MERCURY DE HAAS-VAN ALPHEN AND CYCLOTRON RESONANCE STUDIES

DE HAAS-VAN ALPHEN EFFECT AND AZBEL'-KANER CYCLOTRON RESONANCE STUDIES FOR MERCURY

by

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Previous studies of the mercury Fermi surface by Azbel'-Kaner cyclotron resonance and the de Haas-van Alphen effect have been extended and clarified and several new orbits have been observed. The results are in qualitative agreement with the topologies of current models of the Fermi surface.

Measurements of the temperature dependence of the cyclotron effective mass and the collision relaxation time for an electron lens orbit have provided new experimental evidence for electron-phonon mass enhancement and the influence of Umklapp processes in determining the relaxation time.

Several methods for orienting mercury crystals within the experimental apparatus have been developed.

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ABSTRACT

Azbel-Kaner cyclotron resonance experiments were carried out at 35 GHz with two very flat single crystals of rhombohedral mercury. Mass curves were found for the α , γ , and τ orbits in essential agreement with those of Dixon and Datars, and for several more orbits on the first-zone hole surface. Three of the latter orbits had not previously been observed by any method. A three dimensional interpolation scheme developed for the α orbit masses showed that the second-zone electron lens is tipped three degrees out of the L face toward (111) and that there is a 9% anisotropy of the mass in the L face. The temperature dependence of one α orbit resonance was studied in detail between 1.15 and 2.1°K. The $\omega\tau$ (where τ is the collision relaxation time) for this resonance varied from 20.1 to 10.8, with a temperature dependence for $(\omega\tau)^{-1}$ of $\tau^{4.8}$ instead of the τ^3 dependence predicted by previous work. This dependence is explained in terms of electron-phonon scattering which includes Umklapp processes across the electron lens. The cyclotron mass for this orbit increased (1.3±0.3)% from 1.15 to 2.1°K; this change is attributed to the temperature dependence of the electron-phonon cyclotron mass enhancement and gives new experimental evidence for the mass enhancement predicted by Nakajima and Watabe.

Large amplitude field modulation and fourier analysis techniques were used to extend and clarify the de Haas-van Alphen effect results of Moss and of Brandt

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and Rayne. Two new orbits with sharp cutoffs were observed and yielded new information concerning the sizes of the L, X, and T face openings in the first-zone hole surface.

The observed cyclotron mass and de Haas-van Alphen frequency curves are in general agreement with the topologies of current models of the mercury Fermi surface; however, there remains a need for further band structure calculations to resolve apparent discrepancies between these models and the existing experimental data.

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CHAPTER I

THEORY

A. Introduction

The conduction electrons in a metal may be considered as charged fermions in a potential well having the periodicity and symmetry of the crystal lattice. This potential well is filled to a certain energy, the Fermi energy E_r. The corresponding surface between filled and empty states in momentum space (or \vec{k} space) is called the Fermi surface. It is this surface for mercury, and the de Haas-Van Alphen (dHvA) and Azbel'-Kaner cyclotron resonance (AKCR) methods of determining the Fermi surface geometry and certain properties of electrons at the Fermi surface, which are the subjects of this thesis. Mercury and its Fermi surface will be the subjects of subsequent chapters. In this chapter, the dynamics of electrons in magnetic fields and the information that can be obtained from the AKCR and dHvA methods will be discussed. Fermi surface shapes will be considered to range from spheres (for free electrons) to shapes more complicated than that shown in Figure II-2 for mercury.

