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Field modulation studies of the de Haas-van Alphen effect in nickel

Dennis Ray Stone
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FIELD MODULATION STUDIES
OF THE DE HAAS-VAN ALPHEN EFFECT IN NICKEL

by

Dennis Ray Stone

A Dissertation Submitted to the
Graduate Faculty in Partial Fulfillment of
The Requirements for the Degree of
DOCTOR OF PHILOSOPHY

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[Signatures redacted for privacy]

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Ames, Iowa

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ABSTRACT

We have studied the de Haas-van Alphen effect in spherical nickel crystals in fields up to 63 kOe and at temperatures as low as 1.1° K using a low frequency (≈ 40 Hz) field modulation technique. Our measurements showed that the field modulation technique could be used for ferromagnetic samples provided that the modulation frequency was less than 25 kHz and that the modulation field was parallel to the applied field direction. The observed de Haas-van Alphen oscillations are interpreted as arising from two distinct sheets of Fermi surface -- one, a multiply-connected copper-like sheet centered at the point Π in the zone and the other a set of equivalent fluted ellipsoids situated at the symmetry points X in the zone. We have interpreted these surfaces in terms of the band structure predicted by L. Hodges and have shown that our measurements are in complete accord with these new calculations.
I. INTRODUCTION

In this study of the de Haas-van Alphen effect in ferromagnetic nickel we are concerned with two striking physical phenomena which can only be explained within the framework of the quantum theory of matter. This introductory chapter will therefore be devoted to a brief discussion of the quantum mechanical ideas which account for the ferromagnetism in nickel and for the de Haas-van Alphen effect in metals.

A. Ferromagnetism in Nickel

Nickel is a ferromagnetic transition metal which at absolute zero exhibits a saturation moment of 0.6 Bohr magneton per atom (Weiss and Forrer (1929)). After the small contribution from the orbital motion of the electrons is taken into account (Argyres and Kittel (1953)), the saturation moment of nickel is reduced to 0.55 Bohr magneton per atom. The explanation of this non-integral magneton number has been regarded as one of the great successes of the "itinerant electron" or "band" theories of ferromagnetism.

In order to understand the origin of the ferromagnetism in nickel from the band point of view, we need only consider the distribution of the 10 electrons of the nickel atom which are outside the filled argon core. In the crystalline state, we assume that the wave functions of these outer electrons are unlocalized and that their energy levels form the conduction bands of metallic nickel. For the purpose of illustration, we shall consider the simple schematic relationship of the 4s and 3d bands in nickel shown in Figure 1.1.
Figure 1.1. Schematic relationship of the bands in nickel at absolute zero (after Kittel (1961))
This figure shows the band arising from the atomic 4s levels overlapping a narrow 3d band constructed from the atomic 3d states. This band arising from the atomic 3d states is split into two sub-bands by a spin-dependent interaction between the electrons. Although this interaction is of electrostatic origin, it has no classical analog. This interaction can be considered as a manifestation of the Pauli exclusion principle which prohibits changing the relative spin directions of two electrons without altering their charge distribution (and hence their electrostatic energy). We shall continue the discussion of this "exchange interaction" later but, for the purpose of the present illustration we need only assume the existence of a suitable mechanism for splitting the d band into two sub-bands (one for each spin direction).

We take one of the 3d sub-bands of Figure 1.1 as being entirely filled. The higher energy (spin-down) d sub-band is cut by the Fermi level so that it is filled except for 0.55 hole per atom at the top of the band. In order to achieve charge conservation, we assume that the electrons which could occupy this hole region are distributed in states of the overlapping s band which, in this simple model, is assumed to contain an equal number of both spin-up and spin-down electrons. We are therefore left with an excess of 0.55 unpaired spins per atom in the filled spin-up (lower energy) d sub-band and we see that even this simple model can account for the observed non-integral magneton number in a natural way.

While the simple model presented above is adequate to provide a basis for the discussion of the observed ferromagnetism in nickel, it is a gross
simplification of the detailed band theories which are presently being postulated for the transition metal ferromagnets. We shall see in Chapter III that the density of states is far from uniform in energy; the d band is characterized by a sharply peaked density of states which has an average value of the order of 20 times greater than that of the average s band density of states. Furthermore, current theories do not assume that only the d bands are split by the exchange interaction but consider the interaction as being operative on all of the conduction electrons (i.e. current theories also consider a splitting of the s-like bands albeit much smaller than that for the d bands). The effects of s-d hybridization, the exchange coupling between the s and d electrons and other subtle difficulties have further complicated the present theories (for reviews of this subject see Mott (1964a) and Herring (1966)).

In addition to these difficulties, there is considerable uncertainty as to how one should properly treat the correlation energy from the band theory point of view. The point at issue is the following: since we cannot treat the coulomb interaction between the electrons with an exact many-body theory, the simplest theories for the energy eigenvalues of a transition metal in either the atomic or metallic state involve determinental wave functions which are constructed from single particle wave functions. One then finds integrals of the type

\[ U = \int \frac{\psi^*_{n}(\mathbf{r}) \psi^*_{n'}(\mathbf{r}) \left( \frac{e^2}{\hbar^2} \right) \psi_{n}(\mathbf{r}) \psi_{n'}(\mathbf{r})}{\hbar^2} \, d\tau_1 \, d\tau_2 \sim 10 \text{eV} \quad (1.1) \]

and

\[ J = \int \frac{\psi^*_{n}(\mathbf{r}) \psi^*_{n'}(\mathbf{r}) \left( \frac{e^2}{\hbar^2} \right) \psi_{n}(\mathbf{r}) \psi_{n'}(\mathbf{r})}{\hbar^2} \, d\tau_1 \, d\tau_2 \sim e \text{eV} \quad (1.2) \]
where $\psi_n$ and $\psi_{n'}$ are atomic orbitals in the atomic case and, in the metallic state, Wannier orbitals.

At present, many-body effects are included by altering the magnitude of both $U$ and $J$ in a metal. Treatments by Slater (1936), Friedel (1955), Phillips (1964b) and Ehrenreich et al. (1963) assume that the "direct coulomb energy" $U$ is effectively screened by the conduction electrons and that the "exchange energy" $J$ is responsible for the ferromagnetism in nickel. This assumption leads to an energy difference $\Delta E$ (between the single-particle energy levels corresponding to the two spin directions) which is between 0.5 and 2 eV. Kanamori (1963), Hubbard (1963) and Gutzwiller (1964) take the opposite point of view and assume that the inter-atomic correlation effects are large; this assumption also leads to $\Delta E \sim 0.5$ eV. (See Phillips (1964a) and Mott (1964b) for more comprehensive reviews of this subject.)

In view of the controversial state of the band theory of ferromagnetism which exists at present, knowledge of the Fermi surfaces of both the spin-down and spin-up bands would provide an extremely useful test of any band structure which is proposed. Unfortunately however, our knowledge of the Fermi surfaces for ferromagnetic transition metals was, until recently, quite limited. The discovery of the de Haas-van Alphen effect in iron (Anderson and Gold (1963) ) and measurements on the galvanomagnetic properties of iron and nickel (Fawcett and Reed (1963) ) have shown that these two powerful experimental tools can be used to determine the Fermi surface topology of ferromagnetic materials. (Further discussion of these experiments will be continued in Chapter III.)
Anderson and Gold's measurements (as well as those of Fawcett and Reed) showed that the ordinary theory of the de Haas-van Alphen effect (and the galvanomagnetic properties) for nonmagnetic metals could be used for the ferromagnets provided that one replaced the applied magnetic field intensity, $H$, by the total magnetic induction $B = H + 4\pi M$ within the sample. ($\bar{B}$ is the average magnetic field over the volume of the specimen.)

Kittel (1963) has given a brief theoretical justification for the replacement of $H$ by $B$ as the effective field acting on the momentum of electrons which are subjected to the field of a lattice of point dipoles. Although this point dipole model is inappropriate for an itinerant electron ferromagnet like nickel, the conduction electrons responsible for the oscillatory magnetization move in rather large orbits so that we should, in fact, expect their behavior to be dictated by the spatial average field $B$.

Pippard (1963) and Condon (1966) have considered the somewhat different problem of magnetic interactions in non-magnetic metals (i.e. the interaction between the conduction electrons through oscillatory magnetization, $M_{\text{osc}}$). After a detailed thermodynamic analysis of this problem, they conclude that although the replacement of $H$ by $B$ in the Lifshitz-Kosevich result for the free energy (see Section B) is incorrect, the proper expression for the oscillatory component of the magnetization can in fact, be obtained by replacing $H$ by $B = H + 4\pi M_{\text{osc}}$ in the final equation for $M_{\text{osc}}$. After a brief review of the theory of the de Haas-van Alphen effect, we shall assume that the substitution of $B = H + 4\pi (M + M_{\text{osc}})$ in the final result will give an expression for the oscillatory
component of the magnetization which is appropriate for a ferromagnet.

B. The de Haas-van Alphen Effect

The oscillatory behavior of the magnetic susceptibility in metal single crystals at low temperatures (the de Haas-van Alphen effect) has proved to be one of the most powerful tools for studying the shape of Fermi surfaces in non-magnetic metals (see Shoenberg (1957) for a review of this subject). Onsager (1952) demonstrated that for an arbitrary Fermi surface, the trajectories of the electrons in a plane perpendicular to an applied magnetic field direction should be quantized; the de Haas-van Alphen frequency in $1/H$ is proportional to the extremal cross-sectional area of the Fermi surface normal to the field. Lifshitz and Kosevich (1956) later presented a detailed theory of the amplitude of the de Haas-van Alphen effect which accounted for the effect of a finite temperature. These investigators used statistical mechanics to describe a system of independent electrons (with an arbitrary dispersion law) which were subjected to an applied magnetic field $H$ and they found that the oscillatory free energy for such a system was given by:

$$ F_{osc} \sim 2kT V \sum_{j=1}^{\infty} (-1)^j \left( \frac{eH}{2\pi j e \hbar} \right)^{3/2} \left| \frac{\partial^2 \mathcal{A}_F}{\partial k^2} \right| \left( \frac{\sin \frac{\pi j \mathcal{A}_o(E_F)}{2\pi} \pm \frac{\pi j}{2} \sin \frac{\hbar j \lambda}{\sin \hbar j \lambda}}{2} \right) $$

Here $V$ is the volume of the sample, $\mathcal{A}_o(E_F)$ is the extremal area of the Fermi surface in a plane normal to the magnetic field, ($\pi/4 \pm \pi/4$ are chosen for a maximum or minimum extremal area respectively) and

$$ m^* = \frac{\hbar^2}{2\pi} \frac{\partial \mathcal{A}_o}{\partial E_F} $$
is the cyclotron mass. Here \( \lambda = \frac{2\pi^2 m* c k T}{e \hbar} \) and \( \left| \frac{\partial^2}{\partial x^2} \right| \) is a curvature factor which depends on the rate of change of the area at the extremum.

Equation 1.3 implicitly assumes that the quantized electron energy levels have zero width (i.e. that the electrons are not scattered). Dingle (1952) has shown that each harmonic in the summation should be multiplied by a term of the form

\[
\exp \left( -2\pi^2 j m* k x / e \hbar^2 \right) \tag{1.5}
\]

to take account of the finite lifetime of a particular electronic state. Here \( x = \frac{T}{A_k e \hbar} \) is an effective temperature (the Dingle temperature) and \( \tau \) is the average lifetime of the state.

In order to obtain the magnetization, we will find it convenient to rewrite Equation 1.3 in the form

\[
F_{osc} / V = 2 \sum_{j=1}^{\infty} A_j(\hbar) \sin(2\pi F_j / \hbar) \tag{1.6}
\]

where \( F_j = \frac{j e H_0}{2\pi e} \) and \( A_j(\hbar) \) contains all of the slowly varying terms in Equation 1.3 as well as the Dingle term of Equation 1.5 (we ignore the constant phase factors and \( + \pi / 4 \)). In order to obtain \( M_{osc} \) from Equation 1.6 we use the well-known thermodynamic relationship

\[
M = -\nabla_{\hbar} \left( F_{osc} / V \right)
\]

and only differentiate the rapidly varying sin terms of Equation 1.6 (i.e. we consider \( A_j(\hbar) \) as a constant). Hence, using a spherical coordi-
nate system for which \( H = (H, \theta, \phi) \), we have

\[
M_{osc} = \sum_{j=1}^{\infty} 2 A_j \frac{2 \pi F_i}{H^2} \cos \frac{2 \pi F_i}{H} \left[ \hat{h} - \frac{\hat{\theta}}{F_j} \frac{\partial F_i}{\partial \theta} \hat{\theta}' - \frac{\hat{\phi}}{F_j \sin \theta} \frac{\partial F_i}{\partial \phi} \right] \theta, \phi \tag{1.7}
\]

Here \( \hat{h} \) is a unit vector along \( H \) and \( \hat{\theta} \) and \( \hat{\phi} \) are polar and azimuthal unit vectors respectively.

