{where } \psi_{\underline{m}\underline{k}\sigma} = \frac{1}{\sqrt{N}} \sum_{i,\mu} e^{i\underline{k} \cdot \underline{R}_i} a_{\underline{k}\mu}^{(m)} w_{\mu\sigma}(\underline{r} - \underline{R}_i) \quad (1.3)$$

$$= \frac{1}{\sqrt{N}} \sum_i e^{i\underline{k} \cdot \underline{R}_i} w_{\underline{k}\underline{m}}(\underline{r} - \underline{R}_i) \quad (1.4)$$

where $w_{\mu}(\underline{r} - \underline{R}_i)$ are atomic d functions with μ denoting their symmetry. The largest terms in (1.2), written in a basis of $\underline{k}$ dependent atomic exchange integrals using (1.4), are

$$I_{\underline{k}\underline{m}} = \iint |w_{\underline{k}\underline{m}}(\underline{r}_1)|^2 \frac{e^2}{r_{12}} |w_{\underline{k}\underline{m}}(\underline{r}_2)|^2 d\underline{r}_1 d\underline{r}_2 \quad (1.5a)$$

whose value is of order 20 ev,

$$J_{\underline{k}\underline{m},\underline{k}'\underline{n}} = \iint w_{\underline{k}\underline{m}}^*(\underline{r}_1) w_{\underline{k}'\underline{n}}^*(\underline{r}_2) \frac{e^2}{r_{12}} w_{\underline{k}\underline{m}}(\underline{r}_2) w_{\underline{k}'\underline{n}}(\underline{r}_1) d\underline{r}_1 d\underline{r}_2 \quad (1.5b)$$

$$\text{and } I_{\underline{k}m}(R) = \iint |w_{\underline{k}m}(r_1)|^2 \frac{e^2}{r_{12}} |w_{\underline{k}m}(r_2-R)|^2 dr_1 dr_2 \quad (1.5c)$$

of energy 1 eV and 5 eV respectively, where R is a nearest neighbour distance (Freeman and Watson 1961, Herring 1966b). $I_{\underline{k}m}(R)$ would be much smaller if the screened Coulomb interaction were used to evaluate it.

It should be pointed out that the Hartree-Fock approximation is different if one started with an atomic basis for the interaction. When we later wish to approximate the interaction, only terms representing the interactions between electrons on the same site will be retained. The interaction energies involved are I and J , averages of $I_{\underline{k}m}$ and $J_{\underline{k}m, \underline{k}'n}$ respectively. I is the intra-atomic Coulomb integral representing the interaction between electrons on the same site, in the same orbital, with opposite spin and J is the Hund's Rule term, or the intra-atomic exchange interaction between electrons on the same site, in different orbitals, with parallel spin. The other intra-atomic integral U , is the Coulomb integral between orbitals of different symmetry and is independent of the spin of the electrons.

In order to estimate realistically the magnitude of the exchange splitting ΔE between up and down spin bands,

Slater (1936) used

$$\Delta E = J_{\text{eff}} \Delta n \quad (1.6)$$

where Δn is the difference in the number of up and down spin holes per site and J_{eff} is a suitable average of J_{k_m, k'_n} .

Van Vleck (1953) pointed out that J_{eff} should include some weighting from I_{k_m} , such as

$$J_{\text{eff}} = \frac{1}{5}(4J + I) \quad (1.7)$$

However as I is of order 20 eV and Δn equals 0.6 for nickel, the resulting value for ΔE is much too large, since a reasonable value for ΔE for nickel is 0.4 eV.

This argument, of course neglects the effect of the s electrons which will move to screen I and J and may reduce J_{eff} to the right order. From the atomic spectra Van Vleck argued that

$$E(d^{10}s^1) + E(d^8s^1) - 2E(d^9s^1) = 7.6 \text{ eV} \simeq I \quad (1.8)$$

is the expression for the interaction energy between two holes. $E(d^n s^m)$ is the energy of the atomic configuration $d^n s^m$ containing n 3d electrons and m 4s electrons, so that $d^8 s^1$ has two d holes, $d^9 s^1$ has one d hole and $d^{10} s^1$ has no d holes. Alternatively (Herring 1966b) one can compare

$$E(d^8 s^2) + E(d^{10}) - 2E(d^9 s^1) = 2 \text{ ev} \quad (1.9)$$

In any case it is not clear how relevant these arguments are to a real metal where the s electrons form a broad band.

To summarise there are two viewpoints. Either I is of order 2 ev, which is less than the bandwidth, and the screening by the s electrons is good, or I is of order 8 ev, greater than the bandwidth, and the screening is bad. Of the metals, nickel, manganese and chromium, which we will be discussing in detail, for nickel both approaches lead to the same conclusion. In nickel with about 0.6 d holes, if I is small, there is little correlation and there are often two holes on the same site. If I is large the holes will avoid each other and the d band will be well correlated. To include the effect of correlation one must go beyond the Hartree-Fock Theory. This was done by Kanamori (1963) using an approximation which was appropriate because of the low density of holes. He obtained a new value for I , I_{eff} , which is less than the bandwidth and roughly the same size as I in the weak correlation model.

The Stoner criterion for ferromagnetism within the molecular field model is

$$I_{\text{eff}} N(\epsilon_F) > 1 \quad (1.10)$$

This condition is effectively the same in both the weak and the strong correlation models, but is strict. For (1.10) to be satisfied one must assume that the density of states at the paramagnetic Fermi energy, $N(\epsilon_F)$, is significantly greater than the mean density of states of the band. In nickel the high density of states near ϵ_F comes mainly from one sub-band, at the top of the d band, which is flat in XW direction. This is the justification for considering one-band models (Lowde and Windsor 1970). There are in fact, two hole pockets around X and degenerate at X. The 3d band of nickel is nearly full so that one can picture the band structure as being asymptotically parabolic, with no down spin holes in the ferromagnetic state.

Bandwidths get wider and interaction potentials get weaker as one goes down the 3d transition series from nickel to chromium. Thus for chromium, which has weak magnetic properties considering it has 4 to 5 d holes per atom, the weak correlation model is usually adopted. Vanadium, the element before chromium in the periodic table, is non-magnetic.

Kanamori's theory applies when there is a low density of holes and is correct, like Stoner Theory, in the band limit when $I \rightarrow 0$, but is also correct when $I \rightarrow \infty$. When $I \rightarrow \infty$ the atomic limit is correct no matter what the density of holes. The metal then looks like a lattice of

isolated atoms. Hubbard (1963, 1964a, 1964b, 1966) has worked out a theory, which is correct in the atomic limit. Atomic and bandlike characteristics arise in the following way. For a partially full band the spin on each atom fluctuates with characteristic time $\hbar/b$, where b is a typical hopping energy between sites. However, correlation keeps the spins on one atom aligned so that there is a preponderance of up spins on one atom. For strong correlations the spin becomes associated with the atoms rather than the electrons. Hubbard (1964c) applied his theory to nickel only and, like Kanamori, still got the same sort of exchange splitting as the weak correlation problem, even though the interactions between electrons on the same site are very strong.

For cobalt, iron and manganese there are more than one, two and three holes per atom respectively. Hubbard's theory for these elements is complicated, but Edwards (1970) has suggested a phenomenological theory which retains the essential features of the Hubbard theory. Then the essential feature for a metal with $(r+n)N$ holes, where r is an integer and $n < 1$, is that a band of rN holes is split off from the partially filled bands. These ideas must be generalised when one discusses manganese, which is antiferromagnetic. The alternative approach to manganese, based

on the Hartree-Fock approximation, requires a very large antiferromagnetic exchange field of order 3 to 4 eV (Lowde et al 1970). The use of such a large exchange field seems unjustified in a molecular field approximation. In a later chapter we shall discuss manganese in more detail on the basis of Hubbard's and Edwards' theories.

II Green's Functions, Susceptibility, Neutron Scattering.

In this section the method of Green's functions, used throughout this thesis, will be briefly described. A full account of the technique, which is used widely in magnetism, is given by Zubarev (1960). For example Hubbard in his papers (1963, 1964a,b,c, 1966) used retarded Green's functions to describe the propagation of electrons. These Green's functions, from which the properties of the system can be determined, can be solved from their equations of motion. There is another kind of Green's function, the causal Green's function (Martin and Schwinger 1959, Baym and Mermin 1961) which is useful when one wants to solve problems by means of perturbation theory.

The retarded (+) and advanced (-) Green's functions are defined by

$$\langle\langle A(t); B(t') \rangle\rangle^{\pm} = \mp \frac{i}{\hbar} \theta\{\pm(t-t')\} \langle [A(t), B(t')]_{\eta} \rangle \quad (1.11)$$

(11)

where $A(t) = e^{iHt/\hbar} A e^{-iHt/\hbar}$ and $B(t')$ are two operators,

with $[A, B]_{\eta} = AB + \eta BA$, $\eta = \pm 1$

depending on which ever choice for η is more convenient (usually $\eta = +1$ if A and B are fermions, $\eta = -1$ if A and B are bosons), $\theta(x)$ is the step function

$\theta(x) = 1$ if $x > 0$, $= 0$ otherwise,

and the thermal average $\langle X \rangle$ is defined by

$$\langle X \rangle = \frac{1}{Z} \text{Tr} \{ e^{-\beta H} X \}, \quad Z = \text{Tr} \{ e^{-\beta H} \}$$

Since $\langle\langle A(t); B(t') \rangle\rangle$ is a function of $t-t'$ one can define the Fourier transform

$$\langle\langle A, B \rangle\rangle_{\omega}^{\pm} = \int_{-\infty}^{\infty} \langle\langle A(t); B(0) \rangle\rangle^{\pm} e^{i\omega t} dt \quad (1.12)$$

Then it can be shown (Zubarev 1960) that $\langle\langle A, B \rangle\rangle_{\omega}^{+}$ is analytic in the upper half-plane of the complex ω plane,

$\text{Im} \omega > 0$, and $\langle\langle A, B \rangle\rangle_{\omega}^{-}$ is analytic in the lower half-plane, $\text{Im} \omega < 0$. The discontinuity across the real axis is related to the correlation function $\langle A(t)B(0) \rangle$ by

$$\langle A(t)B(0) \rangle = \frac{i\hbar}{2\pi} \int_{-\infty}^{\infty} \lim_{\epsilon \rightarrow 0^+} \left(\frac{\langle\langle A, B \rangle\rangle_{\omega+i\epsilon} - \langle\langle A, B \rangle\rangle_{\omega-i\epsilon}}{1 + \eta e^{-\beta \hbar \omega}} \right) e^{-i\omega t} d\omega$$

$$\text{and } \langle\langle A; B \rangle\rangle_{\omega-i\epsilon} = (\langle\langle B; A \rangle\rangle_{\omega+i\epsilon})^* \quad (1.13)$$

$\langle\langle A; B \rangle\rangle_{\omega}^{\pm}$ also satisfy the equation of motion

$$\hbar\omega \langle\langle A; B \rangle\rangle_{\omega}^{\pm} = \langle\langle [A, H]; B \rangle\rangle_{\omega}^{\pm} + \langle [A, B]_{\eta} \rangle \quad (1.14)$$

The causal Green's function is defined by

$$G(t, t') = -\frac{i}{\hbar} \langle T \{ A(t) B(t') \} \rangle \quad (1.15)$$

$$\text{where } T \{ A(t) B(t') \} = \theta(t-t') A(t) B(t') + \eta \theta(t'-t) B(t') A(t)$$

with $\eta = 1$ if A and B are Bose operators and $\eta = -1$ if A and B are Fermi operators.

To evaluate the causal Green's function one must evaluate the imaginary time Green's function

$$\mathcal{G}(\tau) = -\frac{i}{\hbar} \langle T \{ A(\tau) B(0) \} \rangle \quad (1.16)$$

where $A(\tau) = e^{\tau H/\hbar} A e^{-\tau H/\hbar}$. For a Hamiltonian $H = H_0 + H_1$, where H_1 is a perturbation, one can write $\mathcal{G}(\tau)$ in the following form, which shows how $\mathcal{G}(\tau)$ can be written as a series of products of imaginary time Green's functions dependent on H_0 alone:

$$G(z) = \frac{-i}{\hbar} \frac{\langle T \{ \exp(-\int_0^{\hbar\beta} H_1(z') dz'/\hbar) A(z) B(0) \} \rangle_0}{\langle T \{ \exp(-\int_0^{\hbar\beta} H_1(z') dz'/\hbar) \} \rangle_0} \quad (1.17)$$

where $\langle X \rangle_0 = \frac{1}{Z_0} \text{Tr} \{ e^{-\beta H_0} X \}$, $Z_0 = \text{Tr} \{ e^{-\beta H_0} \}$

$G(z)$ is defined in the range $0 < |z| \leq \hbar\beta$. So $G(z)$ can be written in a Fourier series with Fourier components

$$G(\omega_n) = \frac{1}{2} \int_{-\hbar\beta}^{\hbar\beta} e^{i\omega_n z} G(z) dz, \quad \omega_n = \frac{\pi n}{\beta\hbar} \quad (1.18)$$

Then

$$G_{\text{per}}(z) = \frac{1}{\hbar\beta} \sum_{n=-\infty}^{\infty} e^{-i\omega_n z} G(\omega_n) \quad (1.19)$$

is defined in the range $-\infty < z < \infty$.

$G(\omega_n)$ is related to the retarded and advanced Green's functions by (Martin and Schwinger 1961)

$$G(\omega_n) = \langle\langle A; B \rangle\rangle_{i\omega_n}^+ \quad \text{for } \omega_n > 0, \quad (1.20)$$

$$G(\omega_n) = \langle\langle A; B \rangle\rangle_{i\omega_n}^- \quad \text{for } \omega_n < 0. \quad (1.21)$$

Thus the causal Green's functions are obtained directly from the retarded and advanced Green's functions, and the analytic continuation of the causal Green's functions

determines the retarded and advanced Green's functions.

In magnetism Green's functions are directly related to two physical quantities, the susceptibility and the differential cross-section for magnetic scattering of neutrons (Zubarev 1960, Izuyama, Kim and Kubo 1965). The interaction energy between an oscillating magnetic field

$$\frac{1}{2} H_{\alpha} e^{-i(\underline{q} \cdot \underline{r}' + \omega t')} e^{\epsilon t'} + \text{complex conjugate}$$

and the spin density $S_{\alpha}(\underline{r}')$ of a system of electrons is

$$-g\mu_B \frac{1}{2} H_{\alpha} \int_V S_{\alpha}(\underline{r}') e^{-i(\underline{q} \cdot \underline{r}' + \omega t')} e^{\epsilon t'} d\underline{r}' + \text{complex conjugate} \quad (1.22)$$

where the integration is over the volume V of the whole crystal. The response of a point $\underline{r}$ of the spin density, at time t , is

$$\begin{aligned} \delta S_{\beta}(\underline{r}, t) &= -(g\mu_B)^2 \text{Re} \left[\langle\langle S_{\beta}(\underline{r}); S_{\alpha}(-\underline{q}) \rangle\rangle_{\omega+i\epsilon} H_{\alpha} e^{-i\omega t} e^{\epsilon t} \right] \\ &= -\left(\frac{g\mu_B}{V}\right)^2 \text{Re} \left[\sum_{\underline{k}} \langle\langle S_{\beta}(\underline{k}+\underline{q}); S_{\alpha}(-\underline{q}) \rangle\rangle_{\omega+i\epsilon} H_{\alpha} e^{-i[(\underline{q}+\underline{k}) \cdot \underline{r} + \omega t]} e^{\epsilon t} \right] \quad (1.23) \end{aligned}$$

where the summation is over reciprocal lattice vectors $\underline{k}$,

and

$$S_{\alpha}(\underline{q}, \omega) = \int_{-\infty}^{\infty} \int_V S_{\alpha}(\underline{r}) e^{i(\underline{q} \cdot \underline{r} + \omega t)} d\underline{r} dt$$

Then

$$S_{\beta}(\underline{k}+\underline{q}, \omega) = \chi_{\beta\alpha}(\underline{k}+\underline{q}, \underline{q}, \omega) H_{\alpha} \quad (1.24)$$

(15)

where we define the dynamical spin susceptibility as

$$\chi_{\beta\alpha}(\underline{k}+\underline{q}, \underline{q}, \omega) = -\frac{(g\mu_B)^2}{V} \ll S_{\beta}(\underline{k}+\underline{q}); S_{\alpha}(-\underline{q}) \gg_{\omega+i\varepsilon} \quad (1.25)$$

The differential neutron cross-section is (Lowde and Windsor 1970)

$$\frac{\partial^2 \sigma}{\partial \Omega \partial \omega} = \left(\frac{e\hbar}{mc} \right)^2 \frac{k'}{k} \sum_{\alpha, \beta} (\delta_{\alpha\beta} - \hat{q}_{\alpha} \hat{q}_{\beta}) M^{\alpha\beta}(\underline{q}, \omega) \quad (1.26)$$

where $\underline{k}$ is the wave vector of the incident neutrons, $\underline{k}'$ is the wave vector of the scattered neutrons, $\underline{k}' = \underline{k} + \underline{q}$, $\hat{q}$ is the unit vector $\underline{q}/q$, $\hbar\omega = \frac{\hbar^2 k'^2}{m} - \frac{\hbar^2 k^2}{m}$ and $M^{\alpha\beta}(\underline{q}, \omega)$ is the correlation function of the magnetisation, which equals

$$\left(\frac{e\hbar}{mc} \right)^2 S^{\alpha\beta}(\underline{q}, \omega)$$

when only the spin magnetisation of the electrons is important in scattering the neutrons, where

$$S^{\alpha\beta}(\underline{q}, \omega) = \frac{1}{2\pi} \int_V d\underline{r} \int_V d\underline{r}' \int_{-\infty}^{\infty} dt e^{i(\underline{q} \cdot \underline{r} + \omega t)} \langle S_{\alpha}(\underline{r}+\underline{r}', t) S_{\beta}(\underline{r}') \rangle \quad (1.27)$$

$$= \frac{1}{2\pi} \int_{-\infty}^{\infty} dt e^{i\omega t} \langle S_{\alpha}(\underline{q}, t) S_{\beta}(-\underline{q}) \rangle \quad (1.28)$$

$S^{\alpha\beta}(\underline{q}, \omega)$ is related to the imaginary part of the susceptibility by (Izuyama, Kilm and Kubo 1968)

$$S^{\alpha\beta}_{\text{sym}}(\underline{q}, \omega) = -\frac{\hbar}{\pi} (1 - e^{-\beta\hbar\omega})^{-1} \text{Im} \ll S_{\alpha}(\underline{q}), S_{\beta}(-\underline{q}) \gg_{\omega}^{\text{sym}} \quad (1.29)$$

(16)

$$= \frac{\hbar}{\pi} (1 - e^{-\beta \hbar \omega})^{-1} \frac{V}{(g/\mu_B)^2} \text{Im} \chi_{\alpha\beta}^{\text{symm}}(\underline{q}, \underline{q}, \omega) \quad (1.30)$$

from (1.13) and (1.25), where symm. denotes a function's symmetric part.

One can therefore see the importance of calculating the Green's function $\ll S_{\alpha}(\underline{k} + \underline{q}); S_{\beta}(-\underline{q}) \gg_{\omega + i\varepsilon}$.

In Hubbard's papers the single particle Green's functions, for which $A = c_i$, $B = c_j^+$ in (1.11), were used.

III Magnons

The immediate consequences of the Heisenberg Hamiltonian $-J \sum_{\langle ij \rangle} \underline{S}_i \cdot \underline{S}_j$, with $J > 0$, are that the ground state is the state $|0\rangle$, with total spin Ns , and there are exact eigenstates $|q\rangle$, of energy

$$\hbar\omega = [J(0) - J(q)]s \simeq Dq^2 + O(q^4) \quad (1.31)$$

with total spin $(N-1)s$ defined by

$$|q\rangle = \frac{1}{\sqrt{2Ns}} S_q^- |0\rangle \quad (1.32)$$

where $S_q^- = \sum_i e^{iq \cdot R_i} S_i^-$. These excited states,

$|\mathbf{q}\rangle$, are called spin waves.

Herring and Kittel (1951) using a phenomenological semi-classical approach, which is independent of any detailed model, argued that these results (1.31), (1.32) may apply outside the range of validity of the Heisenberg model. In their method the Bloch Wall stiffness A , defined for a cubic crystal in terms of the energy increase δE associated with transverse nonuniformity of the magnetisation $M(\mathbf{r})$ by

$$\delta E = A \left[\left(\frac{\partial M}{\partial x} \right)^2 + \left(\frac{\partial M}{\partial y} \right)^2 + \left(\frac{\partial M}{\partial z} \right)^2 \right] / M^2 \quad (1.33)$$

can only be determined theoretically by a quantum mechanical calculation (Herring 1966c, Akhiezer et al 1968). Spin waves, or magnons, are the quantised units of waves in the uniform magnetisation of the magnet. They also conjectured that spin waves would be present in a band model as well as the excited single particle states possible in Stoner Theory. As Stoner Theory and Herring and Kittel's approach use mutually exclusive approximations, a firmer foundation for spin waves was required. Izuyama (1960) and Izuyama, Kim and Kubo (1963) showed how to obtain the complete energy spectrum for a one band model, using equivalent forms of the random phase approximation. They used the Hamiltonian

$$H = \sum_{\mathbf{k}, \sigma} \varepsilon(\mathbf{k}) n_{\mathbf{k}\sigma} + I \sum_i n_{i\uparrow} n_{i\downarrow} \quad (1.34)$$

Within the random phase approximation spin waves are generated by

$$\frac{1}{\sqrt{N}} \sum_{\mathbf{k}} f(\mathbf{k}, \mathbf{q}) c_{\mathbf{k}+\mathbf{q}\downarrow}^+ c_{\mathbf{k}\uparrow} \quad (1.35)$$

where $f(\mathbf{k}, \mathbf{q}) \rightarrow 1$ when $\mathbf{q} \rightarrow 0$. In the limit $\mathbf{q} \rightarrow 0$ the spin wave creation operator given by (1.35) is exact. The spin waves are distinct, for $q < q_{\max}$, from the continuum of Stoner excitations. The RPA formula (1.35) for the spin wave energy Dq^2 is proportional to Aq^2 , when A of equation (1.33) is calculated from the energy difference in the Hartree-Fock approximation between a ferromagnetic and spin density wave arrangement of the itinerant electron spins (Herring 1966c).

We see that for both models, (1.1) and (1.34), $S_0^- |0\rangle$ is an exact eigenstate representing a spin wave of wave vector $q = 0$. Using this fact, Edwards (1962, 1967) was able to obtain the exact formula for the energy $\hbar\omega = Dq^2$ of a spin wave in a strong ferromagnetic metal, using the first terms of an expansion in powers of q . The only condition is that the inverse lifetime of the spin wave tends to zero faster than q , as $q \rightarrow 0$, so that the spin wave is well-defined, and the theory is exact for sufficiently small q . The only other derivation of spin waves from first principles

is that of Abrikosov and Dzialoshinski (1958), who use Fermi liquid theory beyond its true region of validity (Herring 1966c).

Lowde and Windsor (1970) have shown that the model of Izuyama, Kim and Kubo (1963), using the RPA approximation, essentially explains the neutron scattering data on nickel, provided one replaces I by I_{eff} following Kanamori (1963). Edwards (1967) was able to evaluate his formula for D for the Hamiltonian (1.34), approximately, confirming Izuyama, Kim and Kubo's formula for the spin wave energy, with I_{eff} instead of I . Though authors have gone beyond the RPA approximation, (Herring (1966c) reviews the calculations) the RPA approximation for the spin wave energy with an effective interaction I_{eff} seems to be the only practical formula for D in nickel.

Chapter 2

Spin Wave Interactions

I Introduction

One of the striking effects of spin waves in both the Heisenberg model and the itinerant electron model is the effect of spin wave and electron excitations on thermodynamic quantities at low temperatures. There are two contributions to the deviation of the magnetisation from its saturation value in the itinerant electron model (Edwards 1962, Edwards and Wohlfarth 1968). One contribution comes from spin wave excitations and goes as $T^{3/2}$ to lowest order, and the other contribution comes from single particle or Stoner excitations and goes as T^2 . The T^2 term does not arise in the Heisenberg model because there are no single particle electron excitations at low temperatures. Higher powers in T than $T^{3/2}$ and T^2 will occur from interactions between electrons, magnons and other elementary excitations (Izuyama and Kubo 1964, Marshall 1962).

Low temperature measurements of the spin wave energy $\hbar\omega = Dq^2$ in iron, have been interpreted in this way (Phillips 1966, Stringfellow 1968) so that

$$D = D_0 + D_1 T^2 + D_2 T^{5/2} \quad (2.1)$$

where D_1 is the term coming from magnon-electron interactions (Mathon and Wohlfarth 1968) and D_2 is the term coming from magnon-magnon interactions. For the Heisenberg ferromagnet D_1 is zero and D_2 has been extensively investigated. The problem of calculating D_2 was solved by Dyson (1956). the calculation of D_2 for the itinerant electron model is difficult and so far work has been mainly phenomenological. It has been reviewed by Herring (1966a). There is no general agreement about the sign and magnitude of D_2 .

We will discuss these problems within a unified approach using a formula for the spin wave energy obtained by generalising an expression of Ma, Beal-Monod and Fredkin (1968). Thence previous results for the Heisenberg ferromagnet are derived more directly. Cooke and Gersch (1967) and Marshal and Murray (1969) obtain Dyson's results using the equations of motion of Green's functions, whereas Silberglitt and Harris (1968) use the t-matrix equation. The problem of spin wave interactions in the itinerant electron model is put on a more rigorous basis. Previous results on D_2 (Izuyama 1964a, Kawasaki 1964) can be shown to correspond with Dyson's Born approximation, which is the first term in his expression for D_2 . Estimates of D_2 are made for nickel. One is also able to discuss spin wave lifetimes.

II General Formula for the Spin Wave Energy

Any system which has rotational symmetry with regard to spin has the property that its Hamiltonian H commutes with S_0^- , that is

$$[S_0^-, H] = 0 \quad (2.2)$$

where S_0^- is the total spin step-down operator. Any isotropic model of a ferromagnet has this property and also satisfies the condition that the direction of total magnetisation is arbitrary. We assume that there is an infinitesimal magnetic field which fixes the direction of the magnetisation vector in the ground state, so that the majority of the spins are up. The generalised transverse susceptibility $\chi(\underline{q}, \omega)$ is defined by

$$\begin{aligned} \chi(\underline{q}, \omega) &= \langle\langle S_{\underline{q}}^+ ; S_{\underline{q}}^- \rangle\rangle_{\omega} \\ &= -i \int_{-\infty}^{\infty} dt e^{i\omega t} \langle [S_{\underline{q}}^+(t), S_{\underline{q}}^-(0)] \rangle \theta(t) \\ &= -i \int_{-\infty}^{\infty} dt e^{i\omega t} \langle [S_{\underline{q}}^+(0), S_{\underline{q}}^-(-t)] \rangle \theta(t), \end{aligned} \quad (2.3)$$

where $S(\underline{r})$ is the spin density and S_i the total spin in the unit cell centre R_i with

$$\begin{aligned} S_{\underline{q}} &= \int e^{i\underline{q} \cdot \underline{r}} S(\underline{r}) d\underline{r} \\ S_{\underline{q}} &= \sum_i e^{i\underline{q} \cdot \underline{R}_i} S_i \end{aligned} \quad (2.4)$$

or

(23)

according to which model we are investigating. In addition to (2.2) we know that

$$[S_{\underline{q}}^-, V] = 0 \quad (2.5)$$

where V is any part of the Hamiltonian H which is independent of momentum.

From the equations of motion of $\chi(\underline{q}, \omega)$, using (2.2), (2.3) and (2.5) and operating in turn on the operators $S_{-\underline{q}}^+(t)$, $S_{\underline{q}}^-(-t)$, we find

$$\omega^2 \chi(\underline{q}, \omega) = q^2 \chi_J(\underline{q}, \omega) + q \langle [J_{-\underline{q}}^+, S_{\underline{q}}^-] \rangle + \omega^2 \langle S_0^z \rangle \quad (2.6)$$

where $\chi_J(\underline{q}, \omega) = \langle\langle J_{-\underline{q}}^+ ; J_{\underline{q}}^- \rangle\rangle_\omega$, with the spin currents $J_{\pm \underline{q}}^\mp$ defined by

$$q J_{-\underline{q}}^+ = [S_{-\underline{q}}^+, H], \quad J_{\underline{q}}^- = (J_{-\underline{q}}^+)^+ \quad (2.7)$$

The first two terms on the right hand side of equation (2.6) are of order q^2 .

Since the spin wave is the only eigenstate of low energy near the ground state energy involving spin fluctuations, in all the systems we discuss, we can assume that for small q and ω the susceptibility is dominated by the spin wave pole at $\omega = Dq^2$. We therefore write

$$\chi(\underline{q}, \omega) = \frac{2 \langle S^z \rangle (1 + Aq^2)}{\omega - Dq^2} + O(q^2) \quad (2.8)$$

One can compare (2.6) and (2.8) in the region $q/1, q \rightarrow 1$.

Then

$$\begin{aligned} & \lim_{q \rightarrow 0} \lim_{q/\omega \rightarrow 0} \frac{\omega^2}{2\langle S^z \rangle q^2} \left(\chi(q, \omega) - 2\langle S^z \rangle / \omega \right) \\ &= D = \frac{1}{2\langle S_0^z \rangle} \lim_{\omega \rightarrow 0} \lim_{q \rightarrow 0} \left[\frac{1}{q} \langle [J_{-q}^+, S_q^-] \rangle + \chi_J(q, \omega) \right]. \quad (2.9) \end{aligned}$$

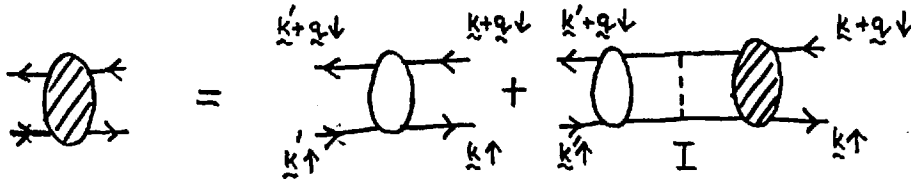
(2.9) is an exact formula for D , valid for any metallic or non-metallic isotropic ferromagnet, or for a non-ferromagnetic material in the presence of a static magnetic field H (Edwards and Fisher 1971). For the latter case we must replace ω , whenever it appears, by $\omega - \omega_L$, where ω_L is the Larmor frequency $g\mu_B H$. Clearly from (2.6) χ_J , as well as χ , contains the spin wave pole, but the limit of χ_J in (2.9) is finite because the residue of the pole in χ_J , $\langle [J_{-q}^+ | q \rangle]^2$, tends to zero as $q \rightarrow 0$. (2.9) is not the same as the general formula derived by Izuyama (1971).