B. Electrons in a Magnetic Field

The effect of a magnetic field, \vec{H} , on a conduction electron quasi-particle with wave vector \vec{k} on the Fermi surface is given by the Lorentz equation

$$\frac{\hbar d\vec{k}}{dt} = \frac{e}{c} \vec{v}_{F} \times \vec{H} , \qquad (I-1)$$

where \vec{v}_F is the real space velocity of the electron; \vec{v}_F is given by

$$\vec{v}_{F} = \frac{1}{n} \nabla_{k} E(\vec{k}) |_{E_{F}} , \qquad (I-2)$$

where $E(\vec{k})$ is the dispersion law (band structure) for the electron energies. Thus $d\vec{k}/dt$ is normal to \vec{H} and to \vec{v}_F (which is itself normal to the Fermi surface), so that \vec{k} is confined to an orbit which is the intersection of the Fermi surface and a plane normal to \vec{H} , for closed Fermi surface sections. The position of the orbital plane may be specified by the projection vector, \vec{k}_H , of \vec{k} onto \vec{H} .

By integrating Equation I-1 with respect to time, the projection of the real space orbit on a plane normal to \vec{H} is seen to have the same shape as the reciprocal space orbit, but to be rotated by $\pi/2$ and scaled by a factor $c\vec{n}/eH$. In real space, the electrons spiral along \vec{H} , with a gyration period T_c , the cyclotron period, and with an average drift velocity, \vec{v}_D , (Harrison, 1960)

$$\vec{v}_{\rm D} = \frac{1}{T_{\rm C}} \int_{0}^{T_{\rm C}} \frac{1}{\Lambda} \nabla_{\rm k} E(\vec{k}) |_{E_{\rm F}} dt = -\frac{\Lambda}{2\pi m_{\rm C}} \frac{\partial A(k_{\rm H})}{\partial k_{\rm H}}, \qquad (I-3)$$

which is non zero unless $\partial A(k_{\rm H})/\partial k_{\rm H} = 0$, where $A(k_{\rm H})$ is the cross sectional area of the \vec{k} space Fermi surface orbit corresponding to $\vec{k}_{\rm H}$. The cyclotron mass, $m_{\rm C}$, will be defined later. Orbits for which $\vec{v}_{\rm D} = 0$ are called stationary orbits. In real space, these stationary orbits are generally neither flat nor normal to \vec{H} , but the electron repeats its path again and again each cyclotron period.

The cyclotron effective mass, $m_{_{\rm C}}$, is defined such that

$$\omega_{c} = \frac{2\pi}{T_{c}} = \frac{eH}{cm_{c}}$$
(I-4)

where c is the velocity of light, ω_{c} is the cyclotron frequency, and m_{c} is a property of the electron orbit. From Equation I-1,

$$\frac{d\vec{k}}{dt} = \frac{eH}{\hbar^2 c} \nabla_{kl} E(\vec{k})$$

where $\nabla_{k_1} E(\vec{k})$ is the projection of $\nabla_k E(\vec{k})$ upon a plane normal to \vec{H} , evaluated at the Fermi surface. Then

$$T_{c} = \phi \, dt = \frac{\hbar^{2}c}{eH} \phi \, \frac{d\vec{k}}{\nabla_{k\downarrow} E(\vec{k})}$$
$$= \frac{\hbar^{2}c}{eH} \frac{1}{\delta E} \phi \, \Delta k_{\downarrow} dk$$
$$= \frac{\hbar^{2}c}{eH} \frac{\partial A}{\partial E} \Big|_{E_{F}},$$

since $\delta A = \oint \Delta_{k\perp} d\vec{k}$ is the annular area between two orbits of energy E and E+ δE . Thus, using Equation I-4,

$$m_{c} = \frac{\hbar^{2}}{2\pi} \frac{\partial A}{\partial E} \Big|_{E_{F}}$$
(1-5)

so that m is a measure of the rate of change of cross

sectional area with energy at the Fermi surface. For general Fermi surfaces, m_c is a function of \vec{k}_H for a given direction of \vec{H} . It is constant, however, over all \vec{k}_H for a given direction of \vec{H} when a Fermi surface section can be described by a local quadratic dispersion law and is therefore ellipsoidal.

