CHAPTER -2

THE THEORY OF DYNAMICAL MAGNETIC SUSCEPTIBILITY
OF TRANSITION METALS

In this chapter the generalized expressions are obtained for the dynamical susceptibility of transition metals in a model band scheme. We start with the Hamiltonian for Bloch electrons in the perturbing magnetic field and linearize the Liouville equation so as to obtain the induced current density in the density matrix approach. The spin and orbital currents induced by the presence of the field are directly related to the susceptibility function. These results are presented in Sec.2.1. The non-interacting model band scheme enables us to separate out the various contributions arising from the intra and interband transitions of electrons. Explicit expressions for each of these contributions to the spin susceptibility of a transition metal are obtained using parabolic band approximation for the energy eigenvalue of the d-subbands. These results are presented in Sec.2.2. The reduced spin susceptibility which is related to the neutron scattering measurements is also formulated and presented in Sec.2.3. The calculations for the orbital susceptibility are presented in Sec.2.4. The results and their limitations are discussed in the last section.
2.1 General Theory

The dynamical magnetic susceptibility is a fundamental quantity required in understanding the dynamics of magnetic substances. In literature various approaches for calculating this quantity have been reported, however we follow the density matrix approach because of its simplicity. The method we employ here is to calculate the induced current density via the single particle density matrix $\rho$. As is well known, $\rho$ satisfies the Liouville equation

$$i\hbar \frac{\partial \rho}{\partial t} = [H, \rho] \quad .(2.1)$$

where $H$ is the Hamiltonian of the system. We begin at infinite times with the metallic system in an equilibrium configuration. A magnetic field $\vec{B}$ varying in space and time is then applied to this system of electrons moving in a constant background of positive charge. The Hamiltonian for such a system is written as

$$H = \frac{1}{2m} \left( \vec{p} + e \vec{A}/c \right)^2 + V(\vec{r}) + (e\hbar/2mc) \vec{\sigma} \cdot \vec{\mathbf{B}} \quad .(2.2)$$

where $V(\vec{r})$ is the periodic lattice potential. $\vec{A}$ is vector potential associated with the magnetic field $\vec{B}$ and $\vec{p}$ is the momentum of electron. $e$ is the electronic charge and $m$ is the mass of electron. $\hbar = h/2\pi$, $h$ being the Planck's constant and $c$ is the velocity of light. $\vec{\sigma}$ is a vector associated with the spin of the electron. The lattice potential $V(\vec{r})$ is
assumed to be independent of magnetic field though the effect of including magnetic field dependence in $V(\mathbf{r})$ may well be interesting.

The theory is conveniently developed in terms of the unperturbed Bloch eigenfunctions of the electrons. It is notationally convenient to write explicitly

$$\langle \mathbf{r} | \mathbf{k} , \nu \rangle = U_{k\nu}(\mathbf{r}) \exp(i \mathbf{k} \cdot \mathbf{r}) = | \mathbf{k} \mu \rangle \quad ..(2.3)$$

where the functions $U_{k\nu}(\mathbf{r})$ are periodic with the periodicity of the lattice and the corresponding eigenvalues are denoted by $E_{k\nu}$. We classify the zero field electron states by wavevector $\mathbf{k}$ and band index $\nu$.

Eq. (2.2) can be written as

$$H = H_0 + H_1 \quad ..(2.4)$$

where $H_0$ is the Hamiltonian for the unperturbed system and $H_1$ is first order perturbation term which involves the applied field ($H$ is linear in the field and 2nd order terms are dropped in the calculation). Correspondingly the density matrix contains an unperturbed term $\rho_0$, the equilibrium distribution and the first order term $\rho_1$, i.e.

$$\rho = \rho_0 + \rho_1 \quad ..(2.5)$$

The unperturbed density matrix $\rho_0$ satisfies:

$$\rho_0 \langle \Psi_{k\mu}(\mathbf{r}) | = \eta_{\mu}(\mathbf{k}) \langle \Psi_{k\mu}(\mathbf{r}) \rangle \quad ..(2.6)$$
where \( n_{\mu}(k) \) is Fermi occupation probability function.

Substituting Eq. (2.4) and Eq. (2.5) in Eq. (2.1), we obtain the zero order and the first order linearized Liouville equation as

\[
\frac{i\hbar}{\partial t} \frac{\partial \rho}{\partial t} = H_0 \rho - \rho H_0
\]

and

\[
\frac{i\hbar}{\partial t} \frac{\partial \rho}{\partial t} = H_1 \rho - \rho H_1 + H_0 \rho_1 - \rho_1 H_0 \quad \ldots (2.7)
\]

where \( \rho_0 \) and \( \rho'_0 \) are matrix products.

Taking the matrix elements between Bloch states \( \psi_{kj}(\vec{r}) \) and \( \psi'_{k'}(\vec{r}) \) in Eq. (2.7) we find:

\[
\langle \psi_{k'}(\vec{r}) | \frac{\partial \rho_1}{\partial t} | \psi_{kj}(\vec{r}) \rangle = (E_{kj} - E_{k'}) \langle \psi_{k'}(\vec{r}) | \rho_1 | \psi_{kj}(\vec{r}) \rangle
\]

\[
- \left( \frac{n_{\mu}(k) - n_{\mu}(k')}{\hbar} \right) \langle \psi_{k'}(\vec{r}) | \rho_1 | \psi_{kj}(\vec{r}) \rangle \quad \ldots (2.8)
\]

If we assume that \( \langle \psi_{kj}(\vec{r}) | \rho_1 | \psi_{k'}(\vec{r}) \rangle \) has the time dependence as \( \exp(i\omega t + \delta t) \), where \( \delta \) is a small positive infinitesimal corresponding to an adiabatic turning on of the perturbing potential, then we have:

\[
\langle \psi_{k'}(\vec{r}) | \rho_1 | \psi_{kj}(\vec{r}) \rangle = \frac{n_{\mu}(k) - n_{\mu}(k')}{E_{kj} - E_{k'} - \hbar \omega + i\hbar \delta} \frac{\langle \psi_{k'}(\vec{r}) | H_1 | \psi_{kj}(\vec{r}) \rangle}{E_{kj} - E_{k'} - \hbar \omega + i\hbar \delta} \quad \ldots (2.9)
\]

The induced current may now be calculated from the relation.

\[
J_{\text{ind}}(\vec{r}) = \text{Tr} \left[ \rho_1 J_0^{\text{op}}(\vec{r}) \right] + \text{Tr} \left[ \rho_1 J_0^{\text{op}}(\vec{r}) \right] \quad (2.10)
\]

where current operator is given as?
and
\[
\frac{\hbar}{e}\mathcal{A}(\mathbf{r}, t) = -\frac{e^2}{mc} \frac{\hbar}{i} \frac{\delta^{\mu\nu}}{E_{k'} - E_k} \left\{ \frac{\Psi_{k'}^*(\mathbf{r})}{\Psi_{k'}(\mathbf{r})} - \frac{\Psi_k^*(\mathbf{r})}{\Psi_k(\mathbf{r})} \right\} \\
- \frac{e^2}{mc} \sum_{k'\mu} \frac{n_k(k)}{\delta^{\mu\nu}} \mathcal{A}(\mathbf{r}, t) \frac{\Psi_{k'}^*(\mathbf{r})}{\Psi_{k'}(\mathbf{r})} \frac{\Psi_k^*(\mathbf{r})}{\Psi_k(\mathbf{r})},
\]
\[
\text{(2.12)}
\]

The field \( \mathbf{B} \) is taken along the \( \hat{z} \) direction and gauge is chosen so that \( \text{div} \mathcal{A} = 0 \). Choosing the time and space dependence of \( \mathbf{B} \) such that
\[
\mathcal{A} = A(q_0, \omega) \exp \left[ i(q_\perp \mathbf{r} - \omega t) \right],
\]
we have
\[
\mathbf{B} = BZ, \quad \mathcal{A} = l \cdot \mathbf{\hat{x}} \quad \text{and} \quad q_\perp = q \mathbf{\hat{y}}
\]
\[
\text{(2.14)}
\]
\( \hat{x}, \hat{y} \) and \( \hat{z} \) are unit vectors and \( \omega \) is the frequency of the applied magnetic field. The frequency \( \omega \) is taken to have a small positive imaginary part corresponding to the adiabatic turning on the perturbing potentials. The induced current in \( x \)-direction will then be considered in order to obtain the diagonal part of the susceptibility tensor. The coupling of the wavevector \( \vec{q} \) with the wavevectors \( \vec{q} + \vec{G} \), where \( \vec{G} \) is reciprocal lattice vector, have been neglected. These so called Umklapp processes give rise to the local field corrections.