For a gas with short-range interactions, for example the one-band model with Hamiltonian given by (1.34),

$$\lim_{q/\omega \rightarrow 0} \chi_J(q, \omega) = \chi_J'(0, \omega) \quad (2.10)$$

where χ_J' is the "irreducible" part of χ_J , consisting of all irreducible diagrams contributing to χ_J . In diagram form the relation between χ_J and χ_J' is (Ma, Beal-Monod and Fredkin 1968)

$$\chi_J(q, \omega) = \chi_J'(q, \omega) + \chi_{JS}' I \chi_{SJ} \quad (2.11)$$



where $\chi_{SJ} = \langle\langle S_{-q}^+, J_q^- \rangle\rangle_\omega$ and χ_{JS}' is the irreducible part of χ_{JS} . The interaction I is independent of momentum transfer. (2.10) follows from (2.11) from the fact that both χ_{SJ} and χ_{JS}' are of $O(q^2)$. This shows that our result (2.9) is a generalisation of that of Ma, Deul-Monod and Fredkin (1966), which has χ_J' instead of χ_J in (2.9). The same formula (2.9) was obtained by Edwards (1967) for a strong ferromagnet with an arbitrary number of bands, at $T=0$. For a short-range one-band model he also showed that in his expression corresponding to $\chi_J(0, \omega)$ one could omit reducible diagrams. Removing these diagrams from χ_J removes the spin wave pole in χ_J .

Our formalism for χ ensures that the corresponding neutron scattering intensity is always proportional to $\langle S^z \rangle$ in the long wavelength limit, a result stressed by Marshall and Murray (1969) for the Heisenberg model and observed in metals by Stringfellow (1968). For a general system, insulating or metallic, of interacting electrons in a periodic potential

$$H = \sum_i \left(\frac{1}{2m} p_i^2 + V(r_i) \right) + \frac{1}{2} \sum_{ij} \frac{e^2}{|r_i - r_j|} \quad (2.12)$$

so that (2.9) can be written in another form

$$D = \frac{1}{(n_{\uparrow} - n_{\downarrow})} \left[\frac{n}{2m} + \lim_{\omega \rightarrow 0} \ll J_0^+; J_0^- \gg_{\omega} \right] \quad (2.13)$$

$$\text{where } J_0^- = \hat{q} \cdot \sum_i \frac{\underline{p}_i \sigma_i^-}{m} = \lim_{q \rightarrow 0} \frac{1}{q} \left[\sum_i \frac{p_i^2}{2m}, \sum_j e^{i\hat{q} \cdot \underline{r}_j} \sigma_j^- \right] \quad (2.14)$$

is the spin current in the direction of $\hat{q}$, $\hat{q} = \underline{q}/q$, and $\underline{p}_i = -i\nabla_i$.

Though we have obtained a formal expression for D at all temperatures below T_c , we are only able to evaluate D exactly at low temperatures. In the cases considered D is real, so that magnon lifetimes appear in higher powers of q and cannot be discussed rigorously by the present method. However $q^2 \chi_J'(q, \omega) / 2\langle S^z \rangle$ in (2.9) is equivalent to an irreducible spin wave self-energy $q^2 \Sigma(q, \omega)$, while $q^2 \chi_J(0, \omega) / 2\langle S^z \rangle$ in (2.9) is equivalent to $q^2 \Sigma(0, \omega)$.

Thus the lifetime τ of a spin wave calculated from the imaginary part of the self-energy is

$$\frac{1}{\tau} = 2q^2 \text{Im} \Sigma(q, Dq^2) = 2q^2 \text{Im} \chi_J(q, Dq^2) / 2\langle S^z \rangle \quad (2.15)$$

or approximately

$$\frac{1}{\tau} = 2q^2 \text{Im} \Sigma(0, Dq^2) = 2q^2 \text{Im} \chi_J(0, Dq^2) / 2\langle S^z \rangle. \quad (2.16)$$

(2.15), which was derived by Ma, Beal-Monod and Fredkin (1968) for an electron gas with short-range interactions, leads to correct results for the Heisenberg model. (2.16) differs

only slightly and may be more useful when it is difficult to define χ_J' the irreducible part of χ_J .

III Heisenberg Ferromagnet

In this section we will make use of equation (2.9) to calculate the energy of spin waves for a Heisenberg Hamiltonian

$$H = -\frac{1}{2N} \sum_{\underline{k}} J(\underline{k}) \underline{S}_{\underline{k}} \cdot \underline{S}_{-\underline{k}} = -\frac{1}{2} J \sum_{\substack{\underline{i}, \underline{j} \\ \text{n.n.}}} \underline{S}_i \cdot \underline{S}_j \quad (2.17)$$

where $J(\underline{k}) = J \sum_{\text{n.n.}} e^{i \underline{k} \cdot \underline{R}_i}$ with the sum over lattice sites which are nearest neighbours to the origin. One finds

$$q J_{-\underline{q}}^+ = [S_{-\underline{q}}^+, H] = \omega_0(\underline{q}) S_{-\underline{q}}^+ + q K_{-\underline{q}}^+, \quad (2.18)$$

$$\text{with} \quad \langle 0 | K_{-\underline{q}}^+ = 0 \quad (2.19)$$

where $|0\rangle$ is the ground state wavefunction with all spins up and

$$\omega_0(\underline{q}) = (J(0) - J(\underline{q})) s \quad (2.20)$$

is the energy of a non-interacting spin wave. The result

$$q K_{-\underline{q}}^+ = q \sum_{\underline{k}_1, \underline{k}_2} \Phi(\underline{k}_1, \underline{k}_2, \underline{q}) S_{-(\underline{k}_1 + \underline{k}_2 + \underline{q})}^- S_{\underline{k}_1}^+ S_{\underline{k}_2}^+ \quad (2.21)$$

$$\text{where} \quad \Phi(\underline{k}_1, \underline{k}_2, \underline{q}) = \frac{1}{2s N^2 \underline{q}} (J(\underline{k}_1) - J(\underline{k}_1 + \underline{q}))$$

is obtained using the relation (Wortis 1959)

$$S_{\underline{q}}^z = sN \delta_{\underline{q},0} - \frac{1}{2sN} \sum_{\underline{k}} S_{\underline{k}+\underline{q}}^- S_{-\underline{k}}^+ + (2.22)$$

(higher order products involving four S^- , S^+ operators).

Higher products are zero for $s = \frac{1}{2}$ and terminating the series after two terms for all s is consistent with working at low temperatures. Defining $\chi_{\underline{k}}(\underline{q}, \omega) = \langle\langle K_{-\underline{q}}^+ ; K_{\underline{q}}^- \rangle\rangle_{\omega}$,

(2.6) to (2.9), with $K_{\underline{q}}$ in place of $J_{\underline{q}}$ and $\omega - \omega_0(\underline{q})$ in place of ω , yield the following expression for the spin wave energy

$$\omega = \omega_0(\underline{q}) + \frac{q^2}{2\langle S^z \rangle} \left[\lim_{\underline{q} \rightarrow 0} \frac{1}{q} \langle [K_{-\underline{q}}^+, S_{\underline{q}}^-] \rangle + \lim_{\omega \rightarrow 0} \langle\langle K_0^+ ; K_0^- \rangle\rangle_{\omega} \right] (2.23)$$

$$\text{where } K_0^+ = -i \frac{J}{2sN^2} \sum_{\underline{i}} \hat{\underline{q}} \cdot \underline{R}_i e^{i \underline{k}_1 \cdot \underline{R}_i} S_{-\underline{k}_1 + \underline{k}_2}^- S_{\underline{k}_1}^+ S_{\underline{k}_2}^+ \quad (2.24)$$

and $\hat{\underline{q}}$ is the unit vector in the direction of $\underline{q}$.

(2.23) gives the low temperature correction to $\omega_0(\underline{q})$ due to magnon-magnon interactions. The thermal averages can be evaluated consistently at low temperatures by transforming from spin space, which has eigenstates of the form $\frac{1}{\sqrt{2sn!}} (S_i^-)^n |0\rangle$, to a boson space, which has corresponding eigenstates $\frac{1}{\sqrt{n!}} (\alpha_i^+)^n |0\rangle$. The ground states $|0\rangle$ and $|0\rangle$ are defined by

$$S_i^+ |0\rangle = 0 \quad \text{and} \quad \alpha_i |0\rangle = 0 \quad (2.25)$$

The spin operators are mapped from the spin space onto the boson space by the Dyson-Maleev transformation (Akhiezer et al, 1968, Maleev 1956)

$$\begin{aligned} S_i^- &\rightarrow \tilde{S}_i^- = \sqrt{2s} a_i^+ \\ S_i^+ &\rightarrow \tilde{S}_i^+ = \sqrt{2s} (1 - a_i^+ a_i / 2s) a_i \\ S_i^z &\rightarrow \tilde{S}_i^z = (s - a_i^+ a_i) \end{aligned} \quad (2.26)$$

The operators $\tilde{S}_i^\pm$, $\tilde{S}_i^z$ satisfy the same commutation relations as $S_i^\pm$, S_i^z .

Thermal averages of the form $\text{Tr} \{G(s)\}$, where the sum is over an orthonormal set of eigenstates in spin space, and $G(s)$ is an arbitrary function of the spin variables of the form $X(s)\exp(-\beta H(s))$, where $H(s)$ is the Heisenberg Hamiltonian, can be evaluated using the formula

$$\text{Tr} \{G(s)\} = \text{Tr}_B \{P G(P \hat{S} P)\} \quad (2.27)$$

where the projection operator P is defined by

$$\begin{aligned} P(a_i^+)^n |0\rangle &= 0 & \text{if } n \geq 2s \\ &= 1 & \text{if } n < 2s \end{aligned} ,$$

and Tr_B is a sum over an orthonormal set of eigenstates in boson space. States $(a_i^+)^n |0\rangle$, with $n \geq 2s$, are non-physical since there are no corresponding states in spin space and are omitted by the use of P . Fortunately there

are no matrix elements of $\tilde{S}$ between physical and non-physical states, so

$$\text{Tr} \{G(s)\} = \text{Tr}_B \{PG(\tilde{s})\} \quad (2.28)$$

Any non-physical state contains more than $2s$ spin deviations on a site and hence would introduce a factor $e^{-\beta 2Js}$ into the trace. Thus

$$\langle X(s) \rangle = \frac{1}{Z} \text{Tr} \{e^{-\beta H(s)} X(s)\} = \frac{1}{Z} \text{Tr}_B \{e^{-\beta H(\tilde{s})} X(\tilde{s})\} + \quad (2.29)$$

(terms of order $\exp -2Js/kT$).

So to evaluate any thermal average one can work entirely in boson space provided one is at low enough temperatures, so that $kT \ll 2Js$. To evaluate $\langle X(s) \rangle$ to the lowest power in kT , when $X|0\rangle = 0$, one may write

$$\langle X(s) \rangle = \langle X(\tilde{s}) \rangle = \frac{1}{N} \sum_{\underline{p}} \langle 0 | a_{\underline{p}} X(\tilde{s}) a_{\underline{p}}^+ | 0 \rangle n_{\underline{p}} \quad (2.30)$$

where $a_{\underline{p}}^+$ is the operator in boson space which creates a non-interacting spin wave and $n_{\underline{p}}$ is the only boson occupation factor in the expression. This lowest order approximation in powers of $k_p T$ comes about (Akhiezer et al, 1968) if one puts

$$\frac{1}{N} \langle a_{\underline{k}}^+ a_{\underline{k}} \rangle = 0, \quad \frac{1}{N} \langle a_{\underline{k}} a_{\underline{k}}^+ \rangle = 1$$

(31)

in any intermediate states of $\langle X \rangle$, and evaluates $\langle X \rangle$ using Wick's Theorem (Gaudin 1960). Any error involves the sum of n_k over all k and hence arises as a correction at least of order $(kT)^{3/2}$. However equation (2.30) is not correct if X involves a δ -function as the δ -function will restrict the summation over k . This problem arises in the calculation of spin wave lifetimes (Harris 1968).

Using (2.21), (2.26) and (2.30) one finds

$$2\langle S^2 \rangle = 2Ns + O(kT)^{3/2} \quad (2.31)$$

$$\lim_{q \rightarrow 0} \frac{1}{2\langle S^2 \rangle_q} \langle [K_q^+, S_q^-] \rangle = \lim_{q \rightarrow 0} \frac{1}{q^2 N^2} \sum_p (0|a_p([a_q, H] - \omega(q)a_q) a_p^+ |0) n_p \quad (2.32)$$

to the lowest power in kT

$$= \lim_{q \rightarrow 0} \frac{2s}{q^2 N^2} \sum_{p, k_1, k_2} \Phi(k_1, k_2, q) (0|a_p a_{-(k_1+k_2+q)}^+ a_{k_1} a_{k_2} a_q^+ a_p^+ |0) n_p \quad (2.33)$$

Using $[a_k, a_q^+] = N\delta_{k,q}$ one can easily evaluate this expression for a simple cubic lattice yielding as the first correction to the magnon energy

$$\omega - \omega_0(q) = -\frac{Ja^4}{6N} q^2 \sum_p p^2 n_p \quad (2.34)$$

This correction term corresponds to Dyson's Born approximation (Dyson 1956). The full $T^{5/2}$ correction comes from evaluating $\langle\langle K_0^+; K_0^- \rangle\rangle_\omega$ in (2.23) and has the form

$$\begin{aligned} \omega - \omega_0(q) &= D_1 T^{5/2} q^2 \\ &= -\frac{Ja^4}{6N} Q(\epsilon) \left(\sum_p n_p p^2 \right) q^2 \end{aligned} \quad (2.35)$$

where $Q(s) = 1$ for $s = \infty$. From (2.35) we see that the Born approximation is exact in the limit of very large spins. The sign of the magnon-magnon correction is negative. The equation of motion for the Green's functions appearing in $\langle\langle K_0^+ ; K_0^- \rangle\rangle_\omega$ is a soluble integral equation, since by (2.30) higher order Green's functions which appear are negligible at low temperatures. Cooke and Gersch (1967) and Marshall and Murray (1969) solve the equivalent equation. However the calculation of D used here is much simpler, since it starts with an exact expression for D and no decoupling of the equations of motion of Green's functions occurs.

For $s = \frac{1}{2}$ (2.35) becomes

$$\omega - \omega_0(q) = -Q(\frac{1}{2}) J q^2 a^2 \zeta(\frac{5}{2}) (k_B T / J)^{5/2} / 16\pi^{3/2} \quad (2.36)$$

where the Riemann zeta function $\zeta(\frac{5}{2}) = 1.341$ and $Q(\frac{1}{2}) = 1.68$. In terms of boson operators K_0^+ becomes

$$K_0^+ = -i \frac{J}{N^2} (2s)^{\frac{1}{2}} \sum_i (\hat{q} \cdot \underline{R}_i) e^{i \underline{k}_1 \cdot \underline{R}_i} a_{-\underline{k}_1 + \underline{k}_2}^+ a_{\underline{k}_1} a_{\underline{k}_2} \quad (2.37)$$

with the properties $\frac{1}{\sqrt{N}} K_0^+ a_{-\underline{p}}^+ |0\rangle = 0$

$\frac{1}{\sqrt{N}} \langle 0 | a_{\underline{p}} K_0^+ = \frac{J}{N} (\frac{s}{2N})^{\frac{1}{2}} \sum_{i, \underline{k}_2} (\hat{q} \cdot \underline{R}_i) (\underline{p} \cdot \underline{R}_i) e^{i \underline{k}_2 \cdot \underline{R}_i} \langle 0 | a_{\underline{k}_2} a_{\underline{p} - \underline{k}_2}$
to the lowest power in p , so that

$$\langle\langle K_0^+ ; K_0^- \rangle\rangle_{\omega=0} = \left(\frac{J}{N}\right)^2 \frac{s}{2N} \sum_{\substack{i, j \\ \underline{p}, \underline{k}_1, \underline{k}_2}} (\hat{q} \cdot \underline{R}_i) (\underline{p} \cdot \underline{R}_i) e^{i \underline{k}_1 \cdot \underline{R}_i} (\hat{q} \cdot \underline{R}_j) (\underline{p} \cdot \underline{R}_j) e^{i \underline{k}_2 \cdot \underline{R}_j} \times \langle\langle a_{\underline{k}_2} a_{\underline{k}_2} ; a_{\underline{k}_1}^+ a_{-\underline{k}_1}^+ \rangle\rangle_{\omega=0} \quad (2.38)$$

$T=0$

We define

$$F(\underline{R}_1, \underline{R}_2) = \frac{J}{N^3} \sum_{\underline{k}_1, \underline{k}_2} e^{i \underline{k}_1 \cdot \underline{R}_1} e^{i \underline{k}_2 \cdot \underline{R}_2} \langle\langle a_{\underline{k}_2} a_{\underline{k}_2}; a_{\underline{k}_1}^+ a_{\underline{k}_1}^+ \rangle\rangle_{\omega=0} \quad (2.39)$$

$T=0$

and from the equations of motion of $\langle\langle a_{\underline{k}_2} a_{\underline{k}_2}; a_{\underline{k}_1}^+ a_{\underline{k}_1}^+ \rangle\rangle_{\omega}$,

$$\begin{aligned} & -2\omega_0(\underline{k}_1) \langle\langle a_{\underline{k}_2} a_{\underline{k}_2}; a_{\underline{k}_1}^+ a_{\underline{k}_1}^+ \rangle\rangle_{\omega=0} \\ & = \sum_{\underline{k}_3} \frac{1}{N} (J(\underline{k}_3) - J(\underline{k}_3 + \underline{k}_1)) \langle\langle a_{\underline{k}_2} a_{\underline{k}_2}; a_{\underline{k}_3}^+ a_{\underline{k}_3}^+ \rangle\rangle_{\omega=0} + 2N^2 \delta_{\underline{k}_1, \underline{k}_2} \end{aligned} \quad (2.40)$$

we get

$$F(\underline{R}_2, \underline{R}_1) + \frac{1}{2Ns} \sum_{\substack{\underline{k}_1 \\ \underline{R}_3}} \frac{(1 - e^{i \underline{k}_1 \cdot \underline{R}_3}) e^{i \underline{k}_1 \cdot \underline{R}_1}}{\sum_i (1 - \cos \underline{k}_1 \cdot \underline{R}_i)} F(\underline{R}_2, \underline{R}_3) = \frac{-1}{sN} \sum_{\underline{k}_1} \frac{e^{i \underline{k}_1 \cdot \underline{R}_1 + \underline{R}_2}}{\sum_i (1 - \cos \underline{k}_1 \cdot \underline{R}_i)} \quad (2.41)$$

$\underline{R}_1, \underline{R}_2, \underline{R}_3$ are nearest neighbours of the origin, so that for a simple cubic lattice this is a 6×6 matrix equation. This equation could then be solved in the usual way for matrices (Cooke and Gersch 1967). We will solve it by a quicker method, originally used by Dyson (1956), for the particular case of a simple cubic lattice (Akhiezer et al, 1968a).

$$\text{We assume } F(\underline{R}_2, \underline{R}_1) = A + B |\underline{R}_1 \cdot \underline{R}_2| \quad (2.42)$$

where A and B will be determined from (2.41). Defining

$$\begin{aligned} \frac{1}{N} \sum_{\underline{k}_1} \frac{(1 - \cos \underline{k}_1 \cdot \underline{R}_3) \cos \underline{k}_1 \cdot \underline{R}_1}{\sum_i (1 - \cos \underline{k}_1 \cdot \underline{R}_i)} &= \Gamma_1 \quad \text{if } \underline{R}_1 = \pm \underline{R}_3 \\ &= \Gamma_2 \quad \text{if } \underline{R}_1 \neq \pm \underline{R}_3 \end{aligned} \quad (2.43)$$

(34)

$$\text{and } \frac{1}{N} \sum_{\underline{k}_1} \frac{\cos \underline{k}_1 \cdot \underline{R}_1}{\sum_i (1 - \cos \underline{k}_1 \cdot \underline{R}_i)} = B_{100} ,$$

$$\text{then } \Gamma_1 + 2\Gamma_2 = 0 \quad (2.44)$$

by summing (2.43) over R_3 and (2.41) becomes

$$A + 6\Gamma_2/s + B|\underline{R}_1 \cdot \underline{R}_2|(1 - 3\Gamma_2/s) = (\Gamma_2 - B_{100})/s - |\underline{R}_1 \cdot \underline{R}_2| 3\Gamma_2/s \quad (2.45)$$

Equating terms independent of and dependent on $|\underline{R}_1 \cdot \underline{R}_2|$ one finds A and B. From (2.45), (2.42) and (2.39)

$$\begin{aligned} \frac{1}{2\langle S^2 \rangle} \langle\langle K_0^+; K_0^- \rangle\rangle_{\omega=0} &= \frac{Ja^4}{N} \sum_{\underline{p}} (p^2 n_{\underline{p}}) ((\hat{\underline{p}} \cdot \hat{\underline{q}})^2 A + 3(\hat{p}_x \hat{q}_x)^2 B) \\ &= \frac{Ja^4}{N} \sum_{\underline{p}} (p^2 n_{\underline{p}}) 2(A+B) \end{aligned} \quad (2.46)$$

$$\begin{aligned} \text{So } Q(s) - 1 &= -2(A+B) \\ &= 4\Gamma_2/s(1 - 3\Gamma_2/s) + 2B_{100}/s \text{ from (2.45)} \\ &= \frac{2}{3} \frac{\Gamma/s}{(1 - \Gamma/2s)} + \frac{2B_{100}}{s} \end{aligned} \quad (2.47)$$

$$\text{where } \Gamma = 6\Gamma_2 .$$

According to Akhiezer et al (1968a), $\Gamma = 0.2$ and $B_{100} = .086$ for a simple cubic lattice and $Q(s)$ obtains its maximum value of about 1.68 near $s = \frac{1}{2}$. Cooke and Gersoh (1967) find values of $Q(\frac{1}{2})$ for f.c.c. and b.c.c. lattices as well.

This result (2.47) in equivalent forms has been obtained by Dyson (1956), Akhiezer et al (1968a), Boyd and

Callaway (1965) by calculating the scattering amplitude of two magnons. The problem is equivalent to that of a low density gas of weakly interacting bosons. Thus Silberglitt and Harris (1968) obtain the same low temperature correction by solving the t-matrix equation for two-body scattering of magnons. Vaks, Larkin and Pikin (1968) find the irreducible part of a temperature dependent Green's function. Cooke and Gersch (1967), Marshal and Murray (1968, 1969), have used a Green's function technique. This method involves the difficulty of obtaining the energy of a spin wave pole from the equations of motion of Green's functions. The exact formula (2.9) for the spin wave energy reproduces all these results for the Heisenberg model in the most rigorous and straightforward manner.

The lifetime τ_q of an interacting spin wave at low temperatures is from (2.15) given by

$$\frac{1}{\tau_q} = -\frac{q^2}{\langle S^2 \rangle} \text{Im} \chi'_K(q, \omega_0(q)) \text{ to the lowest power in } q. \quad (2.48)$$

The simplest contribution to the irreducible part of $\text{Im} \chi_K(q, \omega)$ is that obtained by evaluating $\text{Im} \chi_K(q, \omega)$ in an approximation in which the spin waves are assumed to be non-interacting. This is equivalent to calculating the correction to the spin wave energy in second order

perturbation theory (Akhiezer et al 1968b). As the scattering only involves long wavelength spin waves, we can simplify $K_{\underline{q}}^+$ by writing (2.37) as

$$K_{\underline{q}}^+ = \frac{J(2s)^2}{N^2} \sum_{\underline{k}_1, \underline{k}_2} \alpha^2 \hat{q} \cdot (\underline{k}_1 + \underline{k}_2) a_{-(\underline{k}_1 + \underline{k}_2 + \underline{q})}^+ a_{\underline{k}_1} a_{\underline{k}_2} \quad (2.49)$$

$$\text{Then } \frac{-q^2}{\langle S^2 \rangle} \text{Im} \ll K_{\underline{q}}^+ ; K_{\underline{q}}^- \gg_{\omega_0(\underline{q})} \quad (2.50)$$

$$= \sum_{\underline{k}_1, \underline{k}_2} \frac{4\pi J^2}{N^2} \alpha^4 |\underline{q} \cdot \underline{k}_1 + \underline{k}_2|^2 [n_{\underline{p}}(1+n_{\underline{k}_1})(1+n_{\underline{k}_2}) - (1+n_{\underline{p}})n_{\underline{k}_1}n_{\underline{k}_2}] \delta(\omega(\underline{q}) + \omega(\underline{p}) - \omega(\underline{k}_1) - \omega(\underline{k}_2))$$

with $\underline{p} = \underline{k}_1 + \underline{k}_2 + \underline{q}$, the δ -function becomes $[4Js\alpha^2]^{-1} [\delta(\underline{k}_1 - \underline{p}) - \delta(\underline{k}_1 + \underline{p})]$

where $\underline{p}$ are vectors on a sphere centre $\frac{1}{2}(\underline{p} + \underline{q})$, radius $\frac{1}{2}|\underline{p} - \underline{q}|$. When $B\omega_0(\underline{q}) \gg 1$ one can replace the bose factors in (2.50) by just $n_{\underline{p}}$, as we have already done in (2.30), as all bose factors are small. The angular integration of over the sphere also simplifies to yield

$$\frac{1}{\tau_{\underline{q}}} = \frac{Ja^8}{s(2\pi)^3} \int_0^\infty \int_{-1}^1 n_{\underline{p}} p^4 q^2 x^2 \int_{\underline{p} <}^{\underline{p} >} \frac{2\underline{p}}{|\underline{p} + \underline{q}|} d\underline{p} dx dp \quad (2.51)$$

where $x = \cos \theta_p$, θ_p is the angle between $\underline{p}$ and $\underline{q}$ and

$$\underline{p}^{\geq 2} = \frac{1}{2}(p^2 + q^2) \pm |\underline{p} + \underline{q}| |\underline{p} - \underline{q}|$$

defines the maximum and minimum values of $\underline{p}$. Then

$$\frac{1}{\tau_{\underline{q}}} = \frac{Ja^8}{s(2\pi)^3} \int_0^\infty \int_{-1}^1 n_{\underline{p}} p^4 q^2 |\underline{p} - \underline{q}| dx dp \quad (2.52)$$

The same integral was obtained by Cooke and Gersch (1967),

(Marshal and Murray 1968, 1969) yielding

$$\frac{1}{\tau_q} = \frac{2J}{s} (qa)^3 \zeta(5/2) \left(\frac{1}{\beta 8\pi J s} \right)^{5/2} \quad (2.53)$$

These authors erroneously used the same integral (2.52) to calculate the spin wave damping for the case $\beta \omega_0(q) \ll 1$.

The bose functions in (2.50) are not independent because of the δ -function and satisfy

$$\begin{aligned} & n_p(1+n_p)(1+n_{p-q}) - (1+n_p)n_q n_{p-q} \\ &= n_q n_{p-q}(1+n_p)(e^{\beta \omega_0(q)} - 1) \end{aligned} \quad (2.54)$$

So in the limit $\beta \omega_0(q) \ll 1$ the bose functions are not always small and one must replace n_p in the integral (2.51) by the full expression (2.54), which becomes

$$\beta \omega_0(q) n_q n_{p-q} (1+n_p) \quad (2.55)$$

This integral is discussed fully by Brooks Harris (1968). The dominant term in the spin wave lifetime in this limit, $\beta \omega_0(q) \ll 1$, was originally obtained by Kashcheyev and Krivoglaaz (1961) who evaluated this integral to obtain

$$\frac{1}{\tau_q} = \frac{J}{s} \frac{(qa)^4}{8\pi^3} \frac{1}{(\beta 2Js)^2} \frac{1}{3} \log^2 \left(\frac{1}{\beta 2Js q^2 a^2} \right) \quad (2.56)$$

We have thus seen how (2.48) and (2.23) reproduce known results for the spin wave lifetimes and the

renormalisation of the spin wave energy due to spin wave interactions at low temperatures $T \ll T_c$. This provides powerful confirmation of our method. Vaks, Larkin and Pikin (1968) have shown how to deal with the Heisenberg model at higher temperatures, where the method of this section is not applicable, and were able to calculate the energy of spin waves, within certain limits, right up to the Curie temperature. They find that long wavelength spin waves are well defined in the region $q^2 < (1-T/T_c)^\alpha$ where α is a number of order unity. Our formula (2.23) for the spin wave energy Dq^2 is also valid for all temperatures below T_c for small enough q and so could be used to reproduce Vaks, Larkin and Pikin's results.

IV The Itinerant Electron Model

For a one-band model with short-range interactions, for which $c_{\underline{k}\sigma}^+$, $c_{\underline{k}\sigma}$ are creation and destruction operators of electrons, with spin σ , in the band, and

$$H = \sum_{\underline{k}, \sigma} \epsilon(\underline{k}) n_{\underline{k}\sigma} + \frac{I}{N} \sum_{\underline{k}_1, \underline{k}_2, \underline{k}} c_{\underline{k}_1 + \underline{k}}^+ c_{\underline{k}_1 \uparrow} c_{\underline{k}_2 \uparrow}^+ c_{\underline{k}_2 - \underline{k} \downarrow} c_{\underline{k} \downarrow} \quad (2.57)$$

$$S_{\underline{q}}^- = \sum_{\underline{k}} c_{\underline{k} + \underline{q} \downarrow}^+ c_{\underline{k} \uparrow}, \quad J_{\underline{q}}^- = \sum_{\underline{k}} (\epsilon(\underline{k} + \underline{q}) - \epsilon(\underline{k})) c_{\underline{k} + \underline{q} \downarrow}^+ c_{\underline{k} \uparrow}$$

(2.9) leads directly to a generalisation of Edwards' (1967) previous formula for D , namely

$$Dq^2 = \frac{1}{(n_{\uparrow} - n_{\downarrow})} \left(\sum_{\underline{k}, \sigma} \frac{1}{2} (\underline{q} \cdot \nabla)^2 \varepsilon(\underline{k}) n_{\underline{k}\sigma} + \lim_{\omega \rightarrow 0} \sum_{\underline{k}, \underline{k}'} (\underline{q} \cdot \nabla \varepsilon(\underline{k})) (\underline{q} \cdot \nabla \varepsilon(\underline{k}')) \langle\langle c_{\underline{k}\uparrow}^+ c_{\underline{k}\downarrow} ; c_{\underline{k}'\downarrow}^+ c_{\underline{k}'\uparrow} \rangle\rangle_{\omega} \right) \quad (2.58)$$

Edwards' formula applied to a strong ferromagnet, for which all the electrons have up spin and are therefore non-interacting in the ground state. Formula (2.58) applies at non-zero temperatures and also to a weak ferromagnet, for which the electron spins are only partially aligned and the ground state wave function is not known exactly.