The orbiting electrons are scattered with a mean inter-collision time, τ , by static imperfections, other electrons, and by phonons. The condition

$$\omega_{\tau} \tau \gtrsim 2\pi$$
 (I-6)

must therefore be satisfied if many electrons are to complete an average of one or more cyclotron orbits before being scattered and hence yield information, like m_c, about the Fermi surface.

Finally, for multiply-connected Fermi surfaces, there may be magnetic field directions for which the orbits are not closed but rather extend indefinitely through \vec{k} space. Orbits that do so are called open orbits. In real space these open orbits carry current in a direction perpendicular to both \vec{H} and to the \vec{k} space open orbit direction and have a striking local effect on the angular dependencies of effects, like magnetoresistance, which depend on the conductivity.

C. AZBEL'-KANER CYCLOTRON RESONANCE

C.1. Physical Pictures

Since the electrons spiral along the field direction, \dot{H} , with cyclotron frequency, ω_c , one expects that temporal resonance may be achieved by exciting them with microwave frequencies of comparable frequency, ω . Such resonances can be well defined only when $\omega_c \tau \gtrsim 2\pi$. For typical metals at liquid helium temperatures, $\omega_c \tau \gtrsim 2\pi$ for cyclotron frequencies of the order of 10^{11} radians per second; this frequency corresponds to mean free paths, L, of the order of 10^{-2} cm., microwave skin depths, δ , of the order of 10^{-5} cm., and orbital radii, R, of the order of 10^{-3} cm. for free electrons. Thus resonances are expected only under conditions of the anomalous skin effect (since $\delta <<$ L) and, since $\delta <<$ R, the microwave field acts on the electron only for a brief time when it is in the skin depth region.

The conditions $\delta << R << L$ suggest (Azbel' and Kaner, 1957) that resonances might best be observed with the field accurately aligned parallel to the surface of a very flat sample. Then, even electrons with drift velocities, $\vec{v}_D \sim \vec{v}_F$, along \vec{H} must return each cyclotron period to the skin depth layer, and there can be no complications due to electrons drifting deeper into, or completely out of, the skin depth region. In the following discussion, we shall initially assume the

above "Azbel'-Kaner" geometry. Later it will be shown that, even though a variety of resonances may occur when \vec{H} is tipped out of the sample surface, accurate field parallelism and correspondingly flat samples are generally required if the effective mass is to be measured accurately. Also, to simplify the discussion, all electrons on the Fermi surface will initially be assumed to have a common effective mass (as for an ellipsoidal Fermi surface) and then later the effects of mass spread will be discussed. The following discussion also holds for hole orbits.

In the AKCR geometry, the spiralling electrons return, on the average, $\omega_{c}\tau/2\pi$ times to the skin depth region, and a well developed resonance occurs when these electrons and the microwave fields are in phase for successive revolutions. Such synchronism occurs when the microwave frequency, ω_{c} , is equal to, or an integral multiple of, the cyclotron frequency, ω_{c} , and the resonance condition is

$$\omega = N\omega_{\rm C} \quad (I-7)$$

Generally ω is held constant and the magnitude of \hat{H} varied so that

$$H_{N} = \frac{c \omega m_{C}}{Ne} = \frac{H_{C}}{N}$$
(I-8)

where the resonances at magnetic fields H_c and H_c/N (N>1) are called the fundamental and subharmonic resonances respectively. The fields H_N are periodic in reciprocal field.

In the following physical picture, due to Heine (1957), we consider the effects of a series of pulses experienced by an electron of mass m_c each time it passes through the skin depth layer. Because AKCR occurs under conditions of the anomalous skin effect, a resonant electron presently in the skin depth "remembers" the effects of previous excursions into the skin depth, but its memory of the nth previous pulse fades as $\exp(-nT_c/\tau) = \exp(-2\pi n/\omega_c \tau)$. Also, during each cyclotron period T_c , the electron gains or loses in phase by an amount $(2\pi\omega/\omega_c)$ on the oscillating microwave field. The effect of the previous excursions into the skin depth y a factor

$$F = \sum_{n=0}^{\infty} \exp\left[-\frac{2\pi i \omega n}{\omega_{c}} - \frac{2\pi n}{\omega_{c}\tau}\right]$$
$$= \left\{1 - \exp\left[-\frac{2\pi i \omega}{\omega_{c}} - \frac{2\pi}{\omega_{c}\tau}\right]\right\}^{-1}.$$
 (I-9)