Dingle (1952) has shown that a factor \( \cos \left( j \pi m^* / m \right) \) should be included in each term of Equation 1.7 to take account of the spin splitting of the energy levels. For an itinerant electron ferromagnet, however, the inclusion of this spin term is inappropriate since the spin degeneracy of the energy levels has been removed by the exchange interaction. We therefore simply multiply Equation 1.7 by \( \frac{1}{2} \) for the ferromagnetic case.

In accordance with the discussion at the end of the preceding section, we now replace \( H \) by \( B \) so that

\[
M_{osc} = \sum_{j=1}^{\infty} A_j \frac{2 \pi F_i}{B^2} \cos \frac{2 \pi F_i}{B} \left[ \hat{B} - \frac{\hat{\theta}}{F_j} \left( \frac{\partial F_i}{\partial \theta} \hat{\theta}' \right) \right] \theta, \phi \tag{1.8}
\]

where \( B = H_{ext} + H_d + 4 \pi (M_{AV} + M_{osc}) = (B, \theta, \phi) \); the various contributions to \( B \) will be discussed in detail in Chapter II. Equation 1.8 only gives the contribution to \( M_{osc} \) which arises from a single extremal area \( \mathcal{A}_0 \).

For complicated Fermi surfaces with several extremal areas we shall, in what follows, take the summation index \( j \) to include the contributions from all of the extremal areas as well as the harmonic terms.

From Equation 1.8 we see that not only can the extremal areas, \( \mathcal{A}_0 \), of
the Fermi surface be determined by measuring the $1/B$ periodicity of $\vec{M}_{\text{osc}}$ but the anisotropy of $\vec{A}_0(\theta, \phi)$ can also be deduced by studying the oscillations which occur when only the direction of $\vec{B}$ is changed. $x$ and $m^*$ can be inferred by observation of the field and temperature dependence of the amplitude of $\vec{M}_{\text{osc}}$.

In the next chapter we shall discuss the details of an experimental method (the field modulation technique) which can be used to study the behavior of $\vec{M}_{\text{osc}}$ under the conditions outlined above. This technique uses the fact that the de Haas-van Alphen magnetization is highly non-linear in $\vec{B}$. Thus if $H_{\text{ext}}$ is modulated by a very pure sinusoidal field $h_0 \sin \omega t$, the harmonic content of the voltage induced in a pick-up coil surrounding the sample will be increased by this non-linear magnetization. In the next chapter we shall see how a study of the variation of this harmonic content as a function of both the magnitude and direction of $\vec{B}$ can be used to determine the Fermi surface topology of metals.
II. EXPERIMENTAL TECHNIQUE

A. Choice of the Modulation Frequency

This study of the de Haas-van Alphen effect in nickel employs a low-frequency variation of the modulation technique first developed somewhat independently during 1962-63 by C. G. Grimes\(^1\), Shoenberg and Stiles (1964), and the author. Our original experiments, as well as those of Shoenberg and Stiles, were carried out at modulation frequencies of about 1 MHz, and these experiments demonstrated that the modulation technique is a very sensitive tool for studying the de Haas-van Alphen effect in a variety of metals, e.g., tungsten, molybdenium, tin, lead, and the alkali metals. However, despite patient searching over a period of about two years, no trace of a de Haas-van Alphen effect was ever observed in the ferromagnetic metals iron and nickel when a modulation frequency of 1 MHz was used. Since signal-to-noise ratios of greater than 5000 were observed for various non-ferromagnetic specimens, the lack of results from iron and nickel could not be attributed to any basic insensitivity of the 1 MHz modulation apparatus. On the other hand, when the modulation frequency was decreased to 25 kHz or less it was discovered that a de Haas-van Alphen effect could be observed in both iron and nickel although the overall sensitivity with respect to non-ferromagnetic specimens was greatly diminished. A completely satisfactory explanation of the negative results for ferromagnets with radio-frequency modulation has not been found, but it is suspected that the difficulty may be caused in part by tiny

---

\(^1\)Grimes, C. G., Dept. of Physics, University of California, Berkley, California. Modulation techniques. Private communication. 1963.
magnetic domains which might still exist around impurities and strains in applied field intensities greater than those needed to achieve technical saturation (cf. Aharoni (1962)). While these tiny domains might not affect the internal field appreciably, the motions of their boundaries could give rise to an excessive amount of heating at high modulation frequencies.

Since Shoenberg and Stiles (1964) have given a complete description of the theory of the modulation technique for the high-frequency ("strong skin effect") case, we will not include in this dissertation a detailed discussion of our original 1 MHz experiments or of the apparatus which was used.

While the upper limit of the modulation frequency required for observation of the de Haas-van Alphen effect in ferromagnetic samples seems to be about 25 kHz, most of the data for nickel were in fact obtained using much lower frequencies, typically in the range 10 Hz - 200 Hz. At these low frequencies there is very little change in phase and amplitude of the modulation field throughout the volume of the sample. Under these "weak skin effect" conditions it is possible to exploit an ingenious technique which was first devised by Stark\(^1\); further refinements of this technique have also been introduced by Windmiller and Priestley (1965). This technique allows the experimenter to discriminate between the various de Haas-van Alphen wave-forms when the Fermi surface is particularly complicated, and the details of the

\(^1\)Stark, R., Department of Physics, University of Chicago, Chicago, Ill. Low frequency field modulation. Private communication. ca 1965.
the method will be discussed in the following section.

B. Modulation at Low Frequencies: The "Weak Skin Effect" Limit

Figure 2.1 schematically illustrates the geometrical arrangement of the modulating and pick-up coils for the sample holders which were used in the low-frequency experiments; a detailed description of the sample holders themselves will be given in Section C-2. We consider the case when the frequency of the modulation, $\omega_s$, is sufficiently low so that the phase and amplitude of this a.c. field do not vary throughout the volume of the crystal. Under these conditions the magnetic induction within the sample is given by

$$B = H_0 \sin \omega t + H_d + 4\pi (M_{AV} + M_{osc}) + H_d^-.$$  

(2.1)

Here $H_0$ is the applied magnetic field intensity, and $H_d$ is the demagnetizing field which depends on the shape and orientation of the sample as well as the applied field intensity $H_0$. The magnetization $M_{AV}$ includes the ordinary ferromagnetic magnetization as well as the Landau-Peierls orbital diamagnetism; the latter depends only weakly on the strength of the applied magnetic field intensity, as does the ferromagnetic term above technical saturation (see Freeman et al. (1966) ).

Only for single domain samples whose bounding surface is of the second degree will the demagnetizing field $H_d$ be uniform throughout the interior of the sample (cf. Brown (1962) ). This uniform demagnetizing field is given by

$$H_d = -\overline{B} (M_{AV} + M_{osc}) \frac{Z}{V} M_{AV}.$$  

(2.2)
Figure 2.1. Schematic drawing of pick-up and modulation coil geometry (not drawn to scale)
To Impedance Matching
And Filtering Network

Coil (2) (Compensating Coil)

Coil (l') (Perpendicular Modulating Coil)

Coil (1) (Parallel Modulating Coil)

Coil (3) (Pick-up Coil)

Spacer

Spherical Sample

L

to power amplifiers
and impedance
matching networks
since $|M_{AV}| \gg |M_{osc}|$. Here $\vec{D}$ is the demagnetization tensor which depends only on the shape of the sample (Stoner (1945), Stoner and Wohlfarth (1948)). The quantity

$$B_0 = H_o + M_{AV} - \bar{D}M_{AV}$$

(2.3)

can be calculated by a trivial extension of the Stoner-Wohlfarth theory for various values of $H_o$ and $M_{AV}$ (Schau and Stone (1967)). The simplest sample geometry is evidently a sphere, for which $\bar{D} = (1/3)\bar{I}$, where $\bar{I}$ is the unit tensor. When the magnetocrystalline anisotropy is neglected, the fields $\vec{B}$, $M_{AV}$ and $\bar{D}M_{AV}$ will therefore all be parallel for a spherical sample and the direction of these fields will be independent of the orientation of the crystal axis with respect to $H_o$. Due to the above considerations, and since a sphere is the easiest second degree shape to prepare, spherical samples were used for most of the experiments reported here.

The uniform magnetic moment of a spherical sample of radius $R_o$ can be considered as arising from a simple point dipole of strength $m = \frac{4\pi R_o^3}{3} (M_{AV} + M_{osc})$ placed at its center. The emf induced at the terminals $AB$ of the pick-up coil in Figure 2.1 is then given by

$$V_{AB} = \eta \frac{4\pi R_o^3}{3} \frac{d}{dt} \left[ (M_{AV} + M_{osc}) \cdot \hat{P} \right] + V_{ext}.$$  

(2.4)

where $\eta$ is a constant depending only on the geometry of the pick-up coils. $V_{ext}$ is the voltage induced in the pick-up coil from all sources of time-varying magnetic induction external to the sample. In practice, this voltage $V_{ext}$, which includes voltages induced by the modulating coils,
the superconducting magnet, building vibrations, and external noise sources, can be minimized by connecting the coils (2) and (3) of Figure 2.1 in series opposition and by adjusting the relative numbers of turns for the smallest possible external pick-up.

It will later be shown that the contribution to the pick-up voltage which is due to the $M_{AV}$ term in Equation 2.4 cannot be neglected unless the modulation field $h_0 \sin \omega t$ is parallel to $B_0$. Before showing this result however, we shall proceed with the calculation of Stark's result for the contribution to the pick-up voltage which is due to the $M_{OSC}$ term alone. Although Stark's original derivation of this result was restricted to non-ferromagnetic specimens ($B_0 \parallel H_0$) and parallel modulation ($h_0 \sin \omega t$ parallel to $B_0$), the results which are derived in the following paragraphs are also applicable to ferromagnetic samples and non-parallel modulation.

In order to calculate $M_{OSC}$ consider a spherical coordinate system $(r, \theta, \phi)$ and let the respective unit vectors be denoted by $\hat{r}$, $\hat{\theta}$, and $\hat{\phi}$. We have then, from Chapter I, for $B = (B, \theta, \phi)$

$$M_{OSC} = \sum_{j=1}^{\infty} A_j(B) \cos (2\pi F_j(\theta, \phi)/B)$$

(2.5)

where

$$A_j(B) = \frac{2\pi F_j}{B^2} \left[ \hat{r} - \hat{\theta} \frac{F_j(\theta', \phi')}{\partial \theta'} \right] \hat{\phi} - \frac{\hat{\phi}}{F_j \sin \theta} \frac{F_j(\theta', \phi')}{\partial \phi'}$$

(2.6)

For simplicity the subscript $j$ in the above sum refers to all observed contributions to $M_{OSC}$, i.e. the fundamental de Haas-van Alphen term as well as the harmonic terms ($2F, 3F, ...$ etc.) which occur in the Lifshitz-Kosevich theory (Chapter I), are also included in Equation 2.5.
If we now write

\[ B_o = (B_o, \theta_o, \phi_o), \quad h_o = (h_o, \theta_o + \alpha, \phi_o) \]  

(2.7)

and assume that \( B_o \gg h_o \), then the total magnetic induction \( B \) from Equations 2.1 and 2.3 becomes, to first order in \( h_o/B_o \),

\[ B = (B_o + h_o \cos \alpha \sin \omega t, \theta_o + \frac{h_o \sin \alpha \sin \omega t}{B_o}, \phi_o) \]  

(2.8)

Since \( A_j(B) \) is a slowly-varying function of \( B \), when compared to the rapidly-varying cosine terms in Equation 2.5, we may replace \( A_j(B) \) by \( A_j(B_o) \). We now use Equation 2.8 to substitute for \( B \) in the cosine terms of Equation 2.5 and expand \( F_j(\theta, \phi)/|B| \) in a Taylor series about the point \( (B_o, \theta_o, \phi_o) \), so that we have to first order in \( h_o/B_o \),

\[
M_{osc} \sum_j \frac{\partial}{\partial B} A_j(B_o) \cos \left[ \left( \frac{2\pi}{B_o} \right)^2 F_j(\theta_o, \phi_o) + \frac{\partial F_j(\theta, \phi)}{\partial \theta} \theta_o, \phi_o \frac{h_o \sin \alpha \sin \omega t}{B_o} \right] \\
\times \left[ 1 - \frac{h_o}{B_o} \cos \alpha \sin \omega t \right] \\
= \sum_j \frac{\partial}{\partial B} A_j(B_o) \cos \left[ \frac{2\pi F_j(\theta_o, \phi_o)}{B_o} \right] \\
\times \left[ 1 - \frac{h_o \sin \omega t}{B_o} \cos \alpha - \frac{\partial F_j(\theta, \phi)}{\partial \theta} \theta_o, \phi_o \frac{h_o \sin \alpha}{F_j(\theta_o, \phi_o)} \right] \\
\]  

(2.9)

If we introduce

\[ h_j^{eff} = h_o \left[ \cos \alpha - \frac{\partial F_j(\theta, \phi)}{\partial \theta} \theta_o, \phi_o \frac{h_o \sin \alpha}{F_j(\theta_o, \phi_o)} \right] \]  

(2.10)
Equation 2.8 can be rewritten as

$$L_{osc} = \sum_j A_j(B_o) \left[ \exp \left( \frac{12\pi F_j(\theta_o, \phi_o)}{B_o} \right) \left[ \exp \left( \frac{-12\pi F_j(\theta_o, \phi_o)}{B_o} \right) h_j \sin \omega t \right] \right] (2.11)$$

Equation 2.1 can now be transformed by use of the Bauer expansion (cf. Bowman, Introduction to Bessel Functions, page 89)

$$\exp(-iu \sin x) = \sum_{n=-\infty}^{+\infty} J_n(u) \exp(-inx), \quad (2.12)$$

where $J_n(u)$ is the Bessel function of the first kind and of order $n$.