However (2.58) is not on a convenient form for describing magnon-magnon interactions. Instead, for a strong ferromagnet, we consider the function $\chi_A(\underline{q}, \omega)$ which is defined by (2.3) with $\langle\langle S_{\underline{q}}^+ ; S_{\underline{q}}^- \rangle\rangle_{\omega}$ replaced by $\langle\langle A_{\underline{q}} ; A_{\underline{q}}^+ \rangle\rangle_{\omega}$. Here $A_{\underline{q}}^+$ is the operator which creates a spin wave on the ground state $|0\rangle$. The existence of an operator $A_{\underline{q}}^+$, asymptotically exact to the order q , is implicit in earlier work (Edwards 1967). $A_{\underline{q}}^+$ satisfies

$$A_0^+ = \frac{1}{\sqrt{n}} S_0^- , \quad (2.59)$$

where n is the number of electrons, and $A_{\underline{q}}^+$, in the itinerant model, corresponds to $\frac{1}{\sqrt{N}} a_{\underline{q}}^+$ in the Heisenberg ferromagnet. Following this analogy, $K_{\underline{q}}$ is defined by

$$[A_{\underline{q}}, H] = D_0 q^2 A_{\underline{q}} + q K_{\underline{q}} \quad (2.60)$$

where $D_0 q^2$ is the energy of the spin wave at $T = 0$. Since $\chi_A(q, \omega)$ contains a spin wave pole, we may follow the procedure of section I and obtain an equation corresponding to (2.9). Using (2.9) and (2.60) we get

$$D = D_0 + \lim_{q \rightarrow 0} \frac{1}{q} \langle [K_q, A_q^+] \rangle + \lim_{\omega \rightarrow 0} \langle\langle K_0; K_0^+ \rangle\rangle_\omega \quad (2.61)$$

For a strong ferromagnet the excitations with lowest energy are either spin waves or single particle excitations, without spin flip. To the lowest power in the temperature, these can be assumed to be independent. The third type of excitation that can occur is one in which an electron flips its spin and goes from the up spin band to the down spin band. This means jumping an energy gap, I_n/N , and so does not occur at low temperatures. To obtain the R.P.A.

expression for the spin wave energy (Izuyama, Kim and Kubo 1963, Englert and Antonoff 1964) the Green's function

$\langle\langle c_{k\uparrow}^+ c_{k\downarrow}; c_{k'\downarrow}^+ c_{k'\uparrow} \rangle\rangle_\omega$ in (2.58) is evaluated in an approximation in which the electrons are non-interacting with energy $\epsilon(k)$ in the state $\psi_{k\uparrow}$ and $\epsilon(k) + I(n_\uparrow - n_\downarrow)/N$ in the state $\psi_{k\downarrow}$. Then in this approximation

$$\langle\langle c_{k\uparrow}^+ c_{k\downarrow}; c_{k'\downarrow}^+ c_{k'\uparrow} \rangle\rangle_\omega = \delta_{k,k'} \frac{f_{k\uparrow} - f_{k\downarrow}}{\omega - I(n_\uparrow - n_\downarrow)/N} \quad (2.62)$$

where $f_{k\uparrow, \downarrow}$ are the Fermi occupation numbers of the states $\psi_{k\uparrow, \downarrow}$.

By comparing (2.62) at $T=0$ and at $T \neq 0$, when T is small, one obtains the expansion of (2.58)

$$D = D_0 + D_1 T^2 \quad (2.63)$$

where D_1 is the correction term due to single particle excitations. By using (2.58) D_1 has been worked out without knowing A_q^+ . D_1 has been calculated for a single parabolic band (Izuyama and Kubo 1964) and for a tight-binding s band (Mathon and Wohlfarth 1968). In this latter case the agreement with the experimental values for iron, $D_1/D_0 = -1.4 \times 10^{-6}$ (Phillips 1966), $D_1/D_0 = -5 \times 10^{-6}$ (Stringfellow 1968), is quite good.

To calculate the size of magnon-magnon interactions the operator A_q^+ must be known, and (2.61) is used. The thermal averages are evaluated by using

$$\langle X \rangle = \sum_p \langle 0 | A_p X A_p^+ | 0 \rangle n_p \quad (2.64)$$

in analogy with (2.30). The first correction term to D_0 in (2.61) is then equivalent to

$$\sum_p \langle 0 | A_p A_q [H, A_q^+] A_p^+ | 0 \rangle n_p \quad (2.65)$$

which is the expression used by Izuyama (1964a) and Kawasaki (1964) to calculate magnon-magnon interactions. By comparing (2.65) and (2.32) it is clear that (2.65) is

analogous to Dyson's Born approximation for the Heisenberg ferromagnet. As $[[H, A_q^+], A_p^+]$ is of $O(pq)$ since $A_q^+ = \frac{1}{\sqrt{n}} S_q^- + O(q)$ both terms in (2.61) involve $\sum_p p^2 n_p$ giving

$$D = D_0 + D_2 T^{5/2} \quad (2.66)$$

This justifies the phenomenological argument (Marshall 1962, Izuyama and Kubo 1964) used to obtain the low temperature expansion of D in powers of T though the second term in (2.61) differs from that in Izuyama's (1971) treatment of D_2 . Though A_q^+ is known formally (Edwards 1967) for the strong ferromagnet, the only practical way of evaluating (2.61) and (2.65) is to follow Izuyama and Kawasaki and use the R.P.A. expression for A_q^+ for a parabolic band.

In the R.P.A. approximation one assumes A_q^+ has the form

$$A_q^+ = \sum_k f(k, q) c_{k+q\downarrow}^+ c_{k\uparrow} \quad (2.67)$$

and one finds

$$\begin{aligned} f(k, q) &= \frac{1}{n^{\frac{1}{2}}} \left(\frac{1}{1 + \epsilon(k+q) - \epsilon(k) / T n / N} \right) \\ &= \frac{1}{n^{\frac{1}{2}}} \left(1 - \frac{k \cdot q}{m T n / N} + \frac{1}{m (T n / N)^2} \left\{ \frac{(k \cdot q)^2}{m} - \frac{2}{5} \epsilon_F q^2 \right\} \right) \end{aligned} \quad (2.68)$$

for a parabolic band with effective mass m . The summation in k must be over all k , not just $|k| < k_F$, otherwise condition (2.2) does not apply. This was emphasised by Izuyama (1964b) when criticising Nakamura's approach (1964).

The first term in (2.61), that is (2.65), has been

evaluated using (2.67) and (2.68) to yield

$$D_2 T^{5/2} = \frac{1}{n} \sum_{\underline{p}} \frac{4}{15} \frac{\hbar^2 \varepsilon_F}{m^2 \Delta E^2} p^2 n_p \left(1 - \frac{12}{35} \lambda\right) \quad (2.69)$$

where $\lambda = \varepsilon_F / \Delta E$ and $\Delta E = I n / N$ is the exchange splitting.

It appears that Izuyama's is the correct result. D_2 is positive, in contrast with the equivalent result for the Heisenberg model, and is independent of whether we have electrons or holes. The sign of D_2 is not significant since we have used an approximate expression for $A_{\underline{q}}^+$ and have neglected the second term in (2.61).

An estimate of D_2 has been made for nickel, using (2.69), by assuming that the band structure of nickel consists of six groups of holes of effective mass $5.5m$, exchange splitting $.4\text{ev}$, $\varepsilon_F = .27\text{ev}$, $n = 0.6$ (Herring 1966b) and neglecting the interaction between electron-hole pairs in different groups. Then

$$D_2 / D_0 = 6 \times 10^{-9}$$

This would be increased up to sixfold if the interaction between holes in different groups were included.

We shall now estimate the size of the second term in (2.61). From (2.60), using (2.68),

$$\begin{aligned} \underline{q} \cdot \underline{k}_q^+ A_{\underline{p}}^+ |0\rangle &= [H, A_{\underline{q}}^+] A_{\underline{p}}^+ |0\rangle \\ &= \sum_{\underline{k}, \underline{k}'} -\frac{I}{Nn} \left(\frac{\underline{k} \cdot \underline{q}}{m \Delta E} \right) \left(\frac{\underline{k}' \cdot \underline{p}}{m \Delta E} \right) \sum_{\underline{q}_1} \left(C_{\underline{k}'+\underline{p}, \downarrow}^+ C_{\underline{k}-\underline{q}_1, \uparrow}^+ C_{\underline{k}+\underline{q}-\underline{q}_1, \downarrow}^+ C_{\underline{k}, \uparrow}^+ + C_{\underline{k}+\underline{q}, \downarrow}^+ C_{\underline{k}-\underline{q}_1, \uparrow}^+ C_{\underline{k}'+\underline{p}-\underline{q}_1, \downarrow}^+ C_{\underline{k}', \uparrow}^+ \right) \end{aligned} \quad (2.70)$$

so that

$$\lim_{\omega \rightarrow 0} \langle\langle K_0; K_0^+ \rangle\rangle_{\omega} = -\frac{4}{25n} \left(\sum_{\mathbf{p}} p^2 n_{\mathbf{p}} \right) \frac{\hbar^4 \epsilon_F \lambda}{(m \Delta \epsilon)^2} \quad (2.71)$$

by averaging over momentum transfers involving $\underline{q}_1$, and assuming that the electrons are non-interacting within the approximation used in (2.62). As $\langle\langle K_0; K_0^+ \rangle\rangle_{\omega}$ is a Green's function involving two spin flips created on the ground state, many more diagrams than those discussed by Edwards (1967) in connection with (2.58) could be included. Now there is the possibility of self-energy corrections for up spin electrons or holes, such as those discussed by Izuyama and Kubo (1964) in connection with spin wave interactions. Proceeding with (2.69) and (2.71) in (2.61) gives

$$D_2 T^{5/2} = \frac{1}{n} \frac{4}{15} \left(1 - \frac{33}{35} \lambda \right) \frac{\hbar^4 \epsilon_F}{(m \Delta \epsilon)^2} \sum_{\mathbf{p}} p^2 n_{\mathbf{p}} \quad (2.72)$$

For a strong ferromagnet $\lambda < 1$, so this expression is still positive but possibly by only a small amount. $\lambda = 0.7$ for nickel.

No experimental value for D_2/D_0 has been quoted for nickel, but for iron, Phillips (1966) finds $+1 \times 10^{-8}$ and Stringfellow (1968) finds $+2 \times 10^{-7}$. Therefore both experimentally and theoretically D_2/D_0 is small, of order at least 10^{-2} , so that the temperature dependence of D in the itinerant model is quite different from that in the

Heisenberg model.

The simplest process giving a non-zero contribution to $\text{Im } \chi_J'(\underline{q}, D_0 q^2)$ for a one-band model of a ferromagnet at $T=0$, is the process in which the state $\langle 0 | c_{\underline{k}\uparrow}^+ c_{\underline{k}+\underline{q}\downarrow}$ scatters into the state

$$|\underline{k}_1, \underline{q}_1\rangle = \left(\sum_{\underline{k}_2} f(\underline{k}_2, \underline{q}-\underline{q}_1) c_{\underline{k}_2+\underline{q}-\underline{q}_1\downarrow}^+ c_{\underline{k}_2\uparrow} \right) c_{\underline{k}_1+\underline{q}_1\uparrow}^+ c_{\underline{k}_1\uparrow} |0\rangle \quad (2.73)$$

which is a state consisting of a spin wave with wave vector $\underline{q}-\underline{q}_1$, with an electron excited from an occupied state $\psi_{\underline{k}_1\uparrow}$ of the up spin band to an unoccupied state $\psi_{\underline{k}_1+\underline{q}_1\uparrow}$ of the up spin band. $\text{Im } \chi_J'(\underline{q}, D_0 q^2)$ is non-zero for this process, because energy is conserved, so spin waves have a finite lifetime given by (2.15). One finds

$$\text{Im } \chi_J'(\underline{q}, D_0 q^2) = -\frac{\pi}{n} \sum_{\underline{k}, \underline{q}_1} \left(\frac{\underline{k} \cdot \underline{q}}{m} \right)^2 f_{\underline{k}\uparrow} (1 - f_{\underline{k}+\underline{q}_1\uparrow}) \delta(D_0 q^2 - D_0 (\underline{q}-\underline{q}_1)^2 - \epsilon(\underline{k}+\underline{q}_1) + \epsilon(\underline{k})) \quad (2.74)$$

where the matrix elements in (2.74) contribute by amount

$$|\langle 0 | c_{\underline{k}\uparrow}^+ c_{\underline{k}+\underline{q}\downarrow} | \underline{k}_1, \underline{q}_1 \rangle|^2 = \frac{\delta_{\underline{k}_1, \underline{k}}}{n} (1 + O(q-q_1)) \quad (2.75)$$

Putting (2.74) in (2.15) corresponds exactly with the expression Thompson (1965) obtained for the lifetime τ of spin waves at $T=0$ for a strong ferromagnet. Thompson

found $\tau^{-1} \sim q^6$ so that the spin wave is well defined. All the arguments of this section have been restricted to magnon-magnon interactions in a strong ferromagnet.

Ma, Seal-Monod and Fredkin (1968) find $\tau^{-1} \sim q^2$ for a nearly ferromagnetic material in a magnetic field in an approximation going beyond the R.P.A. approximation. The lifetime of spin waves in a weak ferromagnet is still an open question, though Izuyama (1971) claims τ goes as a higher power than q^2 . Thus spin waves are not so well defined and the phenomenological arguments of Marshall, and Izuyama and Kubo, particularly in connection with the magnon-magnon interaction, are not obviously correct for a weak ferromagnet.

Chapter 3

The Heisenberg Antiferromagnet

I Exact Results

In order to use (2.17) as a model of a ferromagnet, one must assume that the exchange coupling J is positive. However one can also consider the problem of a system with Hamiltonian H , defined by (2.17), with $J < 0$. For example, the hydrogen molecule H_2 is often used to illustrate the exchange interaction between electrons and for the hydrogen molecule, the interatomic exchange interaction J between the two electrons, is negative. If J is negative, semi-classical or mean field arguments suggest that one divides the lattice into two sublattices A and B , on which the direction of spin magnetisation differs. We now define $J = |J|$ to be the exchange interaction between electrons on nearest neighbour atoms, which are in different sublattices and then H in (2.17) becomes

$$H = \frac{1}{N} \sum_{\underline{k}} J(\underline{k}) S_{A\underline{k}} \cdot S_{B-\underline{k}} \quad (3.1)$$

with

$$S_{A,B\underline{q}} = \sum_{i \in A,B} e^{i\underline{q} \cdot \underline{R}_i} S_i \quad (3.2)$$

N is the number of atoms in each sublattice.

The exact ground state for this Hamiltonian is only

known for a one-dimensional magnet, or linear chain (Bethe 1931, Hulthen 1938, des Cloizeaux and Pearson 1962). The exact results are confirmed by theorems (Mattis 1965), valid in three dimensions as well, which state that the ground state is a non-degenerate singlet state. In other words, if S is the quantum number of the total spin operator $\sum_i S_i$, $S = \sum_i S_i^z = 0$ in the ground state.

These results contradict the semi-classical approach which assumes that deviations from the classical ground state, in which all spins on A are up and all spins on B are down, are small. This theory, called the linear theory, was first introduced by Anderson (1952), who explained how the discrepancy occurs.

The ground state of the linear theory is not the exact ground state of the isotropic antiferromagnet. Furthermore, an anisotropy field must be introduced into the Hamiltonian (3.1) to prevent the linear theory breaking down because of a divergence in the amplitudes of the spin wave creation operators α_q^+ , β_q^+ as $q \rightarrow 0$. The choice of direction of this magnetic field is the only thing that fixes the direction of the sublattice magnetisation, so that in the linear theory there is a set of approximately degenerate ground states. As the magnitude of the anisotropy field need only be infinitesimal for a three dimensional magnet, the presence of this set of degenerate states near to the exact

ground state makes the exact ground state irrelevant (Anderson 1964).

Without the anisotropy field, any one of the ground states of the linear theory could reverse its direction of sublattice magnetisation by the emission or absorption of zero wavelength spin waves of zero energy. This merely expresses the fact that the system is rotationally invariant and, after a very long time, will drift around to a reverse spin direction. So, as long as there is a small anisotropy field present, this behaviour can be neglected and the linear theory used. It should be emphasized that the ground state of the linear theory is not the classical ground state and the linear theory, valid for $T=0$ and $T \neq 0$, applies only to a three dimensional antiferromagnet (Ziman 1965).

II Linear Theory

To derive the consequences of the Hamiltonian (3.1), we will consider the generalised susceptibility

$$\chi_{\mu\nu}^{+-}(\underline{q}, \omega) = \ll S_{-\underline{q}\mu}^{+}; S_{\underline{q}\nu} \gg_{\omega} \quad (3.3)$$

where $(\mu, \nu) = (A, A), (A, B), (B, A), (B, B)$ and $S_{A\underline{q}}^{-}, S_{B\underline{q}}^{-}$ are defined by (3.2). Then to find the equations of motion of χ using (3.1), one must evaluate

$$[H, S_{A\underline{q}}^-] = \frac{1}{N} \sum_{\underline{k}} J(\underline{k}) (S_{B\underline{k}}^- S_{A-\underline{k}+\underline{q}}^z - S_{A-\underline{k}+\underline{q}}^- S_{B\underline{k}}^z) \quad (3.4)$$

and $[H, S_{B\underline{q}}^-]$ is obtained by interchanging A and B in (3.4). Following the arguments of (2.26) we write the spin operators in terms of boson operators $b_{\underline{k}}, b_{\underline{k}}^+, a_{\underline{k}}, a_{\underline{k}}^+$, only keeping products of a small number of operators. Then

$$\begin{aligned} S_{A\underline{k}}^z &= sN\delta_{\underline{k},0} - \frac{1}{2sN} \sum_{\underline{k}_1} a_{\underline{k}_1+\underline{k}}^+ a_{-\underline{k}_1}, \\ S_{B\underline{k}}^z &= -sN\delta_{\underline{k},0} + \frac{1}{2sN} \sum_{\underline{k}_1} b_{\underline{k}_1+\underline{k}}^+ b_{-\underline{k}_1}, \end{aligned} \quad (3.5)$$

$$S_{A\underline{k}}^- = (2s)^{\frac{1}{2}} a_{\underline{k}}^+, \quad S_{A\underline{k}}^+ = (2s)^{\frac{1}{2}} a_{\underline{k}} \quad (3.6)$$

$$S_{B\underline{k}}^- = (2s)^{\frac{1}{2}} b_{\underline{k}}^+, \quad S_{B\underline{k}}^+ = (2s)^{\frac{1}{2}} b_{\underline{k}} \quad (3.7)$$

The linear theory of Anderson (1952) and Kubo (1952) arises by keeping only the first terms in (3.5), so that the equations of motion of involve no higher order Green's function. One finds

$$\begin{pmatrix} \omega - H_{an}s - J(0)s & -J(\underline{q})s \\ J(\underline{q})s & \omega + H_{an}s + J(0)s \end{pmatrix} \chi_{\mu\nu} = 2Ns \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} \quad (3.8)$$

$$\text{where } \chi_{\mu\nu} = 2s \ll \begin{pmatrix} a_{\underline{q}} \\ b_{\underline{q}}^+ \end{pmatrix}; \begin{pmatrix} a_{\underline{q}}^+ \\ b_{\underline{q}} \end{pmatrix} \gg_{\omega} \quad (3.9)$$

and we have included a weak anisotropy field $-H_{an}s(S_{A0}^z - S_{B0}^z)$

(51)

into the Hamiltonian (3.1). For the time being we put $H_{0h} = 0$. There exists a unitary transformation U defined by (Herbert 1969, Mattis 1965)

$$\begin{aligned} U^\dagger \begin{pmatrix} a_{\underline{q}}^+ \\ b_{\underline{q}} \end{pmatrix} U &= \begin{pmatrix} \cosh \frac{1}{2} \theta_{\underline{q}} a_{\underline{q}}^+ + \sinh \frac{1}{2} \theta_{\underline{q}} b_{\underline{q}} \\ \cosh \frac{1}{2} \theta_{\underline{q}} b_{\underline{q}} + \sinh \frac{1}{2} \theta_{\underline{q}} a_{\underline{q}}^+ \end{pmatrix} \\ &= \begin{pmatrix} \alpha_{\underline{q}}^+ \\ \beta_{\underline{q}} \end{pmatrix} = T \begin{pmatrix} a_{\underline{q}}^+ \\ b_{\underline{q}} \end{pmatrix} \end{aligned} \quad (3.10)$$

or

$$\begin{pmatrix} a_{\underline{q}}^+ \\ b_{\underline{q}} \end{pmatrix} = T^{-1} \begin{pmatrix} \alpha_{\underline{q}}^+ \\ \beta_{\underline{q}} \end{pmatrix}$$

where $\tanh \theta_{\underline{q}} = -J(\underline{q})/J(0)$ (3.11)

which diagonalises (3.9) for each $\underline{q}$, so that

$$T \chi_{\omega}(\underline{q}, \omega) T^\dagger = 2s \ll \begin{pmatrix} \alpha_{\underline{q}} \\ \beta_{-\underline{q}}^+ \end{pmatrix}; \begin{pmatrix} \alpha_{\underline{q}}^+ \\ \beta_{\underline{q}} \end{pmatrix} \gg_\omega \quad (3.12)$$

$$= 2Ns \begin{pmatrix} [\omega - \omega_s(\underline{q})]^{-1} & 0 \\ 0 & -[\omega + \omega_s(\underline{q})]^{-1} \end{pmatrix} \quad (3.13)$$

where $\omega_s(\underline{q}) = \sqrt{J(0)^2 - J(\underline{q})^2} s \simeq c_{\underline{q}}$ (3.14)

This theory would be exact if the ground state were the classical or Néel state $|0\rangle_N$, defined by

$$a_{\underline{q}}|0\rangle_N = 0 \quad b_{\underline{q}}|0\rangle_N = 0 \quad (3.15)$$

for all $\underline{q}$. However, the ground state for the linear theory is the state $|0\rangle_L$, defined by

$$\alpha_{\underline{q}} |0\rangle_L = 0, \quad \beta_{\underline{q}} |0\rangle_L = 0 \quad (3.16)$$

where $\alpha_{\underline{q}}$ and $\beta_{\underline{q}}$ are the spin wave destruction operators.

(3.16) is satisfied if we choose

$$|0\rangle_L = U^\dagger |0\rangle_N \quad (3.17)$$

so that

$$\alpha_{\underline{q}} |0\rangle_L = U^\dagger a_{\underline{q}} U U^\dagger |0\rangle_N = U^\dagger a_{\underline{q}} |0\rangle_N = 0 \quad (3.18)$$

Unfortunately $U^\dagger$ maps $|0\rangle_N$ onto non-physical states so that the expectation value $\langle X \rangle$ of any operator X in the ground state, must be written

$$\langle X \rangle = \frac{\langle 0 | P X | 0 \rangle_L}{\langle 0 | P | 0 \rangle_L} \quad (3.19)$$

where P is the projection operator onto the physical subspace of the boson space. P was defined by (2.27) in connection with the ferromagnet. Anderson's results (1952) are not quite correct as he uses the unprojected ground state. Herbert (1969) describes how Anderson's results should be modified.

If the unprojected ground state is used to find the sublattice magnetisation $\frac{1}{N} \langle S_{A0}^z \rangle$, using spin waves defined in the linear theory, one finds

$$\begin{aligned} \frac{1}{N} \langle S_{A0}^z \rangle &= S - \frac{1}{2SN^2} \left[\langle 0 | U \left(\sum_{\underline{k}} a_{\underline{k}}^\dagger a_{\underline{k}} \right) U^\dagger | 0 \rangle_N \right. \\ &\quad \left. + \sum_{\underline{f}} n_{\underline{f}} \left\{ \langle 0 | \alpha_{\underline{f}} \left(\sum_{\underline{k}} a_{\underline{k}}^\dagger a_{\underline{k}} \right) \alpha_{\underline{f}}^\dagger | 0 \rangle_L + \langle 0 | \beta_{\underline{f}} \left(\sum_{\underline{k}} a_{\underline{k}}^\dagger a_{\underline{k}} \right) \beta_{\underline{f}}^\dagger | 0 \rangle_L \right\} \right] \quad (3.20) \end{aligned}$$

where $n_{\underline{p}}$ are boson occupation numbers. Then using (3.10) and (3.17)

$$\frac{1}{N} \langle S_{A0}^2 \rangle = s - \frac{1}{2sN} \left[\sum_{\underline{k}} \left(\cosh^2 \frac{1}{2} \theta_{\underline{k}} + n_{\underline{k}} \cosh \theta_{\underline{k}} \right) \right] \quad (3.21)$$

Using (3.11) one finds for $s = \frac{1}{2}$

$$\frac{1}{N} \langle S_{A0}^2 \rangle = \frac{1}{2} - \frac{1}{2N} \sum_{\underline{k}} \left\{ \left(-1 + \frac{J(0)}{2\omega_S(\underline{k})} \right) + n_{\underline{k}} \frac{J(0)}{\omega_S(\underline{k})} \right\} \quad (3.22)$$

This is the linear spin wave result of Anderson and Kubo. Stinchcombe and Cottam (1970) obtain the same result by using a drone fermion representation of the spin operators for spin $\frac{1}{2}$, expanding in powers of $1/z$, where z is the number of spins interacting with any given spin and keeping terms to order $(1/z)^1$. Mean field theory would correspond to keeping terms to order $(1/z)^0$. Vaks, Larkin and Pikin (1968) used the same idea for ferromagnets with a diagram technique, expanding in powers of $1/r_0^3$, where r_0 is the mean interaction radius and Pikalev et al (1969) and Solyom (1969) adapted the idea for antiferromagnets.

(3.22) is important as it illustrates the following points (Ziman 1965). The summation in (3.22) diverges at $\underline{k} = 0$, as $\omega_S(0) = 0$. The presence of an anisotropy field H_{an} means, from (3.8) and (3.14)

$$\omega_S(0) = \sqrt{J(0) H_{an}} \, s \quad (3.23)$$

With H_{an} of order $1/N$ the divergence is removed, so that the anisotropy field need only be infinitesimal. However, even with this infinitesimal anisotropy

$$\int_0^K (1/c_k) dk$$

diverges in one dimension and

$$\int (n_k/c|k|) dk$$

diverges in two dimensions, so that the linear theory only applies to a three dimensional antiferromagnet. The statements made in section I are therefore justified. It is also clear from (3.22) that the sublattice magnetisation never reaches its maximum value, even at $T=0$. Anderson (1952) found that the correction term in (3.22), at $T=0$, was .078 for simple cubic lattices with nearest neighbour interactions.

Another way of obtaining the spin wave spectrum (3.14), going beyond linear theory but still not excluding the non-physical states entirely, as shown in (3.19), is to consider the equations of motion of $\chi_{\mu\nu}$. Using (2.5) to (3.7), (3.4) can be written

$$\begin{aligned} \left[H, \begin{pmatrix} a_{\underline{q}}^+ \\ b_{\underline{q}} \end{pmatrix} \right] &= \begin{pmatrix} J_{a\underline{q}}^+ \\ J_{b\underline{q}} \end{pmatrix} = \begin{pmatrix} J(0) \\ -J(\underline{q}) \end{pmatrix} s \begin{pmatrix} a_{\underline{q}}^+ \\ b_{\underline{q}} \end{pmatrix} + \begin{pmatrix} J(\underline{q}) \\ -J(0) \end{pmatrix} s \begin{pmatrix} b_{\underline{q}} \\ a_{\underline{q}}^+ \end{pmatrix} \\ &- \frac{1}{2sN^2} \sum_{\underline{k}} \begin{pmatrix} J(\underline{k}) \\ -J(\underline{k}-\underline{q}) \end{pmatrix} \left(b_{\underline{k}} \sum_{\underline{k}_1} a_{\underline{k}_1-\underline{k}+\underline{q}}^+ a_{\underline{k}_1} + a_{\underline{k}+\underline{q}}^+ \sum_{\underline{k}_1} b_{\underline{k}_1+\underline{k}} b_{\underline{k}_1} \right) \end{aligned} \quad (3.24)$$

so that the equation of motion of $\chi_{\mu\nu}$, analogous to (2.6), is

$$\omega^2 \chi_{\mu\nu} = \chi_{\mu\nu}^T + \left\langle \left[\begin{pmatrix} a_{\underline{q}} \\ b_{\underline{q}}^+ \end{pmatrix}, \begin{pmatrix} J_{a\underline{q}}^+ \\ J_{b\underline{q}} \end{pmatrix} \right] \right\rangle + \omega N \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} \quad (3.25)$$

(55)

where $\chi_{\mu\nu}^J = \langle\langle J_{\mu-\underline{q}}; J_{\nu\underline{q}} \rangle\rangle_\omega$ and with $s = \frac{1}{2}$. Neglecting $\chi_{\mu\nu}^J$ (3.25) reduces to the form

$$\chi_{\mu\nu} = \omega^{-2} \begin{pmatrix} a + dq^2 & -a \\ -a & a + dq^2 \end{pmatrix} + \omega^{-1} N \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} \quad (3.26)$$

in the limit $q/\omega \ll 1$, where

$$a = J(0)Ns - \frac{1}{2sN} \sum_{\underline{k}} (J(\underline{k}) \langle b_{\underline{k}} a_{\underline{k}} \rangle + J(0) \langle a_{\underline{k}}^+ a_{\underline{k}} \rangle) \quad (3.27)$$

$$\text{and } dq^2 = (1 - J(\underline{q})/J(0)) a, \quad (3.28)$$

using the result (Callen 1963)

$$J(0) \sum_{\underline{q}} J(\underline{k}-\underline{q}) f(\underline{q}) = \sum_{\underline{q}} J(\underline{k}) J(\underline{q}) f(\underline{q}) \quad (3.29)$$

if $f(\underline{q})$ depends only on $|\underline{q}|$. a can be evaluated in a similar way to (3.20), using (3.10) and (3.17), giving

$$a = J(0)Ns (1 + a_1) \quad (3.30)$$

$$a_1 = \frac{1}{N} \sum_{\underline{k}} \left(1 - \frac{2\omega_s(\underline{k})}{J(0)} (1 + 2\eta_{\underline{k}}) \right) \quad (3.31)$$

One can compare (3.26) with (3.13), in the limit $q/\omega \ll 1$. To obtain (3.13), a transformation T was found to diagonalise $\chi_{\mu\nu}$ in the linear theory. In general we assume $\chi_{\mu\nu}$ has the diagonal form

$$\chi_{\mu\nu} = 2Ns \begin{pmatrix} [\omega - cq]^{-1} & 0 \\ 0 & -[\omega + cq]^{-1} \end{pmatrix} = 2Ns \begin{pmatrix} \bar{\omega}^{-1} + \bar{\omega}^{-2} cq & 0 \\ 0 & \bar{\omega}^{-1} + \bar{\omega}^{-2} cq \end{pmatrix} \quad (3.32)$$

in a suitably chosen basis. Then from (3.26) and (3.32)

$$(a + dq^2 + N\omega)(a + dq^2 - N\omega) - a^2 = N^2(c^2 q^2 - \omega^2) \quad (3.33)$$

so that $c = 1/N \sqrt{2ad}$. (3.34)

From (3.31) $cq = \sqrt{2J(0)(J(0) - J(q))}s(1+a_1)$ to $O(q)$ (3.35)

a_1 gives the first correction to the spin wave energy due to spin wave interactions (Pikalev, Savchenko and Solyom 1969). It corresponds to taking diagrams of order $(1/s)^2$ in Stinchcombe and Cottam's (1970b) method.