Taking the Fourier transform of Eq. (2.12) and using the above expressions for magnetic field and the vector potential, the Fourier component of the orbital current for the wavevector \( \vec{q} \) of the applied field is obtained as:

\[
J_x(\vec{q}, \omega) = -\frac{e^2}{mc} \sum_{k} \frac{n_{\mu}(k) A(\vec{q}, \omega) - \sum_{k'} \sum_{\mu} (e^2 \hbar^2 / m^2 c) \frac{-E_{k'} - E_{k} + \hbar \omega + i\epsilon}{E_{k'} - E_{k}}}{\text{det} \text{det}} \times \langle \Psi_{k, \nu}(\vec{r}) | e^{i\vec{q} \cdot \vec{r}} / \partial \vec{x} | \Psi_{k', \mu}(\vec{r}) \rangle \langle \Psi_{k', \mu}(\vec{r}) | e^{i\vec{q} \cdot \vec{r}} / \partial \vec{x} | \Psi_{k, \nu}(\vec{r}) \rangle \rangle A(\vec{q}, \omega)
\]

\[ (2.15) \]

The orbital current is related to the orbital susceptibility by the relation

\[
J_{\text{ind}}(\vec{q}, \omega) = c q^2 \chi_{\text{orb}}(\vec{q}, \omega) A(\vec{q}, \omega)
\]

\[ (2.16) \]
whence the susceptibility function $\chi_{\text{orb}}(q, \omega)$ is given as

\[
\chi_{\text{orb}}(q, \omega) = -\frac{e^2}{m^2c^2} \sum_{k, k'} \frac{n_{\mu}(k) - n_{\nu}(k')} {E_{k\mu} - E_{k'\nu} - i\omega + i\delta}
\times <q^0 \hat{r}_0 \hat{r}_0/\partial x | \Psi_{k\mu}(r) > <q^1 \hat{r} \hat{r}_0/\partial x | \Psi_{k'\nu}(r) > + \text{c.c.}
\]

The induced spin current is calculated from the second bracket in the expression for $j^0_{\text{spin}}(q, \omega)$ (Eq. 2.11a) which is given as

\[
j_{\text{spin}}(q, \omega) = \frac{e^2}{4m^2c^2} \sum_{k, k'} \frac{n_{\mu}(k) - n_{\nu}(k')} {E_{k\mu} - E_{k'\nu} - i\omega + i\delta}
\times <q^0 \hat{r}_0 \hat{r}_0/\partial x | \Psi_{k\mu}(r) > <q^1 \hat{r} \hat{r}_0/\partial x | \Psi_{k'\nu}(r) > + \text{c.c.}
\]

The spin susceptibility is directly calculated using the relation

\[
j_{\text{spin}}(q, \omega) = Cq^2 i(q, \omega) \chi_{\text{spin}}(q, \omega)
\]

Comparing Eq.(2.18) and (2.19) we find

\[
\chi_{\text{spin}}(q, \omega) = \frac{e^2}{m^2c^2} \sum_{k, k'} \frac{n_{\mu}(k) - n_{\nu}(k')} {E_{k\mu} - E_{k'\nu} - i\omega + i\delta}
\times <q^0 \hat{r}_0 \hat{r}_0/\partial x | \Psi_{k\mu}(r) > <q^1 \hat{r} \hat{r}_0/\partial x | \Psi_{k'\nu}(r) > + \text{c.c.}
\]

The Eqs.(2.17) and (2.20) for the dynamical orbital and spin susceptibility are quite general and include excitations between electrons belonging to different bands. If the bands are characterized by the indices 1, m and σ corresponding to
the orbital, magnetic and spin quantum numbers, the Eqs. (2,17) 
and (2, 20) may be written as:

\[
\gamma_{\text{orb}}(q, \omega) = -2 g_e^2 q^2 \frac{2}{q^2} \sum_{k} \frac{n_{\text{lns}}(k)}{E(k) - E(k')} \left( \psi_{\text{lns}}(k) | \bar{\psi}_{\text{lns}}(k') \right) \frac{e^{i \mathbf{q} \cdot \mathbf{r}}}{\hbar} \frac{2m}{\mu} \int \frac{d^3 \mathbf{r}}{(2\pi)^3} \nabla \cdot \frac{\mathbf{r}}{r^3} \left( \psi_{\text{lns}}(k) | \bar{\psi}_{\text{lns}}(k') \right),
\]

and

\[
\gamma_{\text{spin}}(q, \omega) = -2 g_e^2 q^2 \frac{2}{q^2} \sum_{k} \frac{n_{\text{lns}}(k)}{E(k) - E(k')} \left( \psi_{\text{lns}}(k) | \bar{\psi}_{\text{lns}}(k') \right) \frac{e^{i \mathbf{q} \cdot \mathbf{r}}}{\hbar} \frac{2m}{\mu} \int \frac{d^3 \mathbf{r}}{(2\pi)^3} \nabla \cdot \frac{\mathbf{r}}{r^3} \left( \psi_{\text{lns}}(k) | \bar{\psi}_{\text{lns}}(k') \right)
\]

where \( g \) is Lande's splitting factor and \( \mu_0 \) is Bohr magneton.

The summation over \( k \) is for all the occupied electronic states. \( k \) and \( k' = k + q \) are restricted in the first Brillouin zone. In the above expressions, the U-process contributions are neglected. It is quite straightforward to write the general expressions for the susceptibility matrices. The non-diagonal parts of these matrices will give rise to the local field corrections and it may be quite interesting to investigate them. However this leads to prohibitively difficult numerical calculations. There also do not exist any other calculation or experimental measurement to test such calculation. Therefore we limit our calculations to the evaluation of only the diagonal part of the susceptibility matrices given by Eqs. (2.21) and (2.22). Using the ortho-
normality of the spin wavefunctions Eq. (2.22) can be written as
\[ Y(\mathbf{q}, \omega) = \frac{\hbar^2}{2\hbar^2} \sum_{k, k'} \sum_{l, l'} \frac{n_{l'n'}(k) - n_{l'm'}(k)}{E_{l'n'}(k) - E_{l'm'}(k) - \hbar \omega} |\langle \mathbf{J}_{l'n'}(k) | \mathbf{J}_{l'm'}(k) \rangle|^2 \] (2.23)

For a paramagnetic system the spin index \( \sigma \) is dropped and the right hand side is multiplied by a factor of 2 for spin degeneracy.

2.2 The Dynamical Spin Susceptibility

In transition metals, the outermost d-shell is partially filled. As a consequence, the electronic band structure calculations show that the energy eigenvalue and the wavefunction for conduction electrons in these metals have both the s and d-character and these itinerant electrons contribute towards conduction. Therefore it will not be justified to neglect the s or d-character of the conduction electrons while studying the dynamical magnetic response function. Moreover, the separation between the core and the conduction electrons is also not clear. Therefore we resort to a model band structure for a transition metal in which both the s and d-character of conduction electrons is taken into account. In this model it is assumed that the s-electrons are distributed in the partially filled s-band with effective mass \( m_s \) and d-electrons are distributed in the five d-subbands with effective mass \( m_{d,m} \). Some of the d-subbands are completely
filled while some of them are partially filled. The s and d-
subbands are assumed to be non-interacting. This model is
constructed on the basis of the detailed band structure
calculations, which will be discussed in the next chapter for
the metals under consideration. We use the plane wave
approximation for the s-electrons and use the simple tight-
binding wavefunctions for the d-electrons. The parabolic bands
are used for the energy eigen values of both the s and d-
electrons. In this model, the electrons undergo intra and
interband transitions and readjust themselves when magnetic
field is applied. Therefore, we can write the spin suscepti-
bility function as

\[ \chi_{\text{spin}}(\vec{q}, \omega) = \chi_{ss}(\vec{q}, \omega) + \chi_{dd}(\vec{q}, \omega) + \chi_{ds}(\vec{q}, \omega) + \chi_{sd}(\vec{q}, \omega) \]

where the expressions for \( \chi_{ss}(\vec{q}, \omega) \) etc. can readily be
written with the help of Eq. (2.23). \( \chi_{ss}(\vec{q}, \omega), \chi_{dd}(\vec{q}, \omega), \)
\( \chi_{ds}(\vec{q}, \omega) \) and \( \chi_{sd}(\vec{q}, \omega) \) correspond to the transitions from
s-band to s-band, from d-subbands to d-subbands,
from d-subbands to s-band and from s-band to d-subbands,
respectively.