We then have

$$L_{osc} = \sum_j A_j(B_o) \exp \left( \frac{12\pi F_j(\theta_o, \phi_o)}{B_o} \right) \left[ \sum_{m=-\infty}^{+\infty} J_m(x_j) \exp(-im\omega t) \right] (2.13)$$

$$L_{osc} = \sum_j A_j(B_o) \left[ \cos \frac{2\pi F_j(\theta_o, \phi_o)}{B_o} \sum_{m=-\infty}^{+\infty} J_m(x_j) \cos(m\omega t) + \sin \frac{2\pi F_j(\theta_o, \phi_o)}{B_o} \sum_{m=-\infty}^{+\infty} J_m(x_j) \sin(m\omega t) \right] (2.13)$$

where

$$x_j(F_j, h_j, B_o) = 2\pi F_j(\theta_o, \phi_o) h_j / B_o. \quad (2.14)$$

Equation 2.10 may be further simplified through the use of

$$J_{-m}(x) = (-1)^m J_m(x) \quad (2.15)$$

and

$$\cos(\theta + m\pi/2) = \begin{cases} (-1)^m \sin \theta & \text{for } m \text{ an odd integer} \\ (-1)^{m/2} \cos \theta & \text{for } m \text{ an even integer}. \end{cases}$$

We then have
From Equations 2.4 and 2.16 we see that the induced voltage arising from the \( j \)th de Haas-van Alphen component \( F_j \) is a series of terms containing all harmonics \( m \omega \) of the modulation frequency \( \omega \). Thus if a narrow-band detection system is used to select only that voltage component whose frequency is \( m \omega \), the amplitude of these voltage oscillations will have a field dependence which is proportional to

\[
A_j(B_0) \cdot \hat{P} \frac{J_m(x_j)}{B_0} \cos \left( \frac{2\pi F_j(\theta, \phi)}{B_0} + \frac{m \pi}{2} \right)
\]

which is the result first obtained by Stark (1965).

We shall see in Section C-4 that it is possible to control the strength of the effective field \( h_j^{\text{eff}} \) so that it varies directly as \( B_0^2 \). When this is done, the argument of the Bessel function in the expressions 2.14 and 2.16, \( x_j = \frac{2\pi F_j(\theta, \phi)}{h_j^{\text{eff}} / B_0^2} \), becomes a constant quantity. The maximum response to the de Haas-van Alphen frequency \( F_j \), will evidently be achieved by adjusting \( h_j^{\text{eff}} \) so that \( x_j \) corresponds to the first maximum of the \( m \)th order Bessel function, e.g. \( x \sim 5 \) for \( m=4 \) (e.g. Figure 2.2). On the other hand, it is often the case that some strong de Haas-van Alphen term completely swamps all other terms of lower amplitude. When this happens, the strong term can be effectively eliminated by adjusting \( x \) to coincide with any of the zeros of the Bessel function, e.g. \( x=7.58, 11.06, 14.37, \text{etc.} \) for \( m=4 \) (see Figure 2.2). Finally, if the modulation field is small enough so that \( x_j \) is much less than the argument appropriate to the first maximum...
Figure 2.2. The Bessel functions $J_2(x)$, $J_4(x)$ and $J_8(x)$
of $J_m(x_j)$, we may consider the small-argument approximation

$$J_m(x_j) \approx \left(1/m!\right) \left(x_j/2\right)^m \quad (2.17)$$

Then the relative response of the system to two frequencies $F_1$ and $F_2$ will be equal to $\left(\frac{F_1}{F_2}\right)^m$ and higher frequencies are enhanced, the more so the greater $m$.

It is worth pointing out here that for non-parallel modulation ($\alpha \neq 0$), $h_{j_{\text{eff}}}$ is a function of $h_o$, $\alpha$, $F_j$, and $\left(\frac{\partial F_j}{\partial \theta}\right)$ at $h_o, \theta_o$. Thus by varying $\alpha$ and $h_o$, we can adjust the relative response of the system to either enhance or reject one of two components of $\mathbf{M}_{\text{osc}}$ which has $F_1 = F_j$ but for which $\left(\frac{\partial F_j}{\partial \theta}\right) \neq \left(\frac{\partial F_1}{\partial \theta}\right)$. Windmiller and Priestley (1965) have exploited this technique to study the complicated de Haas-van Alphen wave-forms observed in antimony. Unfortunately, this technique cannot be applied to ferromagnetic samples because a large contribution to the pick-up voltage arises from the $\dot{M}_{\text{AV}}$ term in Equation 2.4 whenever $\alpha \neq 0$.

In order to calculate the contribution to the pick-up voltage which is due to this $\dot{M}_{\text{AV}}$ term, we note that since $\dot{M}_{\text{AV}}$ depends only weakly on the applied field intensity above technical saturation (Freeman et al. (1966)).

$$|\dot{M}_{\text{AV}}(\mathbf{H})| \approx |\dot{M}_{\text{AV}}(\mathbf{H})| \approx M_s,$$

where $\mathbf{H} = \mathbf{H}_o + \mathbf{H}_d$, $\mathbf{H}_A = h_o \sin \omega t + \mathbf{H}_o + \mathbf{H}_d$, and $M_s$ is the saturation magnetization (for nickel, $M_s \approx 5.1 \times 10^2$ G). At equilibrium, the energy per unit volume $M_{\text{AV}} \cdot \mathbf{H}$ must be a minimum. Hence, if we assume that
makes a small angle $\delta(t)$ with $H$ so that

$$M_{AV} = \hat{H} M_s \cos \delta + \hat{\theta}_0 M_s \sin \delta , \quad (2.18)$$

we must have

$$\frac{d}{d\delta} \left[ (H + h_o \cos \alpha \sin \omega t) \left( \cos \delta \right) + (h_o \sin \alpha \sin \delta \sin \omega t) \right] = 0.$$

Here $\alpha$ is the modulation angle as in Figure 2.1. Therefore

$$\tan^{-1} \delta = \frac{h_o \sin \alpha \sin \omega t}{H + h_o \sin \omega t \cos \alpha},$$

but since $h_o \ll H$

$$\delta(t) \approx \frac{h_o \sin \alpha \sin \omega t}{H}. \quad (2.19)$$

After substituting Equation 2.19 into 2.18 and again making use of Equations 2.12, 2.15, and 2.17, we have

$$M_{AV} = \hat{H} M_s \left\{ J_0 \left( \frac{h_o \sin \alpha}{H} \right) + 2 \sum_{n=1}^{\infty} J_{2n} \left( \frac{h_o \sin \alpha}{H} \cos 2n \omega t \right) \right\}$$

$$\quad + \hat{\theta}_0 M_s \left\{ 2 \sum_{n=1}^{\infty} J_{2n-1} \left( \frac{h_o \sin \alpha}{H} \sin (2n-1) \omega t \right) \right\}$$

Since $h_o \ll H_0$,

$$M_{AV} = M_s \left\{ \hat{H} \left( 1 + 2 \sum_{n=1}^{\infty} \left( \frac{h_o \sin \alpha}{H} \right)^n \cos 2n \omega t \right) \right\}$$

$$\quad + 2 \hat{\theta}_0 \sum_{n=1}^{\infty} \left( \frac{h_o \sin \alpha}{H} \right)^{2n-1} \sin (2n-1) \omega t \}}.$$

Finally,

$$M_{AV} \cdot \hat{P} = 2M_s \sum_{m=1}^{\infty} m \omega \left( \frac{h_o \sin \alpha}{H} \right)^m \cos \left( \hat{\theta}_0 + \frac{m \pi}{2} \right) \sin \left( m \omega t + \frac{m \pi}{2} \right). \quad (2.20)$$
where $\theta_0$ is the angle of $H$, (or $E$) and $\hat{P}$ is a unit vector parallel to the axis of the pick-up coil as shown in Figure 2.1.

From Equations 2.4 and 2.20 we see that whenever the modulation is not parallel to $H$, the induced voltage arising from $M_{AV}$ is a series of terms again containing all harmonics $m \omega$ of the modulation frequency $\omega$. In order to compare the magnitude of the $m$th harmonic component of Equation 2.16, we consider the case when $x_j << 1$ (so that Equation 2.17 is applicable). The ratio of the contribution arising from $M_{AV}$ to that from $M_{osc}$ is then

$$\sim \left( \frac{M_{gsin} \alpha}{2 \pi |A_j| (F_j/H)} \right)^m$$

For $H \sim 6 \times 10^4$ Oe and $F \sim 10^7$ G, this ratio becomes $(10^{-3} M_{gsin} \alpha)/|A_j|^m$. Since $10^{-3} M_{s}$ is greater than $|A_j|$ for all of the de Haas-van Alphen frequencies observed in nickel, we expect the contribution from $M_{AV}$ to swamp the oscillatory term at large modulation angles $\alpha$. In practice, we indeed found it impossible to detect de Haas-van Alphen signals whenever $\alpha$ exceeded $5^\circ$.

Finally, we note that for an arbitrary time variation of the magnetic induction, $B(t)$, the component of the pick-up voltage associated with the de Haas-van Alphen term $F_i$ has a time dependence proportional to

$$\cos(2\pi F_i/t) = \cos(-\gamma(t) t + \tau(t))$$

(2.21)

where

$$\gamma(t) = 2\pi F_i \frac{d}{dt} \left( \frac{t}{B(t)} \right) \quad \text{and} \quad \tau(t) = 2\pi F_i \frac{d}{dt} \left( \frac{t}{B(t)} \right).$$

However, if we arrange to have
\[ |\mathbf{B}(t)|^2 = C|\mathbf{B}(t)|, \quad (2.22) \]

where \(C\) is a constant, both \(\gamma\) and \(\frac{\gamma}{2}\) are then constants so that Equation 2.21 is strictly periodic in time (with period \(= \frac{1}{2\pi F_C}\)). Therefore, if Equation 2.22 is satisfied, the resultant de Haas-van Alphen signals will be periodic in time and can be analysed by ordinary analog filtering techniques (cf. Sections C-1 and C-4).

C. Experimental Apparatus

1. Superconducting magnet system and cryostat

In order to produce high magnetic field intensities with the degree of homogeneity required for observation of the de Haas-van Alphen effect, a Varian Associates X-4102 superconducting magnet system was chosen for this work. This system is capable of producing magnetic field intensities of 63 kOe while maintaining a field uniformity of 3 parts in \(10^5\) over a spherical volume of 1 cm diameter. To achieve this degree of field uniformity, sixth order compensating coils are wound on the interior of the main field windings. Although these compensating coils are connected in series with the main field windings, the current in the main windings can be independently increased or decreased (from -1 A to +1 A) relative to the current in the compensation windings, through the use of the shimming power unit which is contained in the main power supply frame. By properly adjusting this shimming control, one can presumably compensate for any adverse effects which "trapped flux" might have on the field uniformity.

The power supply for this magnet system provided currents of up
to 25 A with a stability of 1 part in $10^5$ for periods of twenty minutes or more. This power supply can be programmed so that the time-rate-of-change of the current flowing through the magnet is either a constant or proportional to the square of the instantaneous value of the current. The limits of error for the prescribed functions in the above two programmable modes are $\pm 0.1\%$ and $\pm 1.0\%$ respectively. A continuous reading liquid-helium-level monitor and various protective circuits for the superconducting magnet are also contained in the power supply frame.

The magnetic field intensity produced by the superconducting solenoid was calibrated by using the nuclear magnetic resonance (NMR) spectrometer shown in Figure 2.3. The marginal oscillator is a modified version of the oscillator described by Hill and Hwang (1966) and a detailed circuit diagram is given in Figure 2.4. The NMR sample is contained within sample holder SP-2 (cf. Section C-2) and the NMR signals could be monitored at the same time as de Haas-van Alphen signals were being observed. Figure 2.5 shows recorded NMR resonances from Al$^{27}$ which were used to check the frequency of the [100] $\vec{B}$ oscillations in lead. In order to get meaningful results from these measurements, it is important that the NMR probe and the de Haas-van Alphen sample be in close proximity ($\sim 0.3$ inch separation in our experiments).