If $\chi_{\mu\nu}^T(q, \omega)$ were retained in (3.25) a and d would be renormalised and would possibly be complex numbers, but would have the same q and ω dependence as (3.26) in the limit $q/\omega \ll 1$. The possibility that c has an imaginary part corresponding to a spin wave lifetime of order q^{-1} may now arise. Both Solyom (1969) and Stinchcombe and Cottam (1970b) and (1970c) calculate the contribution to the lifetime from the scattering of spin waves with each other, which is dominant at low temperatures, and the scattering of spin waves by fluctuations in S^z , which is dominant at higher

temperatures and exponentially small at low temperatures. They find that the inverse spin wave lifetime always goes as a higher power than q in their approximations, but the lifetime is finite of order q^{-4} even at $T = 0$, because of the zero point oscillations of the ground state.

The results of this section suggest that the method of chapter 2, which applies to any ferromagnet, can be generalised to apply to any antiferromagnet. As we have seen, much work has been done on spin waves in Heisenberg antiferromagnets. Very little has been done on the corresponding problem in a band model. This problem will be dealt with in the next chapter.

Chapter 4

Antiferromagnetism in the Band Model

I Oscillatory Spin Polarisation in the Free Electron Gas

It is easy to see that different choices for the signs and values of the constants J_{ij} in the localised spin Hamiltonian $-\sum_{ij} J_{ij} \underline{S}_i \cdot \underline{S}_j$ lead to different spin density wave configurations in the ground state of the Hamiltonian. Similar spin density wave configurations also occur in the itinerant electron model (Herring 1966). There are three basic spin density configurations (Arrott 1966): the longitudinal linear spin density wave of the form

$$\langle \sigma^{\alpha}(r) \rangle = (0, 0, A \cos qz), \quad (4.1)$$

the transverse linear spin density wave of the form

$$\langle \sigma^{\alpha}(r) \rangle = (0, B \cos qz, 0), \quad (4.2)$$

and the spiral spin density wave of the form

$$\langle \sigma^{\alpha}(r) \rangle = C (\cos qz, \sin qz, 0) \quad (4.3)$$

The term antiferromagnet is usually restricted to spin density waves in which q is an integral multiple of π .

reciprocal lattice vector.

Clearly spiral spin density waves with wave vector $\underline{q}$ are invariant under the group of transformations $T(\underline{t})R(\underline{q},\underline{t})$, where $T(\underline{t})$ is a translation through the lattice vector $\underline{t}$ and $R(\underline{q},\underline{t})$ is a rotation of spin through an angle $\underline{q} \cdot \underline{t}$ about some axis. The one electron Hamiltonian in the band model commutes with transformations of this type, so in analogy with Bloch's theorem, one can define one electron wavefunctions $\phi_{\underline{k}}$, which satisfy

$$T(\underline{t})R(\underline{q},\underline{t})\phi_{\underline{k}} = e^{-i\underline{k} \cdot \underline{t}} \phi_{\underline{k}} \quad (4.4)$$

If the z-axis is the axis of rotation

$$R(\underline{q},\underline{t}) = e^{i\frac{1}{2}(\underline{q} \cdot \underline{t})\sigma_z} = \begin{pmatrix} e^{i\frac{1}{2}(\underline{q} \cdot \underline{t})} & 0 \\ 0 & e^{-i\frac{1}{2}(\underline{q} \cdot \underline{t})} \end{pmatrix} \quad (4.5)$$

so that

$$\phi_{\underline{k}} = \cos \theta_{\underline{k}} \psi_{\underline{k}+\frac{1}{2}\underline{q}\uparrow} + \sin \theta_{\underline{k}} \psi_{\underline{k}-\frac{1}{2}\underline{q}\downarrow}, \quad (4.6)$$

where $\psi_{\underline{k} \pm \frac{1}{2}\underline{q}}$ are wavefunctions of the non-magnetic system.

It is therefore natural to ask whether a determinant of wavefunctions of the form (4.6) have a lower energy in the Hartree-Fock approximation, than a determinant corresponding to a paramagnetic or ferromagnetic ground state.

Overhauser (1959, 1960a) first discussed this problem

in connection with impurity spins in an itinerant electron gas. He later (1966b) suggested that the Hartree-Fock ground state of a one dimensional free electron gas is a spin density wave, however weak the interactions are. This is seen easily because the non-interacting susceptibility $\chi^0(q)$ of the free electron gas becomes infinite as $q \rightarrow 2k_F$. So an arbitrarily weak magnetic field of wave vector $2k_F$ produces an instability in the non-magnetic state, suggesting that a spin density wave is more stable. The divergence in $\chi^0(q)$ occurs because of the very large contribution of the term $n_k - n_{k+q} / \epsilon(k+q) - \epsilon(k)$ in $\chi^0(q)$, when $q = 2k_F$ and k is near the Fermi surface, for a one dimensional system. This effect is referred to as the nesting of energy bands and arises whenever the shifting part of the Fermi surface through a wave vector q brings a large area of contact with another part of the Fermi surface. When it occurs, the coupling of electrons into wavefunctions

$$\cos \theta_k \psi_{k\uparrow} + \sin \theta_k \psi_{k+q\downarrow} \quad \text{or} \quad \cos \theta_k \psi_{k\uparrow} + \sin \theta_k \psi_{k+q\uparrow}$$

will reduce energy even for very weak interactions. Nesting is discussed again in section II and chapter 5 for general band structures, with particular reference to chromium.

For two and three dimensions this divergence does not occur in $\chi^0(q)$. However Overhauser (1962) proved that for

the free electron gas in three dimensions with Coulomb interactions, treated in the Hartree-Fock approximation, the spin density wave state of wave vector $2k_F$ has lower energy than the paramagnetic state. So the interacting susceptibility $\chi(q)$ diverges at $q = 2k_F$ for the unscreened Coulomb interaction. In contrast Izamoto and Sawada (1962) and Fedders and Martin (1966) have made a strong case that the electron gas with screened Coulomb interactions, is not unstable towards antiferromagnetism.

In sections II and III we discuss the consequences of the Slater (1951) alternant molecular orbital model of a one-band antiferromagnet. For this model the lattice can be considered the sum of two sub-lattices, on which the Hartree-Fock exchange field and the spin magnetisation have different directions.

II The Short-range One-band Model

In the simple one-band itinerant electron model of a metal which we will consider, all kinds of magnetic structures are possible. The metal has an antiferromagnetic ordering, or a spin density wave, if its ground state has a periodic spin density which is not the same as the electron density in the lattice.

Two cases when this is likely to occur are well-known:

(1) a particular sort of band structure in which nesting occurs (Lomer 1962). Antiferromagnetism can then occur with quite a small interaction and the resulting antiferromagnetic energy gap is also small. The first quantitative treatment was given by Fedders and Martin (1966), who suggest that this sort of band structure is unlikely to occur in a single band. Parts of two bands, which nest together, form the driving force towards antiferromagnetism though all electrons contribute to the spin polarisation (Asano and Yamashita 1967). However, the treatment for a single band is almost identical to that of a two-band model. Band structure calculations have been made on paramagnetic and antiferromagnetic pure chromium which confirm these ideas fairly well (Loucks 1965, Gupta and Sinha 1970, 1971).

(2) an electron density of nearly one electron per site and a large interaction energy. The limit of one electron per site with a strong repulsion I between electrons on the same site leads to the Heisenberg antiferromagnet. The Heisenberg Hamiltonian arises in second order perturbation theory when hopping from site to site is allowed. In these cases the antiferromagnetic energy gap is wide. The many band generalisation of a model with a wide gap may apply to γ -manganese (Haywood, Lowde et al 1971, Fletcher 1971).

The periodicity $\underline{Q}$ of the spin density may be incommensurate with the reciprocal lattice of the solid. For example, pure chromium has a ground state consisting of three spin density waves with wave vectors $\underline{Q}_x, \underline{Q}_y, \underline{Q}_z$ close to the points $(1, 0, 0), (0, 1, 0), (0, 0, 1)$ of the reciprocal lattice. This structure is complicated by the possibility of second order gaps (Herring 1966) and interactions between the spin density waves, unless the antiferromagnetic energy gap is very much smaller than typical band energies (Asano and Yamashita 1967 and chapter 5 of this thesis). Larger energy gaps may well drive the metal commensurate, so we will assume that we have an antiferromagnetic structure in which the direct lattice can be considered the sum of two sublattices, A and B, with different spin polarisations. This reduces symmetry and for a b.c.c. lattice, the magnetic reciprocal lattice will contain the additional points $(1, 0, 0), (0, 1, 0), (0, 0, 1)$, plus the crystal reciprocal lattice vectors, changing it from a f.c.c. lattice to a s.c. lattice. In chromium the addition of 2% manganese sends $\underline{Q}_x$ from .96(1, 0, 0) to (1, 0, 0) allowing us to adopt this simplification (Hamaguchi, Wollan and Koehler 1964).

We can define Bloch functions $\psi_A(\underline{k}), \psi_B(\underline{k})$ of a tight-binding form on each sublattice separately. This

representation is related to the representation of bands in a paramagnet by

$$\psi(\underline{k}) = \frac{1}{\sqrt{2}}(\psi_A(\underline{k}) + \psi_B(\underline{k})) \quad , \quad \psi(\underline{k}+\underline{Q}) = \frac{1}{\sqrt{2}}(\psi_A(\underline{k}) - \psi_B(\underline{k}))$$

$\psi_A(\underline{k})$ can be written in a Wannier representation as

$$\psi_A(\underline{k}) = \frac{1}{\sqrt{N}} \sum_i e^{i\mathbf{k} \cdot \mathbf{R}_i} \omega(\underline{r} - \underline{R}_i)$$

where the summation is over lattice points in the sublattice

A. The well-known Hamiltonian

$$H = \sum_{\underline{k}, \sigma} \varepsilon(\underline{k}) n_{\underline{k}\sigma} + I \sum_i n_{i\uparrow} n_{i\downarrow} \quad (4.7)$$

where I is the interaction energy of two electrons on the same Wannier function, may be written

$$H = \sum_{\underline{k}, \sigma} \varepsilon_1(\underline{k}) (n_{A\underline{k}\sigma} + n_{B\underline{k}\sigma}) - \varepsilon_2(\underline{k}) (c_{B\underline{k}\sigma}^\dagger c_{A\underline{k}\sigma} + c_{A\underline{k}\sigma}^\dagger c_{B\underline{k}\sigma}) + I \sum_{i \in A, B} n_{i\uparrow} n_{i\downarrow} \quad (4.8)$$

$\varepsilon_1(\underline{k})$ and $\varepsilon_2(\underline{k})$ are defined in terms of the original band energies by

$$\varepsilon_1(\underline{k}) = \frac{1}{2}(\varepsilon(\underline{k}+\underline{Q}) + \varepsilon(\underline{k})) \quad , \quad \varepsilon_2(\underline{k}) = \frac{1}{2}(\varepsilon(\underline{k}+\underline{Q}) - \varepsilon(\underline{k}))$$

and the operators in the first two terms of (4.8) refer to sublattice Bloch states. The second term in (4.8) describes the hopping between the two sublattices.

III Single Particle States and the Ground State

The Hartree-Fock solution to this problem is well-known (Herring 1966). It may be obtained by solving the equations of motion of the one electron Green's function $G_{\alpha\beta}^{\sigma}(\underline{k}, \omega)$ approximately. $G_{\alpha\beta}^{\sigma}(\underline{k}, \omega)$ has the matrix form defined by

$$G_{\alpha\beta}^{\sigma}(\underline{k}, \omega) = \langle\langle c_{\alpha\underline{k}\sigma}; c_{\beta\underline{k}\sigma}^{\dagger} \rangle\rangle_{\omega}$$

where $(\alpha, \beta) = (A, A), (A, B), (B, A)$ and (B, B) , are the elements of the matrix.

The equations of motion can be written

$$W G_{\alpha\beta}^{\uparrow}(\underline{k}, \omega) = \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix} \quad (4.9)$$

$$\text{or} \quad G_{\alpha\beta}^{\uparrow}(\underline{k}, \omega) = W^{-1} \quad (4.10)$$

where

$$W = \begin{pmatrix} \omega - \epsilon_1(\underline{k}) - \text{In} n_{A\downarrow}/N & \epsilon_2(\underline{k}) \\ \epsilon_2(\underline{k}) & \omega - \epsilon_1(\underline{k}) - \text{In} n_{B\downarrow}/N \end{pmatrix}$$

where $n_{A\downarrow}/N$ and $n_{B\downarrow}/N$ are the mean number of down spin electrons per site on sublattices A and B. We obtain new quasiparticle creation operators $\hat{d}_{\underline{k}1\uparrow}^{\dagger}, \hat{d}_{\underline{k}2\uparrow}^{\dagger}$ with energies $\epsilon_1(\underline{k}), \epsilon_2(\underline{k})$ by finding a transformation T which diagonalises W . T satisfies

$$TT^{\dagger} = I, \quad TWT^{\dagger} = \begin{pmatrix} \omega - \epsilon_1(\underline{k}) & 0 \\ 0 & \omega - \epsilon_2(\underline{k}) \end{pmatrix} \quad (4.11)$$

(66)

where $E_2'(k) = E_1(k) \mp \sqrt{g^2 + E_2^2(k)}$ (4.12)

and $\begin{pmatrix} d_{k1\uparrow}^+ \\ d_{k2\uparrow}^+ \end{pmatrix} = T \begin{pmatrix} c_{Ak\uparrow}^+ \\ c_{Bk\uparrow}^+ \end{pmatrix} = \begin{pmatrix} \cos \frac{1}{2} \theta_k c_{Ak\uparrow}^+ + \sin \frac{1}{2} \theta_k c_{Bk\uparrow}^+ \\ \cos \frac{1}{2} \theta_k c_{Bk\uparrow}^+ - \sin \frac{1}{2} \theta_k c_{Ak\uparrow}^+ \end{pmatrix}$ (4.13)

with $\tan \theta_k = E_2(k)/g$ and $g = \frac{1}{2} \langle n_{A\uparrow} - n_{A\downarrow} \rangle I/N$. Then (4.9) is equivalent to

$$TWT^+ \ll \begin{pmatrix} c_{Ak\uparrow} \\ c_{Bk\uparrow} \end{pmatrix} T^+; T \begin{pmatrix} c_{Ak\uparrow}^+ \\ c_{Bk\uparrow}^+ \end{pmatrix} \gg_\omega = \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix} \quad (4.14)$$

or $\ll \begin{pmatrix} d_{k1\uparrow} \\ d_{k2\uparrow} \end{pmatrix}; \begin{pmatrix} d_{k1\uparrow}^+ \\ d_{k2\uparrow}^+ \end{pmatrix} \gg_\omega = \begin{pmatrix} [\omega - E_1(k)]^{-1} & 0 \\ 0 & [\omega - E_2(k)]^{-1} \end{pmatrix}$ (4.15)

using (4.11), (4.12) and (4.13).

For down spin there exist quasiparticle creation operators $d_{k1\downarrow}^+, d_{k2\downarrow}^+$ with energy $E_1(k), E_2(k)$ defined by

$$\begin{pmatrix} d_{k1\downarrow}^+ \\ d_{k2\downarrow}^+ \end{pmatrix} = T^+ \begin{pmatrix} c_{Bk\downarrow}^+ \\ c_{Ak\downarrow}^+ \end{pmatrix} \quad (4.16)$$

The minimum gap between the bands $E_1(k), E_2(k)$ is $2g$, and g vanishes when the ordering vanishes. g is determined selfconsistently from (4.13) since

$$\langle n_{A\uparrow} - n_{A\downarrow} \rangle = \langle n_{A\uparrow} - n_{B\uparrow} \rangle = \sum_k \cos \theta_k (n_{k1\uparrow} - n_{k2\uparrow}) \quad (4.17)$$

so that $R = I/2N \sum_k \frac{n_{k1\sigma} - n_{k2\sigma}}{\sqrt{E_2^2(k) + g^2}} = 1$ (4.18)

$n_{k1\sigma}$ and $n_{k2\sigma}$ are occupation numbers chosen so that the energy at $T = 0$ or the free energy at $T \neq 0$ is a minimum.

One can see that the single particle energy is reduced from $\varepsilon(k)$ to $E_1(k)$ by spreading from the A onto the B sites and vice versa. It is not possible to solve the selfconsistency equations in general. The difficulty is choosing the Fermi surface to minimise the energy (Matsubara and Yokota 1954). To simplify the calculation des Cloizeaux (1959) assumed that the lower band was completely full. Kimball (1969) found the minimum of the free energy by numerical methods. The case of weak antiferromagnetism in nesting bands has been extensively investigated using model band structures (Fedders and Martin 1966, Liu 1980, Sokoloff 1969 a,b). All these are idealistic since they assume perfect nesting, which results in R diverging logarithmically as $g \rightarrow 0$. It is sufficient for $R > 1$, with g set equal to zero, for there to be antiferromagnetic ordering. This is a condition like the Stoner criterion in ferromagnetism.

For one electron per site and $\varepsilon/I = 0$, where 2ε is the bandwidth, the system is in the atomic limit of $2N$ independent atoms. With $\varepsilon/I \ll 1$, hopping between sites is possible but is very restricted. Within second order perturbation theory an effective Heisenberg interaction H_{eff} can be defined (Anderson 1964, Mattis 1965) which

includes the hopping. One has

$$H_{\text{eff}} = -b^2 \sum_{i,j} c_{i\sigma}^+ c_{j\sigma} \frac{1}{V} \sum_{i',j'} c_{i'\sigma'}^+ c_{j'\sigma'} \quad (4.19)$$

where b is the nearest neighbour hopping integral, i and j are nearest neighbours, and hopping between further sites has been neglected. After one hop to an adjacent site an electron hops back to its original site, and higher order terms are negligible since $\varepsilon/I \ll 1$. Defining spin operators

$$S_i^- = c_{i\downarrow}^+ c_{i\uparrow}, \quad S_i^z = \frac{1}{2} (c_{i\uparrow}^+ c_{i\uparrow} - c_{i\downarrow}^+ c_{i\downarrow}) \quad (4.20)$$

yields

$$H_{\text{eff}} = J \sum_{\substack{i < j \\ \text{n.n.}}} \underline{S}_i \cdot \underline{S}_j \quad (4.21)$$

where $J = 4b^2/I$. This has the same form as (1.1). J is positive so the coupling between adjacent sites is antiferromagnetic. The same results can be derived by using single particle Green's function, in the Hartree-Fock approximation, with $g = \frac{1}{2}I \gg \xi(\underline{k})$.

For the case of a nearly half-filled band, $N-n \ll N$, there is the possibility that g may be greatly modified. The dominant corrections are those diagrams (fig.A) in which a down spin electron scatters against up spin holes on the

A sublattice.

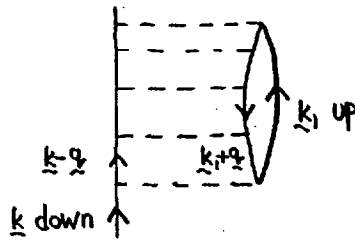


fig. A Dominant diagrams on sublattice A where there is a nearly full band of up spins.

Edwards (1967) has considered the same problem in connection with ferromagnetic spin waves, and finds the exchange splitting between up and down spin bands, when there is a low density of up spin electrons, to be of order

$$\frac{nI}{N} + \frac{I}{N} \left(\frac{nI}{N} \right) \frac{n'}{\epsilon}$$

where $n' = N - n$ and the first term is the exchange splitting in the Hartree-Fock approximation and the second term comes from summing diagrams of fig. A.

So the correction, when there is not exactly one electron per site, is small provided

$$1 \gg \epsilon/I \gg n'/N \quad (4.22)$$

When (4.22) is satisfied, one has a soluble limit which is near to the Heisenberg antiferromagnet. Nagaoka (1966) has investigated the nearly half-filled band and also obtains the condition (4.22) for antiferromagnetism. Hubbard's method (1963), which is an infinite expansion in powers of ϵ/I about the atomic limit, assumes a uniform spin

polarisation per site. Hewson and Lindner (1970) have shown that antiferromagnetism does not occur in Hubbard's simple decoupling, even for $I=\infty$. Presumably the Hubbard theory is correct to the alternative limit

$$1 \gg n'/N \gg \varepsilon/I. \quad (4.23)$$

IV Susceptibility and Spin Waves

Owing to the existence of two sublattices we investigate the reduced susceptibility

$$\chi_{\mu\nu}^{+-}(\underline{q}, \omega) = \langle\langle S_{\underline{q}\mu}^{+}; S_{\underline{q}\nu}^{-} \rangle\rangle_{\omega} \quad (4.24)$$

where $(\mu, \nu) = (A, A), (A, B), (B, A), (B, B)$. The spin density operators $S_{\underline{q}\mu}^{+}, S_{\underline{q}\nu}^{-}$ have their usual representation in terms of Bloch functions i.e.

$$S_{A\underline{q}}^{-} = \sum_{\underline{k}} c_{A\underline{k}+\underline{q}\downarrow}^{+} c_{A\underline{k}\uparrow}, \quad S_{A-\underline{q}}^{+} = \sum_{\underline{k}} c_{A\underline{k}-\underline{q}\uparrow}^{+} c_{A\underline{k}\downarrow} \quad (4.25)$$

Since $S_{\underline{q}}^{-} = \frac{1}{\sqrt{2}}(S_{A\underline{q}}^{-} + S_{B\underline{q}}^{-})$ commutes with H when $\underline{q} = 0$, it is reasonable to assume that χ^{+} will have a simple form containing just two spin wave poles, provided the Fermi level lies in the gap so that spin waves are the only low lying modes. A new orthogonal basis, defined by a transformation P , is

chosen so that the entries in the matrix χ^{+-} in (4.24) are linear combinations of S_{Aq}^- and S_{Bq}^- which generate spin waves. So, just as in (4.10), we choose a similarity transformation which diagonalises χ and separates the two spin wave branches. The transformed χ has the form

$$\bar{\chi}(q, \omega) = P \chi P^+ = \begin{pmatrix} [\omega - \omega_s(q)]^{-1} & 0 \\ 0 & -[\omega + \omega_s(q)]^{-1} \end{pmatrix} \Delta n \quad (4.26)$$

$$\text{or } \left\langle \left(\begin{matrix} S_{A-q}^+ \\ S_{B-q}^+ \end{matrix} \right) P^+ ; P \begin{pmatrix} S_{Aq}^- \\ S_{Bq}^- \end{pmatrix} \right\rangle_{\omega} = \omega^{-1} \begin{pmatrix} 1 + c_q/\omega & 0 \\ 0 & -1 + c_q/\omega \end{pmatrix} \Delta n_0 \quad (4.27)$$

We have assumed

$$\omega_s(q) = c_q + O(q^2), \quad \Delta n = \Delta n_0 + \Delta n_q$$

and have expanded in powers of q/ω with $q/\omega \ll 1$.

At $q = 0$ the transformation will become singular without the presence of a weak anisotropy field. Such a field introduces a term $-h(S_A^z - S_B^z)$ into the Hamiltonian, and stabilises the system against rotations of total spin. Then $\omega_s(0) \neq 0$, however when h is infinitesimal we can still assume ω_s is linear in q . Poles occur at $\pm \omega_s$ representing the creation of two branches of spin waves because the spins on sublattices A and B are in opposite directions. This behaviour is similar to that in the Heisenberg antiferromagnet.

We can compare this expression for $\bar{\chi}$ with the equations of motion of χ and obtain a formula for c . This is analogous

to a recent method of obtaining the spin wave energy of a ferromagnet (Edwards and Fisher 1971). The equations of motion of $\chi(\underline{q}, \omega)$ to the lowest order in q/ω are

$$\chi^{+-}(\underline{q}, \omega) = \omega^{-2} \begin{pmatrix} a + (d+b)q^2 & -a \\ -a & a + (d-b)q^2 \end{pmatrix} + \omega^{-1} \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} \langle n_{A\uparrow} - n_{A\downarrow} \rangle \quad (4.28)$$

a , b and d are given by

$$a = - \lim_{\omega \rightarrow 0} \lim_{q \rightarrow 0} a_{AB}, \quad b = \lim_{\omega \rightarrow 0} \lim_{q \rightarrow 0} \frac{1}{2q^2} (a_{AA} - a_{BB})$$

$$d = \lim_{\omega \rightarrow 0} \lim_{q \rightarrow 0} \frac{1}{2q^2} (a_{AA} + 2a_{AB} + a_{BB}) \quad (4.29)$$

with $a_{\mu\nu} = \langle [[S_{\mu-\underline{q}}^+, H], S_{\nu\underline{q}}^-] \rangle + \ll [S_{\mu-\underline{q}}^+, H]; [H, S_{\nu\underline{q}}^-] \gg_{\omega}$

$$(\mu, \nu) = (A, A), (A, B), (B, A), (B, B) \quad (4.30)$$

The two expressions (4.26) and (4.28) should be the same to within a similarity transformation as the spin wave pole is generated approximately by a linear combination of $S_{A\underline{q}}^-$ and $S_{B\underline{q}}^-$. To find c is equivalent to finding the eigenvalues of the matrix on the right hand side of (4.28). One finds

$$c = \sqrt{2ad} \langle n_{A\uparrow} - n_{A\downarrow} \rangle, \quad \Delta n_0 = \langle n_{A\uparrow} - n_{A\downarrow} \rangle \quad (4.31)$$

a and d can be worked out using expressions already obtained in section III. The commutators $[S_{-\underline{q}}^+, H]$ etc can be written

down in terms of $c_{k\sigma}^+$, $c_{k\sigma}$ etc and then in terms of $d_{k1\sigma}^+$, $d_{k2\sigma}$, using the transformation in (4.13). Sokoloff (1969a equation 6) writes the transformation down in detail though to get his basis one must replace θ_k by $(\pi/2 - \theta_k)$. The resulting Green's functions are worked out in an approximation which assumes that the electrons within the antiferromagnet energy bands are non-interacting. Then

$$a = \sum_k \xi_2(k) \sin \theta_k \cos^2 \theta_k (n_{k1} - n_{k2}) \quad (4.32)$$

$$dq^2 = \sum_k \frac{1}{2} (q \cdot \nabla)^2 \xi_1(k) (n_{k1} + n_{k2}) - \left(\frac{1}{2} (q \cdot \nabla)^2 \xi_2(k) \sin \theta_k + \frac{1}{2g} (q \cdot \nabla \xi_1(k))^2 \cos^2 \theta_k \right) (n_{k1} - n_{k2}) \quad (4.33)$$

$$\langle n_{A\uparrow} - n_{A\downarrow} \rangle = \sum_k \cos \theta_k (n_{k1} - n_{k2}) \quad (4.34)$$

Details of how (4.32) and (4.33) are obtained are in the appendix to this chapter.

In the extreme limit when $\xi_2(k) = 0$, $a = b = 0$. The first term in (4.28) is now diagonal and of order $(q/\omega)^2$. χ now reduces to having two ferromagnetic spin wave poles of equal energy $\omega_s = dq^2 / \langle n_{A\uparrow} - n_{A\downarrow} \rangle$. This is the same spin wave energy as in the R.P.A. approximation. The spin waves exist on each sublattice separately and there is no hopping between the sublattices. In this case we do not need the

restriction that ϵ_F lies in the gap as there are no other low lying modes. This limit arises in Lidiard's model (1954).

Our expression for c can be simplified when there is a wide energy gap. Putting in $\cos \theta_k = 1$, $\sin \theta_k = (I\varphi)^{-1} 2\epsilon_2(k)$ and $\varphi = n/N$ one gets

$$c^2 = (N\varphi)^{-2} \left(\sum_k n_k \frac{2\epsilon_2^2(k)}{I\varphi} \right) \left(\sum_k n_k \frac{|\nabla \epsilon_2(k)|^2}{I\varphi} + \frac{1}{2} \frac{\Omega N}{(2\pi)^3} \int \nabla \epsilon_1 dS_F \right) \quad (4.35)$$

The condition that the Fermi surface lies in the gap means the integral over the Fermi surface vanishes and $\varphi = 1$. For a band in a simple cubic lattice with hopping between nearest neighbours only, (4.31) reduces to

$$c^2 q^2 = 2J(0)(J(0) - J(q))s^2 \quad (4.36)$$

where $J(q) = \sum_{i \text{ n.n.}} J e^{iq \cdot R_i}$ and $J = 4b^2/I$.

using (4.32) to (4.35). b is the hopping integral between nearest neighbour sites. This is the formula for spin waves in a spin $\frac{1}{2}$ Heisenberg antiferromagnet with exchange coupling J . We have already in section III, (Anderson 1964), how it is possible to obtain an effective Heisenberg Hamiltonian, with a positive exchange coupling J equal to $4b^2/I$, for a system with one electron per site and a strong repulsion between electrons on the same site.

Sokoloff (1969a) only manages to obtain the Heisenberg limit by putting an interatomic exchange term into the Hamiltonian and considering the zero bandwidth limit. The reason for the discrepancy between Sokoloff's results and those of this thesis lies in the different approximations used. Sokoloff writes the spin density operators $S_{A\mathbf{q}} \pm S_{B\mathbf{q}}$ in terms of the antiferromagnetic energy bands. He is thus able to write down the susceptibility in an approximation in which the electrons in the antiferromagnetic bands are non-interacting. By linearising the equations of motion of the susceptibility within a basis defined by these bands he obtains a generalised enhanced susceptibility. He is thus working all the time in a subspace restricted by these bands. On the contrary (4.31) is general and we only put in the approximation to the ground state to work out the correlation functions in (4.31). (The disadvantage of this approach is that one does not work out from first principles a formula for the susceptibility.)