A. Evaluation of \( \chi_{ss}^0(\vec{q}, \omega) \)

The expression for the s-s part of susceptibility
\( \chi_{ss}^0(\vec{q}, \omega) \) from Eq. (2.23) is written as
As discussed earlier we use the free electron approximation for the electrons in the s-band and the parabolic band approximation for the energy eigen values i.e.
\[
\langle \mathbf{k}' | \mathbf{L} | \mathbf{k} \rangle = \frac{1}{\sqrt{\mathcal{N}}} e^{i \mathbf{k} \cdot \mathbf{r}} \quad \ldots (2.26)
\]
and
\[
E_s (k) = E^*_s + \frac{\hbar^2 k^2}{2m_s} \quad \ldots (2.27)
\]
where \( k_F S \) is the Fermi momentum of s-electrons and \( E^0_s \) is the energy at \( k = 0 \). \( N \) is the number of unit cells in the crystal and \( \mathcal{N} \) is the volume of the unit cell.

Using Eqs.(2.26) and (2.27), the matrix elements in Eq.(2.25) reduce to
\[
\langle \mathbf{q}, \omega | \mathbf{L} | \mathbf{q} \rangle \rightarrow \delta_{\mathbf{k}, \mathbf{k} + \mathbf{q}} \quad \ldots (2.28)
\]
\[
\langle \mathbf{q}, \omega | \mathbf{L} | \mathbf{q} \rangle \rightarrow \delta_{\mathbf{k}, \mathbf{k} + \mathbf{q}} \quad \ldots (2.29)
\]
Therefore Eq.(2.25) simplifies as
\[
\chi^0_{ss}(\mathbf{q}, \omega) = \sum_k \frac{n_s(k) - n_s(k+q)}{E_s(k+q) - E_s(k) - i\omega + i\delta} \quad \ldots (2.30)
\]
The real and imaginary parts of $\chi_{ss}^0 (\mathbf{q}, \omega)$ are separated using the identity

$$\frac{1}{x \pm i\delta} = \frac{1}{x} \mp i\frac{\delta}{\pi} \ln \delta(x) \quad \text{ ... (2.31)}$$

Replacing the sum over $k$ by integration, and evaluating the integral analytically, we get the real and imaginary parts of $\chi_{ss}^0 (\mathbf{q}, \omega)$ in the following form:

$$\text{Re} \chi_{ss}^0 (\mathbf{q}, \omega) = \frac{g^2 \mu_B^2}{4\pi^2} m_s k_{FS} \left\{ \frac{1}{2} + \frac{1}{8\lambda} \left[ \frac{1}{1 - (m_s W/\lambda - \lambda)^2} \ln \left[ \frac{1 - m_s W/\lambda - \lambda}{1 + m_s W/\lambda + \lambda} \right] \right] \right. \times \ln \left[ \frac{1 - m_s W/\lambda + \lambda}{1 + m_s W/\lambda - \lambda} \right] - \frac{1}{8\lambda} \left[ \frac{1}{1 - (m_s W/\lambda + \lambda)^2} \ln \left[ \frac{1 - m_s W/\lambda + \lambda}{1 + m_s W/\lambda - \lambda} \right] \right]$$

and

$$\text{Im} \chi_{ss}^0 (\mathbf{q}, \omega) = -\frac{g^2 \mu_B^2}{32\pi \lambda} m_s k_{FS} \left\{ \left[ 1 - (m_s W/\lambda - \lambda)^2 \right] \theta(1 - (m_s W/\lambda - \lambda)^2) - \left[ 1 - (m_s W/\lambda + \lambda)^2 \right] \theta(1 - (m_s W/\lambda + \lambda)^2) \right\} \quad \text{ ... (2.32)},$$

where $\lambda = q/2k_{FS}$, $W = \omega/2k_{FS}^2$.

$\theta(x)$ is a step function which is unity if $x > 0$ and zero otherwise. Eqs. (2.32) and (2.33) are similar to those obtained by Doniach except that the effective masses and Fermi momenta for the $s$-electrons are defined in the non-interacting band scheme. In the limit $q \to 0$, $\omega \to 0$ the above result (2.32) reduces to

$$\chi_{ss}^0 (0, 0) = \left( g^2 \mu_B^2 N_n e / 8\pi^2 m_s \right) k_{FS} \quad \text{ ... (2.34a)}.$$
which is similar to the Pauli Spin susceptibility\textsuperscript{56}.

In $\omega \to 0$ limit expression (2.32) reduces to

$$\chi_{ss}^0(q,\omega) = \frac{2\hbar^2}{4\pi^2} n_s k_F \left[ \ln \frac{1+\frac{\lambda}{4\omega}}{1-\frac{\lambda}{4\omega}} \right]. \quad (2.34b)$$

This expression is similar to that obtained by Hebborn and March\textsuperscript{7} for the static susceptibility.

**B. Evaluation of $\chi_{dd}^0(q,\omega)$**

$$\chi_{dd}^0(q,\omega) = \frac{\mu_B^2}{2} \sum_{k,k'} \sum_{n,n'} \frac{n_{dn}(k) - n_{dn}(k')}{E_{dn}(k) - E_{dn}(k') - i\omega + i\delta}$$

$$\times \left| \langle \Psi_{dn}(k) | e^{i\mathbf{q}\cdot\mathbf{L}} | \Psi_{dn}(k') \rangle \right|^2. \quad (2.35)$$

The simple tightbinding wavefunctions for electrons in the $d$-subbands and parabolic band approximations for the energy eigenvalues are written as

$$\psi_{dn}(\mathbf{k}) = \frac{1}{\sqrt{N}} \sum_{\mathbf{L}} e^{i\mathbf{k}\cdot\mathbf{L}} \Phi_{dn}(\mathbf{r} - \mathbf{L}), \quad (2.36)$$

and

$$E_{dn}(k) = E_{dn}^0 + \hbar^2 k^2 / 2m_{\text{dn}}. \quad (2.37)$$

where $\mathbf{L}$ is a lattice vector and $\Phi_{dn}(\mathbf{r} - \mathbf{L})$ are the atomic orbitals.

Using Eqs. (2.36) in the matrix elements in Eq. (2.35) we get
\[
\langle \hat{L}_{\text{dm}} (k) | e^{i \mathbf{q} \cdot \mathbf{r}} | \hat{L}_{\text{dm'}} (k') \rangle = \delta_{k, k+q} \Delta_{\text{dm}, \text{dm'}} (q) \quad \text{..(2.38)}
\]

and
\[
\langle \hat{\mathcal{L}}_{\text{dm'}} (k') | e^{i \mathbf{q} \cdot \mathbf{r}} | \hat{\mathcal{L}}_{\text{dm}} (k) \rangle = \delta_{k, k+q} \Delta^*_{\text{dm}, \text{dm'}} (q) \quad \text{..(2.39)}
\]

where
\[
\Delta_{\text{dm}, \text{dm'}} = \int \tilde{\psi}_{\text{dm}} (r) e^{-i \mathbf{q} \cdot \mathbf{r}} \tilde{\psi}_{\text{dm'}} (r) \, d^3r
\quad \text{..(2.40)}
\]

The explicit form of \( \Delta_{\text{dm}, \text{dm'}} (q) \) is given in the paper by Prakash and Joshi \textsuperscript{21}. These have been evaluated using the radial part of the atomic orbitals in the form
\[
\tilde{\psi} (r) = \sum a_i e^{i \alpha_i r}
\quad \text{..(2.41)}
\]

where \( a_i \) and \( \alpha_i \) parameters are determined by fitting the numerically tabulated Hartree-Fock wavefunctions\textsuperscript{57} and these are taken from the values tabulated by Clementi\textsuperscript{58}.