Unfortunately the field inhomogeneity caused by the presence of our ferromagnetic nickel sample was found to be sufficient to destroy the NMR signal. Thus, for the Ni measurements it was necessary to rely on a calibration of magnetic field intensities in terms of the currents in the solenoid. Here two coefficients are involved, and we write
Figure 2.3. A block diagram of the NMR spectrometer used for field intensity calibration purposes
NOTES:
1 - UG-626 COAX FITTING "DOPED" WITH TOR-SEAL TO PROVIDE VACUUM SEAL
2 - RG-58U COAX CABLE
3 - TRANSMISSION LINE: 50\" OF RG-58U WITH CU BRAID REPLACED BY 3/16\" 321 STAINLESS STEEL TUBING
4 - R.F. COIL: 8 TURNS 36 AWG CLOSE WOUND ON 3/16\" DIA.
5 - NMR SAMPLE: 1/4\" X 3/16\" GLASS TUBE FILLED WITH 154 AI POWDER
6 - D HV A MODULATION COIL
7 - PLUG CAN BE PLACED IN EITHER A OR E
Figure 2.4. Circuit diagram of the marginal oscillator used in the NMR spectrometer. (Dashed lines indicate r.f. shielding; all resistances given in ohms, inductances in microhenries, and capacitances in microfarads unless otherwise noted.)
Figure 2.5. Typical data obtained from NMR spectrometer with A127 sample. (Upper trace shows the [100] oscillations which were simultaneously recorded with the NMR signal.)
DHVA SIGNAL ([100] \( \beta \) OSCILLATIONS IN Pb)

OUTPUT FROM NMR SPECTROMETER

\( \text{Al}^{27} \text{ RESONANCE } \neq 11.1119 \text{ MHz} / 10^4 \text{ Oe} \)

40.8139 MHz

36.7299 K0e

41.7855 MHz

37.6043 K0e
where \( I_m \) and \( I_s \) are the main and shimming currents respectively; we shall discuss their measurement shortly. NMR measurements showed that the coefficients \( C_1 \) and \( C_2 \) were not constant even above the troublesome low field \( (H < 20 \text{ kOe}) \) hysteretic region; the coefficients were found to depend on \( I_s \), although apparently not on the main current \( I_m \). The values of \( C_1 \) and \( C_2 \) for a particular shimming current, were determined by making a least-squares fit of Equation 2.23 to field points obtained from 100 NMR frequencies. All of the nickel data were taken with \( I_s = 0.086 \text{ A} \) and for this shimming current the coefficients were found to be

\[
C_1 = (3.045 \pm 0.002) \text{ kOe/A} \quad \text{and} \quad C_2 = (274 \pm 2) \text{ Oe}.
\]

These coefficients were obtained by using the NMR spectrometer in the absence of the nickel sample and we must tacitly assume in what follows that the introduction of the ferromagnetic sample does not alter the field intensity produced by the superconducting magnet (e.g. by image effects).

The values of the currents \( I_m \) and \( I_s \) were determined by measuring the potential difference which appeared across 0.001 ohm standard resistors which are provided in the power supply for monitoring purposes. These potential differences were measured with a Leads and Northrop type 5657 potentiometer which has a limit of error given by \( \pm (0.04\% \text{ of reading} + 3\mu \text{V}) \). Since the frequency stability of the marginal oscillator was always greater than a few parts in \( 10^6 \), and since the resonance could be centered on the oscilloscope screen to at least \( \frac{1}{2} \text{ Oe} \), a typical uncertainty of 0.06% due to the potentiometer is a conservative
limit of error for the magnet calibration. Hysteresis effects, if present, were much smaller than the .06% measurement error for fields above 25 kOe, and the magnet calibration did not change when the magnet was cycled between room temperature and 4.2° K.

The liquid helium cryostat used in these experiments was furnished by Varian Associates as part of their superconducting magnet system. This cryostat was capable of holding 15 liters of liquid helium in the magnet chamber while the maximum capacity of the sample isolation chamber was 2 liters.

Temperatures as low as 1.1° K were attained by pumping on the liquid helium contained in the sample isolation chamber by means of a Heraeus-Rootes type vacuum pump (CVC model VPRG-1000A) backed by a Kinney fore-pump (CVC model E225). Temperatures were determined from the vapor pressure of liquid helium by ordinary mercury and oil manometers.

2. Sample holders

One of the major experimental problems was to design and construct an accurate and reliable mechanism for changing the orientation of a sample with respect to the axis of the superconducting solenoid. Two radically different sample holder systems which were used for the majority of the nickel experiments are shown in Figures 2.6a, 2.6b, and 2.7; we now describe each in turn.

The first of these (sample holder SP-1) employed a 0.01 in. diameter, type 321 stainless steel control wire and nylon wheel to accomplish the sample rotation. The spherical sample was glued (with G.E. #7031 adhesive) into a small "black nylon" wheel which was contained in a "key
Figure 2.6a. A pictorial view of the sample rotation mechanism of sample holder SP-1. (The perpendicular modulation coils are not included in this drawing for clarity.)
STAINLESS STEEL CONTROL WIRE WRAPPED ONCE AROUND SAMPLE WHEEL

PICK-UP COILS
SAMPLE WHEEL
SPHERICAL SAMPLE
SPACER
MODULATING COIL

IDLER WHEEL
TEFLON BUSHING
STAINLESS STEEL AXLE

"BLACK NYLON"

TEFLON BUSHINGS

DETAIL "A"
Figure 2.6b. Over-all view of sample holder SP-1
(Cryostat vacuum seals have omitted for clarity.)
Figure 2.7. Schematic drawing of rotation mechanism for sample holder SP-2 (not drawn to scale)
TO MARGINAL OSCILLATOR

$\text{Al}^{27}$ NMR SAMPLE

SPHERICAL SAMPLE

SAMPLE SUPPORT

BRASS WORM

KEL-F BODY

KEL-F WORM GEAR

COILS
hole" shaped slot in the pick-up coil former (cf. Figure 2.6a). A loop of stainless steel control wire, under approximately 4 lbs. tension (provided by a spring at the top of the support tube) was looped around both the sample wheel and an idler wheel located below the pick-up coil. The ends of this control wire loop were connected to the top of the sample holder through the support rod and wrapped twice around a pulley. This pulley was actuated by an anti-backlash worm gear which was driven by a synchronous motor and gear box. The angular position of this worm gear was monitored by using a Borg-Warner revolution counter connected to the worm shaft.

The revolution counter could not be used for the purpose of obtaining an absolute angular orientation of the sample because of backlash in the system caused by stretching of the stainless steel control wire. However, after this backlash had been taken up, the system provided an extremely reliable, smooth, and vibration-free mechanism for sample rotation. Since the sample could be rotated a total of 600 degrees in either direction, the backlash difficulties did not in any way limit the usefulness of this system. Relative angular positions of the sample could be determined with a limit of error of ± 0.5 degree. Total elimination of backlash could presumably be effected by using either a control wire with a lower modulus of elasticity or by shortening the length of the control wire (by placing the worm gear nearer to the sample wheel).

The perpendicular and parallel modulating coils shown in Figure 2.6a contained 400 and 1000 turns of number 40 AWG copper wire respectively. The parallel modulating coil was wound on a $\frac{1}{2}$ in. diameter by 1 in. long
cylindrical spool and the perpendicular coil was wound in the form of
two multi-turn loops located outside of the parallel coil. The pick-up
coils were connected in series-opposition so that their total mutual
inductance with respect to the parallel modulating coil was reduced by
a factor of 5000 below that for a single coil. In order to accomplish
this high degree of compensation, the phase sensitive detection system
discussed in Section C-4 was used to detect the out-of-balance voltage
during the final adjustment of the relative turns ratio. The sample
holder was also immersed in liquid nitrogen while making these measure-
ments.

Sample holder SP-2 shown diagrammatically in Figure 2.7 exploited a
simple worm gear arrangement for simultaneous rotation of a modulation
coil, a pick-up coil and the sample. An additional fixed coil for parallel
modulation was provided by winding 1000 turns of number 40 AWG copper
wire over the Kel-F body of this device. This fixed coil provided modu-
lating fields for both the de Haas-van Alphen sample and for the NMR
spectrometer sample which are situated about 0.3 in. from each other.
This system of sample rotation allows absolute positioning of the sample
with a limit of error of ±0.3 degree. Since the pick-up coil rotates
with respect to the axis of the solenoid, use can be made of the \( \mathbf{k} \cdot \hat{\mathbf{p}} \)
term in Equation 2.4 to provide additional de Haas-van Alphen frequency
discrimination.

Unfortunately, however, this second rotation device was very suscep-
tible to vibration caused by friction between the worm and the sample
wheel. This vibration induces sporadic noise into the pick-up voltage.
The vibration seemed to be proportional to the amount of solid air which accumulated in the sample chamber after several days of experimentation.

A Polaroid x-ray camera was used to align a particular crystallographic plane of the spherical sample with the plane of rotation of the sample holder. Initially the sample was loosely held in the appropriate sample retainer (i.e. the sample wheel of SP-1 or the sample support cone of SP-2) through the use of special fixtures mounted on the x-ray camera. After the desired sample orientation had been achieved, the sample was glued into the retainer. The orientation was then checked (after the glue had dried) while the sample retainer remained in situ on the x-ray camera. The estimated limit of error in the alignment of a particular crystallographic plane by this method was ± 1°.

3. Samples and sample preparation

Although a number of different nickel samples were investigated throughout the course of this work, most of the data were obtained from two 0.18 in. diameter single-crystal spheres which have been designated NI-3 and NI-7. Both of these spherical samples were spark-cut from a ¼ in. diameter electron-beam zone-refined rod which was obtained from Dr. W. A. Reed of the Bell Telephone Laboratories. This rod had a residual resistance ratio ($R_{300}/R_{4.2}$) = 980, as measured in the presence of the earth's magnetic field; however, due to the contributions of the domain structure this value of $R_{300}/R_{4.2}$ is lower than that which would be obtained for a single domain sample (Dheer (1967)). The samples were spark-cut under kerosene with a Servo-Met spark cutter and lathe attachment.
The sample NI-3 was carefully polished by Mr. Joseph A. Corriveau\textsuperscript{1}. This sample had a measured sphericity of 2 parts in $10^5$ before a thin polycrystalline surface layer was etched off. The sphericity measurements were performed by Mr. Corriveau using a Swedish amplifying gauge which was capable of giving dimensions to an accuracy of one ten-millionth of an inch.

The sample NI-7 was lightly polished with 10/μ diamond paste in this lab and had an estimated sphericity of at least 1 part in 100. This second sample (NI-7) was annealed at a temperature of $1400^\circ$ C in a quartz crucible for a period of two days. A pressure of $10^{-5}$ Torr was maintained during the annealing process and the sample was cooled at a maximum rate of $30^\circ$/hour.

4. Modulation and signal detection circuitry

Figure 2.8 shows a schematic diagram of the modulation and signal recovery equipment used in these experiments. This equipment is housed in three six-foot Emcor cabinets which are equipped with a filtered, forced-air cooling system. Two of these cabinets are electrically isolated from the remaining one in order to eliminate a.c. ground currents which can flow between the modulation and signal detection portions of the equipment.

a. Modulation circuitry In Section B we discussed the desirability of controlling the amplitude of the modulation, $h_o$, so that $h_o \propto B_o^2$. We proceed now to a detailed discussion of the circuitry which implements this functional dependence between $h_o$ and $B_o$.

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The switches shown in Figure 2.8 allow the modulation circuits to function in either of two modes of operation which are dictated by the modulation frequency and the type of experiment being performed. When
Figure 2.8. Block diagram of electronics equipment used in these experiments
AMPLIFIER SECTION
OF OPTOUATION AC*1S
POWER SOURCE

OSCILLATOR SECTION
OF OPTOUATION AC*1S
POWER SOURCE

BROWN
SERVO AMP

ATTENUATOR

PHASE SHIFTER

HEWLETT PACKARD
AMPLIFIER

PHILBRICX KW2
OPERATIONAL
AMPLIFIER

WIDE BAND
FREQUENCY
MULTIPLIER

PA R LOCK-IN
AMPLIFIER

SERVO
REVISION
INVERTER

VARIAN K-402
POWER SUPPLY

TO SUPERCONDUCTING
MAGNET

D.C. OFFSET
UNIT

LEEDS & NORTHRUP
POTENTIOMETER

TO ANGLE MONITOR TELEMETERING
ON SAMPLE HOLDER

HEWLETT PACKARD
DIGITAL PRINTER

DYMEK DIGITAL
VOLT METER

HEWLETT PACKARD
DIGITAL CLOCK

DINER DIGITAL
VOLT METER

HEWLETT PACKARD
DIGITAL PRINTER

TO PARALLEL
WINDING COIL

TO PERPENDICULAR
WINDING COIL

TO PICK-UP COIL

FILTERING & IMPEDANCE
MATCHING NETWORKS

MOSELY 2 PEN
STRIP CHART
RECORDER

MOSELY X-Y RECORDER

VARIAN SWEEP UNIT

TO SUPERCONDUCTING
MAGNET

VARIAN
"DIGITAL CLOCK"

TO ANGLE MONITOR TELEMETERING
ON SAMPLE HOLDER

SWITCH POSITIONS
1=OPEN LOOP
2=CLOSED LOOP

TO ANGLE MONITOR TELEMETERING
ON SAMPLE HOLDER
the modulation frequency is less than 100 Hz, all of the switches are placed in the "open-loop" position and a high-purity sine-wave oscillator (total harmonic distortion less than 0.05%) is connected directly across the voltage divider $R_1$. The output of this voltage divider is shunted by a $10^5$ ohm servo-driven analog function potentiometer (Servotrol type H155). This wire-wound analog function potentiometer was constructed so that its resistance is a parabolic function of the wiper position. The servo-positioner (shown in detail in Figure 2.9) senses the current which is flowing in the superconducting magnet and automatically orients the wiper shaft of the function potentiometer to an angular position which is proportional to the magnetic induction, $B_0$, in the interior of the sample under investigation. Therefore, the amplitude of the sinusoidal voltage, which appears between the wiper and ground, is proportional to the square of the magnetic induction, $B_0$ -- the constant of proportionality being set by the voltage divider $R_1$. To avoid loading the function potentiometer by the inputs of the power-amplifier stages, it was necessary to isolate the potentiometer with a Philbrick K-2W operational amplifier connected as a $10^7$ ohm input impedance voltage follower. The two low-distortion power amplifiers, an Optomation AC-15 and a Hewlett Packard 467A, amplify the high purity a.c. signal and separately excite the parallel and perpendicular modulation coils through appropriate impedance matching and filtering networks.