We can write our results in a third basis to demonstrate the form of the neutron scattering. Defining

$$\chi_{A \pm B, A \pm B}^{+-}(\mathbf{q}, \omega) = \frac{1}{2} \ll S_{A-\mathbf{q}}^{+} \pm S_{B-\mathbf{q}}^{+}; S_{A\mathbf{q}}^{-} \pm S_{B\mathbf{q}}^{-} \gg_{\omega}$$

we find

$$\chi_{A+B, A+B}^{+-}(\mathbf{q}, \omega) = \frac{1}{2} \sqrt{\frac{d}{2a}} \frac{1}{q} \langle n_{A\uparrow} - n_{A\downarrow} \rangle ([\omega - cq]^{-1} - [\omega + cq]^{-1}) \quad (4.37)$$

$$\chi_{A-B, A-B}^{+-}(\mathbf{q}, \omega) = \frac{1}{2} \sqrt{\frac{2a}{d}} \frac{1}{q} \langle n_{A\uparrow} - n_{A\downarrow} \rangle ([\omega - cq]^{-1} - [\omega + cq]^{-1}) \quad (4.38)$$

The neutron scattering (Sokoloff 1969a) depends on

$$(1 - e^{-\beta\omega})^{-1} \text{Im } \chi^+(\underline{q}, \omega)$$

From (4.37) one sees that the neutron scattering near nuclear reciprocal lattice vectors ($q \ll kT \ll g$) is small of order kT . Whereas from (4.38) near magnetic reciprocal lattice vectors ($q \ll kT \ll g$) the neutron scattering is large of order kT/q . In the region ($kT \ll q \ll g$) positive energies are weighted much more heavily than the negative (Cottam and Stinchcombe 1970).

V Spin Waves with Fermi Level not in the Gap

When ϵ_F does not lie in the gap the argument of the previous section no longer works since there are other low lying excitations besides spin waves. These are single particle excitations within the antiferromagnetic energy bands. In the region $q/\omega \ll 1$ they only contribute to the susceptibility to $O(q/\omega)^2$, as shown below, and do not introduce singularities into the limiting processes.

We therefore consider the case when ϵ_F lies in the lower band, and assume transitions between upper and lower bands require energy much greater than spin wave energies ($g \gg \omega_s$). Despite this assumption single particle excitations will still contribute to the susceptibility in the small (q, ω) region.

We assume that

$$\chi - \chi^{SP} = \chi^{SW} \quad (4.39)$$

where χ^{SW} is the part of the susceptibility coming from spin waves, which is of the form given by (4.26) and χ^{SP} is the susceptibility worked out in the Hartree-Fock approximation and so includes intraband single particle transitions only. χ^{SP} has been worked out by Sokoloff (1969a equation 55). Using (4.66) and (4.67) of the appendix to this chapter, one finds

$$\chi_{A+B, A+B}^{SP}(\underline{q}, \omega) = \frac{1}{2} \sum_{\underline{k}} \sin^2 \frac{1}{2}(\theta_{\underline{k}+\underline{q}} + \theta_{\underline{k}}) [\chi_0''(\underline{q}, \underline{k}, \omega) + \chi_0^{22}(\underline{q}, \underline{k}, \omega)] + O(1/g) \quad (4.40)$$

$$\chi_{A-B, A-B}^{SP}(\underline{q}, \omega) = \frac{1}{2} \sum_{\underline{k}} \sin^2 \frac{1}{2}(\theta_{\underline{k}+\underline{q}} - \theta_{\underline{k}}) [\chi_0''(\underline{q}, \underline{k}, \omega) + \chi_0^{22}(\underline{q}, \underline{k}, \omega)] + O(1/g) \quad (4.41)$$

$$\chi_{A+B, A-B}^{SP}(\underline{q}, \omega) = \frac{1}{2} \sum_{\underline{k}} \sin \frac{1}{2}(\theta_{\underline{k}+\underline{q}} - \theta_{\underline{k}}) \sin \frac{1}{2}(\theta_{\underline{k}+\underline{q}} + \theta_{\underline{k}}) [\chi_0''(\underline{q}, \underline{k}, \omega) - \chi_0^{22}(\underline{q}, \underline{k}, \omega)] + O(1/g) \quad (4.42)$$

where

$$\chi_0^{\mu\nu}(\underline{q}, \underline{k}, \omega) = \frac{n_{\underline{k}\mu} - n_{\underline{k}+\underline{q}\lambda}}{\omega - E_{\lambda}(\underline{k}+\underline{q}) + E_{\mu}(\underline{k})}$$

Other terms would arise in χ^{SP} involving χ_0^{12} , χ_0^{21} but are $O(1/g)$ and can be neglected because of our assumption $\omega_s \ll g$. Expanding χ^{SP} to the lowest powers in $\underline{q}/\omega$ we find that only $\chi_{A+B, A+B}^{SP}(\underline{q}, \omega)$ contains a term of the form $\Delta(\underline{q}/\omega)^2$.

(78)

Then χ^{SP} has the form

$$\chi^{SP} = \frac{1}{2} \Delta \left(\frac{q}{\omega} \right)^2 \begin{pmatrix} 1 & 1 \\ 1 & 1 \end{pmatrix} \quad (4.43)$$

so that from (4.28), (4.39) becomes

$$\chi^{SW} = \omega^{-2} \begin{pmatrix} a + (d - \frac{1}{2}\Delta + b)q^2 & -(a + \frac{1}{2}\Delta q^2) \\ -(a + \frac{1}{2}\Delta q^2) & a + (d - \frac{1}{2}\Delta + b)q^2 \end{pmatrix} + \omega^{-1} \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} \langle n_{A\uparrow} - n_{A\downarrow} \rangle \quad (4.44)$$

$\omega = \pm \omega_q$ are the roots of the equation obtained when the determinant of the matrix on the right hand side of (4.44) is set equal to zero. Our formula for c is now slightly changed from (4.31) and is

$$c = \sqrt{2a(d-\Delta)} / \langle n_{A\uparrow} - n_{A\downarrow} \rangle \quad (4.45)$$

No imaginary terms occur to order $(q/\omega)^2$ and so ω_S is real to $O(q)$.

The scattering of a spin wave into a pair $d_{\underline{k}+\underline{q},\downarrow}^+ d_{\underline{k},\uparrow}$ can occur since it may be possible to satisfy energy and momentum conservation laws for this process for some values of $\underline{k}$ near the Fermi surface. In such circumstances this may be a dominant term in the damping of the spin wave. In our method this scattering would give an imaginary part to the Green's functions of (4.30) in the small (q, ω) region. However, we expand in powers of q/ω to order $(q/\omega)^2$ and in this limit the imaginary part is zero. In the same limit the suscep-

tibilities defined by (4.40), (4.41) and (4.42) have no imaginary parts. These results imply that c^2 is real. This conclusion is open to some doubt as the form of the susceptibility in (4.26) and (4.27) already assumes that spin waves (and the intraband transitions) are well defined excitations.

We will compare our conclusions with Sokoloff's (1969a) method. He also makes corrections involving (4.40), (4.41) and (4.42) because of intraband transitions when the Fermi energy lies below the top of band 1. He still obtains the dispersion relation $\omega = cq$, but with c now renormalised. He does not mention that c may now have an imaginary part because of the scattering process discussed above. We would claim the spin wave is not so heavily damped, with a lifetime proportional to a higher power than $1/q$.

The damping in either case will be small in certain circumstances. If the Fermi surface lies in regions of the Brillouin zone where there is strong spin polarisation, the weights of the intraband transitions in the susceptibility χ , which involve small momentum transfers, are proportional to $\sin^2 \theta_{k_F}$ or less (from (4.40) to (4.42)), and are therefore small. Also intraband transitions about parts of the Fermi surface in paramagnetic regions are presumably not coupled very strongly to the spin wave.

We will now consider how Δ affects the real part of the

spin wave energy. We find it has the value

$$\Delta = \frac{1}{6} \sum_{\underline{k}} \nabla(\sin^2 \theta_{\underline{k}} \nabla E_1(\underline{k})) n_{\underline{k}1} \quad (4.46)$$

At zero temperature this can be written as a surface integral

$$\Delta = \frac{2N}{6(2\pi)^3} \int \sin^2 \theta_{\underline{k}} \underline{n} \cdot \nabla E_1(\underline{k}) dS_F \quad (4.47)$$

When $\mathcal{E}_F$ lies in the gap $\chi^{SP} = 0$ and $\Delta = 0$. With $\mathcal{E}_F$ not in the gap we will pay particular attention to the cases when the damping is thought to be small. When the Fermi surface lies entirely in the region strongly distorted by the antiferromagnetism, $\sin \theta_{k_F} \ll 1$ and $\Delta \ll d$, so that c is still given by (4.31), or (4.35) for an antiferromagnet with a wide gap. If the Fermi surface lies entirely in paramagnetic parts of the Brillouin zone away from the nesting surfaces, $\sin \theta_{k_F}$ is almost one, so that

$$\Delta = \frac{2N}{6(2\pi)^3} \int \underline{n} \cdot \nabla E_1(\underline{k}) dS_F \quad (4.48)$$

One can then write d as the sum of a volume and a surface integral so that

$$d - \Delta = \frac{1}{6} \sum_{\underline{k}} n_{\underline{k}1} (\nabla E_2(\underline{k}) \nabla(\sin \theta_{\underline{k}}) - g^{-1} |\nabla E_1(\underline{k})|^2 \cos^3 \theta_{\underline{k}}) \quad (4.49)$$

In this form one sees that electrons which contribute only weakly to the magnetic moment contribute weakly to α . Using (4.49) more realistic models of weak antiferromagnets than those with perfect nesting bands could be investigated.

We can investigate our formula (4.45) for α , with d given by (4.49), as $I \rightarrow 0$ remembering to remain in the region $g/\omega_s \gg 1$. Supposing $\xi_2(\underline{k})$ is of order δ on the Fermi surface, as $I \rightarrow \delta +$ we have a region where $\delta \gg g$. From (4.32) and (4.49) we find $d - \Delta$ is $O(Ng^2/\delta^3)$ and

$$\alpha / \langle n_{\uparrow\uparrow} - n_{\downarrow\downarrow} \rangle^2 \leq g / \langle n_{\uparrow\uparrow} - n_{\downarrow\downarrow} \rangle = \frac{1}{2} I / N \quad (4.50)$$

Therefore α is $O(g/\delta)$ from (4.45) showing that the spin wave velocity tends to zero as the energy gap tends to zero. For $I < \delta$ there will be a breakdown in the antiferromagnetism because the selfconsistency equation (4.18) cannot be satisfied. So the spin wave velocity as well as the energy gap is a measure of when antiferromagnetism occurs in a model with nearly perfect nesting.

This argument does not apply to the idealistic case of perfect nesting for which $\delta = 0$, and (4.31) still applies. An example is a half full s band with nearest neighbour hopping in a b.c.c. crystal. The Fermi surface is a cube on which $\xi_2(\underline{k}) = 0$. A rough approximation for α can be made by considering the model of Fedders and Martin (1966) in

which there is a spherical Fermi surface near which

$$\varepsilon_2(k) = v_F(k - k_c)$$

We find

$$c = \frac{1}{\sqrt{3}} v_F \frac{\sqrt{\log 2v_F k_c/g - 5/2}}{\log 2v_F k_c/g - 3/2} \quad (4.51)$$

where k_c is the radius of the sphere, v_F the Fermi velocity and $\omega_s \ll g \ll v_F k_c$. Then

$$c \rightarrow v_F / \sqrt{3 \log 2v_F k_c/g} \quad \text{as} \quad \sqrt{\log 2v_F k_c/g} \rightarrow \infty \quad (4.52)$$

One can see that c decreases very slowly as g decreases. This arises because the perfect nesting of bands greatly favours antiferromagnetism. This result contrasts with the result of Sokoloff (1969a) who found c increased as $2v_F k_c/g$ increased. The numerical factors cannot be accurate because of the approximations made, but c is of the order of v_F in agreement with experiment (Sinha, Liu et al 1969).

We have now considerable confirmation that our formula for the spin wave velocity c , for an antiferromagnetic itinerant metal when the Fermi energy lies in the gap, is correct. Firstly the formula for spin waves in a Heisenberg antiferromagnet is obtained as an example of when there is a wide antiferromagnetic energy gap. In the model of Fedders

and Martin (1966) there is a weak energy gap. For this case our expression for the spin wave velocity is of the order of magnitude of the Fermi velocity, in agreement with experiment and other theoretical results.

We have extended our formula to the case when the Fermi level is not in the gap. The basic assumption used is that part of the magnetic response is dominated by spin waves. Then, even with the Fermi level not in the gap we argue that c is real. Furthermore, when the Fermi level lies only in regions strongly distorted by the antiferromagnetism, our original formula can be retained. Not surprisingly we find that c is a measure of the stability of the antiferromagnetic state in weak coupling models, and decreases to zero as the energy gap tends to zero. The work differs from that of Sokoloff (1969a, b) by not using a restricted basis set. Sokoloff, for example, was not able to obtain the same Heisenberg limit.

Appendix

In this appendix we give further details of how the general formula for c in (4.31) is evaluated. From (4.8) one finds

$$[H, S_{A\bar{q}}^-] = \sum_{\underline{k}} (\epsilon_1(\underline{k}+\underline{q}) - \epsilon_1(\underline{k})) C_{A\underline{k}+\underline{q}\downarrow}^+ C_{A\underline{k}\uparrow} + \epsilon_2(\underline{k}) C_{A\underline{k}+\underline{q}\downarrow}^+ C_{B\underline{k}\uparrow} - \epsilon_2(\underline{k}+\underline{q}) C_{B\underline{k}+\underline{q}\downarrow}^+ C_{A\underline{k}\uparrow} \quad (4.53)$$

$$[H, S_{B\bar{q}}^-] = \sum_{\underline{k}} (\epsilon_1(\underline{k}+\underline{q}) - \epsilon_1(\underline{k})) C_{B\underline{k}+\underline{q}\downarrow}^+ C_{B\underline{k}\uparrow} + \epsilon_2(\underline{k}) C_{B\underline{k}+\underline{q}\downarrow}^+ C_{A\underline{k}\uparrow} - \epsilon_2(\underline{k}+\underline{q}) C_{A\underline{k}+\underline{q}\downarrow}^+ C_{B\underline{k}\uparrow} \quad (4.54)$$

One can write (4.53) and (4.54) in terms of the antiferromagnetic energy bands by inverting (4.13) and (4.16), so that

$$\begin{pmatrix} C_{A\underline{k}\uparrow}^+ \\ C_{B\underline{k}\uparrow}^+ \end{pmatrix} = T^+ \begin{pmatrix} d_{\underline{k}1\uparrow}^+ \\ d_{\underline{k}2\uparrow}^+ \end{pmatrix}, \quad \begin{pmatrix} C_{A\underline{k}\downarrow}^+ \\ C_{B\underline{k}\downarrow}^+ \end{pmatrix} = T \begin{pmatrix} d_{\underline{k}1\downarrow}^+ \\ d_{\underline{k}2\downarrow}^+ \end{pmatrix} \quad (4.55)$$

We define

$$a = a_1 + a_2, \quad b = b_1 + b_2, \quad d = d_1 + d_2 \quad (4.56)$$

where a_1 , b_1 and d_1 arise from the correlation functions in (4.30) and a_2 , b_2 and d_2 from the Green's functions in (4.30).

As

$$\langle [[S_{A-\bar{q}}^+, H] S_{A\bar{q}}^-] \rangle = \langle [[S_{B-\bar{q}}^+, H] S_{B\bar{q}}^-] \rangle$$

we find $b_1 = 0$, and from (4.29) and (4.30)

$$\begin{aligned} a_1 &= -\lim_{q \rightarrow 0} \langle [[S_{A-\bar{q}}^+, H] S_{B\bar{q}}^-] \rangle = \sum_{\underline{k}} \epsilon_2(\underline{k}) 2 \langle C_{A\underline{k}\uparrow}^+ C_{B\underline{k}\uparrow} \rangle \\ &= \sum_{\underline{k}} \epsilon_2(\underline{k}) \sin \theta_{\underline{k}} (n_{\underline{k}1} - n_{\underline{k}2}) \end{aligned} \quad (4.57)$$

$$d_1 q^2 = q^2 \lim_{q \rightarrow 0} \frac{1}{q^2} \langle [S_{A-q}^+, H] S_{Aq}^- + S_{Bq}^- \rangle \quad (4.58)$$

$$= \sum_{\underline{k}} \frac{1}{2} (\underline{q} \cdot \nabla)^2 \varepsilon_1(\underline{k}) (n_{\underline{k}1\uparrow} + n_{\underline{k}2\uparrow}) - \frac{1}{2} (\underline{q} \cdot \nabla)^2 \varepsilon_2(\underline{k}) 2 \langle c_{A\underline{k}\uparrow}^+ c_{B\underline{k}\uparrow} \rangle \quad (4.59)$$

$$= \sum_{\underline{k}} \frac{1}{2} (\underline{q} \cdot \nabla)^2 \varepsilon_1(\underline{k}) (n_{\underline{k}1} + n_{\underline{k}2}) - \frac{1}{2} (\underline{q} \cdot \nabla)^2 \varepsilon_2(\underline{k}) (n_{\underline{k}1} - n_{\underline{k}2}) \sin \theta_{\underline{k}}$$

These correlation functions have been worked out in the antiferromagnetic ground state.

To evaluate the Green's functions we define

$$F_{\mu\nu}(\underline{q}, \omega) = \langle\langle [S_{\mu-\underline{q}}^+, H]; [H, S_{\nu\underline{q}}^-] \rangle\rangle_{\omega}, \quad 2F_{AB} = F_{A+B, A+B} - F_{A-B, A-B} \quad (4.60)$$

$$2F_{AA} = F_{A+B, A+B} + F_{A-B, A-B} + 2F_{A+B, A-B}, \quad 2F_{BB} = F_{A+B, A+B} + F_{A-B, A-B} - 2F_{A+B, A-B}$$

In terms of the antiferromagnetic energy bands, one may write (4.53) and (4.54) as

$$[H, S_{A+B\underline{q}}^-] = \sqrt{2} \sum_{\underline{k}} (\underline{q} \cdot \nabla) \varepsilon_1(\underline{k}) \cos \theta_{\underline{k}} (d_{\underline{k}2\downarrow}^+ d_{\underline{k}1\uparrow} + d_{\underline{k}1\downarrow}^+ d_{\underline{k}2\uparrow}) \quad (4.61)$$

$$[H, S_{A-B\underline{q}}^-] = \sqrt{2} \sum_{\underline{k}} 2\varepsilon_2(\underline{k}) \sin \theta_{\underline{k}} (d_{\underline{k}2\downarrow}^+ d_{\underline{k}1\uparrow} + d_{\underline{k}1\downarrow}^+ d_{\underline{k}2\uparrow}) \quad (4.62)$$

to the lowest power in q , neglecting terms involving intraband transitions. When the Fermi level does not lie in the gap one should include intraband transitions in (4.61) and (4.62). However, intraband terms do not contribute to the Green's functions $F_{\mu\nu}(\underline{q}, \omega)$ in the limit $q/\omega \rightarrow 0$, in the approximation in which electrons within the antiferromagnetic bands are non-interacting. So putting (4.61) and (4.62) into (4.60) we get

$$\begin{aligned}
 d_2 q^2 &= q^2 \lim_{\omega \rightarrow 0} \lim_{q \rightarrow 0} \frac{1}{q^2} F_{A+B, A+B} \\
 &= \frac{1}{2} g \sum_{\underline{k}} |\underline{q} \cdot \nabla \varepsilon(\underline{k})|^2 \cos^3 \theta_{\underline{k}} (n_{\underline{k}1} - n_{\underline{k}2})
 \end{aligned} \tag{4.63}$$

$$\begin{aligned}
 a_2 &= -\frac{1}{2} \lim_{\omega \rightarrow 0} \lim_{q \rightarrow 0} F_{A-B, A-B} \\
 &= - \sum_{\underline{k}} \varepsilon_2(\underline{k}) \sin^3 \theta_{\underline{k}} (n_{\underline{k}1} - n_{\underline{k}2})
 \end{aligned} \tag{4.64}$$

$$b_2 = \lim_{\omega \rightarrow 0} \lim_{q \rightarrow 0} \frac{1}{q^2} F_{A+B, A-B} \tag{4.65}$$

but this term does not contribute to the spin wave energy to $O(q)$ and may be neglected. From (4.57), (4.58), (4.63) and (4.64) we obtain (4.32) and (4.33) of the text.

To obtain (4.40) to (4.42) of the text, one needs the following expressions for $S_A^-(\underline{q}) \pm S_B^-(\underline{q})$ in terms of the antiferromagnetic bands:

$$\begin{aligned}
 S_{A\underline{q}}^- + S_{B\underline{q}}^- &= \sum_{\underline{k}} \cos \frac{1}{2}(\theta_{\underline{k}+\underline{q}} + \theta_{\underline{k}}) (d_{\underline{k}+\underline{q}1\downarrow}^+ d_{\underline{k}2\uparrow} + d_{\underline{k}+\underline{q}2\downarrow}^+ d_{\underline{k}1\uparrow}) \\
 &\quad + \sin \frac{1}{2}(\theta_{\underline{k}+\underline{q}} + \theta_{\underline{k}}) (d_{\underline{k}+\underline{q}1\downarrow}^+ d_{\underline{k}1\uparrow} - d_{\underline{k}+\underline{q}2\downarrow}^+ d_{\underline{k}2\uparrow})
 \end{aligned} \tag{4.66}$$

$$\begin{aligned}
 S_{A\underline{q}}^- - S_{B\underline{q}}^- &= \sum_{\underline{k}} \cos \frac{1}{2}(\theta_{\underline{k}+\underline{q}} - \theta_{\underline{k}}) (d_{\underline{k}+\underline{q}1\downarrow}^+ d_{\underline{k}2\uparrow} - d_{\underline{k}+\underline{q}2\downarrow}^+ d_{\underline{k}1\uparrow}) \\
 &\quad + \sin \frac{1}{2}(\theta_{\underline{k}+\underline{q}} - \theta_{\underline{k}}) (d_{\underline{k}+\underline{q}1\downarrow}^+ d_{\underline{k}1\uparrow} + d_{\underline{k}+\underline{q}2\downarrow}^+ d_{\underline{k}2\uparrow})
 \end{aligned} \tag{4.67}$$

Chapter 5

Spin Waves in Chromium

I The Two Band Approximation

In the previous chapter a formula was obtained for the energy ϵ_q of long wavelength spin waves in a one-band model of an antiferromagnetic metal. Chromium is the only metal, to which a purely itinerant electron model applies, and which has an antiferromagnetic ground state. However, chromium in a paramagnetic state, has a complicated band structure and hence the structure, in an antiferromagnetic state, is equally complicated. Nevertheless, in this chapter a formula for c is calculated and comparison is made with experimental values for c . This is possible because the two band approximation introduced by Asano and Yamashita (1967), to be referred to as A.Y., greatly simplifies any treatment of the antiferromagnetic ground state.

Using this approximation, the contribution to the total magnetisation from the spins in the different bands can be estimated. As emphasised by A.Y. the bands which are not strongly polarised cannot be neglected, since there is so much of them they are the dominant contribution to the magnetisation. However, these weakly magnetised bands are usually neglected when calculations of c are made. The

method of this chapter tries to treat these bands properly and hence is an improvement on earlier calculations (Fedders and Martin 1966, Sokoloff 1969, Liu 1970).

We go further than A.Y. in discussing the conditions for this two band approximation to apply. It appears that the approximation breaks down once the spin density waves in chromium are no longer independent, but this may coincide with the transformation from the spin density wave state (S.D.W.) to the perfect antiferromagnetic state (P.A.F.). This transition S.D.W. to P.A.F. would not be reflected in the paramagnetic susceptibility which is often discussed in this connection (Gupta and Sinha 1970, 1971, Hamaguchi 1965).

The two band approximation has greatest applicability to a P.A.F. state, and experiments are most easily performed on Cr-Mn alloys which take up a perfect antiferromagnetic configuration. Hence c , the spin wave velocity, is calculated for this sort of alloy. The method of this calculation relies entirely on the formalism of the previous chapter and the two band approximation of A.Y.

II The Spin Density State of Chromium

It was first suggested by Lomer (1962) that the ground state of chromium was a spin density wave. Arrot et al

(1965 and 1966) discuss neutron scattering experiments on chromium. The elastic magnetic scattering from the electrons is proportional to

$$S^{\alpha\beta}(\underline{q}, \omega) = \delta(\omega) \langle S_{\underline{q}}^{\alpha} \rangle \langle S_{-\underline{q}}^{\beta} \rangle$$

in (1.26) and (1.28), with scattering wave vectors $\underline{q} = \underline{Q} + \underline{K}$ for spin densities of the form $\underline{A} \cos \underline{Q} \cdot \underline{r}$ provided $\underline{A}$ is not parallel to $\underline{Q}$. For chromium there are three different basic wave vectors $\underline{Q}$ of the form

$$\underline{Q}_x = \frac{\pi}{a}(1-e, 0, 0) \quad , \quad \underline{Q}_y = \frac{\pi}{a}(0, 1-e, 0) \quad , \quad \underline{Q}_z = \frac{\pi}{a}(0, 0, 1-e)$$

giving three different spin density waves.

Chromium is body centered cubic so that the basis of the face centered cubic reciprocal lattice is

$$\frac{\pi}{a}(1, 1, 0) \quad , \quad \frac{\pi}{a}(0, 1, 1) \quad , \quad \frac{\pi}{a}(1, 0, 1)$$

$2a$ is the side of the cube in the direct lattice with atoms at $(0, 0, 0)$ and (a, a, a) . In the presence of a magnetic field along the x -axis only the $\underline{Q}_x$ spin density wave will be represented. Below 115°K the reflections are of the form

$$\frac{\pi}{a}(1-e, 0, 0) \pm \frac{\pi}{a}(1, 1, 0) \quad , \quad \frac{\pi}{a}(1-e, 0, 0) \pm \frac{\pi}{a}(1, 0, 1)$$

but not $\pm \pi/a(1 \pm e, 0, 0)$. Above 115°K but below the Néel temperature 38.5°C , one has the additional points

$\pm \pi/a(1 \pm e, 0, 0)$. This is interpreted to mean below 115°K

(called the spin flip temperature) the polarisation $\underline{A}$ of the $\underline{Q}_x$ spin density wave is longitudinal i.e. $\underline{A}$ is parallel to $\underline{Q}_x$, whereas above it is transverse.

We see that the magnetic scattering occurs around the

point $H = \pi/a (1, 0, 0)$ of the first Brillouin zone. Temperatures and dilute concentrations of impurity metals, which increase or decrease the effective electron concentration, change e . Hamaguchi et al (1964) discuss chromium-manganese and chromium-vanadium. For pure chromium e is $\pi/21a$ at liquid air temperatures and is $\pi/27a$ at the Néel temperature (Herring 1966). For alloys with 2% manganese a purely antiferromagnetic structure with $e = 0$, is stable at all temperatures below the Néel temperature. Hamaguchi et al plot the different phases on a temperature versus effective electron concentration diagram.

Lomer's (1962) explanation of the antiferromagnetism in chromium relied on there being two closed pockets in the band structure, one enclosing electrons at the point Γ of the Brillouin zone and one enclosing holes at the point H . Both have approximately the same volume. This band structure, favourable for the occurrence of a spin density wave of the form described above, was derived from considering the band structure of iron. Since then, calculations have been done on paramagnetic chromium e.g. Loucks (1965), Asano and Yamashita (1967) and Gupta and Sinha (1970). These calculations have the same favourable feature i.e. the shifting of one of these pockets by $\pi/a (0.96, 0, 0)$ or $\pi/a (1.04, 0, 0)$ along one of the cubic axis approximately brings a large area of contact between the pockets. This

produces an antiferromagnetic coupling between the electrons, which can be estimated using the Hartree-Fock approximation.

It would be hard to solve the Hartree-Fock equations in general. However, the assumption that the antiferromagnetic coupling between the electrons is weak except for those in the pockets around Γ and H involved in the nesting, produces considerable simplification in theoretical work, and is justified experimentally. Thus much progress has been made using model band structures (Pedders and Martin 1966, Kimball 1969).

The method of looking at peaks in the paramagnetic non-interacting susceptibility $\chi^0(\underline{q})$ in order to determine the wave vector of the spin density wave, has been discussed by various authors (Tachiki and Nagomiya 1963, Herring 1966). Gupta and Sinha have looked at $\chi^0(\underline{q})$ including the effects of many bands and trying to include the effects of matrix elements in the susceptibility. The interacting susceptibility is enhanced by a factor of the form $[1 - 2J_{\text{eff}} \chi^0(\underline{q})]^{-1}$, so that a peak in $\chi^0(\underline{q})$ at the wave vector $\underline{Q}$ may make $\chi(\underline{Q})$ diverge, indicating that the ground state is a spin density wave of wave vector $\underline{Q}$. Gupta and Sinha's calculation is not sufficiently good to allow a detailed discussion of the width of the peak, or of its change in position with small changes in electron concentration. Hamaguchi et al (1965) assume a form for the

susceptibility to explain their alloy results on the basis of the rigid band approximation, but they also remark on the need for detailed information on the band structure.

The variation of Q_x with effective electron concentration for chromium-manganese is the following: Q_x approaches π/a very steeply once $Q_x > .96 \pi/a$, and this transformation from the spin density state, with $Q_x \neq \pi/a$, to the perfect antiferromagnetic state, with $Q_x = \pi/a$, depends on the temperature (Hamaguchi et al 1965). Asano and Yamashita discuss a mechanism for this to happen, assuming that the gap in the perfect antiferromagnetic state is twice that in the spin density state. The rigid band approach is also relevant since the increase of electron concentration should cause the electron surface to expand and the hole surface to contract increasing Q_x . Asano and Yamashita obtain the formula

$$Q_x = \pi/a (.95 + .4n , 0 , 0) \quad (5.1)$$

on this basis, where n is the number of excess electrons per atom. For Cr-Mn this change is far too slow, but Cr-Fe does follow this formula (Ishikawa et al 1967).

In section III we will show that it is unreasonable to associate peaks in the paramagnetic susceptibility with values of the ordering wave vector Q_x close to π/a , where the rigid band approach breaks down. Anyway, as Gupta and

Sinha remark, the transition in chromium to the spin density state is a first order transition so Q will be in a position where $\chi(q)$ has a steep slope rather than at a maximum of $\chi(q)$.

According to Rice et al (1968) the experimental value of the antiferromagnetic energy gap is .14ev. This is very small compared with band energies. A large portion of the Fermi surface must be truncated by these gaps for them to have such a crucial importance on the magnetic structure of the metal. Rice quotes an estimate that 50% of the Fermi surface is truncated. The density of states at the Fermi surface is low at .5 states/ev atom according to the band structure calculations mentioned above. We can thus estimate that at most, only .035 electrons/atom are fully polarised contributing a moment of $.035\mu_B$, whereas the total spin polarisation is about $.6\mu_B$. The addition of 3.5% vanadium removes the antiferromagnetism altogether. This corresponds to removing about .017 electrons/atom from parts of the Fermi surface contributing to the antiferromagnetism.