Thus, since the surface impedance in the extreme anomalous skin effect limit varies as $\sigma^{-1/3}$, the surface impedance Z(H) is

$$Z(H) = \alpha Z(0) \left\{ 1 - \exp\left(-\frac{2\pi i \omega}{\omega_{c}} - \frac{2\pi}{\omega_{c}\tau}\right) \right\}^{1/3}$$
 (I-10)

where Z(0) is the extreme anomalous surface impedance at zero field, and α is a constant of order unity. The resonances are due to oscillations in the resonance factor

F, which will be shown in section B.2 of this chapter to arise in more detailed theoretical calculations. The field derivative dR/dH of the real part of Z(H) is the usual experimental observable; computed variations of dR/dH versus H with changing wt are illustrated later in Figure I-1.

For general Fermi surfaces, m and $\omega_{\rm c}$ are functions of $k_{_{\rm H}}$ for fixed $\ddot{\text{H}},$ and contributions to the resonance signal from different parallel slices of the Fermi surface, each normal to $\dot{\vec{H}}$, are not necessarily in phase. As a result, many of the contributions cancel, and the dominant contribution to the resonance signal comes from regions where $m_{c}(k_{H})$ is stationary (either a maximum or a minimum) as a function of k_H. However, as for the magnetoacoustic effect (Pippard, 1960), resonances due to Fermi surface sections for which $m_{c}(k_{\mu})$ is not strictly extremal may be observed by AKCR. This is because the number of observable subharmonics (or the phase $2\pi\omega/\omega_{c}$) is so small that orbits which change slowly in area and mass before an abrupt cutoff may still contribute constructively in a Fresnel-type integral (or Cornu spiral) representing the summation of the resonance effects over ku. The strength of such a resonance, and of resonances corresponding to mass extrema, depends on the Fermi surface curvature which governs the width of the band of orbits which can contribute in phase to the resonance signal.

Under anomalous skin effect conditions, only electrons travelling within an angle of order δ/L of the sample surface contribute effectively to the conductivity. The energy picked up by a resonant electron (and thus the resonance amplitude) therefore depends on the component of the microwave polarization, \dot{j}_{rf} , parallel to the velocity of the resonating electron as it passes through the skin depth. Resonances for electrons on stationary orbits therefore will be largest for $\mathbf{j}_{rf} \mathbf{k}$ and much smaller for $\mathbf{j}_{rf} \mathbf{k}$. Similarly, electrons near limiting points (where $\vec{v}_{D} \,|\, |\vec{H})$ will yield strong resonances for $\vec{j}_{rf} | | \vec{H}$ and disappear for $\vec{j}_{rf} | \vec{H}$. Resonance strengths will, however, be comparable for both polarizations for nearly ellipsoidal Fermi surface sections where electrons with wide ranges of drift velocities contribute in phase to the resonance. These examples demonstrate that, by comparing signal strengths for both polarizations, one may gain information to aid in determining the Fermi surface orbits responsible for a given resonance signal.

The resonance strength also depends on the time an electron spends in interaction with the microwave r.f. field in the skin depth region. Thus, a very elliptical orbit, for example, will contribute much more strongly to the resonance if the electrons pass through the skin depth while on the flat portion of their real space orbits than if they do so while on the sharp end of the ellipse. The latter case may be said to have an unfavourable "geometrical factor" (Spong and Kip, 1965).