For modulation frequencies greater than 100 Hz, the switches shown in Figure 2.8 must be placed in the "closed-loop" position. This mode of operation is required because at high time frequencies the inductance
Figure 2.9. Schematic diagram of the 60 Hz servo positioner unit
(All resistances given in ohms, inductances in microhenries, and capacitances in microfarads unless otherwise noted.)
of the analog function potentiometer becomes important and its parabolic characteristic is not obeyed for a.c. signals. When the "closed-loop" mode of operation is used, a d.c. voltage, \( V_1 \), is generated which is proportional to the square of the magnetic induction. This is accomplished by replacing the high-purity sine-wave oscillator, discussed above, with a 22.5 volt battery and by connecting the Philbrick amplifier as a d.c. voltage follower. This d.c. voltage, \( V_1 \), is compared with a second d.c. voltage, \( V_2 \), which is proportional to the a.c. current flowing in the parallel modulating coil. The difference, \( V_2 - V_1 \), between these two voltages is sensed by a Brown servo-amplifier. The output of the Brown amplifier is connected to a servo-motor which controls the amplitude of a high-purity sine wave which appears across the input of the power amplifier stages. Since the connection of the servo-motor is such that the voltage difference, \( |V_2 - V_1| \), always tends to zero, the amplitude of the a.c. current which flows in the parallel modulation coil is always proportional to the square of the magnetic induction, \( B_0 \). Therefore, this mode of operation not only avoids the inductive effects of the analog potentiometer but also controls the a.c. current actually flowing in the modulation coil.

It is desirable to control the current flowing in the modulation coil (rather than the voltage across the coil as is done in the "open-loop" case) since the magneto-resistance of the modulating coil does not then affect the \( B_0^2 \) amplitude dependence of the modulating field. Unfortunately, however, the electro-mechanical components used in this "closed-loop" circuit proved to be rather unreliable and the simpler
"open-loop" mode of operation was used wherever the experimental conditions allowed.

For either of the above two modes of operation, the adjustment of $R_1$ and the relative gains of the two power amplifiers allow the magnitude and direction of the modulation field to be varied. The magnitude of the modulation field is thereafter automatically controlled so that

$$\frac{h_{\text{eff}}}{B_0} \propto R_1.$$  

One can then take advantage of the frequency discrimination techniques outlined in Section B.

b. Signal detection circuitry

The impedance matching and filtering network which couples the pick-up coil to the preamplifier in Figure 2.8 serves a twofold purpose. This network not only transforms the impedance of the pick-up coil into the optimum source impedance for the preamplifier but it also eliminates any spurious signals which might overload the input stages of the preamplifier. Since the impedance of the pick-up coil is a function of both frequency and coil inductance, several different matching schemes were dictated by the various pick-up and modulation frequencies used in these experiments. Figures 2.10 and 2.11 show the details of two typical networks which are used at 50 kHz and 160 Hz respectively. The 50 kHz filter is basically a five-pole Butterworth band-pass filter (Reference Data for Radio Engineers (1961)) which uses the pick-up coil as one of its circuit elements. This filter actually attenuates sufficiently outside of its 2 kHz pass band to permit the use of a totally uncompensated pick-up coil without overloading of
Figure 2.10. A 5 pole Butterworth filter with 90 db/octave response (outside of the 2 kHz bandpass region located at 50 kHz). Coils are wound on $1\frac{3}{8}$ in. diameter polystyrene forms -- capacitors are silver-mica types.
The diagram shows a circuit with the following components:

- **PICK-UP COIL**
  - $L_p \sim 2.2 \text{ mH}$
  - $C_p \sim 132 \text{ pfd}$

- Capacitors:
  - $38 \text{ pfd}$
  - $394 \text{ pfd}$
  - $284 \text{ pfd}$
  - $505 \text{ pfd}$

- Inductors:
  - $L \sim 0.8 \text{ mH}$, $Q = 100$ at $50 \text{ KHz}$

The circuit leads to a pre-amp.
Figure 2.11. 80 Hz constant-k band reject filter (500 mH coil is cooled in a liquid nitrogen bath)
500 mH; Q ~ 50 @ 80 Hz

PICK-UP COIL

INPUT TRANSFORMER
OF PAR-HR8
LOCK IN AMPLIFIER
the preamplifier at the modulation frequency. The 160 Hz filtering arrangement is basically a constant-k 80 Hz band-reject filter (Reference Data for Radio Engineers (1961)) which uses the pick-up coil and the low noise input transformer of the preamplifier as circuit elements.

A Princeton Applied Research model HR-8 lock-in-amplifier was used to recover the desired de Haas-van Alphen information contained in the amplitudes of minute a.c. signals. These signals were often buried deep in noise caused by vibration, stray pick-up, the presence of solid oxygen near the pick-up coil, and the inherent Johnson noise of the input circuits. The lock-in technique cross-correlates the input signal (which contains both desired signal and noise) with a reference signal which is both frequency and phase locked to the desired signal. The result of this cross-correlation is to produce a d.c. voltage which is proportional to the amplitude of the desired a.c. signal and to average the uncorrelated noise to zero. Since the equivalent noise bandwidth of the system is directly proportional to the averaging time employed in the cross-correlation (i.e. the time constant of the low-pass filter in the HR-8 detector), the time-rate of change of the amplitude of the desired signal sets a practical limit on the ultimate noise rejection of the system. In practice integration times on the order of one second would permit the detection of about $2 \times 10^{-7}$ V from sample holder SP-1 and about $5 \times 10^{-8}$ V from holder SP-2.

The wide-band frequency multiplier circuit shown in Figure 2.12 is used to provide the reference voltage required for operation of the lock-in-amplifier at harmonics of the modulation frequency. Frequency
Figure 2.12. Wide band (1 to 100 kHz) frequency multiplier
(All resistances given in ohms and all capacitances
given in microfarads unless otherwise noted.)
multiplication is accomplished through the use of three frequency doublers which can be operated in cascade for even harmonic generation. Odd harmonics are generated by using a diode square-wave clipper followed by the doubler circuits. This device is capable of generating up to twenty-first harmonic reference voltages in a frequency range from 1 Hz to 100 kHz.

5. Data recording

In order to obtain absolute de Haas-van Alphen frequencies for a particular orientation of the crystal, the magnet current was varied using one of the two programmable modes of the power supply. The slowly varying d.c. voltage which appeared at the output terminals of the lock-in-amplifier during these field sweeps was passed through an ultra-low-frequency filter (cf. Figure 2.8) and recorded in either analog or digital form as a function of time.

a. Analog data recording The analog records were made by either simultaneously recording the magnet current and the output of the lock-in-detector as ordinates on a Moseley 7101A two pen strip chart recorder, or by displaying the lock-in-detector output as the ordinate on a Moseley 7001 X-Y recorder with the magnet current as the abscissa. Particular care was exercised during these field sweeps to avoid the effects of time lags between the instantaneous values of the magnetic induction at the sample and the current in the solenoid. This time lag is caused both by eddy currents in the magnet spool, and to a lesser extent, in the dewar tail; the lag prevents precise calculation of the magnetic induction at the sample when the magnet current is changing. Therefore,
precise current measurements were made only for fixed values of the magnet current. Any additional cycles of de Haas-van Alphen oscillation which appeared at the end of a sweep could easily be counted on the strip chart records. Since these additional oscillations occurred at the same current value they could not be resolved on X-Y records and the strip chart recorder proved to be the most useful of the two analog recording techniques.

For particularly simple analog data records, such as the one shown in Figure 2.13, the de Haas-van Alphen frequency can be determined directly from the data record by using the relation

\[ F = \frac{n (B_2 - B_1)}{B_2 - B_1} \]

where \( n \) is the number of cycles (estimated to 0.1 cycle in typically 70 cycles) between the measured static field points \( B_1 \) and \( B_2 \).

b. Digital data recording  In some complex cases, such as the analog record shown in Figure 2.14, it is tedious to utilize the inherent frequency discrimination capabilities of the apparatus for the resolution of a complex de Haas-van Alphen wave forms into separate components. A digital data processing scheme developed by P. T. Panousis (1967) was therefore often used for the analysis of such wave forms.

To exploit Panousis's digital analysis it was necessary to obtain a digital record of the output voltage of the lock-in-detector as a function of time. This digital record was obtained by using a Dymec 2401 A digital voltmeter which was gated with an accurate one second timing pulse to convert the output voltage of the lock-in-detector into
Figure 2.13. Typical de Haas-van Alphen field sweep obtained from nickel sphere with field along a [100] axis (Diagonal line is proportional to solenoid current; time for sweep = 28 min.; $\mathcal{E} = 1.00 \times 10^7$ G.)
INITIAL STATIC CURRENT 17.305 A

FINAL CURRENT 11.818 A
Figure 2.14. Typical analog de Haas-van Alphen data for nickel which was simultaneously recorded with the digital data shown in Figure 2.15

① = First digitized point
② = Last digitized point
FINAL STATIC CURRENT = 16.380 A

INITIAL STATIC CURRENT = 19.209 A
digital form at the rate of one conversion per second.

The reading of the digital voltmeter was printed on paper tape at the end of each conversion by a Hewlett Packard 562A digital printer. In order to avoid the previously mentioned time lags which are inherent in the system, the printer was not started until the response of the detector indicated that the field was sweeping. The first point of the digital record was then assumed to be the field intensity corresponding to the initial static current. Similarly the printer was not stopped until after the ramp had been completed. Since all of this digital information was obtained when using the linear current sweep mode of the power supply, field intensities at each of the digitized points were obtained by linearly extrapolating between the two static end points. A plot of the digital data which was simultaneously recorded with the analog data in Figure 2.14 is shown in Figure 2.15. Figure 2.16 shows the de Haas-van Alphen frequency spectrum obtained by using the Panousis data analysis scheme for the digital data of Figure 2.15.

c. Rotation diagrams After a particular de Haas-van Alphen frequency component had been isolated by the proper choice of pick-up frequency and modulation amplitude, the dependence of this frequency on crystallographic orientation could be determined by rotating the crystal in a constant field intensity. Equation 2.5 shows that each time the frequency \( F(\Theta_0, \Phi_0) \) changes by the magnitude of the magnetic induction \( B_0 \), the detected signal will go through one cycle of oscillation. Figure 2.17 shows a typical rotation diagram obtained for rotation in the (100) crystallographic plane of nickel. The markers which are at
Figure 2.15. Typical digital data recording for nickel (as plotted by the computer)

1 = First digitized point
2 = Last digitized point  (cf. Figure 2.14)
Figure 2.16. Frequency spectrum of the data shown in Figures 2.14 and 2.15 (obtained by using the Panousis analysis scheme; lower curve is at an intermediate stage of the computer analysis)
Figure 2.17. Typical rotation diagram for the (100) plane in nickel (symmetry point which is evident in the data is the [100] axis; the separation of the angle markers is 3.3°)
the bottom of this recording are obtained from an angle telemetering
arrangement in the motor drive mechanism of the sample holder. Most of
the rotation diagrams were obtained with sample holder SP-1 and the
angular scale was determined by a linear extrapolation between the apparent
symmetry points of the data. This sample holder could be rotated through
an angular range greater than 360° so that the consistency of this
linear extrapolation could be checked at various symmetry directions.
Figure 2.18 is a compressed rotation diagram for the (110) plane which
shows the various symmetry directions which were used to calibrate the
angular scale.
Figure 2.18. A compressed rotation diagram for nickel in the (110) plane
III. THE FERMI SURFACE OF FERROMAGNETIC NICKEL

In this chapter we shall consider the results of several experiments which have recently shed considerable light on the topology of the Fermi surface of ferromagnetic nickel. After a brief review of previous galvanomagnetic, optical, and de Haas-van Alphen experiments, we shall discuss the results of recent energy band calculations for ferromagnetic nickel. Finally, the results of our de Haas-van Alphen experiments will be presented; we shall further show how these data can be used in conjunction with band calculations of Hodges et al. (1966) to provide a model of the Fermi surface of nickel which is consistent with all of the experimental information presently available.