This suggests that only a very few electrons are coupled strongly and are strongly polarised. Even if all the electrons were fully polarised in the electron pocket around Γ , this would not be sufficient to explain the observed antiferromagnetic magnetisation, since the pocket occupies only a fraction of the Brillouin zone. The crucial

parameter for describing the polarisation is g/ε_2 , where g is a typical gap energy and ε_2 is a typical band energy of the form $\frac{1}{2}(\varepsilon_{k_m} - \varepsilon_{k+Qn})$. Most of the amplitude of the spin density wave must come from weakly polarised electrons, well away from the nesting surfaces, characterised by $g/\varepsilon_2 \ll 1$. This was already suggested in Lomer's original paper. Asano and Yamashita show that the energy difference between the spin density wave state and the paramagnetic state is of order $(g/\varepsilon_2)^3$ in this region, suggesting that the energy difference comes from the strongly polarised electrons only. We will assume the occupied zone can be divided into two regions, I and II, where $g/\varepsilon_2 \gg 1$ in region I and $g/\varepsilon_2 \ll 1$ in region II. This is a very useful simplification.

III The Ground State

The general form of the spin density in a spin density wave is $\langle \underline{S}(\underline{r}) \rangle = \underline{A}_0 \cos \underline{Q} \cdot \underline{r}$, plus harmonics, that is terms of the form $\underline{A}_{\underline{K}} \cos \underline{Q} \cdot \underline{r} + \underline{K} \cdot \underline{r}$, where $\underline{K}$ is a reciprocal lattice vector. One can neglect harmonics if one adopts the "rigid spin" approximation (Overhauser 1962).

If $\underline{S}(\underline{r}-\underline{R}_i)$ is the spin density in the cell $\underline{R}_i$, $\rho(\underline{r}-\underline{R}_i)$ is the electron density in the cell $\underline{R}_i$, then in the rigid spin approximation

$$\underline{S}(\underline{r}-\underline{R}_i) = \rho(\underline{r}-\underline{R}_i) \underline{S}_i \quad (5.2)$$

where $\underline{S}_i$ is the total spin operator of the cell $\underline{R}_i$. $\underline{S}_i$ has its usual representation in terms of atomic wave functions, namely

$$S_i^2 = \frac{1}{2}(n_{i\uparrow} - n_{i\downarrow}), \quad S_i^- = \sum_{\mu} c_{i\mu\downarrow}^+ c_{i\mu\uparrow} \quad (5.3)$$

with $n_{i\sigma} = \sum_{\mu} c_{i\mu\sigma}^+ c_{i\mu\sigma}$.

μ denotes the symmetry of the atomic wavefunctions and

$c_{i\mu\sigma}^+$, $c_{i\mu\sigma}$ are related to the creation and annihilation operators of Bloch states $c_{\underline{k}\sigma}^+$, $c_{\underline{k}\sigma}$ by

$$c_{\underline{k}\sigma} = \frac{1}{\sqrt{N}} \sum_{\mu} a_{\underline{k}\mu}^{(n)} c_{i\mu\sigma} e^{i\underline{k} \cdot \underline{R}_i}$$

and $c_{\underline{k}\sigma}^+ = \frac{1}{\sqrt{N}} \sum_{\mu} a_{\underline{k}\mu}^{(n)\dagger} c_{i\mu\sigma}^+ e^{-i\underline{k} \cdot \underline{R}_i} \quad (5.4)$

Spin operators now need be labelled by their site only.

If one further assumes that the polarisation is the same for each atomic wavefunction, so that

$$\begin{aligned} n_{i\mu\uparrow} &= n_{i\mu} + \frac{1}{10}(n_{i\uparrow} - n_{i\downarrow}) \\ n_{i\mu\downarrow} &= n_{i\mu} - \frac{1}{10}(n_{i\uparrow} - n_{i\downarrow}) \end{aligned} \quad (5.5)$$

then the only spin dependent term in the short-range interaction is

$$J_{\text{eff}} \sum_i n_{i\uparrow} n_{i\downarrow} \quad (5.6)$$

where $J_{\text{eff}} = \frac{1}{5}(I+4J)$, and I and J are defined in chapter 1.

This interaction (5.6) can be rewritten

$$\frac{1}{N} J_{\text{eff}} \sum_{\underline{q}} S_{\underline{q}}^z S_{-\underline{q}}^z \quad (5.7)$$

where
$$S_{\underline{q}}^z = \sum_i e^{i\underline{q} \cdot \underline{R}_i} S_i^z \quad (5.8)$$

$$= \sum_{\underline{k}, n, m} F_{nm}(\underline{k}+\underline{q}, \underline{k}) \frac{1}{2} (c_{n\underline{k}+\underline{q}\uparrow}^+ c_{m\underline{k}\uparrow} - c_{n\underline{k}+\underline{q}\downarrow}^+ c_{m\underline{k}\downarrow}) \quad (5.9)$$

and
$$F_{nm}(\underline{k}+\underline{q}, \underline{k}) = \sum_{\mu} a_{\underline{k}+\underline{q}\mu}^{(n)} a_{\underline{k}\mu}^{(m)\dagger} \quad (5.10)$$

For a given $\underline{q}$, region I is the part of the Brillouin zone for which there is strong mixing between two bands, II and II'. The generalisation of (4.10) to two bands is

$$\ll \begin{pmatrix} c_{N\underline{k}+\underline{q}\sigma} \\ c_{M\underline{k}\sigma} \end{pmatrix}; \begin{pmatrix} c_{N\underline{k}+\underline{q}\sigma}^+ \\ c_{M\underline{k}\sigma}^+ \end{pmatrix} \gg_{\omega} = \begin{pmatrix} \omega - \epsilon_{\underline{k}+\underline{q}N} & \Gamma_{NM}(\underline{q}) \\ \Gamma_{NM}^*(\underline{q}) & \omega - \epsilon_{\underline{k}M} \end{pmatrix}^{-1} \quad (5.11)$$

where
$$\Gamma_{NM}(\underline{q}) = \frac{1}{N} J_{\text{eff}} \langle S_{\underline{q}}^z \rangle F_{NM}(\underline{k}+\underline{q}, \underline{k}). \quad (5.12)$$

The antiferromagnetic energy bands $\psi_{\underline{k}1}^I, \psi_{\underline{k}2}^I$ in region I are formed from linear combinations of $\psi_{\underline{k}+\underline{q}N}, \psi_{\underline{k}M}$. There are small corrections to $\psi_{\underline{k}1,2}^I$, because of weak coupling of $\psi_{\underline{k}1,2}^I$ with electrons in region II.

$\langle S_{\underline{q}}^z \rangle$ in (5.12) is derived selfconsistently, using

$$\langle S_Q^z \rangle = \langle S_Q^z \rangle^I + \langle S_Q^z \rangle^{II} \quad (5.13)$$

where S_Q^z is defined by (5.9), and I refers to regions where the summation over k, n, m lies entirely in region I, and II refers to the rest of the summation. Using (5.11), one finds

$$\begin{aligned} & \frac{1}{2} \langle C_{Nk+Q\uparrow}^+ C_{Mk\uparrow} - C_{Nk+Q\downarrow}^+ C_{Mk\downarrow} \rangle^I \\ &= \Gamma_{NM}(Q) \left(|\Gamma_{NM}(Q)|^2 + \frac{1}{4} (\epsilon_{k+Qn} - \epsilon_{km})^2 \right)^{-\frac{1}{2}} (n_{k1}^I - n_{k2}^I) \end{aligned} \quad (5.14)$$

In region II there is coupling between electrons in all the bands. However, as the coupling is weak we work to first order perturbation theory in the interaction, so that the coupling between bands m and n , is independent of the coupling between bands m and n_2 . The new bands in region II have energy (Kimball 1969)

$$E_{km}^{II} = \epsilon_{km} - \sum_n \frac{|\Gamma_{nm}(Q)|^2}{\epsilon_{k+Qn} - \epsilon_{km}} (n_{km} - n_{k+Qn}) \quad (5.15)$$

where n_{km}, n_{k+Qn} are occupation numbers of the paramagnetic bands of region II. $\langle S_Q^z \rangle^{II}$, in (5.13), is derived from (5.11) with

$$\frac{1}{2} \langle C_{k+Qn\uparrow}^+ C_{km\uparrow} - C_{k+Qn\downarrow}^+ C_{km\downarrow} \rangle^{II} = \Gamma_{nm}(Q) (\epsilon_{k+Qn} - \epsilon_{km})^{-1} (n_{km} - n_{k+Qn}) \quad (5.16)$$

(5.16) is the same as (5.14) to the first order in the interaction energy.

With a magnetic field $(0, 0, H \cos \theta, R)$ on, one has to replace $\Gamma_{nm}(\underline{Q})$ by

$$\Gamma_{nm}(\underline{Q}) + g\mu_B H \frac{1}{2} F_{nm}(\underline{k} + \underline{Q}, \underline{k}) \quad (5.17)$$

Then if the whole of the band structure were in region II the metal would be paramagnetic, that is

$$\langle S_{\underline{Q}}^z \rangle_{H=0} = 0 \quad (5.18)$$

The paramagnetic susceptibility $\chi(\underline{Q})$ is thus from (5.9), (5.16) and (5.17)

$$g\mu_B \frac{\partial \langle S_{\underline{Q}}^z \rangle}{\partial H} \Big|_{H=0} = (g\mu_B)^2 \frac{\chi^0(\underline{Q})}{1 - 2(J_{\text{eff}}/N)\chi^0(\underline{Q})} \quad (5.19)$$

$$\text{where } \chi^0(\underline{Q}) = \frac{1}{2} \sum_{\underline{k}, n, m} |F_{nm}(\underline{k} + \underline{Q}, \underline{k})|^2 \frac{n_{\underline{k}m} - n_{\underline{k} + \underline{Q}n}}{\epsilon_{\underline{k} + \underline{Q}n} - \epsilon_{\underline{k}m}} \quad (5.20)$$

One can compare this result with the result one obtains for a point interaction $J_{\text{eff}} n_{\uparrow} n_{\downarrow} \Omega^2$, for which one must solve a matrix equation for the susceptibility $\chi(\underline{Q} + \underline{K}, \underline{Q})$ (Gupta and Sinha 1971). This is

$$[\delta_{\underline{K}, \underline{K}'} - 2 J_{\text{eff}}/N \chi^0(\underline{Q} + \underline{K}, \underline{Q} + \underline{K}')] \chi(\underline{Q} + \underline{K}', \underline{Q}) = \chi^0(\underline{Q} + \underline{K}, \underline{Q}) \quad (5.21)$$

$$\text{where } \chi^0(\underline{Q} + \underline{K}, \underline{Q}) = \frac{1}{2} \sum_{\underline{k}, n, m} f_{nm}(\underline{k} + \underline{K} + \underline{Q}, \underline{k}) f_{nm}^*(\underline{k} + \underline{Q}, \underline{k}) \frac{n_{\underline{k}m} - n_{\underline{k} + \underline{Q}n}}{\epsilon_{\underline{k} + \underline{Q}n} - \epsilon_{\underline{k}m}} \quad (5.22)$$

and
$$f_{nm}(\underline{k} + \underline{Q}, \underline{k}) = \int \psi_{\underline{k}m}^* e^{i\underline{Q} \cdot \underline{r}} \psi_{\underline{k}+ \underline{Q}n} d\underline{r} \quad (5.23)$$

These authors make an averaging approximation $f_{nm}(\underline{k} + \underline{Q}, \underline{k}) = f(\underline{Q})$ to turn the matrix equation into an ordinary equation, giving a simple enhancement to the susceptibility. One can solve the selfconsistency equation for this type of interaction using a similar approximation for the matrix elements.

From (5.16) one finds

$$\langle S_{\underline{Q}}^z \rangle^{\Pi} = \frac{2 J_{\text{eff}}}{N} \chi_{\Pi}^{\circ} \langle S_{\underline{Q}}^z \rangle \quad (5.24)$$

where χ_{Π}° is the paramagnetic susceptibility of the electrons in region II, defined by (5.20). Thus from (5.13), (5.14) and (5.24), one finds the following selfconsistency equation for $\langle S_{\underline{Q}}^z \rangle$,

$$1 - \frac{2 J_{\text{eff}}}{N} \chi_{\Pi}^{\circ}(\underline{Q}) = \frac{1}{2} \frac{J_{\text{eff}}}{N} \sum_{\underline{k}} \frac{|F_{NM}(\underline{k} + \underline{Q}, \underline{k})|^2 (n_{\underline{k}1}^{\text{I}} - n_{\underline{k}2}^{\text{I}})}{\sqrt{|\Gamma_{NM}(\underline{Q})|^2 + \frac{1}{4}(\epsilon_{\underline{k}+ \underline{Q}N} - \epsilon_{\underline{k}M})^2}} \quad (5.25)$$

$\chi_{\Pi}^{\circ}(\underline{Q})$ may be reasonably flat as a function of $\underline{Q}$, showing that only part of the susceptibility, that involving the nesting, is important for determining $\underline{Q}$.

The dramatic effects of the pressure on the Neel temperature discussed by Rice et al (1969) can be interpreted in a new way. Assuming $F_{NM}(\underline{k} + \underline{Q}, \underline{k})$ is approximately 1 and

approximating the nesting bands by parabolic bands (Fodders and Martin 1966) one has the approximate formula for the Néel temperature,

$$T_N = T_B \exp -1/\lambda \quad (5.25)$$

with kT_B of the order of a band energy and

$$\lambda = N(0) J_{\text{eff}} / 1 - J_{\text{eff}} \chi_{\text{II}}(\underline{Q}) \quad (5.26)$$

The rapid decrease in T_N with pressure, may be due to a decrease in $\chi_{\text{II}}(\underline{Q})$ rather than a variation in the density of states $N(0)$ of the nesting surfaces. The smallness of $\chi_{\text{II}}(\underline{Q})$ may also explain why molybdenum and tungsten are not antiferromagnetic though they should have a similar band structure to chromium.

To derive (5.25) we assumed that in region I the dominant mixing is from the nesting between different parts of two bands for each spin density wave. The nesting regions important for each wave are independent, so the only interactions between spin density waves are weak ones. In region II, because of perturbation theory, one can include the coupling of all the spin density waves. One can investigate how the bands in (5.11) are altered, by including all the interactions.

We assume that the antiferromagnetic energy gaps have a constant value

$$g = \Gamma_{NM}(\underline{Q}) \quad (5.27)$$

Until now, in region I, we have assumed that $|\varepsilon_{\underline{k}M} - \varepsilon_{\underline{k}+\underline{Q}N}|$ is of order g , and is very much less than $|\varepsilon_{\underline{k}M} - \varepsilon_{\underline{k}-\underline{Q}N}|$ so that the two band approximation applies. From (5.11) the energy bands in region I are

$$E_{1,2}^I(\underline{k}) = \frac{1}{2}(\varepsilon_{\underline{k}M} + \varepsilon_{\underline{k}+\underline{Q}N}) \pm \sqrt{g^2 + \frac{1}{4}(\varepsilon_{\underline{k}M} - \varepsilon_{\underline{k}+\underline{Q}N})^2} \quad (5.28)$$

If we include the mixing with $\psi_{\underline{k}-\underline{Q}N}$ in (5.11) the bands are determined by solving

$$\det \begin{pmatrix} \omega - \varepsilon_{\underline{k}+\underline{Q}N} & g & 0 \\ g & \omega - \varepsilon_{\underline{k}M} & g \\ 0 & g & \omega - \varepsilon_{\underline{k}-\underline{Q}N} \end{pmatrix} = 0 \quad (5.29)$$

The lowest root gives the lowest energy band $E_{MIN}^I(\underline{k})$, which is, for $|\varepsilon_{\underline{k}M} - \varepsilon_{\underline{k}+\underline{Q}N}| \ll |\varepsilon_{\underline{k}M} - \varepsilon_{\underline{k}-\underline{Q}N}|$

$$E_{MIN}^I(\underline{k}) = \frac{1}{2}(\varepsilon_{\underline{k}M} + \varepsilon_{\underline{k}+\underline{Q}N}) - \sqrt{g_1^2 + \frac{1}{4}(\varepsilon_{\underline{k}M} - \varepsilon_{\underline{k}+\underline{Q}N})^2} \quad (5.30)$$

where
$$g_1^2 = g^2 \left(1 + \frac{E_1^I(\underline{k}) - \varepsilon_{\underline{k}+\underline{Q}N}}{E_1^I(\underline{k}) - \varepsilon_{\underline{k}-\underline{Q}N}} \right) \quad (5.31)$$

One sees from (5.30) that the correction to g is small, of order g/ε_2 , where ε_2 is roughly the band width, showing that the bands given by (5.28) are approximately correct. This

justifies taking the two band approximation. Going to second order in g , when Q is incommensurate with the lattice, produces second order energy gaps, of order g^2/ε_2 , in the band structure.

However as Q tends to $\pi/a(1, 0, 0)$, $|\varepsilon_{kM} - \varepsilon_{k+QN}|$ becomes of order g , so (5.29) must be solved more generally. If we assume that Q_x is still not too close to π/a , so that ψ_{k-2QN} , ψ_{k-2QN} etc. need not be included in (5.29), (5.29) becomes approximately

$$(\omega - \varepsilon_{k+QN})(\omega - \varepsilon_{kM}) - 2g^2 = 0 \quad (5.32)$$

The lower energy band solving this equation can be compared with (5.29) when $Q_x = \pi/a$, for which $\psi_{k-QN} \equiv \psi_{k+QN}$ and which has the solution

$$(\omega - \varepsilon_{k+QN})(\omega - \varepsilon_{kM}) - 4g^2 = 0 \quad (5.33)$$

In this case the gap is doubled but no higher order gaps occur. Either (5.32) or (5.33) may give the lower energy, since $|\varepsilon_{k+QN} - \varepsilon_{kM}|$ is $O(g)$.

Hence for some Q_x such that $|Q_x - \pi/a| < \delta q$, where δq is very small, the perfect antiferromagnetic state has a lower energy than the spin density state and a first order phase transition takes place. This apparently happens in Cr-Mn alloys where $\delta q = .03 \pi/a$. For $|Q_x - \pi/a| > \delta q$ the variation of Q of the spin density wave depends on the

changing shape of the bands, however, in the range $|\mathbf{q} - \pi/a| < \delta q$ the spin density state does not occur. In any case in this range the spin density wave state should include the effects of second and third order gaps and the interactions between the spin density waves, and would be very complicated. In this range, where higher order effects are important, $\chi^0(\mathbf{q})$ has no relevance in determining the ordering.

IV Spin Waves

In chapter 4 we have shown how to treat spin waves in a model without necessarily having perfect nesting of the bands. In this chapter we have described the spin density state of chromium. We shall now estimate the energy $\hbar\omega = \hbar c q$ of spin waves for dilute Cr-Mn alloys, on which experiments have been done (Sinha et al 1969, Als-Nielsen and Dietrich 1969). For 8% manganese this alloy has a perfect antiferromagnetic structure. One can still argue that the energy gap is small compared with typical band energies. Hence the simplification of the weak coupling model discussed in this chapter still applies.

For a one-band itinerant model of an antiferromagnet, defined by (4.8), the spin wave velocity is

$$c = \sqrt{2ad} / \hbar \langle S^z_{\mathbf{q}} \rangle \quad (5.34)$$

where
$$\alpha = \sum_{\underline{k}} \varepsilon_2(\underline{k}) \sin \theta_{\underline{k}} \cos^2 \theta_{\underline{k}} n_{\underline{k}}, \quad (5.35)$$

$$d = \frac{1}{6} \sum_{\underline{k}} \left(\nabla \varepsilon_2(\underline{k}) \nabla (\sin \theta_{\underline{k}}) - \frac{1}{2g} |\nabla \varepsilon_1(\underline{k})|^2 \cos^3 \theta_{\underline{k}} \right) n_{\underline{k}}, \quad (5.36)$$

ε_1 , ε_2 and $\theta_{\underline{k}}$ are defined in terms of the non-magnetic band by

$$\varepsilon_{1,2}(\underline{k}) = \frac{1}{2} (\varepsilon(\underline{k}) \pm \varepsilon(\underline{k} + \underline{Q})) , \quad \tan \theta_{\underline{k}} = \varepsilon_2(\underline{k})/g \quad (5.37)$$

where $g = I/2N \langle S_Q^z \rangle$ satisfies (4.18).

It has been assumed that ε_F lies in parts of the lower antiferromagnetic energy band which are nearly paramagnetic. The only intraband transitions occur in these parts of the band. In addition $g \gg \hbar \omega_q$ so that transitions between upper and lower bands require energies far greater than typical spin wave energies.

In chromium we have shown that, though region I is strongly polarised, most of the moment comes from the large number of electrons in region II. We can use the same idea in the one-band model, by assuming that a large part of the band is in region II with just a small part, consisting of the nesting region, in region I. From (5.35)

$$\alpha = \frac{I}{N} \langle S_Q^z \rangle^2 \quad (5.38)$$

since $\sin \theta_k \simeq 1$ in region II, and the volume of region II is very much greater than the volume of region I. Hence one finds

$$\sqrt{a} \langle S_Q^z \rangle = \sqrt{I/N} \quad (5.39)$$

Then from (5.34)

$$c = \frac{1}{\hbar} \sqrt{\frac{2Id}{N}} \quad (5.40)$$

d is of order g^2/ε_2^3 in region II. So, assuming the volume of region I is greater than $(g/\varepsilon_2)^2 \times$ volume of region II, d is dominated by the energy bands in region I, and we need only integrate (5.36) for values of k over the nesting surfaces.

Though (5.40) applies only to a one-band model, it can be applied to chromium. The integration over k , in d , is now over the parts of the bands II and III involved in the nesting. We therefore define

$$\varepsilon_{1,2}(k) = \frac{1}{2}(\varepsilon_M(k) \pm \varepsilon_N(k+Q)) \quad , \quad I = J_{\text{eff}} \quad (5.41)$$

and $F_{NM}(k+Q, k) = 1.$

Assuming that one can approximate the nesting surfaces by parabolic bands (Pedders and Martin 1966), one can write

$$\varepsilon_1(k) = 0 \quad , \quad \varepsilon_2(k) = \frac{\hbar^2}{2m}(k^2 - k_F^2) \simeq \hbar v_F(k - k_F) \quad (5.42)$$

in region I, near the Fermi surface whose radius is k_F . Then (5.36) and (5.37) give

$$d \simeq \frac{1}{6} \frac{\Omega N}{(2\pi)^3} \int_0^{k_F} \frac{(\hbar v_F)^2 g^2 4\pi k^2 dk}{(\hbar^2 v_F^2 (k - k_F)^2 + g^2)^{3/2}} \simeq \frac{1}{3} \frac{\Omega N}{(2\pi)^2} \hbar v_F k_F^2 \quad (5.43)$$

since $g \ll \hbar v_F k_F$ or $g \ll \mathcal{E}_2$ in region II. So from (5.40) and (5.43)

$$\hbar \omega = \hbar c q = \sqrt{\frac{2}{3} J_{\text{eff}} \hbar a v_F \left(\frac{a k_F}{\pi} \right)} q \quad (5.44)$$

where $\Omega = \frac{1}{8}(2a)^3$ is the volume of the unit cell. The spin wave energy depends on the volume of the nesting surfaces through $a k_F / \pi$. If the electron pocket occupies about half the first Brillouin zone, then equating the number of electrons in the pocket to $n/2$ gives $k_F = \sqrt{3/4\pi} \pi/a$. In fact the Fermi surface is not a sphere, so that numerical factors can only be approximate. Putting $k_F = \pi / \sqrt{2} a$ for convenience, (5.44) becomes

$$\hbar \omega = \sqrt{\frac{1}{3} J_{\text{eff}} a \hbar v_F} q \quad (5.45)$$

One can compare this result with others obtained experimentally and theoretically for Cr-Mn alloys. Theoretically (Pedders and Martin 1966, Sokoloff 1969, Liu 1970) a spin wave velocity c of order of magnitude the Fermi velocity of the nesting surfaces, is obtained. These authors

use a completely different derivation of c . Their formula for c is independent of J_{eff} in the simplest approximation. Their method only applies when there are perfect nesting bands. From neutron scattering experiments Sinha et al (1969) obtain $c = 1.29 \times 10^7$ cm/sec, and Als-Nielsen and Pietrich (1969) obtain $c = .69 \times 10^7$ cm/sec. Using $J_{\text{eff}} = 1$ ev, $v_F = 5.1 \times 10^7$ cm/sec (Sinha et al 1969) and $a = 1.5$ Å, (5.45) yields

$$\hbar c = 1,300 \text{ meV } \text{\AA}$$

$$\text{or} \quad c = 2 \times 10^7 \text{ cm/sec} \quad (5.46)$$

Thus our result is in fair agreement with experiment.

Chapter 6

Spin Waves in a System of Itinerant Electrons and Local Moments

I Local Moments in Alloys and Rare Earths

There are a number of different cases in which the spin density wave state of itinerant electrons is associated with interactions between the itinerant electrons and local moments. Firstly, the properties of rare earths can be described by assuming the f shell electrons act as local moments interacting via the s band. Secondly, Overhauser's (1960) original investigation of the spin density wave state was based on ideas of local moments in alloy systems like Cu-Mn etc. Marshall (1960) was able to explain the low temperature peak in the specific heat, which prompted Overhauser's suggestion, without assuming that the conduction band was in a long-range antiferromagnetic state. However, the ideas of Overhauser may well be applied to a nearly antiferromagnetic metal. This is a metal, analogous to palladium which is nearly ferromagnetic, that has a band structure and electron-electron interaction which nearly satisfy the conditions for antiferromagnetism.

We will consider a one band itinerant electron gas, containing local moments and with a preference for a spin density wave ground state. The wave vector $\underline{Q}$ of the spin

density wave is not necessarily commensurate with the lattice i.e. $\underline{Q} \neq \frac{1}{2}\underline{K}$ where $\underline{K}$ is a base vector of the reciprocal lattice. If $\underline{Q} = \frac{1}{2}\underline{K}$ one would have a perfect antiferromagnetic structure in which the spin polarisation is in opposite directions on the two sublattices. The Hamiltonian we consider is

$$H = \sum_{\underline{k}} \epsilon(\underline{k}) n_{\underline{k}\sigma} + I \sum_i n_{i\uparrow} n_{i\downarrow} + J \sum_i \underline{\sigma}_i \cdot \underline{S}_i c_i \quad (6.1)$$

where $n_{i\sigma}$ is the density of the σ spin itinerant electrons on the $\underline{R}_i$ of the lattice in a Wannier representation, $\underline{\sigma}_i$ is the spin density of the itinerant electrons and $\underline{S}_i$ is the spin of the local moment on the site $\underline{R}_i$. $c_i = 0$ or 1 depending on whether there is a local moment on the site. For the rare earths $c_i = 1$ since there are f electrons on every site. For dilute alloys in which the solute forms local moments, c_i is predominantly 0.

The form of the interaction between the local moments and itinerant electrons is known as the s-d or s-f interaction, $H_{sd(f)}$ (Kasuya 1966). Ideally the operator $\underline{S}_i$ in (6.1) should describe the moment of the impurity atom at $\underline{R}_i$ and be independent of the matrix in which the impurity lies and independent of the temperature. Though this condition does not apply, much experimental work (Arrot 1966, Van der Berg 1965) has been done on alloys of 3d transition

metals in non-magnetic metals such as Cu, Ag, Au and elements of the 4d transition series. The rigorous theoretical treatments of impurities in metals (Clogston 1962, Anderson 1961) allow the possibility of a virtual bound state forming, associated with the impurity atom. Thus the s-d model is a convenient approximation, though in this case, J is only phenomenological. The f-shell radii of the rare earth atoms are so small that they do not have significant overlap. In this case there is a sounder basis for putting

$$J = \iint \left(\omega_s(r_1) \omega_{4f}(r_2) \frac{e^2}{r_{12}} \omega_s(r_2) \omega_{4f}(r_1) \right) dr_1 dr_2 \quad (6.2)$$

where the integral is over a unit cell.

Positive and negative values of J , corresponding to parallel and antiparallel alignment of the itinerant and localised electrons, arise quite naturally in the Anderson model. The form of the interaction in (6.1) and (6.2) occurs when overlaps between nearest neighbour sites are neglected, and then H_{sd} commutes with the Fourier components of the total spin density operators, that is

$$\left[H_{sd} , \sum_i (S_i c_i + \sigma_i) e^{iq \cdot R_i} \right] = 0 \quad (6.3)$$

Doniach and Wohlfarth (1967) use the form of H_{sd} given by (6.1) when discussing ferromagnetic Pd-Fe in the R.P.A. approximation.

They obtain a formula $\hbar\omega = Dq^2$ for acoustic spin waves in Pd-Fe, but D can also be obtained from the general formula for D , discussed in chapter 2, using (6.3) (Edwards 1970).