The line shapes for AKCR vary spectacularly when the field \vec{H} is tipped an angle α out of the sample surface. For $\alpha \leq 1^{\circ}$, the resonance lines (dR/dH) are found (Koch, Stradling, and Kip 1964, for example) to broaden, shift to higher or lower fields, or even to split in two, with increasing α . Except near $\alpha = 0$ (where the tip dependence lessens due to surface irregularities) the observed shifts are linear in α (with rates of 0 to 40% per degree), and generally symmetrical for positive and negative tip angles. Usually the higher harmonics are less affected than the lower harmonics. For $\alpha \ge 1^{\circ}$, the resonances are generally inverted (Koch, Stradling, and Kip, 1964) but sometimes double in number (Spong and Kip, 1965). Large, apparently normal (neither inverted nor doubled) resonances associated with cylindrical $(\partial^2 A / \partial k_{\mu}^2 = 0, v_{p} = 0)$ Fermi surface sections have also been reported (Koch, Stradling, and Kip, 1964) at very large tip angles.

Aside from being interesting in their own right, the small angle tipping effects, by their symmetry for positive and negative tip angles, provide very sensitive indicators with which to align \overline{H} parallel to the sample surface accurately, and to set limits on the surface flatness. Such accurate alignment (and samples flat to the order of several minutes of arc) are necessary in order to measure accurate AKCR masses. This is because generally the lower harmonics are found to be more sensitive to tipping effects than the higher harmonics, thus causing errors in determining the mass corresponding to a resonance series which may depend sensitively on the tip angle (or surface flatness) and on the number of observable subharmonics.

The low angle ($\alpha < 1^{\circ}$) field tipping effects all appear (Smith, 1967) to be due to the Doppler shifting mechanism proposed by Koch, Stradling, and Kip (1964). This Doppler shifting arises because an electron, which on successive traversals of the skin depth appears at different depths, sees an effectively Doppler shifted external frequency, because the r.f. field changes both in amplitude and phase with depth into the sample. Since the r.f. field E(z), at depth z, in a good conductor is approximately

 $E(z) = E(0) \exp(-(1+i) z/\delta),$

the phase change $\Delta \phi_c$, per cyclotron period, for an electron drifting with velocity $v_D \sin \alpha$ into the sample, is

 $\Delta \phi_{c} = - \frac{i T_{c} v_{D} \sin \alpha}{\delta} = - \frac{2 \pi i v_{D} \sin \alpha}{\delta \omega_{c}} .$

When this phase change is included in the discussion leading to Equation I-9, it is seen that the effectively Doppler shifted frequency, ω_{D} , observed by the drifting electron is

$$\omega_{\rm D} = \omega + \frac{v_{\rm D} \sin \alpha}{\delta} \tag{I-11}$$

where ω is the frequency of the external field. From Equation I-11, the resonance condition for electrons with drift velocity v_{p} is seen to change by an amount

$$\frac{\Delta H_{N}}{H_{M}} = \frac{\Delta \omega}{\omega} = \frac{v_{D} \sin \alpha}{\omega \delta} \sim \frac{v_{D} \alpha}{\omega \delta}$$
(I-12)

which is linear in α , as observed. Here δ is the effective skin depth for the anomalous skin effect. It is expected that the shift should deviate from linearity in α when limiting point electrons can drift out of the skin depth, thus causing increases in the effective skin depth. For free electrons the limiting angle is $\alpha < \delta/L \sim 10^{-3}$ radians, assuming $\omega_{c}\tau = 2\pi$ and $v_{D} = v_{F}$ (then $T_{c}v_{D}\sin\alpha = \delta$).

Equation I-12 is directly applicable to limiting point resonances for which v_D is essentially constant for all the contributing electrons. Because v_D is large, the normal AKCR resonances shift rapidly with α and then quickly disappear as the electrons begin to drift out of the skin depth. Koch, Stradling, and Kip (1964) report one such resonance which shifted ~40% per degree (an extrapolated value) for the second harmonic and which could only be observed for tip angles less than 15 minutes of arc. Clearly very flat samples are required for the observation of such resonances.