Using Eqs.\textsuperscript{(2.38)} - \textsuperscript{(2.40)} in Eq. \textsuperscript{(2.35)}, \( \chi_{\text{dd}}^\sigma (q, \omega) \) simplifies as:
\[
\chi_{\text{dd}}^\sigma (q, \omega) = \lim_{\epsilon \to 0} \epsilon^{\frac{3}{2}} \sum_{m,n} \left[ \frac{n_{\text{dm}} (k) - n_{\text{dm'}} (k+q)}{E_{\text{dm}} (k+q) - E_{\text{dm}} (k) - \hbar \omega + i \epsilon} \right] \times \left[ \Delta_{\text{dm}, \text{dm'}} (q) \right] \quad \text{..(2.42)}
\]

An exact expression for \( \Delta_{\text{dm}, \text{dm'}} (q) \) involves the overlapping between d-wavefunctions on the same atomic site and the overlapping on different atomic sites, where the latter involves the calculation of multicentral integrals. Since the intra-atomic overlapping is the leading term as compared to the interatomic overlapping and also for simplicity we consider
only the intra-atomic overlapping. This reduces exactly to what we call the atomic form factors. We call the first factor in the parenthesis in Eq. (2.42), the band structure part and the second factor in the square brackets, the overlap integral part.

For the intra d-subband transitions i.e. when \( m = m' \), we evaluate \( \chi^{\sigma}_{dd} (q, \omega) \) exactly in the same manner as \( \chi^{\sigma}_{ss} (q, \omega) \). The expressions for the real and imaginary parts are as follows:

\[
\chi^{\sigma}_{dd}(\text{intra})_m = \sum (I_1 + iI_2) I_{dm} \quad \text{(2.43)}
\]

where

\[
I_1 = \frac{e^2}{4\pi^2} \sum_{n_d} n_d F_{dn} \left\{ \frac{1}{2} + \frac{1}{8\lambda'} \left[ 1 - \left( n_{dm} \frac{W}{\lambda'} + \lambda' \right)^2 \right] \right\}
\]

\[
\ln \left[ \frac{1 - n_{dm} \frac{W}{\lambda'} + \lambda'}{1 + n_{dm} \frac{W}{\lambda'} - \lambda'} \right] - \frac{1}{8\lambda'} \left[ 1 - \left( n_{dm} \frac{W}{\lambda'} + \lambda' \right)^2 \right] \ln \left[ \frac{1 - n_{dm} \frac{W}{\lambda'} + \lambda'}{1 + n_{dm} \frac{W}{\lambda'} - \lambda'} \right] \quad \text{(2.44)}
\]

and

\[
I_2 = -\frac{e^2}{32\pi} \sum_{n_d} n_d \frac{k_{Fdn}}{\lambda'} \left\{ \left[ 1 - \left( n_{dm} \frac{W}{\lambda'} + \lambda' \right)^2 \right] \theta(1 - \left( n_{dm} \frac{W}{\lambda'} + \lambda' \right)^2) \right. \]

\[
\left. - \left[ 1 - \left( n_{dm} \frac{W}{\lambda'} + \lambda' \right)^2 \right] \theta(1 - \left( n_{dm} \frac{W}{\lambda'} + \lambda' \right)^2) \right\} \quad \text{(2.45)}
\]
\( \chi = q/2k_{Fdm} \), \( W' = \omega/2k_{Fdm}^2 \). 
\( I_{dm} = \triangle_{dm, dm}' \triangle_{dm, dm}' \) \( \ldots (2.46) \)

\( k_{Fdm} \) is Fermimoment for the \( d \)-subband with magnetic quantum number \( m \). In the limits \( q \to 0 \) and \( \omega \to 0 \) the expression \( (2.43) \) reduces to

\[ \chi^0 (\omega, \omega) = (e^2/4\pi^2 \beta/8\pi^2) N \sum_m m_{dm} k_{Fdm} \ldots (2.47) \]

which is nothing but the density of states of \( d \)-electrons at Fermi surface. The imaginary part reduces to zero for the above limiting cases which is otherwise feasible. It was pointed out by Gupta and Sinha\(^{14}\) and by Prakash and Singh\(^{59}\) that the overlap matrix elements reduce the magnitude of the susceptibility and also change the nature of the susceptibility function.

For the inter \( d \)-subband transitions i.e. when \( m \neq m' \) the analytical expressions for the real and imaginary parts of the susceptibility function, we obtain as follows:

\[ \chi^{(i)} (\omega, \omega) = \sum_{m \neq m'} (I_1 + i I_2') \triangle_{dm, dm}' \triangle_{dm, dm}' \ldots (2.48) \]

where

\[ I_1' = -\frac{e^2}{4\pi^2} \frac{N_x}{\gamma} \left[ \delta_{dm}(\frac{k_{Fdm}}{\gamma} - x_1 = \frac{1}{4\xi^2} Y_1) + \delta_{dm}(\frac{k_{Fdm}}{\gamma} - x_2 = \frac{1}{4\xi^2} Y_2) \right] \ldots (2.49) \]
\begin{align*}
    x_1 &= \left[ \frac{k_{Fdn}^2}{4q} - \frac{q}{4q} \left(1 + \frac{2/m_{dn}}{\omega} \right) \right] \ln \left[ \frac{2k_{Fdn}q + \frac{1}{2}k_{Fdn}^2 + q^2 - 2m_{dn}}{2k_{Fdn}q + \frac{1}{2}k_{Fdn}^2 - q^2 + 2m_{dn}} \right] \\
    x_2 &= \left[ \frac{k_{Fdn}^2}{4q} - \frac{q}{4q} \left(1 + \frac{2/m_{dn}}{\omega} \right) \right] \ln \left[ \frac{2k_{Fdn}q - \frac{1}{2}k_{Fdn}^2 + q^2 + 2m_{dn}}{2k_{Fdn}q + \frac{1}{2}k_{Fdn}^2 - q^2 - 2m_{dn}} \right] \\
    \text{if } \Delta < 0 \quad &
    \left\{ \begin{array}{l}
    \frac{2}{V\Delta} \left( \tan^{-1} \left[ 2(\xi_{k_{Fdn}+q}/V\Delta) \right] + \tan^{-1} \left[ 2(\xi_{k_{Fdn}-q}/V\Delta) \right] \right) \\
    \frac{1}{V-\Delta} \left( \ln \left| \frac{2\xi_{k_{Fdn}+2q-V\Delta}}{2\xi_{k_{Fdn}}+2q+V\Delta} \right| + \ln \left| \frac{2\xi_{k_{Fdn}}-2q-V\Delta}{2\xi_{k_{Fdn}}+2q-V\Delta} \right| \right)
    \end{array} \right.
\end{align*}
\[-31-
\]

\[
Y_2 = \Delta' \left\{ \begin{array}{ll}
\sqrt{2} \tan^{-1} \left[ 2 \left( \tau'_1 k_{F, dm} + q \right) / \sqrt{\Delta'} \right] + \tan^{-1} \left[ 2 \left( \tau'_2 k_{F, dm} - q \right) / \sqrt{\Delta'} \right]
\end{array} \right.
\]

\[
if \Delta' > 0
\]

\[
1/\sqrt{\Delta} \left\{ \ln \left[ \frac{2 \tau'_1 k_{F, dm} + 2 q - \sqrt{\Delta'}}{2 \tau'_2 k_{F, dm} + 2 q + \sqrt{\Delta'}} \right] + \ln \left[ \frac{2 \tau'_2 k_{F, dm} - 2 q - \sqrt{\Delta'}}{2 \tau'_1 k_{F, dm} - 2 q + \sqrt{\Delta'}} \right] \right\}
\]

\[
if \Delta' < 0
\]