A. Previous Experimental Information

1. Galvanomagnetic properties

Fawcett and Reed (1963) measured the galvanomagnetic properties of ferromagnetic nickel in the high field limit ($\omega \gamma > 1$). They found that for a general field direction, $\gamma$, the magnetoresistance saturated, $\gamma$ the Hall voltage was linear with the total magnetic induction, $\gamma$ the Hall constant was consistent with a carrier concentration of one electron per atom. These results were at variance with the systematic rules for the observed galvanomagnetic properties of non-magnetic metals, from which one expected nickel to behave like a compensated metal\(^1\), having a quadratic

\(^1\)A metal is said to be "compensated" if the number of electrons equals the number of holes; for non-magnetic metals with crystal structures which allow one atom/unit cell, an even atomic number is a sufficient condition for compensation (see Fawcett (1964)).
magnetoresistance and a vanishingly small Hall coefficient at sufficiently high fields. Fawcett and Reed noted, however, that if the ferromagnetism in nickel was itinerant in origin, the spin degeneracy would be removed; one should therefore distinguish between the two sheets of Fermi surface which contain electrons of opposite spin (see Chapter I). After introducing the concept of a "spin zone" which was defined as "the assembly of quantum states in a Brillouin zone of one sign of spin," Fawcett and Reed were able to show that for ferromagnetic metals an even atomic number did not necessarily imply a compensated behavior. They further showed that all of their experimental results could be explained on the basis of a simple model which assumed only that one spin-up sheet of the Fermi surface in a single spin zone of the d band had electron character. These experiments gave the first experimental evidence which suggested that the magnetic carriers in nickel were itinerant. If this were not true, and some carriers had such low mobilities that they were effectively localized, the effective number of carriers would not have been equal to one electron/atom and the magnetoresistance would not have shown the observed uncompensated behavior.

The similarity between the anisotropy of the magnetoresistance in nickel and that for copper (Klander and Kunzler (1960)) led to the conclusion that nickel has one sheet of its Fermi surface which is essentially a sphere pulled out to contact the Brillouin zone faces along the [111] directions. In nickel however, the contact areas are considerably smaller. Assuming that the multiply-connected sheet contains electrons of a single sign of spin, the Hall data give $[7.2 \pm 1.4]$ degrees
for the angle subtended by the neck diameter at the center of the zone. This value of the angular width is consistent with that obtained from the anisotropy of the magnetoresistance data (and with de Haas-van Alphen data — see Section 2).

2. Previous de Haas-van Alphen measurements

The de Haas-van Alphen effect in ferromagnetic nickel was first discovered by Joseph and Thorsen (1963), (1964). More recent investigations have been carried out by Gold (1964), Tsui and Stark (1966) and the author. Since our measurements, as well as those of Tsui and Stark, will be discussed in detail in Section C, we consider here only the results of the earlier investigations by Joseph and Thorsen, and Gold.

Joseph and Thorsen studied the low field de Haas-van Alphen effect by using a disk shaped sample (0.20 in. dia. x 0.030 in. thick) of nickel in conjunction with a null deflection torque magnetometer. They found that the oscillations were periodic in 1/B (within the limits of their experimental error) and could be attributed to neck orbits on a copper-like Fermi surface segment (with necks protruding in the [111] directions). Over the range of their measurements the anisotropy of the de Haas-van Alphen frequency in the (110) plane could be attributed to an energy surface having the form of a hyperboloid of one sheet, i.e.,

\[ E = \frac{1}{2} \hbar^2 \left( \frac{k_x^2 + k_y^2}{m_t} - \frac{k_z^2}{m_1} \right), \quad (3.1) \]

where \( \mathbf{k} \) is the wave vector, \( E \) is measured from the symmetry point \( L \) on the hexagonal face of the zone, (the \( k_z \) axis is taken along the neck)
and $m_t$ and $m_\perp$ are the "transverse and longitudinal" effective mass parameters relative to the $k_z$ axis. The angular variation of the de Haas-van Alphen frequency and cyclotron mass corresponding to Equation 3.1 is given by

$$F(\phi)/F(0) = m(\phi)/m(0) = \left\{\cos \phi \left[1 - \left(\frac{m_t}{m_\perp}\right) \cdot \tan^2 \phi\right]^{\frac{1}{2}}\right\}^{-1}$$

(3.2)

where $\phi$ is the angle measured from the [111] axis in the (110) plane.

Joseph and Thorsen found good agreement with the observed anisotropy of the de Haas-van Alphen frequency by choosing the ratio $m_t/m_\perp$ as $0.40 \pm 0.05$ and using the measured value $F(0) = 2.74 \times 10^6$ G. By using the measured values $F(110)$, $F(111)$ and $m(110)$, Joseph and Thorsen used Equation 3.2 to calculate $m_t = (0.26 \pm 0.05)m_0$ and $m_\perp = (0.65 \pm 0.1)m_0$. The temperature dependence of the de Haas-van Alphen amplitudes yielded $m_t = (0.26 \pm 0.04)m_0$, in excellent agreement with the calculated value.

Gold (1964) also reported the observation of de Haas-van Alphen oscillations of higher frequencies in hollow nickel whiskers. These data were obtained by using the high field (250 kOe) impulsive technique but interpretation of the results was complicated by the demagnetizing effects inherent in the unusual sample geometry. Oscillations with a frequency of about $1 \times 10^7$ G were found near the [100] crystal axis; the temperature dependence of their amplitude yielded an effective mass of $(0.7 \pm 0.1)m_0$. Although Thorsen and Joseph (1964) attributed these high frequency oscillations to orbits on the hyperbolic necks discussed in the preceding paragraph, the results of the present study (see Section C) show that Gold's oscillations are in fact associated with a hole
surface located at the point \( X \) of the zone.

3. Optical properties

Ehrenreich, Phillip and Olechna (1963) studied the dielectric constants obtained from reflectance data for ferromagnetic nickel at room temperature using photon energies exceeding 11 eV. For photon energies above 4 eV, the spectral dependence in the reflectance of nickel which were obtained in these experiments showed similarity to data which had previously been obtained for copper. Below 4 eV however, there were marked differences between the spectral dependence in the reflectance of nickel and that of copper. Ehrenreich and his collaborators interpreted this structure at 0.3 and 1.4 eV in terms of interband transitions between the d bands and the Fermi surface. They proposed a model of the band structure for ferromagnetic nickel which was a simple adaptation of the paramagnetic band calculations of Hanus (1963). The Fermi surfaces predicted by these calculations consisted of three electron sheets and several small hole pockets. One surface, corresponding to spin-down carriers was copper-like but the remaining electron surfaces did not contact the zone faces. The results of this band structure were thus in agreement with Fawcett and Reed's galvanomagnetic measurements and further gave semiquantitative agreement with other measured properties which were known at that time (optical properties, the magneton number, and the electronic specific heat).

Ehrenreich, Cooper, and Hodges (1964), and Phillips (1964) have examined the spectral dependence of the off-diagonal components \( \epsilon_m = \epsilon_m^{\nu \nu} + i \epsilon_m^{\nu \tau} \) of the dielectric tensor which had been derived from
measurements of the ferromagnetic Kerr effect in nickel (Krinchik and Muralieva (1959), Krinchik (1964) and Martin et al. (1964)). Any sharp structure in $\varepsilon_m$ was interpreted as arising from regions in $k$-space where the Fermi level is such that vertical interband transitions between filled and unfilled energy bands are possible. The experimental data of Krinchik et al. is shown in Figure 3.1a. Ehrenreich and his co-workers interpreted the structure in $\varepsilon_m$ which occurs above 0.25 eV in terms of the band structure shown in Figure 3.1c. (This band structure is very similar to that predicted earlier by Ehrenreich, Phillip and Olechna (1963) and Phillips (1964). The first sharp minimum was assumed to be caused by interband transitions between the spin-down $L_2^1$ level (this band was presumed to be responsible for the copper-like necks of one sheet of the nickel Fermi surface) and a parabolic spin-down $L_{32}$ band (this band was assumed to lie beneath $L_2^1$ at the symmetry point $L$ of the zone).

The "on-set" frequency for the anomaly in the spectral dependence of $\varepsilon_m$ could then be interpreted to be the energy difference between the spin-down $L_2^1$ band and the $L_{32}$ band where the $L_2^1$ band crosses the Fermi energy. By using a combination of parameters from de Haas-van Alphen measurements (Joseph and Thorsen (1963)) and the results of Hanus' (1962) calculations to compute the energy band curvatures, Ehrenreich et al. were able to fix the position of the spin-down bands relative to the Fermi level. After this placement of the spin-down bands, the interband contribution to $\varepsilon_m$ was calculated using formulae derived by Argyres (1955). The results of this calculation are depicted by the solid lines in Figure 3.1b. The dashed lines in Figure 3.1b correspond to assumed
Figure 3.1. Explanation of the ferromagnetic Kerr effect given by Ehrenreich, Cooper, and Hodges (1964)

(a) Experimental data for the spectral dependence of \( \varepsilon_m \)

(b) Theoretical predictions for \( \varepsilon_m \)

(c) The assumed band structure near L with energies close to \( E_F \). \( E_g = 0.08 \) eV, Model 3A: \( E_A = 0.32 \) eV, \( E_B = 0.75 \) eV; Model 3B: \( E_A = 0.20 \) eV, \( E_B = 0.48 \) eV
MON Experiment
- Krinchik
- Martin, Doniach, and Neal

Theory (band Model 3A total)

\( \epsilon_m^{(1)} \)

Experiment
- Krinchik
- Martin, Doniach, and Neal

MDN Experiment

Theory
- band
- total

Model 3A

Model 3B

(C)

\( \Gamma \) -- \( L \) -- \( W \)
interband transitions between the analogous bands of the spin-up electrons. Ehrenreich (1965) later used a more realistic model of the bands obtained from self-consistent pseudo-potential calculations (Hoiges, Ehrenreich and Lang (1966)) to explain these ferromagnetic Kerr effect results and they found essential agreement with the earlier interpretation for the assumed transitions between the minority (spin-down) carriers at L.

Krinchik and Canshina (1966) have recently pointed out, however, that the position of the \( L_2 \) bands relative to the \( L_{32} \) band cannot be determined from band calculations (i.e. the precision of present band calculations is not adequate to decide if \( E(L_2) - E(L_{32}) > 0 \) or if \( E(L_{32}) - E(L_2) > 0 \)). Krinchik calculated the spectral dependence of assuming that \( E(L_{32}) - E(L_2) > 0 \) and found excellent agreement with experiment (see Figure 3.2c). This ordering of the bands at L is further supported by our de Haas-van Alphen measurements and we shall return to this point later in Section C.

B. A Review of Recent Band Calculations for Nickel

In the past few years, sophisticated first-principles band calculations for the paramagnetic state of nickel have been performed by various workers, (e.g. Hanus (1962), Yamoshita, Fukuchi, and Wakoh (1964)). The results of these calculations have been used to predict reasonable band structures for ferromagnetic nickel (Snow, Waiber and Switendick (1966), Ehrenreich (1965), Phillips (1964), Wakoh and Yamashita (1964)). While the Fermi surfaces predicted by these calculations were in qualitative agreement, such treatments of the ferromagnetic state necessarily suffer
Figure 3.2. Explanation of the ferromagnetic Kerr effect given by Krinchik and Canshina (1966)

(a) The assumed minority spin band structure near L with energies near $E_F$

(b) The assumed majority spin band structure near L with energies near $E_F$

(The dashed lines are the curves not taking into consideration the s-d hybridization)

(c) Spectral dependence of $\varepsilon_m$
from the difficulty that the correlations among the d electrons are entirely ignored. Recently Hodges, Ehrenreich and Lang (1966) have developed an interpolation scheme which is based on a representation of the d bands by a linear combination of atomic orbitals (LCAO), while the conduction bands are represented by orthogonalized plane waves. Hybridization effects between the s and d bands are included through the use of \( \mathbf{k} \) dependent matrix elements, and correlation effects are accounted for by using a suitably generalized band Hamiltonian.

All but two of the 12 parameters involved in the interpolation scheme were determined by choosing the parameters to obtain the best agreement with the Hanus bands for paramagnetic nickel. The remaining two parameters, which they denote by \( U^{d-d}_{\text{eff}} \) and \( J^{s-d}_{\text{eff}} \) were then determined self-consistently by using available experimental data for ferromagnetic nickel. The parameter \( U^{d-d}_{\text{eff}} \) was determined by the requirement that the magneton number of ferromagnetic nickel should equal 0.55 per atom. The remaining parameter, \( J^{s-d}_{\text{eff}} \) was found by fitting the experimental information about the energy bands near the point L in the zone (i.e. by using the size of the copper-like neck from the de Haas-van Alphen results of Thorsen and Joseph, and the interpretation of the ferromagnetic Kerr effect data by Ehrenreich et al.).