For general Fermi surfaces, the conductivity is a weighted average of contributions from orbits having different values of v_{D} ; without working out the conductivity in detail it is difficult to determine which orbits will dominate. Koch, Stradling, and Kip (1964) have shown qualitatively that the simple Doppler shifting ideas above can account for peak splitting for Fermi surfaces which have non-stationary mass extrema on either side of stationary central orbits. The simple model cannot, however, readily account for the relative insensitivity of the higher harmonics to small angle tipping or the shifts to higher or lower fields which are unaccompanied by splitting. Smith (1967) has shown, however, that, when incorporated in detailed calculations of dR/dH, the concept of Doppler shifted frequencies can qualitatively explain peak splitting even for the free electron model and peak shifts both to higher and lower fields for more general surfaces for which $\textbf{m}_{_{\mathbf{C}}}$ varies along $\textbf{k}_{_{_{\mathbf{H}}}}$ for fixed $\vec{\textbf{H}}.$ His calculation and the effects arising at larger tip angles will be discussed in the next section.

C.2. Azbel'-Kaner and Related Theories

The theories for Azbel'-Kaner cyclotron resonance involve solving for the microwave surface impedance of a metal, under anomalous skin effect conditions, when a steady

magnetic field is applied parallel to the surface of a very flat sample. Azbel' and Kaner (1958) calculated the form of the field dependent changes in the anomalous surface impedance directly; their calculation has formed the basis of line shape calculations by Chambers (1965), and the effects of tipping \vec{H} out of the sample surface by Smith (1967). Chambers (1965) made a significant contribution in recognizing that the resonant electrons do not dominate the overall conductivity, but rather only perturb the nonresonant background due to all the electrons. Alternately, Heine (1957), with modifications by Chambers (1965) has achieved essentially the same results by showing that the resonant electrons contribute a resonant term, like the F in Equation I-9, inside the integral expression for the effective conductivity resulting from Pippard's "ineffectiveness concept" (ie. only electrons with velocities within an angle $\pm\beta\delta/2L$, where $\beta \sim 1$, of the sample surface contribute effectively to the conductivity). Many of the details have recently been discussed by Chambers (1969). Here, the Azbel'-Kaner theory will be summarized together with the Chambers non-resonant contribution, and reference will be made to other pertinent studies.

Azbel' and Kaner (1958) solved for the surface impedance tensor, Z_{uv} , which relates the r.f. field E(0)

at the surface (z = 0) of the metal and the total r.f. current J by

$$E_{\mu}(0) = \sum_{\nu=1}^{2} Z_{\mu\nu} J_{\mu}$$

where $Z_{uv} = R_{uv} + i X_{uv}$

and
$$\mu, \nu = x, y$$
.

They formulated the problem in terms of Maxwell's equations and the Boltzman transport equation, imposing diffuse reflection of the carriers at the surface as a boundary condition. The system of equations solved was

$$\vec{J} = -\frac{2e}{h^3} \int \vec{v} f_1 dp_x dp_y dp_z ,$$

$$i\omega f_1 + v_z \frac{\partial f_1}{\partial z} - \frac{e}{c} (\vec{v} \times \vec{H}) \cdot \frac{\partial f_1}{\partial p} + \left(\frac{\partial f_1}{\partial t} \right)_{coll.} = e\vec{E} \cdot \vec{v} \frac{\partial f_0}{\partial \epsilon}$$

$$f_0(\epsilon) = \frac{1}{1 + exp\{(\epsilon - E_F)/kT\}}$$

and

where $f_1(z=0, v_z>0) = 0$ is the condition for diffuse reflection at the surface. Here, \vec{v} , \vec{p} , and ε are the velocity, quasi-momentum, and energy of the electron, and $(\partial f_1/\partial t)_{coll}$. is the collision integral for collisions of the electron with phonons, electrons, and lattice defects. Under anomalous skin effect conditions, they show that a relaxation time τ may always be introduced. Hence, $\left(\begin{array}{c} \partial f_1\\ \partial t\end{array}\right)_{coll} = \frac{f_1}{\tau}$.