\[
\Lambda = -4 \left[ q^2 (\xi + 1) - 2m_{dm}, \omega \xi \right]
\]

\[
\Lambda' = -4 \left[ \xi_i^2 (\xi + 1) + 2m_{dm}, (\xi_i \xi) \right]
\]

\[
\xi' = \frac{m_{dm}}{m_{dm}} - 1; \quad \xi_i' = \frac{m_{dm}}{m_{dm}} - 1
\]

and

\[
I_2 = -\left[ e^{2} L \frac{2}{N \sqrt{v/16 \pi q}} \left( \Lambda_1 + Z_2 \right) \right]
\]

\[
k_{F, dm}^2 - k_i^2 \quad if \quad k_1 < k_{F, dm} < k_2; \quad \Lambda_1 > 0
\]

\[
k_2 - k_i^2 \quad if \quad k_2 < k_{F, dm} \quad ; \quad \Lambda_1 > 0
\]

\[
0 \quad if \quad k_{F, dm} < k_1 \quad ; \quad \Lambda_1 > 0
\]

\[
0 \quad if \quad \Lambda_1 < 0
\]

\[
Z_1 = m_{dm} \left\{ \begin{array}{ll}
k_{F, dm}^2 - k_i^2 \quad if \quad k_1 < k_{F, dm} < k_2; \quad \Lambda_2 > 0
\end{array} \right.
\]

\[
k_2 - k_i^2 \quad if \quad k_2 < k_{F, dm} \quad ; \quad \Lambda_2 > 0
\]

\[
0 \quad if \quad k_{F, dm} < k_1 \quad ; \quad \Lambda_2 > 0
\]

\[
0 \quad if \quad \Lambda_2 < 0
\]

\[
Z_2 = m_{dm} \left\{ \begin{array}{ll}
k_{F, dm}^2 - k_i^2 \quad if \quad k_1 < k_{F, dm} < k_2; \quad \Lambda_2 > 0
\end{array} \right.
\]

\[
k_2 - k_i^2 \quad if \quad k_2 < k_{F, dm} \quad ; \quad \Lambda_2 > 0
\]

\[
0 \quad if \quad k_{F, dm} < k_1 \quad ; \quad \Lambda_2 > 0
\]

\[
0 \quad if \quad \Lambda_2 < 0
\]

\[
\Lambda = -\Delta / 4m_{dm}^2
\]

..(2.53)

..(2.54)

..(2.55)

..(2.56)

..(2.57)

..(2.58)

..(2.59)

..(2.60)
These expressions seem to be similar to those obtained by Czachor for the interband transitions, while evaluating the dielectric function for a semi metal. In the limit $q \rightarrow 0$, $\chi^{0}_{dd(\text{inter})}(q, \omega)$ reduces to zero as $\Delta_{\text{dm}, \text{dm}'} = 0$, because of the orthogonality of the $d$-wavefunctions.

C. Evaluation of $\chi^{0}_{ds}(q, \omega)$

For the transitions from the $d$-subbands to the $s$-band the expression as written from Eq.(2.23), is

$$
\chi^{0}_{ds}(q, \omega) = \frac{\Delta^{'}}{E_{s}(k') - E_{d}(k) - h\omega + i\epsilon}
$$

Using Eqs.(2.26), (2.27), (2.36) and (2.37) for the eigenfunctions and energy eigenvalues of $s$ and $d$-electrons, Eq.(2.66) simplifies as
The real and imaginary parts are separated and evaluated in the manner described in Appendix A. The explicit expressions are as follows:

\[ \text{Re} \chi^0_{ds}(q, \omega) = 2e^2 \mu_0^2 \frac{n_s(4\pi)^2}{n} \sum_{i,j} a_i a_j \Sigma(-1)^m \int_0^k \sum_{n} F_1(k) \, dk \]  \hspace{1cm} \text{(2.68a)}

and

\[ \text{Im} \chi^0_{ds}(q, \omega) = -2e^2 \mu_0^2 \frac{n_s(4\pi)^2}{n} \sum_{i,j} a_i a_j \Sigma(-1)^m \int_0^k \sum_{n} F_2(k) \, dk \]  \hspace{1cm} \text{(2.68b)}

where

\[ F_1(k) = \frac{k^6}{(k^2 + \alpha_1^2)^4 (k^2 + \alpha_2^2)^4} \left[ D^2_{om} p^2_{o-m} I^1_o + (D^2_{im} p^2_{1-m} + p^2_{im} p^2_{1-m}) I^1_i \right] \]  \hspace{1cm} \text{(2.69a)}

\[ F_2(k) = \frac{k^6}{(k^2 + \alpha_1^2)^4 (k^2 + \alpha_2^2)^4} \left[ D^2_{om} p^2_{o-m} I^2_o + (D^2_{im} p^2_{1-m} + p^2_{im} p^2_{1-m}) I^2_i \right] \]  \hspace{1cm} \text{(2.69b)}
$I_n^m$ are the elements of the rotation matrix with argument $(-\gamma, -\beta, -\alpha)$ while $\alpha, \beta, \gamma$ are the Euler's angles.

The other quantities are as follows:

\[
\begin{align*}
I_0^1 &= 5/4 \left( 1/2 I_{n0} - 3 I_{n2} + 9/2 I_{n4} \right) & \text{(2.70)} \\
I_1^1 &= 15/4 \left( -I_{n2} + I_{n4} \right) & \text{(2.71)} \\
I_2^1 &= 15/8 \left( 1/2 I_{n0} - I_{n2} + 1/2 I_{n4} \right) & \text{(2.72)} \\
I_{n0} &= -1/b \ln \left| (b-a)/(b+a) \right| & \text{(2.73)} \\
I_{n2} &= -1/b \left( 2a/b + a^2/b^2 \right) \ln \left| (b-a)/(b+a) \right| & \text{(2.74)} \\
I_{n4} &= -1/b \left( 2a/b + 2a^3/b^3 + a^4/b^4 \right) \ln \left| (b-a)/(b+a) \right| & \text{(2.75)} \\
I_0^2 &= \begin{cases} 5/8b \left( 3a^2/b^2 - 1 \right)^2 & \text{if } (1-a^2/b^2) > 0 \\ 0 & \text{if } (1-a^2/b^2) < 0 \end{cases} & \text{(2.76)} \\
I_1^2 &= \begin{cases} 15/4b a^2/b^2 \left( a^2/b^2 - 1 \right) & \text{if } (1-a^2/b^2) > 0 \\ 0 & \text{if } (1-a^2/b^2) < 0 \end{cases} & \text{(2.77)} \\
I_2^2 &= \begin{cases} 15/16b \left( a^2/b^2 - 1 \right)^2 & \text{if } (1-a^2/b^2) > 0 \\ 0 & \text{if } (1-a^2/b^2) < 0 \end{cases} & \text{(2.78)} \\
\alpha &= \left( 1-m_s/m_{dn} \right) k^2 + \zeta^2 - 2m_s \omega & \text{(2.79)} \\
\text{and} & \\
b &= 2kq & \text{(2.80)}
\end{align*}
\]
The radial integration over \( k \) in Eqs. (2.68a) and (2.68b) is carried out numerically. The rotation matrices and the parameters of the 3d radial wavefunction are taken from the paper by Irakash and Joshi. \( \chi_{ds}^0(q,\omega) \) should reduce to zero in the \( q \to 0 \) limit if an orthogonal set of eigenfunctions is used for s and d conduction electrons. However in our calculations, the s and d wavefunctions are not orthogonal, therefore \( \chi_{ds}^0(q,\omega) \) does not reduce exactly to zero at \( q = 0 \) but we have taken it to be zero at \( q = 0 \) and evaluated it exactly for finite values of \( q \).