The energy band structure, the density of states and the proposed Fermi surfaces which were predicted by these calculations are shown in Figures 3.3, 3.4, and 3.5 respectively.

Although this calculation contains many approximations and over-simplifications, it appears to be, at the present time, the most real-
Figure 3.3. Energy band structure for ferromagnetic nickel predicted by the interpolation scheme of Hodges et al. (1966)
Figure 3.4. The density of states predicted by the interpolation scheme of Hodges et al. (1966)
Figure 3.5. Fermi surfaces predicted by the interpolation scheme of Hodges et al. (1966)
FERROMAGNETIC NICKEL

MAJORITY SPINS

MINORITY SPINS

POSSIBLE HOLE POCKETS?
istic model for ferromagnetic nickel which is available. We shall therefore make use of this model for an interpretation of the de Haas-van Alphen data discussed in the next section. We shall see that only slight changes in the parameters of Hodges' model are necessary to obtain agreement with new de Haas-van Alphen results to be discussed.

C. Experimental Results of the Present Investigation

Figures 3.6, 3.8, and 3.9 show the angular variation of the de Haas-van Alphen frequencies which were observed in our field modulation studies of spherical crystals of ferromagnetic nickel. These results are interpreted as arising from two distinct sheets of the Fermi surface — one, a multiply-connected copper-like sheet centered at \( \Gamma \) in the zone and the other a set of equivalent "fluted ellipsoids" situated at the symmetry points \( X \) in the zone. A similar independent study of the de Haas-van Alphen effect in nickel by Tsui and Stark (1966) is in complete agreement with our results and the present interpretation.

1. Copper-like necks

The angular dependence of the low frequency oscillations in the (110) plane which are shown in Figure 3.6 agree with the original data by Joseph and Thorsen (1963). A comparison with their data is made by plotting both their experimental points (open circles) and the expected angular variation for hyperboloidal necks (Equation 3.2 is plotted as a solid line using \( m_t/m_\perp = 0.40 \)) along with our data in Figure 3.6. Whereas Joseph and Thorsen observed the amplitude of these oscillations to become vanishingly small at approximately 18° on either side of [100] in
Figure 3.6. The observed angular dependence of the low frequency neck-oscillations in the (110) plane.
Nickel (110) Plane

$\Delta \nu$: Rotation of Sphere

○: Field Sweep, Sphere
○: Field Sweeps, Disc

(Joseph & Thorsen)

$F(10^6 \text{ G})$

$\frac{F(0)}{F(\theta)} = \cos \theta \left[ 1 + \frac{M_1}{M_I} \tan^2 \theta \right]^{1/2}$

$M_1 / M_I = 0.40$

[001] [111] [110]
a (110) plane, we have been able to follow the oscillations nearer to [100]. Figure 3.7 is a rotation diagram (i.e., rotation of the crystal in constant field — see Chapter II) which shows the vanishing of these oscillations at (7 ± 0.5)° from [100]. Hodges¹ has computed the angle at which the neck oscillations should vanish by finding the angle at which the necks predicted by his model will no longer support extremal orbits. His calculation shows that the oscillations should indeed vanish at (8 ± 1.5)° from [100], in good agreement with our experimental results.

Although most of the points in Figure 3.6 were obtained by using rotation diagrams, field sweeps at [111] indicate the minimum cross sectional area of the necks to be 8.13 x 10⁻³ (in units of (2π/a)²). This result was obtained by using the experimentally observed minimum de Haas-van Alphen frequency of 2.72 x 10⁶ G and an extrapolated lattice constant a = 3.5166 Å at 0° K (Heumann (1944)). The temperature dependence of these oscillations yield an effective mass of (0.26 ± 0.03)m₀, in agreement with the value obtained by Joseph and Thorsen (1963) and Tsui and Stark (1966).

2. Hole pockets at X

The results of our study of the high frequency oscillations (originally discovered near [100] by Gold (1964)) are shown in Figures 3.8 and 3.9. The angular anisotropy of the frequency of these oscillations exhibit the symmetry expected from small elongated pieces of Fermi

¹Hodges, L., Physics Department, Iowa State University, Ames, Iowa. Size of copper-like necks in nickel. Private communication. 1967.
Figure 3.7. Rotation diagram for (110) plane which shows the vanishing of the neck oscillations. (The slowly varying oscillations which exhibit a turning point at [001] are due to the fluted ellipsoids at X. The rapidly varying oscillations which are superimposed for angles $\sim 7^\circ$ on each side of [001] are due to the copper-like necks.)
Rotation of Nickel sphere in (110) plane
Figure 3.8. The observed angular dependence of the higher frequency oscillations (associated with the fluted ellipsoids at X) in the (110) plane.
HIGH FREQUENCY BRANCH VANISHES ~15° FROM [T10]

NICKEL (110) PLANE
△ FIELD SWEEP (SPHERE)
• ROTATION IN CONSTANT FIELD (SPHERE)
□ HOLLOW WHISKER RESULTS
 (IMPULSIVE FIELD: GOLD (1964))

\[ F \times 10^7 \text{ G}^{-1} \]

\[ 1.0 \leq F \leq 2.5 \]

\[ \text{ANGLE FROM [001] IN DEGREES (θ)} \]
Table 3.1. Summary of observed de Haas-van Alphen frequencies for nickel

<table>
<thead>
<tr>
<th>Direction</th>
<th>( F ) (Present Work)(^1)</th>
<th>( F ) (Tsui and Stark)(^2)</th>
<th>( F ) (Thorsen and Joseph (1963))</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>((X) hole pocket)</td>
<td>((X) hole pocket)</td>
<td>((X) hole pocket)</td>
</tr>
<tr>
<td>([100])</td>
<td>(1.003 \times 10^7 \text{ G})</td>
<td>(1.01 \times 10^7 \text{ G})</td>
<td>not observed</td>
</tr>
<tr>
<td></td>
<td>(2.433 \times 10^7 \text{ G})</td>
<td>(2.49 \times 10^7 \text{ G})</td>
<td>not observed</td>
</tr>
<tr>
<td>([111])</td>
<td>(2.72 \times 10^6 \text{ G})</td>
<td>(2.68 \times 10^6 \text{ G})</td>
<td>(2.74 \times 10^6 \text{ G})</td>
</tr>
<tr>
<td></td>
<td>(1.59 \times 10^7 \text{ G})</td>
<td>(1.58 \times 10^7 \text{ G})</td>
<td>not observed</td>
</tr>
<tr>
<td>([110])</td>
<td>((X) hole pocket)</td>
<td>((X) hole pocket)</td>
<td>((X) hole pocket)</td>
</tr>
<tr>
<td></td>
<td>(3.81 \times 10^6 \text{ G})</td>
<td>(3.85 \times 10^6 \text{ G})</td>
<td>(3.77 \times 10^6 \text{ G})</td>
</tr>
<tr>
<td></td>
<td>(2.18 \times 10^7 \text{ G})</td>
<td>(2.19 \times 10^7 \text{ G})</td>
<td>not observed</td>
</tr>
<tr>
<td></td>
<td>(2.21 \times 10^7 \text{ G})</td>
<td>(2.41 \times 10^7 \text{ G})</td>
<td>not observed</td>
</tr>
<tr>
<td></td>
<td>(1.41 \times 10^7 \text{ G})</td>
<td>(1.42 \times 10^7 \text{ G})</td>
<td>not observed</td>
</tr>
</tbody>
</table>

\(^1\) Absolute frequency data subjected to large (\(\pm 2\%\)) scatter -- see Chapter IV for explanation.


\(^3\) Frequency discrepancy at \([110]\) assumed to be due to slight misalignment from \((100)\) plane.
surfaces located at the equivalent points X in the zone (e.g., for the (110) plane: two frequency branches with turning points at [110] and [100] and a crossing point at [111] (see Figure 3.8)). Early attempts to interpret the observed frequency anisotropy in terms of closed surfaces of revolution at X were hampered by the observation that the extremal cross sectional area of the relevant piece of surface must increase faster than that expected for a cylinder with its axis along the \( \Gamma \rightarrow X \) direction (cf. solid line plotted in Figure 3.8). A closed surface of revolution which exhibits such a rapid variation must be hyperboloidal near point X — this implies the existence of two extremal areas perpendicular to the \( \Gamma \rightarrow X \) direction (viz. a dumb-bell shaped surface) — which is at variance with the observed de Haas-van Alphen frequencies at [100]. This dilemma was resolved in part when we realized that the relevant pieces of Fermi surface responsible for the high frequency oscillations need not be surfaces of revolution, but must in fact be "fluted ellipsoids" whose dimensions depend upon the direction of the magnetic field \( B \).

The band structure for ferromagnetic nickel proposed by Hodges et al. (1966) (see Figure 3.3) predicts that the Fermi surface of nickel should consist of three large electron sheets of surface centered at the point \( \Gamma \) in the zone, a small hole pocket at \( L \), and two small hole pockets at \( X \). The results of our experiments as well as those of Tsui and Stark (1966) suggest that there is only one hole pocket at \( X \) and there is also no evidence for the small hole pocket at \( L \). Hodges et al. (1966) have noted that the absence of a second hole pocket at \( X \) indicates that their \( X_2 \) energy level should be depressed below the Fermi level (i.e.
Figure 3.9. The observed angular dependence of the higher frequency (associated with the fluted ellipsoids at X) in the (100) plane. The solid lines are extrapolations to the scattered field sweep points.
NICKEL SPHERE (100) PLANE
○ FIELD SWEEP
○ ROTATION IN CONSTANT FIELD

\[ F \times 10^{-7} \text{e}^{-1} \]

ANGLE FROM [100] IN DEGREES
about 0.005 Ry below the position indicated in Figure 3.3. We therefore suppose that the fluted ellipsoids which were discussed in the preceding paragraph are centered on the point X and associated with the \( X_5 \) level as shown in Figure 3.3. We note that the \( X_5 \) level shown in Figure 3.3 is degenerate at X; in actual fact, however, the spin-orbit interaction will lift this degeneracy so that the X hole pocket and the large electron surface centered at \( \Gamma \) are separated by a small energy gap. (Figure 3.5 shows the two pieces of surface arising from this degenerate level to touch at a point on the line \( \Gamma \rightarrow X \).) In the course of our experiments, we observed the high frequency branch of the oscillations (associated with the \( X_5 \) hole pocket) to vanish over a narrow angular region (0.2°) situated about 2° on either side of a \([110]\) direction in a (110) plane (see Figure 3.8). Stark\(^1\) has pointed out that this behavior is probably the result of magnetic break-down of the orbit on the X hole pocket to the large electron surface which is separated from the pocket by a small energy difference along the direction \( \Gamma \rightarrow X \) due to the spin-orbit interaction. The observation of these break-down effects therefore provides further justification for the assignment of the observed fluted ellipsoids to the \( X_5 \) hole pocket.

At first it appeared difficult to explain the lack of evidence for the hole pockets at L which are predicted for the minority spins by Hodges et al. (1966). One cannot simply make adjustments of the parameters which will depress the \( L_{32} \) level beneath the minority spin Fermi

\(^1\)Stark, R., Department of Physics, University of Chicago, Chicago, Ill. Magnetic break-down of nickel. Private communication. 1967.
level because such an adjustment would also carry the flat d-like band in the region W—L beneath the Fermi level (see Figure 3.3). Since the density of states associated with this d-like band is rather large, such an adjustment of the band structure would lead to serious disagreement between the predicted and observed magneton number.

We therefore propose that Krinchik's (1966) recent interpretation of the ferromagnetic Kerr effect data is in fact correct; i.e. \( E_{L^{32}} - E_{L^{2}} = 0 \). According to this revised ordering of the bands at L (see (b), Figure 3.2) the copper-like necks must be assigned to states near the \( L^{32} \) band, and the flat d-like band is associated with the upper energy level (see Figure 3.2). We are now at liberty to vary the lower \( L_{2} \) energy level without upsetting the magneton number, and Krinchik suggests that \( E_{L_{2}} \) lies some 0.2 eV below the Fermi level to account for the Kerr effect results. Krinchik's proposed ordering eliminates the need for hole pockets at L and is thus in complete accord with our experiments.

Hodges\(^1\) has recently made minor adjustments of the parameters in his interpolation scheme in order to force agreement with the band structure at L proposed by Krinchik. The energy bands which are predicted by this new calculation are shown in Figures 3.10 and 3.11. Again \( U_{\text{eff}}^{d-d} \) has been fitted to the magneton number of 0.55 Bohr magnetons/atom.