In the resonance region, Azbel' and Kaner (1958) found that $Z_{\mu\nu}$ depended on terms like

$$A_{\mu\nu} = \frac{8e^2}{3h^3} \sum_{0} \oint_{0}^{2\pi} \frac{n_{\mu}n_{\nu}}{K(\phi)} \frac{d\phi}{1 - \exp\left(-\frac{2\pi i\omega}{\omega_{c}(\phi)} - \frac{2\pi}{\omega_{c}(\phi)\tau}\right)} \quad (I-13)$$

where the integration is over the equatorial contour of the Fermi surface for which the electron velocity \vec{v} is parallel to the sample surface. The position of any point on this contour is specified by the angle ϕ between \vec{v} and \vec{H} , $n_{11} = \cos \phi$, and $n_{12} = \sin \phi$. The integration is only over the equatorial contour because, under anomalous skin effect conditions, only electrons travelling nearly parallel to the surface contribute effectively to the surface conductivity; the number of such effective electrons is proportional to $1/K(\phi)$, where $K(\phi)$ is the Gaussian curvature at ϕ in the equatorial plane. The effect of microwave polarization is included by the terms $\boldsymbol{n}_{_{\boldsymbol{U}}}$ and $\boldsymbol{n}_{_{\boldsymbol{V}}}\text{,}$ and the effect of mass spread by the dependence of $\omega_{C}(\phi)$ on ϕ . The summation is over disconnected parts of the Fermi surface, and $\mathtt{A}_{_{\scriptstyle\rm UV}}$ depends intimately on the shape of the Fermi surface.

By appropriate orientation of the x-y axes in the sample surface, $A_{\mu\nu}$ may be diagonalized (Azbel' and Kaner, 1958). Then

 $Z_{ii} = R_{ii} + i X_{ii} \propto \exp(i\pi/3) (A_{ii})^{-1/3}$, (I-14) if some small, essentially constant, magnetoresistance terms are neglected. For an ellipsoidal Fermi surface,

 $m_{c}(\phi)$ is constant and

$$Z(H)_{XX,YY} = Z(0)_{XX,YY} \left[1 - \exp \left(-\frac{2\pi i \omega}{\omega_{c}} - \frac{2\pi}{\omega_{c}\tau} \right) \right]^{+1/3}$$
(I-15)

where $Z(0)_{xx,yy}$ are the zero field anomalous surface impedances for microwave polarizations along x or y and are complex, each containing the factor $\exp(+i\pi/3)$. Note that Equations I-10 and I-15 are essentially equivalent.

Plots of the field derivative, dR/dH, of the real part of the surface impedance, as computed by Kip, Langenberg, and Moore (1961), using Equation I-15 are shown in Figure I-1.

Chambers (1965) has shown that there may be nonresonant contributions to Equation I-13 when the Fermi surface is not a simple, closed convex surface. His argument is illustrated in Figure I-2, where points 1, 2, 3 and 4 are all points in the equatorial plane. Point 1 yields a resonant contribution since with 1 in the skin depth, electrons on that orbit can complete their orbits without striking the surface, and hence resonate. Points 2, 3, and 4 however, give non-resonant contributions since, if the orbit is displaced so that one of these points is in the

Figure I-1.