D. Evaluation of \( \chi_{sd}^0(q,\omega) \)

The contribution \( \chi_{sd}^0(q,\omega) \) to the susceptibility function is again written from Eq. (2.23) as

\[
\chi_{sd}^0(q,\omega) = \lim_{q\to 0} \frac{n_s(k') - n_{dn}(k')}{\frac{1}{2} \sum_{k,k'} \frac{F_{dn}(k') - F_s(k)}{E_{dn}(k') - E_s(k)} \omega + i\hbar} \times |\langle \chi_{sd}^0 | \psi_{s}^{1} \rangle |^2
\]

The real and imaginary parts of \( \chi_{sd}^0(q,\omega) \) are evaluated exactly in the same manner as for \( \chi_{ds}^0(q,\omega) \). In this case the transitions are to be considered from the partially filled s-band to partially filled d-subbands. The explicit expressions for the real and imaginary parts are as follows:

\[
\text{Re} \chi_{sd}^0(q,\omega) = \frac{2g_l^2}{\pi} \sum_{ij} a_i a_j |\langle \chi_{sd}^0 | \psi_{d}^{1} \rangle |^2 \int \frac{F_{s}(k) dk}{F_5(k)}
\]

\[
\text{Im} \chi_{sd}^0(q,\omega) = \frac{2g_l^2}{\pi} \sum_{ij} a_i a_j |\langle \chi_{sd}^0 | \psi_{d}^{1} \rangle |^2 \int \frac{F_{s}(k) dk}{F_5(k)}
\]
and

\[ I_m \sum_{b_d}^2 (q, \alpha) = -2 \varepsilon_i^2 \varepsilon_j^2 N(48)^2 \sum_{b_d} a_i a_j a_m a_n (1 - 1)^n n_{d_m} \int_0^F \mathcal{F}_4^2 \, dk \]

where

\[ P_3(k) = k^2 \left[ \varepsilon_d^2 \varepsilon_{d-1} \varepsilon_0 \varepsilon_{0-1} I_0^3 + (D_{1m}^2 D_{1-m}^2 + D_{1m}^2 D_{1-m}^2) I_1^3 \right] \]

\[ + (D_{2m}^2 D_{2-m}^2 + D_{2m}^2 D_{2-m}^2) I_2^3 \]

\[ P_4(k) = k^2 \left[ \varepsilon_d^2 \varepsilon_{d-1} \varepsilon_0 \varepsilon_{0-1} I_0^4 + (D_{1m}^2 D_{1-m}^2 + D_{1m}^2 D_{1-m}^2) I_1^4 \right] \]

\[ + (D_{2m}^2 D_{2-m}^2 + D_{2m}^2 D_{2-m}^2) I_2^4 \]

\[ I_0^3 = \frac{5}{8} \int_1^1 (3k^2 + 2bt + 2a^2 - k^2)^2 / [S'(a' + bt)] \, dt \quad \ldots (2.84) \]

\[ I_1^3 = \frac{15}{4} \int_1^1 k^2 (t^2 - 1) (kt + q)^2 / [S'(a' + bt)] \, dt \quad \ldots (2.85) \]

\[ I_2^3 = \frac{15}{16} \int_1^1 k^4 (t^2 - 1)^2 / [S'(a' + bt)] \, dt \quad \ldots (2.86) \]

\[ I_0^4 = \left\{ \begin{array}{ll}
\frac{5}{8b} \left[ 3(a^2/b^2 - 1/3) k^2 + 2q^2 - 2a^2 \right] / [S' \left( a^2/b^2 \right)] & \text{if } (1 - a^2/b^2) > 0 \\
0 & \text{if } (1 - a^2/b^2) < 0 \\
\end{array} \right. \quad \ldots (2.87) \]

\[ I_1^4 = \left\{ \begin{array}{ll}
\frac{15}{8b} \left[ k^2 (a^2/b^2 - 1) (k^2 a^2/b^2 + q^2 - a^2) \right] / [S' \left( a^2/b^2 \right)] & \text{if } (1 - a^2/b^2) > 0 \\
0 & \text{if } (1 - a^2/b^2) < 0 \\
\end{array} \right. \quad \ldots (2.88) \]
\[ I_2^4 = \begin{cases} \frac{15}{16b} \left( \frac{a_1^2}{b^2} - 1 \right)^2 / s & \text{if } (1-a_1^2/b^2) > 0 \\ 0 & \text{if } (1-a_1^2/b^2) < 0 \end{cases} \]

...(2.89)

\[ a' = \left( 1 - \frac{m_{dm}}{m_g} \right) k^2 + q^2 - 2m_{dm} \omega \]

...(2.90)

\[ s = (k^2 + q^2 + a_{j-a'}^2)^4 \left( k^2 + q^2 + a_{j-a'}^2 \right)^4 \]

...(2.91)

and

\[ s' = (k^2 + q^2 + a_{j-b}^2)^4 \left( k^2 + q^2 + a_{j-b}^2 \right)^4 \]

...(2.92)

The integrations in the expression for \( I_0^3, I_1^3 \) and \( I_2^3 \) are carried out using the method of partial fractions. The expressions are very lengthy and therefore these are not given in the text. \( \chi_{sd}^0(\vec{q},\omega) \) also does not reduce to the correct limit for \( \vec{q} \to 0 \) but expressions are exact for finite values of \( \vec{q} \). However in the numerical calculations we have taken

\[ \chi_{sd}^0(\vec{q},\omega) = 0 \text{ for } \vec{q} = 0. \]

2.3 The Reduced Dynamical Spin Susceptibility

Izuyama, Kim and Kubo\(^8\) pointed out that the reduced susceptibility can be related to the neutron scattering measurements. Therefore with a view to compare our results with the experiment we also calculate the reduced spin susceptibility. Following Lowde and Windsor\(^6\), we extract the atomic form factor outside the summations in case of intra and inter d-subband transitions. If we take the atomic form factor to be
equal to unity, then our results for intra and inter d-subband transitions become equivalent to the reduced spin susceptibility. These results become equivalent in principle to those obtained by Allan et al.²⁰ and Lowde and Windsor.⁶ The contribution from the s-s intra-band transitions is directly the reduced susceptibility since in this case the form factor itself is unity. In brief

\[ \chi_{\text{SS}}^0 (q, \omega) = \chi_{\text{SS}}^0 (q, \omega) \]  

\[ \chi_{\text{dd(intra)}}^0 (q, \omega) = \sum_m (I_1 + iI_2) \]  \( \text{(2.94)} \)

\[ \chi_{\text{dd(inter)}}^0 (q, \omega) = \sum_m (I_1' + iI_2') \]  \( \text{(2.95)} \)

where \( I_1, I_2, I_1', I_2' \) are the same as given by Eqs. (2.44), (2.45), (2.49) and (2.57). Because of the \( k \) dependence of the overlap integrals in Eq. (2.67) it is not possible to separate the band structure and overlap integral parts of \( \chi_{\text{ds}} (q, \omega) \). However, to have an expression for the reduced \( \chi_{\text{ds}} (q, \omega) \) we use some simplifications. We take \( k = k_{Fdn} \) in the overlap integrals and take them out of the integral over \( k \) in Eq. (2.67). This approximation is justified because maximum response of electrons is due to the electrons in the vicinity of Fermi Surface. However this may underestimate the reduced \( \chi_{\text{ds}} (q, \omega) \), yet the final results may not be seriously affected because the contribution of
\(\chi_{ds}(q,\omega)\) is itself very small as found in the calculation of the static susceptibility and dielectric function of nickel\(^{21}\). Putting these redefined form factors equal to unity, the expressions for the real and imaginary parts of the reduced susceptibility obtained are as follows:

\[
\begin{align*}
\text{Re}\, \chi_{ds}(q,\omega) &= -\frac{\epsilon^2}{4\pi^2} \sum \frac{m_n m_g}{n} (k_{\text{Fdm}}/\xi' - X_3 - 1/4\xi''^2 Y_3)  \\
\text{Im}\, \chi_{ds}(q,\omega) &= -\frac{g^2}{16\pi q} \begin{cases} 
\frac{k_{\text{Fdm}}^2 - k_1^2}{2} & \text{if } k'_1 k_{\text{Fdm}} < k''_2, \mu' > 0 \\
\frac{k''_2 - k_1^2}{2} & \text{if } k'_1 k_{\text{Fdm}} < k''_2, \mu' < 0 \\
0 & \text{if } k_{\text{Fdm}} < k'_1, \mu' > 0 \\
0 & \text{if } \mu' < 0
\end{cases}  
\end{align*}
\]