II The Ground State

One can write (6.1) as

$$H = \sum_{\underline{k}} \varepsilon(\underline{k}) n_{\underline{k}\sigma} + \frac{I}{N} \sum_{\underline{k}} \sigma_{\underline{k}}^z \sigma_{-\underline{k}}^z + \frac{J}{N} \sum_{\underline{k}} \underline{S}_{\underline{k}} \cdot \underline{\sigma}_{-\underline{k}} \quad (6.4)$$

where the summation is over the whole Brillouin zone, and the Fourier transforms $\sigma_{\underline{k}}^z$, $\underline{S}_{\underline{k}}$ of the itinerant and localised spins are defined by

$$\begin{aligned} \sigma_{\underline{k}}^z &= \frac{1}{2} \sum_{\underline{k}_1} (c_{\underline{k}_1+\underline{k}\uparrow}^\dagger c_{\underline{k}_1\uparrow} - c_{\underline{k}_1+\underline{k}\downarrow}^\dagger c_{\underline{k}_1\downarrow}) \\ \text{and} \quad \underline{S}_{\underline{k}} &= \sum_i \underline{S}_i c_i e^{i\underline{k} \cdot \underline{R}_i} \end{aligned} \quad (6.5)$$

If we assume that the system is in a longitudinal spin density wave state, the Hartree-Fock approximation, as in (4.10) and (5.11), is equivalent to approximating the interaction by

$$\sigma_{\underline{k}}^z \sigma_{-\underline{k}}^z = \langle \sigma_{\underline{k}}^z \rangle \sigma_{-\underline{k}}^z + \langle \sigma_{-\underline{k}}^z \rangle \sigma_{\underline{k}}^z \quad (6.6)$$

where $\langle \sigma_{\underline{k}}^z \rangle = 0$ unless $\underline{k} = \pm \underline{Q}$. For spiral spin density waves (Arrott 1966) one makes the following

approximation to the interaction

$$\sigma_{\underline{k}}^+ \sigma_{-\underline{k}}^- = \langle \sigma_{\underline{k}}^+ \rangle \sigma_{-\underline{k}}^- + \langle \sigma_{-\underline{k}}^- \rangle \sigma_{\underline{k}}^+ \quad (6.7)$$

For the spiral spin density wave $\langle \sigma_{\underline{k}}^+ \rangle = 0$ and $\langle \sigma_{-\underline{k}}^- \rangle = 0$ unless $\underline{k} = \underline{Q}$. Spiral spin densities occur frequently in the rare earths and have the special property that the polarisation on each site has the same magnitude, though not the same direction. This is not the case for incommensurate linear spin density waves. The following analysis deals with linear spin density wave states but we will mention how to get results for other types of spin ordering. One also approximates the local moment interaction by

$$\underline{S}_{\underline{k}} \cdot \underline{S}_{-\underline{k}} = \langle S_{\underline{k}}^z \rangle S_{-\underline{k}}^z + \langle S_{-\underline{k}}^z \rangle S_{\underline{k}}^z \quad (6.8)$$

Defining $g_1 = \frac{J}{2N} \langle S_{\underline{Q}}^z \rangle$, $g = \frac{I}{N} \langle \sigma_{\underline{Q}}^z \rangle$ (6.9)

and assuming $\langle \sigma_{\underline{Q}}^z \rangle = \langle \sigma_{\underline{Q}}^z \rangle$, $\langle S_{\underline{Q}}^z \rangle = \langle S_{-\underline{Q}}^z \rangle$ we get

$$(\omega - \epsilon_{\underline{k}}) \langle\langle C_{\underline{k}\sigma}; C_{\underline{k}\sigma}^+ \rangle\rangle_{\omega} = (g + g_1) \left(\langle\langle C_{\underline{k}\underline{Q}\uparrow}; C_{\underline{k}\uparrow}^+ \rangle\rangle_{\omega} + \langle\langle C_{\underline{k}+\underline{Q}\uparrow}; C_{\underline{k}\uparrow}^+ \rangle\rangle_{\omega} \right) + \dots \quad (6.10)$$

Further equations of motion will couple $\psi_{\underline{k}}$ and $\psi_{\underline{k}+2\underline{Q}}$ etc. To solve the problem we make the same assumptions as in chapter 5. For some $\underline{k}$, the coupling between $\psi_{\underline{k}\uparrow}$ and $\psi_{\underline{k}+\underline{Q}\uparrow}$ is strong so we may neglect the coupling between $\psi_{\underline{k}\uparrow}$ and $\psi_{\underline{k}+2\underline{Q}\uparrow}$. For other $\underline{k}$, the coupling is weak so the coupling

of $\psi_{k\uparrow}$ with $\psi_{k+Q\uparrow}$ is independent of the coupling of $\psi_{k\uparrow}$ with $\psi_{k-Q\uparrow}$. The equations of motion can then be written in the form

$$G_{\alpha\beta}^{\uparrow}(k, \omega) = \begin{pmatrix} \omega - \varepsilon(k) & g + g_1 \\ g + g_1 & \omega - \varepsilon(k+Q) \end{pmatrix}^{-1} \quad (6.11)$$

where $G_{\alpha\beta}^{\uparrow}(k, \omega) = \ll \begin{pmatrix} c_{k\uparrow} \\ c_{k+Q\uparrow} \end{pmatrix}; \begin{pmatrix} c_{k\uparrow}^+ \\ c_{k+Q\uparrow}^+ \end{pmatrix} \gg_{\omega}$

By diagonalising $G_{\alpha\beta}^{\uparrow}$ one obtains the new antiferromagnetic bands ψ_{k_1}, ψ_{k_2} of energy $E_{1,2}(k)$ where

$$\begin{aligned} \psi_{k_1} &= \cos \frac{1}{2}(\pi_2 - \theta_k) \psi_{k\uparrow} + \sin \frac{1}{2}(\pi_2 - \theta_k) \psi_{k+Q\uparrow} \\ \psi_{k_2} &= \cos \frac{1}{2}(\pi_2 - \theta_k) \psi_{k+Q\uparrow} - \sin \frac{1}{2}(\pi_2 - \theta_k) \psi_{k\uparrow} \end{aligned} \quad (6.12)$$

where $\tan \theta_k = \varepsilon_2(k)/g + g_1$, and $\varepsilon_2(k) = \frac{1}{2}(\varepsilon(k+Q) - \varepsilon(k))$

$$E_{1,2}(k) = \frac{1}{2}(\varepsilon(k) + \varepsilon(k+Q)) \pm \sqrt{\varepsilon_2^2(k) + (g + g_1)^2} \quad (6.13)$$

The phases $\pi/4$ are put in so that θ_k will correspond to the definition we used in chapter 4. For spiral spin densities one would investigate the same equations but would include the coupling between $\psi_{k\uparrow}$ and $\psi_{k+Q\downarrow}$.

From the correlation functions of $G_{\alpha\beta}$ in (6.11) one obtains the following selfconsistency equation, just as in

(4.18),

$$g = \frac{I}{N} \langle \sigma_{\underline{Q}}^z \rangle = \frac{I}{2N} \sum_{\underline{k}} \frac{(n_{\underline{k}1} - n_{\underline{k}2})(g + g_1)}{\sqrt{(g + g_1)^2 + \varepsilon_2^2(\underline{k})}} \quad (6.14)$$

The occupation numbers $n_{\underline{k}1,2}$ of the antiferromagnetic bands are chosen to minimise the energy.

Defining
$$\chi_{\text{SDW}} = \frac{1}{4N} \sum_{\underline{k}} \frac{(n_{\underline{k}1} - n_{\underline{k}2})}{\sqrt{(g + g_1)^2 + \varepsilon_2^2(\underline{k})}}$$

one gets
$$g = \frac{I \chi_{\text{SDW}}}{1 - 2I \chi_{\text{SDW}}} 2g_1 \quad (6.15)$$

If the coupling between $\psi_{\underline{k}}$ and $\psi_{\underline{k}+\underline{Q}}$ is always weak so that $g + g_1 \ll |\varepsilon(\underline{k}+\underline{Q}) - \varepsilon(\underline{k})|$ then (6.15) reduces to

$$\langle \sigma_{\underline{Q}}^z \rangle = \frac{\chi^0(\underline{Q})}{1 - 2I \chi^0(\underline{Q})} J \langle S_{\underline{Q}}^z \rangle \quad (6.16)$$

where
$$\chi^0(\underline{Q}) = \chi_{\text{SDW}} (g + g_1 = 0)$$

$$= \frac{1}{2N} \sum_{\underline{k}} \frac{n_{\underline{k}} - n_{\underline{k}+\underline{Q}}}{\varepsilon(\underline{k}+\underline{Q}) - \varepsilon(\underline{k})}$$

is the non-interacting paramagnetic susceptibility, or response, of the itinerant electrons. So we see that in this case the electrons respond linearly to the local moments.

For a band with no interactions between the itinerant electrons, the difference in the band energy between the antiferromagnetic state and the paramagnetic state is

$$\delta E = 4 \sum_{\mathbf{k}} (n_{\mathbf{k}_1} - n_{\mathbf{k}_2}) (\sqrt{g_1^2 + \epsilon_2^2(\mathbf{k})} - \epsilon_2(\mathbf{k})) \quad (6.17)$$

$$\approx \sum_{\mathbf{Q}_1 = \pm \mathbf{Q}} \frac{1}{N} \chi_0(\mathbf{Q}_1) J^2 \langle S_{\mathbf{Q}_1}^z \rangle^2 \quad (6.18)$$

in terms of the reduced susceptibility of the itinerant electrons. A factor 2 arises because we have two spin density waves of wave vector $\mathbf{Q}$ and $-\mathbf{Q}$, and half the local moments are in one spin density wave and half in the other. So

$$\langle S_{\mathbf{Q}}^z \rangle = \frac{1}{2} N S \bar{c} \quad (6.19)$$

where $\bar{c}$ is the concentration of local moments and S is the value of the local moment. For the perfect antiferromagnetic state ($\mathbf{Q} = \frac{1}{2}\mathbf{K}$) the factor 2 in δE would not be there but

$$\langle S_{\mathbf{Q}}^z \rangle = N S \bar{c} \quad (6.20)$$

Hence the energy reduction in the perfect antiferromagnetic state is twice that in the spin density wave state. This occurs in linear but not spiral spin density wave states.

The result (6.18) can also be obtained by using the Ruderman-Kittel-Yosida interaction (Matis 1965) in which the local moments interact through an effective Heisenberg interaction, whose exchange constant is $J_{\text{RKY}}(\mathbf{q}) = J^2 \chi^0(\mathbf{q})$. This is the usual Hamiltonian for discussing the rare earths. For rare earths with a local moment on every site, the wave

vector $\underline{Q}$ of the spin density wave ground state configuration will be the one for which $\chi^0(\underline{q})$ is a maximum. This gives a general explanation for the ordering in the heavy rare earths.

There are two refinements to using the R.K.Y. interaction. Firstly, g_1 can become quite large and therefore the polarisation of the itinerant electrons becomes quite important and the Fermi surface is distorted. Elliott and Wedgwood (1964) therefore go beyond second order perturbation theory, to which the R.K.Y. interaction is equivalent, by investigating the spin density wave state via the Hartree-Fock states of the conduction electrons, as we have done in chapters 4, 5 and this chapter. The conduction electrons are assumed to be free electron like and I is zero. Elliott and Wedgwood include the energy lowering due to gaps at superzone boundaries $\underline{K} + \underline{Q}$, and suggest that a large portion of the Fermi surface is destroyed by these gaps.

Secondly, the R.K.Y. interaction assumes that the response of the itinerant electrons is static. One ought to include the lifetime τ_e or the mean free path λ of the itinerant electrons. The lifetime τ_e would be dependent on the spin disorder scattering of the itinerant electrons with the local moments. The effect of the finite lifetime is to introduce an exponential factor $e^{-R/\lambda}$ (de Gennes 1962) into the exchange integral $J_{RKY}(R)$. This is however, only a first step to including the dynamics of the itinerant

electrons.

For dilute alloys like Cu-Mn the distribution of the local moments is random and so is the effective field due to them. Although the interaction between the local moment and itinerant electrons may be described by an R.K.Y. interaction (Marshall 1960) the maximum in $\chi^0(q)$ must be very sharp indeed for just one spin density wave of wave vector q to produce the energy minimum. The range of the polarisation about an impurity is of order Δq^{-1} , where Δq is the width of the peak in $\chi^0(q)$. As Herring (1966) illustrates, a random field will have lower energy, provided the concentration of impurities is very small. Thus it is more likely that the system is without long-range order (Arrott 1966).

There may however be dilute alloys in which the long-range order does occur. Doniach and Wohlfarth (1967) have investigated the dynamic properties of Pd-Fe. Because palladium satisfies the condition, $1 \gtrsim \text{Im}(\epsilon_F)$, it is nearly ferromagnetic. It becomes magnetised even at very low concentrations of iron impurities, and when magnetised carries its own spin waves. Doniach (1967) has also shown that the relaxation time of palladium is many times slower than that of a metal of non-interacting electrons, so that the response of palladium to fluctuations of local spins cannot be assumed to be static.

Doniach and Wohlfarth (1967) investigate the dynamic

R.P.A. susceptibility of the palladium and find that the spin wave spectrum may be derived in terms of a frequency dependent R.K.Y. interaction between the iron spins. The spin wave spectrum is given by

$$\omega = \frac{1}{N} J^2 \langle S_0^z \rangle [\chi(0,0) - \chi(q,\omega)] \quad (6.21)$$

where $\chi(q,\omega)$ is the transverse component of the susceptibility of the palladium with an enhancement I . The frequency dependence of the interaction in (6.21) changes the spin wave spectrum drastically, leading for example to an optical branch. Being able to write the R.P.A. approximation in this way is a fortuitous feature for the ferromagnet. For the ferromagnet, the basis of single particle states in the R.P.A. approximation is the same as for the paramagnet. The energies of the up and down states are just split. For an antiferromagnet the single particle basis is a k dependent mixture of ψ_k and ψ_{k+Q} depending on the polarisation throughout the crystal. Thus the R.P.A. treatment of the response in an antiferromagnet used by Sokoloff (1969), is not a good approximation. For the antiferromagnet one is not able to get a simple result analogous to (6.21) for the spin wave spectrum.

We will investigate an alloy system in which a spin density wave state occurs. Suppose there is a weak concen-

-tration of impurities in a host metal which is nearly antiferromagnetic (Moriya 1970). Then $g_1 \ll 1$ and $c \ll 1$ but for some Q

$$1 > 2 I \chi^0(Q) = 1 - \delta(Q), \quad \delta(Q) \ll 1 \quad (6.22)$$

So $g \simeq 1/\delta(Q)$ from (6.15). At low temperatures, if the local spins are fully aligned, $g_1 = J\bar{S}$, so that an approximate condition for long-range ordering is that $J\bar{S}/I \delta(Q)$ be of order 1. In this case the enhancement due to I compensates for there not being a local moment at every site. If the left hand inequality in (6.22) is relaxed, the host itinerant electrons have long-range order, even when there are no local moments. The condition (6.22) on the host metal is quite severe, but there have been suggestions that it is satisfied by scandium and yttrium, containing rare earth impurities like gadolinium and terbium (Wohllengen 1968).

It should be emphasised that the relationship between the following theory of spin waves and the rare earths is only qualitative. The rare earths have a hexagonal lattice structure with more than one atom to a cell. Also, it is usual to assume that the s -band in rare earths is free electron-like, whereas we have assumed the itinerant band is d -like with a tight-binding form. According to Wohllengen, the long-range spin correlations in scandium is associated

with the d-electrons.

III Spin Wave Energy

We have already calculated, in chapter 4, the energy of spin waves in an itinerant electron model of a perfect antiferromagnet. In order to apply the formulae in chapter 4 directly, we assume $\underline{Q} = \frac{1}{2}\underline{K}$. It should be possible to generalise our results for other $\underline{Q}$. Since the s-d interaction in (6.1) commutes with $S_{\underline{Q}}^z + \sigma_{\underline{Q}}^z$ we can take over the procedure used in chapter 4 exactly. This time however, we look at the generalised susceptibility

$$\ll S_{\mu-\underline{Q}}^+ + \sigma_{\mu-\underline{Q}}^+ ; S_{\nu\underline{Q}}^- + \sigma_{\nu\underline{Q}}^- \gg_{\omega} \quad (6.23)$$

involving the Fourier transforms of the total spin on the sublattices A and B. Then the formula for the energy $\hbar\omega = \hbar\epsilon_{\underline{Q}}$ of spin waves in the antiferromagnet, is derived by looking at the poles of the susceptibility (6.23) in the region $\omega/\epsilon_{\underline{Q}} \ll 1$, with the result

$$c = \sqrt{2a(d-\Delta)} / \langle \sigma_{\underline{Q}}^z + S_{\underline{Q}}^z \rangle \quad (6.24)$$

a, d, Δ are written in terms of the antiferromagnetic energy bands and are defined by

$$d = \frac{1}{6} \sum_{\underline{k}} (\nabla^2 \varepsilon_1(\underline{k}) + \nabla^2 \varepsilon_2(\underline{k}) \sin \theta_{\underline{k}}) (n_{\underline{k}_1} + n_{\underline{k}_2}) - \frac{1}{g+g_1} |\nabla \varepsilon_1(\underline{k})|^2 \cos^3 \theta_{\underline{k}} (n_{\underline{k}_1} - n_{\underline{k}_2}) \quad (6.25)$$

$$\Delta = \frac{1}{6} \sum_{\underline{k}} \nabla (\sin^2 \theta_{\underline{k}} \nabla \varepsilon_1(\underline{k})) (n_{\underline{k}_1} + n_{\underline{k}_2}) \quad (6.26)$$

$$a = \sum_{\underline{k}} \varepsilon_2(\underline{k}) \sin \theta_{\underline{k}} \cos^2 \theta_{\underline{k}} (n_{\underline{k}_1} - n_{\underline{k}_2}) \quad (6.27)$$

where $\tan \theta_{\underline{k}} = \varepsilon_2(\underline{k})/g+g_1$

and $\varepsilon_{1,2}(\underline{k}) = \frac{1}{2}(\varepsilon(\underline{k}+\underline{Q}) - \varepsilon(\underline{k}))$.

The only difference with the previous expression (4.45) for a , is that now $\langle \sigma_{\underline{Q}}^z + S_{\underline{Q}}^z \rangle$ appears in the denominator, instead of just $\langle \sigma_{\underline{Q}}^z \rangle$, and $g + g_1$ replaces g . It is assumed the Fermi level lies in the lower energy band only. Δ is the contribution arising from intraband single particle excitations in the region $q/\omega \ll 1$. $d - \Delta$ may be simplified as

$$d - \Delta = \frac{1}{6} \sum_{\underline{k}} (\nabla \varepsilon_2(\underline{k}) \nabla (\sin \theta_{\underline{k}}) - \frac{1}{g+g_1} |\nabla \varepsilon_1(\underline{k})|^2 \cos^3 \theta_{\underline{k}} + \nabla (\cos^2 \theta_{\underline{k}} \nabla \varepsilon_1(\underline{k}))) n_{\underline{k}_1} \quad (6.28)$$

The usual procedure is to assume that the Fermi level lies in the antiferromagnetic energy gap when it is in regions of strong polarisation, $g + g_1 \gg \varepsilon_2(\underline{k})$, so that the only contribution to the last term in (6.28) involving ε_1 comes from regions of weak polarisation where $g + g_1 \ll \varepsilon_2(\underline{k})$. Since this term in (6.28) is of order $(g + g_1 / \varepsilon_2)^2$ it can often be neglected. In for example the rare earths, a large

section of the Fermi surface may be cut by the antiferromagnetic energy gaps, and electrons near these gaps are dominant in the spin wave velocity. Hence in this situation we can write

$$d - \Delta = \frac{1}{6} \sum_{\underline{k}} \left(\nabla \varepsilon_2(\underline{k}) \nabla (\sin \theta_{\underline{k}}) - \frac{1}{g+g_1} |\nabla \varepsilon_1(\underline{k})|^2 \cos^3 \theta_{\underline{k}} \right) n_{\underline{k}} \quad (6.29)$$

and the formula for c given by (6.24), (6.27) and (6.29) is a simple generalisation of (4.49). Now both the itinerant and the localised electrons carry the spin wave.

There are two disadvantages in using the R.K.Y. interaction as an effective Heisenberg Hamiltonian for the localised spins to calculate spin waves. Firstly, the R.K.Y. interaction (6.18) is only correct to the second order in the local moment interaction, that is to order J^2 . One calculates the polarisation of the conduction electrons to the first order in g/ε_2 only, where ε_2 is the mean value of $\varepsilon_2(\underline{k})$. Secondly, it uses the static response of the itinerant electrons and so ignores any excitations of these electrons. This condition may be written

$$\tau_s^{-1} \ll c q \ll \tau_e^{-1} \quad (6.30)$$

where τ_e , τ_s are the lifetimes of low energy excitations of the itinerant and localised electrons respectively (Doniach 1967).

The approach used above treats the localised and itinerant electrons on the same footing. We can compare our formula (6.24) with the R.K.Y. formula for the spin wave energy in the limit where the R.K.Y. interaction should apply i.e. $g_1 \ll \xi_2(\underline{k})$ for all $\underline{k}$. In this limit the full weights of the spin wave and the single particle excitations, d and Δ , in (6.24) must be retained. We will assume that the inclusion of Δ is an adequate correction for the single particle excitations. This is a different approximation to the "static" approximation (6.30). We neglect correlations in the conduction band by putting $I=0$, and assume there is a local moment on most sites. Then setting

$$\begin{aligned} d - d_0 &= \frac{1}{6} \sum_{\underline{k}} \left(\nabla \xi_2(\underline{k}) \nabla (\sin \theta_{\underline{k}}) - \frac{1}{g+g_1} |\nabla \xi_1(\underline{k})|^2 \cos^3 \theta_{\underline{k}} \right) n_{\underline{k}1} \\ &= \frac{4}{3} (g+g_1)^2 \sum_{\underline{k}}^{\text{h.z.}} \frac{\nabla \xi(\underline{k}+\underline{Q}) \nabla \xi(\underline{k})}{(\xi(\underline{k}+\underline{Q}) - \xi(\underline{k}))^3} (n_{\underline{k}} - n_{\underline{k}+\underline{Q}}) \end{aligned} \quad (6.51)$$

$$\text{and } d_0 - \Delta = \frac{1}{6} \sum_{\underline{k}} \nabla (\cos^2 \theta_{\underline{k}} \nabla \xi_1(\underline{k})) n_{\underline{k}1} \quad (6.52)$$

$$= \frac{4}{3} (g+g_1)^2 \sum_{\underline{k}} (\xi(\underline{k}+\underline{Q}) - \xi(\underline{k}))^{-2} \left[\nabla^2 \xi(\underline{k}) - 2(\xi(\underline{k}+\underline{Q}) - \xi(\underline{k}))^{-1} (\nabla \xi(\underline{k}) \nabla \xi(\underline{k}+\underline{Q}) + |\nabla \xi(\underline{k})|^2) \right] n_{\underline{k}}$$

where we have put $\underline{k}1 = \underline{k}$ for $\xi(\underline{k}) < \xi(\underline{k} + \underline{Q})$, $\underline{k}1 = \underline{k} + \underline{Q}$ for $\xi(\underline{k} + \underline{Q}) < \xi(\underline{k})$ and h z denotes that the summation is over the half-zone only, we get

$$d - \Delta = \frac{4}{3} (g+g_1)^2 \sum_{\underline{k}} n_{\underline{k}} \left(\frac{\nabla^2 \xi(\underline{k})}{(\xi(\underline{k}+\underline{Q}) - \xi(\underline{k}))^2} - \frac{|\nabla \xi(\underline{k})|^2}{(\xi(\underline{k}+\underline{Q}) - \xi(\underline{k}))^3} \right) \quad (6.53)$$

and
$$a = (g + g_1) \langle \sigma_Q^z \rangle \quad (6.34)$$

to the lowest power in $g + g_1/\varepsilon_2$. Putting $g = 0$ since $I = 0$, $g_1 = J/2N \langle S_Q^z \rangle$ and

$$\langle \sigma_Q^z \rangle = J \chi^0(Q) \langle S_Q^z \rangle$$

(6.24) becomes

$$C = \frac{J^2 \langle S_Q^z \rangle}{N} (1 + J \chi^0(Q))^{-1} \sqrt{\frac{\chi^0(Q)}{3N} \sum_k n_k \frac{\frac{1}{2} \nabla^2 \varepsilon(k)}{(\varepsilon(k+Q) - \varepsilon(k))^2} - \frac{|\nabla \varepsilon(k)|^2}{(\varepsilon(k+Q) - \varepsilon(k))^3}} \quad (6.35)$$

One can see that C is proportional to $1/N \langle S_Q^z \rangle$ and hence to the concentration of impurities. When the concentration becomes small, long-range order no longer exists. (6.35) is not the same as the corresponding result in a Heisenberg antiferromagnet using $J_{RKY}(R)$ as the exchange interaction. For the Heisenberg model (Sokoloff 1969), the spin wave energy is

$$C^2 q^2 = \left(\frac{1}{N} \langle S_Q^z \rangle \right)^2 (J_{RKY}(0) - J_{RKY}(Q)) (J_{RKY}(Q+Q) - J_{RKY}(Q)) \quad (6.36)$$

$$= \left(\frac{1}{N} \langle S_Q^z \rangle J^2 \right)^2 (\chi^0(0) - \chi^0(Q)) (\chi^0(Q+Q) - \chi^0(Q)) \quad (6.37)$$

$$= \frac{1}{3} q^2 \left(\frac{1}{N} \langle S_Q^z \rangle J^2 \right)^2 (\chi^0(Q) - \chi^0(0)) \left(\frac{1}{N} \sum_k \frac{n_{k+Q}}{|\varepsilon(k+Q) - \varepsilon(k)|^2} \left(\frac{1}{2} \nabla^2 \varepsilon(k) - \frac{|\nabla \varepsilon(k)|^2}{\varepsilon(k+Q) - \varepsilon(k)} \right) \right) \quad (6.38)$$

The difference between (6.35) and (6.38) in the denominator arises because the R.K.Y. interaction is only of second

order in g or J . The condition for the "static" approximation when (6.38) applies, is that low energy itinerant electron excitations have a lifetime much shorter than the period of the spin waves under consideration. (6.35) applies in the limit when low energy itinerant electrons have a long lifetime. In the method of this chapter one expands in powers of q/ω to get (6.35). Whereas in the R.K.Y. approximation to get (6.38) one takes the limit $\omega/q \rightarrow 0$ for the itinerant electrons, to remove any dependence the itinerant electrons may have on frequency. One cannot interchange the two limits since both d and Δ have a totally different form in the limit $\omega/q \rightarrow 0$ and the limit $q/\omega \rightarrow 0$.

We will now investigate the spin wave velocity for the dilute alloy problem. The condition for a dilute alloy with local moments, to have long-range order is given by (6.22). It is satisfied if the enhancement due to I compensates for there not being a local moment on every site. (6.24), (6.33) and (6.34) now apply but with g in terms of g_1 given by (6.15), with $I \neq 0$, so that $g_1 \ll g$. We also assume that there is no region of nesting in the band, so that $g \ll \varepsilon_2(\underline{k})$. However $I/\varepsilon_2(\underline{k})$ is of order 1 throughout the band. Thus

$$g + g_1 \simeq g = \frac{I}{N} \langle \sigma_{\underline{Q}}^z \rangle, \quad \langle \sigma_{\underline{Q}}^z + S_{\underline{Q}}^z \rangle \simeq \langle \sigma_{\underline{Q}}^z \rangle \quad (6.39)$$

From (6.24), (6.33) and (6.34), applying the approximation

(6.39), the spin wave velocity now becomes

$$c^2 = \frac{8}{3} \frac{I}{N} \sum_{\underline{k}} \left(\frac{I/N \langle \sigma_Q^z \rangle}{\varepsilon(\underline{k}+\underline{Q}) - \varepsilon(\underline{k})} \right)^2 \left(\frac{1}{2} \nabla^2 \varepsilon(\underline{k}) - \frac{|\nabla \varepsilon(\underline{k})|^2}{\varepsilon(\underline{k}+\underline{Q}) - \varepsilon(\underline{k})} \right) n_{\underline{k}} \quad (6.40)$$

We see that c is of order S/ε_2 , where ε_2 is the mean value of $\varepsilon_2(\underline{k})$, which is a measure of the polarisation of the itinerant electrons.

In this chapter we have extended our theory of spin waves to a system of itinerant and local spins. We have shown that it is possible to treat them on the same footing. Two specific examples are investigated. In one case there is a local moment on every site but no correlation between the itinerant electrons. The other case is a nearly antiferromagnetic system with dilute impurities. The spin wave velocity has been calculated in each case for a perfect antiferromagnet. In the former, the spin wave velocity (6.35) is of order $1/N\varepsilon_2 \langle S_Q^z \rangle$, where $\langle S_Q^z \rangle/N$ is the sublattice magnetisation per atom of the local moments. In the latter, the spin wave velocity (6.39) is of order $1/N\varepsilon_2 \langle \sigma_Q^z \rangle$, where $\langle \sigma_Q^z \rangle/N$ is the sublattice magnetisation per atom of the itinerant electrons. The results do not agree with those based on the R.K.Y. Hamiltonian but this disagreement is shown to arise because of the different approximations used. The R.K.Y. Hamiltonian is correct if the conduction electrons are

static compared with the period of typical spin wave frequencies, whereas the theory in this thesis applies when the dynamics of the conduction electrons are included.

Chapter 7

Spin Correlations in Gamma Manganese

I The Magnetisation in Gamma Manganese

The only other element of the 3d transition series besides chromium, for which there is strong evidence of antiferromagnetism is γ -manganese. By extrapolating their results on Mn-Ni and Mn-Pd alloys, Hicks, Pepper and Smith (1968) obtain a value of $2.3\mu_B$ for the sublattice moment per atom. The results of Bacon et al (1967), on Mn-Cu, yield $2.4\mu_B$ for the sublattice moment. If one discusses γ -manganese in terms of the spin density wave model, as we have done for chromium, then one must have a wide energy gap in the band structure (of order 3 to 4 eV) to produce the large sublattice polarisation of $2.3\mu_B$ per atom (Lowde et al 1971). Band structure calculations have been done, in which a large, selfconsistent field has been introduced into the Hamiltonian, with a periodicity different from that of the lattice (Fletcher 1971, Asano, Yamashita and Wakoh 1968). However, a wide energy gap suggests that for γ -manganese, one is in the limit of a strong correlation model, and Hartree-Fock theory and band theory cannot be immediately applied.

With such large gaps, arguments to do with nesting can not be important. Another difference with chromium is that the alloys of Mn, discussed above, are either f.c.c. or

have a tetragonal distortion, with alternating ferromagnetic planes. In contrast 98%Cr-2%Mn alloys are b.c.c. with clearly defined nearest neighbours of opposite spin. It is not unreasonable to try to describe χ -manganese in terms of a model which includes the strong correlations between the electrons. Hubbard's theory (1963, 1964) of strong correlations, is complicated and he has only applied it to nickel, effectively a one band problem. Edwards (1970) has shown how the theory, in its simplest form, can be applied to Co, Fe and Mn.

In Edwards' theory the band structure is split into two. For a metal with $10-r-n$ d electrons per atom, where r is an integer and $n < 1$, there are r localised electron states per atom of high energy, which are empty, and a band with $(10-r)N$ states containing nN holes of low energy. The interaction between the two types of band, itinerant and localised, is assumed to be a phenomenological s-d interaction. We will work out the ground state of χ -manganese without this last assumption. We can compare our approach with that using the s-d interaction, which is not so likely to produce the strong polarisation necessary to fit the observed sublattice magnetisation. Either way one finds that not only are the localised and itinerant holes aligned antiferromagnetically within their respective bands, they are also aligned in opposite directions with respect to each other on the same site.

The reason for the antiferromagnetism is that there is less restriction on the hopping of the localised holes if the localised holes are aligned antiferromagnetically.