While the over-all features of Hodges' revised band structure are only slightly changed from those of the previous calculation, the Fermi surfaces predicted from these new bands are significantly different. The

\(^1\)Hodges, L., Physics Department, Iowa State University, Ames, Iowa. Krinchik's ordering of the bands. Private communication. 1967.
Figure 3.10. The results of Hodges' new calculation for the minority spin bands
Figure 3.11. The results of Hodges' new calculations for the majority spin bands.
interpolation scheme now predicts that the Fermi surfaces should consist
of three large electron sheets centered at the point $\Gamma$ in the zone (one
of these, from the majority spin band, being copper-like with $[111]$ necks)
and a single hole pocket at the equivalent points $X$ in the zone.

Although Hodges' new calculation predicts Fermi surfaces which are
in qualitative agreement with our de Haas-van Alphen results, the extre­
mal cross sectional areas predicted for the hole pockets are about 10
per cent larger than our observations indicate. In an attempt to obtain
better agreement with our results, Hodges has recently modified his
interpolation scheme to take into account the spin-orbit splitting of
the degenerate $X_5$ level. Preliminary results obtained from this calcu­
lation indicate that the amount, $\Delta E$, by which the degenerate $X_5$ level is
split depends upon the direction of the internal field. This surprising
result is illustrated in Figure 3.12. Hodges finds for $B$ parallel to the
$[100]$ axis, the splitting is $\sim 50$ times greater along the $[100]$ and $[\bar{1}00]$ direc­tions, than it is along the $[010]$, $[\bar{1}0\bar{1}]$, $[001]$, and $[\bar{1}0\bar{1}]_F$ directions.

Since the dimensions of the $X$ hole pocket are very sensitive to the energy
difference, $\Delta E$, between the two levels at $X$, we see that for a particular
field direction the dimensions of the hole pockets at the various points
$X$ in the zone will be different. This means that the relevant pieces of
Fermi surface which are responsible for the de Haas-van Alphen frequency
variation shown in Figures 3.8 and 3.9 can not be regarded as "rigid
objects." The relevant hole pocket must be considered as surfaces whose
dimensions depend upon the direction of $B$. For the field in the $[10\bar{1}]$
direction, Hodges' calculation indicates that the "parallel" pockets
Figure 3.12. A schematic drawing of the minority spin energy bands near the point X (including spin-orbit coupling; B parallel to the [100] direction.)
\[ \Delta E_{||} \sim 0.005 \text{ Ry} \]

\[ \Delta E_{\perp} \sim 0.0001 \text{ Ry} \]
(elongated dimension parallel to the field) are smaller than the "perpendicular" pockets (those with the elongated dimension perpendicular to $B$). As the field direction is rotated away from $[100]$ the parallel pockets will tend to grow while the perpendicular pockets tend to shrink. Therefore the extremal cross sectional area of the parallel hole pockets should exhibit an extremely rapid increase as the field is rotated away from $[100]$. The preliminary results of Hodges' calculation are at present insufficient to allow a quantitative comparison between the observed anisotropy of cross sectional area shown in Figures 3.8 and 3.9 and those predicted by the band structure. The inclusion of the spin-orbit term does, however, appear to give a plausible explanation of the observed rapid variation of the de Haas-van Alphen frequency near $[100]$. We did not observe any high frequency ($F \gtrsim 10^8$ G) oscillations which could be attributed to orbits on large electron surfaces centered at the point $\bar{7}$ in the zone. In addition to measurements in the spherical samples, a search for these high frequency oscillations was conducted in a variety of other crystals (e.g. a single crystal rod which had been "pulled" from a 99.999% Ni melt, and in several irregularly shaped nickel whiskers). While the amplitudes of the oscillations observed near $[111]$ in the pulled rod were nearly five times greater than that observed for the spheres, no trace of any new high frequency oscillations was observed. Tsui and Stark\textsuperscript{1} have recently extended their measurements down to tempera-

\textsuperscript{1}Tsui, D. and Stark R. Department of Physics, University of Chicago, Chicago, Illinois. Lack of evidence for high frequency oscillations. Private communication. 1967.
tures as low as 0.3° K and they, too, find no evidence for the higher frequency oscillations. Even though one expects the oscillation amplitudes to be low in a ferromagnetic metal (since the Fermi surfaces are only occupied by electrons of one sign of spin and because of possible magnon-electron scattering effects), the lack of any evidence for the high frequency oscillations is, at present, blamed on either a lack of crystal perfection or on an unusually large cyclotron mass associated with the high frequency oscillations.
IV. SATURATION MAGNETIZATION AND ABSOLUTE FREQUENCY MEASUREMENT

The accuracy with which we can determine the absolute value of a particular de Haas-van Alphen frequency is no better than the precision involved in our calculations of the magnetic induction \( B \) at the endpoints of our field sweeps. Unfortunately, however, the published experimental data for the low-temperature saturation magnetization \( 4\pi M_s \) for nickel are rather meager and of dubious accuracy (e.g. \( M_s = (512 \pm ?) \) G, Kondorski et al. (1959); \( M_s = (508 \pm ?) \) G, Argyle et al. (1963)). The low temperature values which are often quoted (e.g. Kittel (1961), Bozarth (1951); \( M_s = 510 \) G) have been obtained from an extrapolation of the room temperature results of Weiss and Forrer (1929). These room temperature results were originally thought to be accurate to a few parts in \( 10^3 \), but it is now realized (Danan (1958)) that the Weiss-Forrer data were subjected to a rather large systematic error caused by "image effects" in their magnet, so that their published values of \( M_s \) may be low (by as much as \( 2\% \)). Because of this uncertainty we have attempted to determine the value of \( (2/3) 4\pi M_s \) indirectly from the periodicity of our de Haas-van Alphen oscillations.

Several long field sweeps were made for the oscillations which have a frequency of \( \sim 1 \times 10^7 \) G near the [100] crystallographic direction (cf. Figure 2.13). These oscillations are very strong and there is little interference from other frequencies here. Since our spherical samples allow precise determination of the demagnetizing field, we should be able to obtain a precise value of \( 4\pi M_s \) by either using the method of
Anderson and Gold (1963) or by simply choosing $M_s$ to obtain the best fit to the relation

$$F(100)\left(\frac{H_{\text{ext}}}{H_{\text{ext}} + (2/3)4\pi M_s}\right) = n/2$$

Here $H_{\text{ext}}$ is determined from the value of the superconducting magnet current at the turning-points of the oscillations and $n$ is a set of consecutive integers. For a particular set of data we found that the values of $(2/3)4\pi M_s$ which were determined by either of the above methods contained a standard deviation of only about 10 G. Unfortunately however, we found that the absolute magnitude of $(2/3)4\pi M_s$ would vary as much as 250 G for different data sets (taken over different field regions and on different days). The value of $(2/3)4\pi M_s$ thus determined was $(4260 \pm 250)$ G i.e. $M_s = 508 \pm 30$ G.

The random scatter which was observed in these determinations of $M_s$ was also reflected in a similar $\pm 2\%$ variation of our absolute de Haas-van Alphen frequency determinations. We often found that when sweeping between fixed two magnet current values, which were spaced about 10 A ($\sim 30$ kOe) apart, the number of oscillations obtained when the current was increasing would differ from that obtained for decreasing current by as much as 1.5%. Since our NMR measurements showed that the magnetic field intensity could be predicted by the magnet current to better than 0.05% (cf. Chapter II), it is difficult to explain the scatter in our long field sweep data on the basis of hysteresis effects in the field versus current relationship of the magnet system. One might, however, blame the scatter between the various data sets (obtained on different
days) on "image effects" in the magnet since these might be rather sensitive to the sample position, relative to the center of the solenoid. On the other hand, it is difficult to understand how "image" arguments can account for the differences observed between increasing and decreasing fields.

Late in the course of this investigation, measurements were made to determine the field dependence of the homogeneity of our magnet system. These experiments used a Varion Associates "Flux Ball" which consists of a search coil and a stable time-integration circuit. As the search coil is passed through the bore of the solenoid, any changes in magnetic intensity will induce a voltage in the coil. This voltage is integrated (with respect to time) and recorded as the ordinate of an x-y plot; the abscissa is proportional to the position of the coil. Due to the special construction of the search coil, this device is capable of detecting field inhomogeneities as low as 10^-6 over 1 cm distances. Therefore, by using this device, we could determine the field profile of our magnet system with a precision which far exceeded the homogeneity measurements previously obtained with the Al^{27} NMR spectrometer.\(^1\)

Preliminary results obtained from these measurements indicate that the homogeneity of our magnet system is very dependent on the value of the main magnet current. Although appropriate adjustments of the shimming

\(^1\)Since the line width of the Al^{27} resonance is \(\sim 80\) and since the sample volume is rather large, the NMR measurements give the average of the magnetic intensity over the sample volume and are thus relatively insensitive to small magnetic field inhomogeneities. We found it impossible to detect field inhomogeneities smaller than 3 parts in \(10^4\) over 1 cm with the NMR probe.
current will always adjust the field profile so that $(\Delta H/H) \sim 3 \times 10^{-5}$ over 1 cm, changing the magnet current causes a steady degradation of this homogeniety (i.e. at $\sim 50$ kOe the shimming current was adjusted so that $(\Delta H/H) \sim 10^{-5}$ over 1 cm -- the same value of the shimming current gave only $(\Delta H/H) \sim 10^{-6}$ at 45 kOe). The magnitude of this degradation was also found to be a function of sweep direction (the effect is most pronounced when the current is decreased).

At the time of this writing, the homogeniety data are insufficient to allow us to make an assessment of the effects of the homogeniety degradation on our de Haas-van Alphen results. The preliminary data do, however, suggest that the lack of consistency in our field sweep data could be caused by these effects.

We intend to determine the behavior of the field homogeniety as a function of both the main and shimming currents. Depending on the results of this study, it is hoped that the power supply can be modified to allow dynamic compensation of the field homogeniety (i.e. to automatically control the shimming current to its optimum value at all fields).
V. CONCLUSIONS AND SUGGESTIONS FOR FURTHER STUDY

A. Conclusions

Our investigations have shown that the field modulation technique can be successfully used to study the de Haas-van Alphen effect in ferromagnetic specimens provided that the modulation frequency is low enough and that the modulation is only applied in a direction which is parallel to the magnetic field. While we did not observe oscillations which could be associated with the 3 large electron surfaces centered at the point Γ in the zone, our measurements have extended the angular range of earlier measurements for the copper-like necks and have also determined the angular anisotropy of the extremal cross sectional area of hole pockets located at the equivalent points X in the zone.

The information provided by our de Haas-van Alphen measurements of these relatively small pieces of Fermi surface (e.g. the size of the fluted ellipsoids at X and the lack of evidence for hole pockets at L) has allowed L. Hodges to further modify the parameters of his interpolation scheme so that it predicts Fermi surfaces which are in excellent agreement with our observations. The bands predicted by Hodges new calculations should now be more reliable (at least near the Fermi level) and should provide realistic quantitative predictions as to the sizes and shapes of the still unobserved large sheets of Fermi surface in ferromagnetic nickel.

B. Suggestions for Further Study

Resolution of the homogeneity difficulties discussed in Chapter IV
is of primary importance. After determining a functional relationship between the optimum shimming current and the main current, operational function modules should be used to dynamically shim the magnet. Although these rather drastic measures may be difficult to implement, any absolute frequency data obtained with our superconducting magnet system (or perhaps any other) will be of dubious accuracy unless these difficulties are resolved.

We propose, in the near future, to search again for the elusive high frequency de Haas-van Alphen oscillations associated with large electron sheets of the Fermi surface. It is hoped that a real-time digital data acquisition system which has recently been incorporated in the 250 kOe impulsive field de Haas-van Alphen apparatus can be used in conjunction with the Panousis data analysis scheme to enhance the signal-to-noise ratio to the extent that the high frequency oscillations can be observed. Failing this, attempts should again be made to obtain specimens with a better crystal perfection (e.g. by pulling them from the melt, by using an ultra high vacuum electron-beam zone refiner or perhaps a combination of these two techniques).

Finally, when the information becomes available, it would be of interest to make a detailed comparison between the frequency variation for both the fluted ellipsoids and the necks which we observed in these experiments and those which are predicted by Hodges new calculations.
VI. ACKNOWLEDGMENTS

It is a pleasure to thank my major professor, Dr. A. V. Gold, for his encouragement and guidance during my graduate studies.

The understanding counsel of Dr. D. J. Zaffarano has also been greatly appreciated.

Especial thanks are due to Mr. R. A. Johnson for his helpful advice on electronic circuitry, and to Mr. P. T. Panousis for the use of his data analysis programs.

The author also wishes to thank Dr. R. Stark and Dr. E. P. Wohlfarth for many stimulating and valuable discussions.

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The author is also indebted to Dr. W. A. Reed for supplying the crystal used in this investigation and to Mr. R. A. Phillips for his advice on pulling crystals from the melt.

I would especially like to thank my wife, Karen, and my parents, Floyd and Eleanor Stone. Their encouragement has helped make my graduate career both a rewarding and enjoyable experience.
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