where

\[
X_3 = \left[\frac{k_{\text{Fdm}}^2}{4q} - \frac{q}{4\xi''} (1+2/\xi') + \frac{m_n m_g}{n} \ln \left|\frac{2k_{\text{Fdm}} q - \xi''^2 k_{\text{Fdm}}^2 + q^2 - 2n \omega}{2k_{\text{Fdm}} q + \xi''^2 k_{\text{Fdm}}^2 - q^2 + 2n \omega}\right| \right] 
\]

\[
Y_3 = \left[ \frac{2/\sqrt{\Delta''}}{\tan^{-1} \left[ 2(\xi'' k_{\text{Fdm}} q - q^2)/\sqrt{\Delta''} \right]} + \tan^{-1} \left[ 2(\xi'' k_{\text{Fdm}} q - q^2)/\sqrt{\Delta''} \right] \right] 
\]

\[
\begin{cases} 
\frac{2\sqrt{\Delta''}}{\tan^{-1} \left[ 2(\xi'' k_{\text{Fdm}} q - q^2)/\sqrt{\Delta''} \right]} & \text{if } \Delta'' > 0 \\
1/\sqrt{-\Delta''} \left( \ln \left| \frac{2\xi'' k_{\text{Fdm}} q + q^2 - (\Delta'')^{1/2}}{2\xi'' k_{\text{Fdm}} q + q^2 + (\Delta'')^{1/2}} \right| + \ln \left| \frac{2\xi'' k_{\text{Fdm}} q + q^2 - (\Delta'')^{1/2}}{2\xi'' k_{\text{Fdm}} q + q^2 + (\Delta'')^{1/2}} \right| \right) & \text{if } \Delta'' < 0
\end{cases}  
\]

\(\Delta''\) is a function of \(q\) and \(\omega\).
The expressions for the real and imaginary parts of the reduced susceptibility \( \chi^{\text{sd}}_{\text{red}} \) are also obtained in the same manner as for \( \chi^{\text{sd}}_{\text{red}}(q, \omega) \) and these are given as follows:

\[
\text{Re} \chi^{\text{sd}}_{\text{red}}(q, \omega) = -\frac{\varepsilon_{0}^{2} 2^{2} \varepsilon_{0}^{2}}{4 \pi^{2} m} \sum_{\text{dm}} \left( k_{\text{FS}} / \varepsilon'' - k_{1}' - \frac{1}{4 \varepsilon'_{F}} \varepsilon'_{F} Y_{1} \right) \quad (2.105)
\]

\[
\text{Im} \chi^{\text{sd}}_{\text{red}}(q, \omega) = -\frac{\varepsilon_{0}^{2} 2^{2} \varepsilon_{0}^{2}}{16 \pi q} \sum_{\text{dm}} \begin{cases} 
\frac{k_{2}}{k_{1}^{2}} \left( k_{1}'' \right)^{2} & \text{if } k_{1}'' < k_{\text{FS}}, \mu' > 0 \\
\frac{k_{2}''}{k_{1}''} \left( k_{1}'' \right)^{2} & \text{if } k_{2}'' < k_{\text{FS}}, \mu'' > 0 \\
0 & \text{if } k_{\text{FS}} < k_{2}'' , \mu'' > 0 \\
0 & \text{if } \mu' < 0 
\end{cases} \quad (2.106)
\]
2.4 The Dynamical Orbital Susceptibility

We also study the orbital response function given by Eq. (2.21) in the non-interacting model band picture for a transition metal. Separating out the contributions arising from the intra and interband transitions in the s and d-subbands, we can rewrite Eq. (2.21) as follows:
\chi^0_{\text{orb},ss}(q,\omega) = \chi^0_{\text{orb},dd}(q,\omega) + \chi^0_{\text{orb},ds}(q,\omega) + \chi^0_{\text{orb},sd}(q,\omega) \quad \ldots(2.114)

where \chi^0_{\text{orb},ss}, \chi^0_{\text{orb},dd}, \chi^0_{\text{orb},ds} and \chi^0_{\text{orb},sd} arise due to transitions from s-band to s-band, from d-subbands to d-subbands, from d-subbands to s-band and from s-band to d-subbands, respectively.

These contributions are evaluated following the same procedure as was adopted in Sec. 2.2.

A. Evaluation of \chi^0_{\text{orb},ss}(q,\omega)

The expression for \chi^0_{\text{orb},ss}(q,\omega) for a paramagnetic system is written from Eq.(2.21) in the form

\chi^0_{\text{orb},ss}(q,\omega) = -\frac{e^2}{\hbar^2} \frac{L^2}{q^2} \left[ 2n_s/\hbar^2 \frac{\partial n_s}{\partial q} \frac{\partial n_s}{\partial q'} \frac{\partial n_s}{\partial q} \right] \frac{\epsilon_{s,\text{orb}}(k) - \epsilon_{s,\text{orb}}(k')}{E_{s,\text{orb}}(k) - E_{s,\text{orb}}(k') - \hbar \omega + i\delta} 

\times \left\langle \hat{\psi}_s(k) \right| \frac{\partial}{\partial \phi} \left| \hat{\psi}_s(k') \right\rangle \left\langle \hat{\psi}_s(k') \right| e^{i\mathbf{q} \cdot \mathbf{r}} \delta(q+x) \left| \hat{\psi}_s(k) \right\rangle \quad \ldots(2.115)

Using Eqs. (2.26) for the wavefunctions of s-electrons the matrix elements in the above expression simplify as:

\left\langle \hat{\psi}_s(k) \right| e^{i\mathbf{q} \cdot \mathbf{r}} \frac{\partial}{\partial x} \left| \hat{\psi}_s(k) \right\rangle = i \frac{k_x}{k^2} \frac{\delta(q,\mathbf{k})}{k^2} \quad \ldots(2.116)

and

\left\langle \hat{\psi}_s(k) \right| e^{i\mathbf{q} \cdot \mathbf{r}} \frac{\partial}{\partial x} \left| \hat{\psi}_s(k) \right\rangle = i \frac{k_x}{k^2} \frac{\delta(q,\mathbf{k})}{k^2} \quad \ldots(2.117)
Substituting Eq.(2.116) and Eq.(2.117) in Eq.(2.115) we obtain

\[
\chi_{\text{orb}, ss}^{(q, \omega)} = -\frac{\varepsilon_{B}^{2} \rho_{B}^{2} n_{s} k_{F}}{32 \pi^{2}} \left[ \frac{2 n_{s}^{2} k_{F}}{\hbar^{2}} \right] + \frac{2 k_{F}}{k_{F}} \sum_{k} \frac{n_{s}(k) - n_{s}(k+q)}{k_{F}^{2} E_{s}(k) - \hbar \omega + i \varepsilon}
\]

...(2.118)

Using Eq.(2.27) for the energy eigenvalues of s-electrons and replacing the summation over \( k \) by integration, we get the following expressions for the real and imaginary parts of dynamical orbital susceptibility.

\[
\text{Re} \chi_{\text{orb}, ss}^{(q, \omega)} = \frac{\varepsilon_{B}^{2} \rho_{B}^{2} n_{s} k_{F}}{32 \pi^{2}} \left\{ 1 + \frac{1}{4} \left[ 1 - \left( \frac{m_{s} W_{A}}{\lambda} + \lambda \right)^{2} \right]^{2} \ln \left[ \frac{1 - \frac{m_{s} W_{A}}{\lambda} + \lambda}{1 + \frac{m_{s} W_{A}}{\lambda} + \lambda} \right] - \frac{1}{4 \lambda^{2}} \left[ 1 - \left( \frac{m_{s} W_{A}}{\lambda} + \lambda \right)^{2} \right]^{2} \right\}
\]

...(2.119)

\[
\text{Im} \chi_{\text{orb}, ss}^{(q, \omega)} = \frac{\varepsilon_{B}^{2} \rho_{B}^{2} n_{s} k_{F}}{128 \pi^{3} \lambda^{3}} \left\{ 1 - \left( \frac{m_{s} W_{A}}{\lambda} + \lambda \right)^{2} \theta(1 - \left( \frac{m_{s} W_{A}}{\lambda} + \lambda \right)^{2}) \left[ 1 - \left( \frac{m_{s} W_{A}}{\lambda} + \lambda \right)^{2} \right]^{2} \right\}
\]