II Strong Correlation Model

In order to calculate the ground state of γ -manganese, we formulate the problem of strong correlations in multiple energy bands in a simple way. We are concerned with the electrons making up the outer 3d, 4s bands of γ -manganese. We assume that one is able to describe the system in terms of atomic-like orbitals, which are orthogonal and as localised as possible. If R_i and R_j are lattice sites and μ and ν label orbitals, then the Hamiltonian is (Hubbard 1963, 1964a, b, c)

$$H = \sum_{i,j} \sum_{\mu,\nu} b_{ij}^{\mu\nu} c_{i\mu}^+ c_{j\nu} + V \quad (7.1)$$

where
$$b_{ij}^{\mu\nu} = \int \omega_{\mu}^*(\underline{r}-\underline{R}_i) \left(-\frac{1}{2m} \nabla^2 + V_0(\underline{r}) \right) \omega_{\nu}(\underline{r}-\underline{R}_j) d\underline{r}$$

$V_0(\underline{r})$ is the periodic potential of the lattice and V is the part of the Hamiltonian coming from interactions between electrons. The purpose of band structure calculations is to find wavefunctions

$$\psi_{\underline{k}n} = N^{-1/2} \sum_{i\mu} a_{\underline{k}\mu}^{(n)} \omega_{\mu}(\underline{r}-\underline{R}_i) e^{i\underline{k} \cdot \underline{R}_i} \quad (7.2)$$

with band index n , which diagonalise $b_{ij}^{\mu\nu}$, so that

$$\sum_{\nu} b_{\underline{k}}^{\mu\nu} a_{\underline{k}\nu}^{(n)} = \epsilon_n(\underline{k}) a_{\underline{k}\mu}^{(n)} \quad (7.3)$$

where $b_{ij}^{\mu\nu} = \frac{1}{N} \sum_{\underline{k}} b_{\underline{k}}^{\mu\nu} e^{i\underline{k} \cdot (\underline{R}_i - \underline{R}_j)}$

and $\epsilon_n(\underline{k})$ is the energy of band n . In an atomic basis

$$V = \frac{1}{2} \sum_{\substack{\mu, \nu \\ s, \tau}} \sum_{\substack{i, j \\ k, l}} \sum_{\sigma_1, \sigma_2} \langle i\mu, j\nu | \frac{e^2}{r_{12}} | k\tau, l\sigma \rangle c_{i\mu\sigma_1}^+ c_{j\nu\sigma_2}^+ c_{l\tau\sigma_1} c_{k\sigma_2} \quad (7.4)$$

where $\langle i\mu, j\nu | \frac{e^2}{r_{12}} | k\tau, l\sigma \rangle = e^2 \iint \frac{\omega_{i\mu}^*(\underline{r}) \omega_{j\nu}^*(\underline{r}') \omega_{k\tau}(\underline{r}) \omega_{l\sigma}(\underline{r}')}{|\underline{r} - \underline{r}'|} d\underline{r} d\underline{r}'$

As discussed in chapter 1, the magnitudes of some terms in this interaction are large for d-like orbitals. We assume that one can clearly distinguish electrons in the system which are in d orbitals. As one goes down the d transition series from Ni to Cr, $b_{ij}^{\mu\nu}$ becomes more important and V less important. The major terms in the interaction, are those between d orbitals on the same site, so V becomes approximately

$$V = I \sum_{\mu} n_{i\mu\uparrow} n_{i\mu\downarrow} - J \sum_{\substack{\mu \neq \nu \\ \sigma}} n_{i\mu\sigma} n_{i\nu\sigma} + U \sum_{\substack{\mu \neq \nu \\ \sigma_1, \sigma_2}} n_{i\mu\sigma_1} n_{i\nu\sigma_2} \quad (7.5)$$

+ constants independent of spin,

where I is the intra-atomic Coulomb integral, J is the Hund's Rule integral and U is the intra-atomic Coulomb

integral between atomic functions of different symmetry. I , J and U are assumed to be independent of the symmetry of the $3d$ orbitals. In this chapter the effects arising because the d orbitals have different symmetry will be largely neglected so as to make the resulting model solvable.

For widely separated atoms the Hamiltonian (7.1) reduces to the atomic representation in which $b_{ij}^{\mu\nu} = 0$ for $i \neq j$.

Then one can choose a basis so that

$$\sum_{\substack{\mu, \nu \\ i, \sigma}} b_{ii}^{\mu\nu} c_{i\mu\sigma}^\dagger c_{i\nu\sigma} \approx \varepsilon_d \sum_{\substack{\mu \\ i, \sigma}} n_{i\mu\sigma} \quad (7.6)$$

where it is assumed the $3d$ atomic energy levels, labelled by μ , are approximately degenerate with energy ε_d . The energy of holes, in the Hartree-Fock approximation using (7.5), is

$$E_{\uparrow, \downarrow} = \varepsilon_d + \frac{1}{5} I n_{\downarrow, \uparrow} - \frac{4}{5} J n_{\uparrow, \downarrow} + \frac{4}{5} U (n_{\uparrow} + n_{\downarrow}) \quad (7.7)$$

where $n_{\uparrow, \downarrow}$ are the mean number of up and down spin holes per site. Alternatively this can be written

$$E_{\uparrow, \downarrow} = \varepsilon_d + U_{\text{eff}} (n_{\uparrow} + n_{\downarrow}) + J_{\text{eff}} n_{\downarrow, \uparrow} \quad (7.8)$$

where $J_{\text{eff}} = \frac{1}{5} I + \frac{4}{5} J$ and $U_{\text{eff}} = \frac{4}{5} (U - J)$

Only the last term in (7.8) is spin dependent.

In the strong correlation model both J_{eff} and U_{eff} are large compared with hopping energies b_{ij}^{xy} , $i \neq j$. For a metal containing $(r + n)/N$ holes, their effect is to ensure that there are predominantly r or more aligned holes per site, with fluctuations from r to $r + 1$ holes or from $r + 1$ to r holes. In the extreme case when $n = 0$ there are r aligned holes on most sites and the energy required to excite an electron into one of these holes is large. These holes may be treated as localised, as suggested by Edwards (1970).

Within second order perturbation theory the energy correction, if hopping between nearest neighbours is allowed, per nearest neighbour pair, is

$$-b^2/r(U_{\text{eff}} + J_{\text{eff}}) \quad (7.9)$$

for antiferromagnetic coupling between the localised holes and

$$-(1 - r/5)b^2/rU_{\text{eff}} \quad (7.10)$$

for ferromagnetic coupling between the localised holes. U_{eff} is usually much greater than J_{eff} , so for $r = 3$, antiferromagnetic coupling gives the greater reduction in energy. A similar argument was used in chapter 4 to obtain (4.21). The factor $1 - r/5$ arises because an up spin hole, in a given orbital, jumping onto a site with r up spin holes

already on it, will only have a probability $1 - r/5$ of finding an orbital occupied. The orbitals occupied by the r localised holes, on a given site, are changing continuously.

We assume that the direct lattice can be divided up into two sublattices A and B with the localised spins on A up, and the localised spins on B down. When $n \neq 0$ one can consider the propagator or Green's function $G_{ij}^{\mu\nu}(\omega)$ of the n additional itinerant holes

$$G_{ij\sigma}^{\mu\nu}(\omega) = \langle\langle c_{i\mu\sigma}; c_{j\nu\sigma}^+ \rangle\rangle_{\omega}$$

$$= Z^{-1} \sum_P \langle P | c_{i\mu\sigma} \frac{1}{\omega - (H - E_P)} c_{j\nu\sigma}^+ + c_{j\nu\sigma}^+ \frac{1}{\omega + (H - E_P)} c_{i\mu\sigma} | P \rangle e^{-\beta E_P} \quad (7.11)$$

where $c_{j\nu\sigma}^+$ creates a hole with spin σ , in the orbital ν , on the site R_j ; $|P\rangle$ is a configuration of the localised holes, which has energy E_P , subject to the condition that on the sublattice A, the spins of the localised holes are up and on B they are down.

Within the atomic representation defined by (7.7) or (7.8)

$$G_{ij\sigma}^{\mu\nu}(\omega) = \delta_{\mu\nu} \delta_{ij} G_{\alpha\beta\sigma}^{\text{atomic}}(\omega) \quad (7.12)$$

where

$$G_{\alpha\beta\sigma}^{\text{atomic}}(\omega) = \begin{pmatrix} (1-r/5)(\omega - \epsilon_0 - J_{\text{eff}} n_{AV})^{-1} & 0 \\ 0 & (\omega - \epsilon_0 - r J_{\text{eff}} - J_{\text{eff}} n_{AV})^{-1} \end{pmatrix} \quad (7.13)$$

$(\alpha, \beta) = (A, A), (A, B), (B, A)$ and (B, B) when (R_i, R_j) lie in sublattices (A, A), (A, B), (B, A) and (B, B)

respectively, $\epsilon_0 = \epsilon_d + U_{\text{eff}}(r + n)$ and $n_{A\downarrow}$, $n_{B\downarrow}$ are the mean number of itinerant down spin holes per site, on the sublattices A and B. We have assumed that there are always either r or $r + 1$ holes on every site.

To include the kinetic energy term, T , in the Green's function one assumes that just one band m is involved in the hopping of the itinerant electrons. We define

$$G_{\alpha\beta}^{\sigma}(\underline{k}, \omega) = \ll \begin{pmatrix} c_{A\underline{k}\sigma} \\ c_{B\underline{k}\sigma} \end{pmatrix}; \begin{pmatrix} c_{A\underline{k}\sigma}^+ \\ c_{B\underline{k}\sigma}^+ \end{pmatrix} \gg_{\omega} \quad (7.14)$$

where

$$c_{A,B\underline{k}\sigma} = \frac{1}{\sqrt{2}}(c_{m\underline{k}\sigma} \pm c_{m\underline{k}+\underline{Q}\sigma})$$

$$c_{m\underline{k}\sigma} = N^{-\frac{1}{2}} \sum_{i,\mu} a_{\underline{k}\mu}^{(m)} c_{i\mu\sigma} e^{i\underline{k} \cdot \underline{R}_i} \quad (7.15)$$

diagonalises T , and $\underline{Q}$ is the spin density wave vector of the localised holes. Then, the perturbation expansion

$$G_{\alpha\beta}^{\uparrow}(\underline{k}, \omega) = \left(G_{\alpha\beta}^{\text{atomic}}(\underline{k}, \omega)^{-1} - T \right)^{-1} \quad (7.16)$$

becomes

$$G_{\alpha\beta}^{\uparrow}(\underline{k}, \omega) = \begin{pmatrix} (1-r)^{-1}(\omega - \epsilon_0 - I_{\text{eff}}n_{A\downarrow}) - \epsilon_1(\underline{k}) & \epsilon_2(\underline{k}) \\ \epsilon_2(\underline{k}) & \omega - \epsilon_0 - (rI_{\text{eff}} + I_{\text{eff}}n_{B\downarrow}) - \epsilon_1(\underline{k}) \end{pmatrix}^{-1} \quad (7.17)$$

where

$$\epsilon_{1,2}(\underline{k}) = \frac{1}{2}(\epsilon_m(\underline{k}+\underline{Q}) \pm \epsilon_m(\underline{k})) \quad (7.18)$$

For $r = 0$ (7.17) reduces to (4.10). One can also reduce

J_{eff} to I_{eff} in (7.17) because of correlation between the holes as described in chapter 1. We can write (7.17) as

$$G_{\alpha\beta}^{\uparrow}(\underline{k}, \omega) = \begin{pmatrix} 1-\frac{1}{5} & 0 \\ 0 & 1 \end{pmatrix} \begin{pmatrix} \omega - (1-\frac{1}{5})(t_1(\underline{k}) - \varepsilon) - J_{\text{eff}}n_{A\downarrow} & t_2(\underline{k}) \\ t_2(\underline{k}) & \omega - (t_1(\underline{k}) - \varepsilon) - (J_{\text{eff}} + I_{\text{eff}}n_{B\downarrow}) \end{pmatrix}^{-1} \quad (7.19)$$

where we have absorbed unnecessary constants into ω , ε is half the bandwidth, the energy zero is at the midpoint of the band structure and we have defined $\varepsilon_1(\underline{k}) = t_1(\underline{k}) - \varepsilon$ (t_1 is positive) and $t_2(\underline{k}) = \varepsilon_2(\underline{k})$.

For manganese $r = 3$. Clearly the itinerant holes will line up antiparallel to the localised holes, if the up spin band with a majority of spins on B has lower energy than the up spin band with a majority of spins on A. The condition for this, is from (7.19), approximately

$$\begin{aligned} (3J_{\text{eff}} - \varepsilon) &< -\frac{2}{5}\varepsilon \\ J_{\text{eff}} &< \frac{1}{5}\varepsilon \end{aligned} \quad (7.20)$$

Edwards (1970) has suggested that this is a reasonable condition for manganese since $J_{\text{eff}}/\varepsilon$ decreases down the d transition series. (7.19) may be written

$$G_{\alpha\beta}^{\uparrow}(\underline{k}, \omega) = \begin{pmatrix} \frac{2}{5} & 0 \\ 0 & 1 \end{pmatrix} \begin{pmatrix} \omega - d & c \\ b & \omega - a \end{pmatrix}^{-1} \quad (7.21)$$

$$\text{so that } G_{AA}^{\uparrow} = \frac{2}{5D} \left(\frac{-(a-d)+D}{2\omega-(a+d)+D} + \frac{D+(a-d)}{2\omega-(a+d)-D} \right) \quad (7.22)$$

$$G_{BB}^{\uparrow} = \frac{1}{D} \left(\frac{-(a-d) + D}{2\omega - (a+d) - D} + \frac{(a-d) + D}{2\omega - (a+d) + D} \right) \quad (7.23)$$

$$G_{AB}^{\uparrow} = G_{BA}^{\uparrow} = \frac{2}{5} \frac{t_2}{D} \left(\frac{2}{2\omega - (a+d) + D} - \frac{2}{2\omega - (a+d) - D} \right) \quad (7.24)$$

where $D = \sqrt{(d-a)^2 + 4bc}$. There are two antiferromagnetic energy bands, of energy

$$E_{1,2}(k) = \frac{1}{2}(a+d) \mp \frac{1}{2}D \quad (7.25)$$

The contribution to the sublattice moment from the itinerant holes is $\frac{1}{2}(n_{B\uparrow} - n_{A\uparrow})g/\mu_B$, where

$$\begin{aligned} n_{B\uparrow} &= -\frac{1}{\pi N} \int^{\varepsilon_F} \text{Im} \sum_{\underline{k}} G_{BB}^{\uparrow}(\underline{k}, \omega) d\omega \\ &= \frac{1}{N} \sum_{\underline{k}} n_{k1} \frac{1}{2D} (-(a+d) + D) + n_{k2} \frac{1}{2D} (a-d + D) \end{aligned} \quad (7.26)$$

$$\begin{aligned} n_{A\uparrow} &= -\frac{1}{\pi N} \int^{\varepsilon_F} \text{Im} \sum_{\underline{k}} G_{AA}^{\uparrow}(\underline{k}, \omega) d\omega \\ &= \frac{1}{N} \sum_{\underline{k}} n_{k1} \frac{1}{5D} (a-d + D) + n_{k2} \frac{1}{5D} (-(a+d) + D) \end{aligned} \quad (7.27)$$

$\frac{1}{2}(n_{B\uparrow} - n_{A\uparrow})$ is obtained, from (7.26) and (7.27), by solving the selfconsistency equation

$$\begin{aligned} g &= \frac{1}{2} I_{\text{eff}} (n_{B\uparrow} - n_{A\uparrow}) \\ &= \frac{I_{\text{eff}}}{2N} \left[\sum_{\underline{k}} (n_{k1} - n_{k2}) \frac{7}{10} \frac{\left[g - \frac{3}{2} I_{\text{eff}} - \frac{3}{10} (t_1(k) - \varepsilon) \right]}{\sqrt{\left[g - \frac{3}{2} I_{\text{eff}} - \frac{3}{10} (t_1(k) - \varepsilon) \right]^2 + \frac{2}{5} t_2^2(k)}} + \sum_{\underline{k}} (n_{k1} + n_{k2}) \frac{3}{10} \right] \end{aligned} \quad (7.28)$$

Clearly $\frac{1}{2}(n_{\uparrow} - n_{\downarrow})g\mu_B > 3n\mu_B$, so that the total sublattice moment, including the localised spins, is less than $(3 - 3n)\mu_B$. The total moment depends on the size of the first term in (7.28). A reduction in the number of itinerant holes increases the total moment until it reaches $3\mu_B$ for $n = 0$.

This kind of behaviour is confirmed by the experimental results of Hicks, Pepper and Smith (1968), who found for Mn-Pd alloys, that the total moment increased from $2.3\mu_B$ for pure Mn, to $3.0\mu_B$ for 80% Mn-20% Pd. One can argue that the three extra electrons per Pd atom just fill up the itinerant band.

Though this analysis is very crude, it does illustrate how the presence of a split off band of holes reduces the orbitals available to the remaining holes, and how this favours antiferromagnetism for the itinerant holes. Edwards (1970) introduced a phenomenological s-d interaction $J' \sum_i \underline{S}_i \cdot \underline{\sigma}_i$, $J' = J_{\text{eff}} - \epsilon/5$ to describe the interaction between the localised spins $\underline{S}_i$, and the itinerant spins $\underline{\sigma}_i$. For this interaction the selfconsistency equation (7.28) would be (see (6.14))

$$g = \frac{J_{\text{eff}}}{2N} \sum_{\underline{k}} \frac{(n_{\underline{k}1} - n_{\underline{k}2}) [g - \frac{3}{2}J_{\text{eff}} - (\epsilon_1(\underline{k}) - \frac{3}{10}\epsilon)]}{\sqrt{[g - \frac{3}{2}J_{\text{eff}} - (\epsilon_1(\underline{k}) - \frac{3}{10}\epsilon)]^2 + t_2^2(\underline{k})}} \quad (7.29)$$

This equation is far less likely than (7.28) to produce a

large polarisation, of order $0.6\mu_B$, in the itinerant band.

In order to make further progress one requires a simple phenomenological Hamiltonian which describes roughly the dynamics of the system. For example, experiments are already being done to find spin wave energies in γ -manganese (Lowde et al 1971). Edwards' phenomenological Hamiltonian does not describe correlations properly, nor include the interaction between localised holes which we claim is the cause of the antiferromagnetism. However, his Hamiltonian is rotationally invariant, which is essential if one is to calculate spin wave energies. There does not appear to be an obvious candidate for a Hamiltonian which satisfies all these conditions.

However, it is interesting to note that γ -manganese lies in the atomic limit of our theory of spin waves in itinerant electron models of antiferromagnetism. This is the opposite limit to chromium. Future work may enable, at least a qualitative, calculation of the spin wave energy in manganese to be made.

References

Chapter 1

- Abrikosov A A and Dzialoshinski I E, 1958, Soviet Phys JETP, 8, 535.
- Akhiezer A I, Bar'yakhtar V G and Peletminskii S V , 1968, Spin Waves (North Holland) ch 1.
- Baym G and Mermin N D , 1961, J Math Phys, 2, 232
- Bloch F , 1929, Z Physik, 57, 545
- Callaway J, 1964, Energy Band Theory (Academic Press)
- Dirac P, 1929, Proc Roy Soc A, 123, 714
- Edwards D M, 1970, Phys Letters, 24A, 350
- Edwards D M, 1962, Proc Roy Soc, A269, 338
- Edwards D M, 1967, Proc Roy Soc, A300, 373
- Fletcher G C, 1952, Proc Phys Soc, A65, 192
- Freeman A J and Watson R E, 1961, Phys Rev, 124, 1439
- Herring C, 1966a, Magnetism IIB, ed G T Rado and H Suhl (Academic Press)
- Herring C, 1966b, Magnetism IV, ed G T Rado and H Suhl (Academic Press)
- Herring C, 1966c, Magnetism IV, ed G T Rado and H Suhl (Academic Press), ch 14.
- Herring C and Kittel C, 1951, Phys Rev, 81, 869
- Hubbard J and Dalton N W, 1968, J Phys C, 1, 1637
- Hubbard J, 1963, Proc Roy Soc, A276, 238
- Hubbard J, 1964a, Proc Roy Soc, A277, 237

- Hubbard J, 1964b, Proc Roy Soc, A281, 401
- Hubbard J, 1964c, Proc Phys Soc, 84, 455
- Hubbard J, 1966, Proc Roy Soc, A296, 82
- Izuyama T, 1960, Prog Theor Phys, 23, 969
- Izuyama T, Kim D J and Kubo R, 1963, J Phys Soc Japan, 18, 1025
- Kanamori J, 1963, Prog Theor Phys Japan, 30, 275
- Lowde R D and Windsor C G, 1970, Adv Physics, 19, 813
- Lowde R D, Haywood B C G, Stringfellow M W and Weber W B, 1971, J Physique, 22, 1186
- Martin P C and Schwinger J, 1959, Phys Rev, 115, 1342
- Slater J C, 1936, Phys Rev, 49, 537
- Stoner E C, 1947, Rept Prog Phys, 11, 43
- Van Vleck J H, 1953, Rev Mod Phys, 25, 220
- Wohlfarth E P, 1953, Rev Mod Phys, 25, 211
- Yamashita J, Fukuchi M and Nakoh S, 1963, J Phys Soc Japan, 18, 999
- Zubarev D N, 1960, Soviet Phys Usp, 3, 320

Chapter 2

- Akhiezer A, Bar'yakhtar V and Peletminskii S, 1968a, Spin Waves (North Holland), ch 8.29 and 8.30; 1968b, ch 8.31
- Boyd R G and Callaway J, 1966, Phys Rev, 139, 1621
- Cooke J and Gersch H, 1967, Phys Rev, 153, 641
- Dyson F J, 1956, Phys Rev, 102, 1217 and 1234
- Edwards D M, 1962, Proc Roy Soc, A269, 338

- Edwards D M, 1967, Proc Roy Soc, A300, 373
 Edwards D M and Wohlfarth E P, 1968, Proc Roy Soc, A303, 127
 Edwards D M and Fisher B, 1971, J Physique, 32, 697
 Englert F and Antonoff M M, 1964, Physica, 30, 429
 Gaudin M, 1960, Nucl Phys, 15, 89
 Harris A B, 1968, Phys Rev, 175, 674
 Herring C, 1966a, Magnetism IV, ed G T Rado and H Suhl
 (Academic Press) ch 14 ; 1966b p356.
 Izuyama T, 1971, J Physique, 32, 809
 Izuyama T, 1964a, Phys Letters, 9, 293
 Izuyama T, 1964b, Phys Rev Letters, 12, 585
 Izuyama T and Kubo R, 1964, J Appl Phys, 35, 1074
 Izuyama T, Kim D J and Kubo R, 1963, J Phys Soc Japan, 18, 1025
 Kashcheev V N and Krivoglaaz M A, 1961, Soviet Phys Solid
 State, 3, 1117
 Kawasaki K, 1964, Phys Rev, 135, A1371
 Ma S, Béal-Monod M and Fredkin D, 1968, Phys Rev, 174, 227
 Maleev S V, 1956, Soviet Phys JETP, 6, 776
 Marshall W, 1962, Proc 8th Intern Conf Low Temp Physics
 (Butterworths, London 1963) p 215.
 Marshall W and Murray G, 1968, J Appl Phys, 39, 380
 Marshall W and Murray G, 1969, J Phys Soc C, 2, 539
 Mathon J and Wohlfarth E P, 1968, Proc Roy Soc, A302, 409
 Nakamura T, 1964, Phys Rev Letters, 12, 279
 Phillips T G, 1966, Proc Roy Soc, A292, 224

- Silberglitt R and Harris A B , 1968, Phys Rev, 174, 640
 Stringfellow M W , 1968, J Phys Soc C, 1, 950
 Vaks V, Larkin A and Pikin S , 1968, Soviet Phys JETP, 26, 647
 Wortis M, 1959, PhD Thesis, Harvard University

Chapter 3

- Anderson P W , 1952, Phys Rev, 86, 694
 Anderson P W , 1964, Concepts in Solids (W A Benjamin) ch 3.
 Bethe H, 1931, Z Physik 71, 205
 Callen H B , 1963, Phys Rev, 130, 690
 Des Cloizeaux J and Pearson J J , 1962, Phys Rev, 128, 2131
 Herbert D C , 1969, J Math Phys, 10, 2269
 Hulthén L, 1938, Arkiv Mat Astron Fysik 26A, Hall
 Kubo R, 1952, Phys Rev, 87, 568
 Lieb E and Mattis D C , 1952, J Math Phys, 3, 749
 Mattis D C , 1965, Theory of Magnetism (Harper and Row) ch 6.
 Pikalev E M, Savchenko M A and Solyom J , 1969, Soviet Phys JETP , 28, 734
 Solyom J, 1969, Soviet Phys JETP, 28, 1251
 Stinchcombe R B and Cottam M G , 1970a, J Phys C, 3, 2283;
 1970b, J Phys C, 3, 2305; 1970c, J Phys C, 3, 2326
 Vaks V G, Larkin A I and Pikin S A , 1968, Soviet Phys JETP, 26, 647
 Ziman J M, 1965, Theory of Solids (Cambridge Univ Press) ch 10

Chapter 4

- Anderson P W , 1964, Concepts in Solids (W A Benjamin) ch 5
- Arrott A, 1966, Magnetism IIB , ed G T Rado and H Suhl
(Academic Press)
- Asano S and Yamashita J , 1967, J Phys Soc Japan, 25, 714
- Des Cloizeaux J , 1959, J Phys Radium , 20, 606 & 751
- Cottam M G and Stinchcombe R B , 1970, J Phys C, 3, 2305
- Edwards D M, 1967, Proc Roy Soc, A300, 373
- Edwards D M and Fisher B, 1971, J Physique, 32, 697
- Fedders P A and Martin P C, 1966, Phys Rev, 143, 245
- Fletcher G C, 1971, J Phys F , 1, 177
- Gupta R P and Sinha S K , 1970, J Appl Physics, 41, 915; 1971
(to be published)
- Hamaguchi Y, Wollan E O and Koehler W C , 1965, Phys Rev, 138,
A737
- Haywood B C G, Lowde R D, Stringfellow M W and Weber W B, 1971,
J Physique, 32, 1186
- Herring C, 1966, Magnetism IV, ed G T Rado and H Suhl,
(Academic Press)
- Hewson A C and Lindner U , 1970, Physica Stat Sol, 42, K186
- Hubbard J, 1963, Proc Roy Soc, A276, 238
- Hubbard J, 1966, Proc Roy Soc, A296, 82
- Iwamoto F and Sawada K , 1962, Phys Rev, 126, 887
- Kimball J C, 1969, Phys Rev, 183, 533
- Lidiard A B, 1954, Proc Roy Soc, A224, 161

- Liu S H, 1970, Phys Rev B, 2, 2664
- Lomer W M , 1962, Proc Phys Soc, 80, 489
- Loucks T L, 1965, Phys Rev , 139, A1181
- Matsubara T and Yokota T ,1954, Proc Int Conf Theoret Phys
Kyoto, Japan 1953, 693
- Nagaoka Y, 1966, Phys Rev, 147, 892
- Overhauser A W, 1959, Phys Rev Letters, 3, 414
- Overhauser A W, 1960a, Phys Chem Solids, 13, 71
- Overhauser A W, 1960b, Phys Rev Letters, 4, 462
- Overhauser A W, 1962, Phys Rev, 128, 1437
- Sinha S K, Liu S H , Muhlstein L D and Wakabayashi N, 1969,
Phys Rev Letters, 23, 311
- Slater J C , 1951, Phys Rev, 82, 538
- Sokoloff J B, 1969a, Phys Rev, 185, 770
- Sokoloff J B, 1969b, Phys Rev, 185, 783

Chapter 5

- Als-Nielsen J and Dietrich O W , 1969, Phys Rev Letters, 22,
290
- Arrott A, Ferner S A and Kendrick H, 1966, Phys Rev Letters,
14, 1022
- Arrott A, 1966, Magnetism IIB, ed G T Rado and H Suhl
(Academic Press)
- Asano S and Yamashita J , 1967, J Phys Soc Japan, 23, 714
- Feäders P A and Martin P C , 1966, Phys Rev, 143, 245

- Gupta R P and Sinha S K , 1970, J Appl Phys, 41, 915;
1971 (to be published)
- Hamaguchi Y, Wollen E O and Koehler V C , 1965, Phys Rev,
138, A737
- Herring C, 1966, Magnetism IV, ed G T Rado and H Suhl
(Academic Press) , ch 13
- Ishikawa Y , Hoshino S and Endoh Y , 1967, J Phys Soc Japan,
22, 1221
- Kimball J C, 1969, Phys Rev , 183, 533
- Liu S H, 1970, Phys Rev B, 2, 2664
- Lomer W M, 1962, Proc Phys Soc, 80, 489
- Loucks T L, 1965, Phys Rev, 139, A1181
- Overhauser A W, 1962, Phys Rev, 128, 1437
- Rice T M, Barker A S and Halperin B I, 1968, Phys Rev Letters,
20, 384
- Rice T M, Barker A S, Halperin B I and McWhan D B , 1969,
J Appl Physics, 40, 1337
- Sinha S K, Liu S H, Mahlestein L D and Wakabayashi N, 1969,
Phys Rev Letters, 23, 311
- Sokoloff J B, 1969, Phys Rev, 185, 770
- Sokoloff J B, 1969, Phys Rev, 185, 783
- Tachiki M and Nagomiya T , 1968, Phys Rev Letters, 3, 214

Chapter 6

- Anderson P W, 1961, Phys Rev, 124, 41
- Arrott A, 1966, Magnetism IIB, ed G T Rado and H Suhl
(Academic Press)
- Van der Berg G J, 1965, Proc 9th Int Conf Low Temp Physics,
ed J A Daunt et al (Plenum Press)
- Clogston A M, 1962, Phys Rev, 125, 439
- Doniach S, 1967, Proc Phys Soc, 91, 86
- Doniach S and Wohlfarth E P , 1967, Proc Roy Soc, A296, 442
- Edwards D M, 1970, (private communication)
- Elliott R J, 1965, Magnetism IIA, ed G T Rado and H Suhl
(Academic Press)
- Elliott R J and Wedgwood F A , 1964, Proc Phys Soc, 84, 63
- De Gennes P G, 1962, J Physique, 23, 630
- Herring C, 1966, Magnetism IV, ed G T Rado and H Suhl
(Academic Press) ch 13
- Kasuya T, 1966, Magnetism IIB, ed G T Rado and H Suhl
(Academic Press)
- Marshall W, 1960, Phys Rev, 118, 1520
- Mattis D C, 1965, Theory of Magnetism (Harper Row)
- Moriya T, 1970, Phys Rev Letters, 24, 1433
- Overhauser A W, 1960, Phys Rev Letters, 4, 462
- Overhauser A W, 1962, Phys Rev, 128, 1437
- Wohlfarth E P, 1970, Phys Rev Letters, 24, 1433

Chapter 7

- Asano S, Yamashita J and Wakoh S , 1968, J Appl Phys, 39, 1274
- Bacon G E, Donnan I W, Smith J H and Street R , 1957, Proc Roy Soc, A241, 223
- Edwards D M, 1970, Phys Letters, 24A, 350
- Fletcher G C, 1971, J Phys F, 1, 177
- Hicks T J, Pepper A R, Smith J R, 1968, J Phys C, 1, 1683
- Hubbard J, 1963, Proc Roy Soc, A276, 238
- Hubbard J, 1964a, Proc Roy Soc, A277, 237
- Hubbard J, 1964b, Proc Roy Soc, A281, 401
- Hubbard J, 1964c, Proc Phys Soc, 84, 455
- Lowde R D, Haywood B C G, Stringfellow M W and Weber W B, 1971, J Physique, 32, 1186