...(2.120)

These expressions are similar to those obtained by S'zabo and by Hebborn and March except that the effective mass and Fermi momentum are involved in these expressions. In the limit \( q \to 0, \omega \to 0 \), the above Eq.(2.119) reduce to

\[
\text{Re} \chi_{\text{orb}, ss}^{(q, \omega)} (0, 0) = -\frac{\varepsilon_{B}^{2} \rho_{B}^{2} n_{s} k_{F}}{32 \pi^{2}}
\]

...(2.121)
which is similar to the expression for Landau magnetism. In the limit \( \omega \to 0 \), the result (2.119) simplifies as:

\[
\chi_{\text{orb},ss}^0 (q,0) = -\frac{\mu B^2}{12 \pi^2} \int_0^\infty \frac{m_s e}{k \beta^2} \left[ 1 + \frac{1}{\lambda^2} \ln \left| \frac{1+\lambda}{1-\lambda} \right| \right]
\]

which is static orbital susceptibility. This expression is similar to that obtained by Hebborn and March\(^7\) and by Haldensperger\(^6\). It is interesting that it reflects the sharp Fermi surface through the factors \( \ln|1-m_s W/\lambda-\epsilon/(1+m_s W/\lambda-\lambda)| \) and \( \ln|1-m_s W/\lambda+\epsilon/(1+m_s W/\lambda+\lambda)| \) occurring in Eq. (2.119) and (2.32).

B. Evaluation of \( \chi_{\text{orb},dd}^0 (q,\omega) \), \( \chi_{\text{orb},ds}^0 (q,\omega) \), and \( \chi_{\text{orb},dd}^0 (q,\omega) \)

The expression for \( \chi_{\text{orb},dd}^0 (q,\omega) \) part of orbital susceptibility is:

\[
\chi_{\text{orb},dd}^0 (q,\omega) = 2g^2 \mu_B^2 q^2 \int \frac{d\Omega}{4\pi} \sum_{k,m,m'} \frac{E_{\text{dm}}(k)}{E_{\text{dm}}(k)-\epsilon_{\text{dm}}(k)-\omega+i\Sigma}
\]

\[
\times \langle \frac{\delta \bar{\psi}_{\text{dm}}(k)}{\delta \bar{q}} | \bar{\psi}_{\text{dm}}(k) \rangle \langle \bar{\psi}_{\text{dm}}(k) | e^{i\bar{q}.\bar{r}_\epsilon/\delta x} \bar{\psi}_{\text{dm}}(k) \rangle
\]

\( \ldots \) (2.123)
Using Eq. (2.36) for the wavefunction \( \psi_{dm}(k) \), the matrix elements reduce to

\[
\langle \psi_{dm}(k) | e^{i \mathbf{q} \cdot \mathbf{r}} \psi_{dm}(k) \rangle = \delta_{k, k+q} \Delta_{dm, dm}^{(q)} \ldots (2.124)
\]

\[
\langle \psi_{dm}(k)' | e^{i \mathbf{q} \cdot \mathbf{r}} \psi_{dm}(k) \rangle = \delta_{k, k+q} \Delta_{dm, dm}''(q) \ldots (2.125)
\]

where \( \Delta_{dm, dm}^{(q)} = \int \mathbf{r} \psi_{dm}(r) e^{i \mathbf{q} \cdot \mathbf{r}} \frac{\partial}{\partial \mathbf{r}} \psi_{dm}(r) \, dr \ldots (2.126) \)

and \( \Delta_{dm, dm}''(q) = \int \mathbf{r} \psi_{dm}(r) e^{i \mathbf{q} \cdot \mathbf{r}} \frac{\partial}{\partial \mathbf{r}} \psi_{dm}(r) \, dr \ldots (2.127) \)

when we substitute for the most general atomic orbital

\[
\Phi_{lm}(r) = R_1(r) Y_{l}^{m}(\theta, \varphi) \ldots (2.128)
\]

and integrate out the expression, the angular integration which involves sine and cosine of \( \theta \) and \( \varphi \) renders the integrand for radial integration zero, thereby reducing the matrix elements to zero. Thus the contribution \( \chi_{dd}^{(q, \omega)} \) becomes zero. This looks feasible even physically, since the tightly bound electrons should not contribute towards orbital susceptibility.

Similarly the \( d-s \) and \( s-d \) interband contributions to the orbital susceptibility are written from Eq. (2.21)

\[
\chi_{orb, ds}^{(q, \omega)} = 2 e^2 r^2 q^2 \sum_{k, k'} \sum_{m} \frac{n_{dm}(k) - n_{dm}(k')}{E_{d}(k) - E_{dm}(k) - \omega + i\varepsilon}
\]

\[
\times \left( \psi_{dm}(k) \left| \psi_{dm}(k') \right> \right) \left< \psi_{dm}(k) \left| \psi_{dm}(k') \right> \right) \left< \psi_{dm}(k') \left| \psi_{dm}(k) \right> \right> \ldots (2.129)
\]
and

$$\chi_{\text{orb}}^{\circ}(q, \omega) = 2g^2 \frac{q^2}{E_{q} E_{q'}} \sum \frac{n_g(k) - n_d(k')}{E_{dn}(k) - E_{s}(k) - i\hbar \omega}$$

$$\times \langle \Psi_{dn}^{\circ}(k)| \frac{\partial}{\partial x} \frac{\partial}{\partial y} \Psi_{s}^{\circ}(k') \rangle \langle \Psi_{dn}^{\circ}(k')| \frac{\partial}{\partial x} \frac{\partial}{\partial y} \Psi_{s}^{\circ}(k) \rangle$$

..(2.130)

The matrix elements in the above expressions, under the approximation manifested in Eqs.(2.26) and (2.36), reduce to

$$\langle \Psi_{dn}^{\circ}(k)| \frac{\partial}{\partial x} \frac{\partial}{\partial y} \Psi_{s}^{\circ}(k') \rangle =$$

$$i k_x \int_{0}^{\infty} \frac{1}{s} \int_{0}^{\infty} \frac{1}{s'} d \frac{1}{s'} \frac{1}{s} \langle \Psi_{dn}^{\circ}(k)| \frac{\partial}{\partial x} \frac{\partial}{\partial y} \Psi_{s}^{\circ}(k) \rangle$$

and

$$\langle \Psi_{s}^{\circ}(k')| \frac{\partial}{\partial x} \frac{\partial}{\partial y} \Psi_{dn}^{\circ}(k) \rangle =$$

$$i k_x \int_{0}^{\infty} \frac{1}{s} \int_{0}^{\infty} \frac{1}{s'} d \frac{1}{s'} \frac{1}{s} \langle \Psi_{dn}^{\circ}(k)| \frac{\partial}{\partial x} \frac{\partial}{\partial y} \Psi_{s}^{\circ}(k) \rangle$$

..(2.131)

Again the angular integration makes the interand for radial function zero thereby the contributions from $\chi_{\text{orb}}^{\circ}(q, \omega)$ and $\chi_{\text{orb}}^{\circ}(q, \omega)$ to the orbital susceptibility vanishes.

### 2.5 Discussion

In the evaluation of dynamical susceptibility, we use a model band structure which has reduced the computational efforts as it made possible the evaluation of many expressions analytically. In principle one should use wavefunction for s-electrons which is orthogonal to core and d-wavefunctions. An orthogonalized plane wave is a suitable choice, but it has
been found that the orthogonalization corrections are very small and therefore the use of simple plane wave for s-electrons is justified. The d-bands are in fact flat near the zone boundary, which gives the structural features in the susceptibility function therefore the parabolic band approximation for d-electrons is however crucial.

The present derivation is carried out in the Hartree approximation. The many body effects are completely neglected. However these will be introduced in the numerical calculation in the following chapter in a phenomenological manner. The spin-orbit interaction has also been neglected as it was pointed out by Hebborn and March that its contribution is too small in a transition metal like nickel.

The numerical results for Ni, Pd and Ir will be presented in the following chapter.