Computed variations of dR/dH versus H when all electrons have common ω_c (after Kip et al, 1961). These traces were computed from Equation I-15 for fixed ω ; H_c denotes the field at which the fundamental is expected to occur. As $\omega \tau$ decreases, fewer subharmonics are observed, the resonances are less sharp, the overall amplitudes decrease approximately as $(\omega \tau)^2$, and the resonances shift to higher fields.





skin depth, electrons on that orbit collide with, and are scattered by, the sample surface within the first cyclotron period, T_c . Chambers (1965) also computed AKCR line shapes (for dR/dH) near resonance, for $\omega_c \tau > 50$, from equations, like Equations I-14 and I-13 (diagonalized), but including nonresonant contributions. These line shapes depended on whether m_c was a maximum, minimum, or relatively constant near the mass extremum. The Chambers line shapes compare well with measured line shapes for high purity ($\omega_{c}\tau > 50$) metals (Chambers, 1965, Moore, 1966, and Goy and Weisbuch, 1969). These results show that, even at resonance, the non-resonant background due to other parts of the Fermi surface is significant when relatively few electrons participate in the resonance.

Chambers' (1965) calculations also show that, when relatively few electrons participate in a resonance and $\omega_{\rm c}\tau$ > 50, the fractional line width, $\delta H/H,$ of a resonance line is proportional to $(\omega\tau)^{-1}$ and the shape, expressed in units of $\mu = \omega \tau \delta H/H$, remains unchanged. Physically this means that the non-resonant background is sufficiently large that the skin depth δ does not change appreciably at resonance and the line broadening is due only to wt. Moore (1966) has found orbits in gallium which satisfy these conditions, and used these orbits to measure the relaxation time t and its temperature dependence. In relatively simple metals, however, a significant number of electrons may participate in a given resonance, the resonance line is broadened due to decreases in the skin depth near resonance, and there is no simple relationship between the line width and τ . For such conditions, or when $\omega_{_{\rm C}}\tau$ < 50, τ may be estimated using the Haussler-Welles method (Haussler and Welles, 1966) which will now be discussed.

The Haussler-Welles method is based on Equation I-15 for which the amplitudes, $B_{\rm N}$, of the peaks in dR/dH vary as

$$B_{N} \propto N^{2} \exp(-2\pi N/\omega\tau) \qquad (I-16)$$

provided $2\pi N > \omega \tau$. Hence, $\omega \tau$ may be estimated from the slopes of plots of $\ln (B_N/N^2)$ versus N. The result (Haussler and Welles, 1966) is an effective relaxation time, τ_{eff} ,

$$\frac{1}{\tau_{eff}} = \frac{1}{\tau} + \frac{1}{\tau_{m}} + \frac{1}{\tau_{s,H}} .$$
 (I-17)

Here τ is the true relaxation time, and τ_m and $\tau_{s,H}$ are temperature independent contributions due, for τ_m , to mass spread about the mass extremal studied, and, for $\tau_{s,H}$, to surface curvature of the sample or poor alignment of the field, \vec{H} , parallel to the sample surface. The method is strictly valid only for ellipsoidal Fermi surfaces for which Equation I-15 applies; for more general Fermi surfaces it appears that this method is applicable only when the $\ln(B_N/N^2)$ versus N plots are found to be very nearly linear. Then, since τ_m is unknown, the method only sets a lower limit on τ and is primarily useful for measuring the temperature dependence of the relaxation time.

Smith (1967) has investigated field tipping effects theoretically by generalizing the Azbel'-Kaner treatment to include the Doppler shifted frequency ω_{p} rather than ω . He assumes the equivalent of $A_{\mu\nu}$ to be diagonal, and ignores both the small, almost constant, magnetoresistance terms from the Azbel'-Kaner theory, and the Chambers non-resonant contributions. His results rival in their variety the effects observed experimentally but are in general qualitative agreement with them. Some of the results of Smith's (1967) calculations, and of others', are as follows:

First, consider the small angle tipping regime (Lsin $\alpha < \delta$) where, even for the free electron model, no electrons can drift out of the skin depth, δ , so that the effective skin depth remains virtually unchanged and the effects of Doppler shifting predominate. Here Smith (1967) found that, even for the free electron model and for Fermi surface sections with little mass spread, the resonance lines split into two asymmetrical parts, for both parallel and perpendicular microwave polarization. However, for Fermi surface sections with considerable mass spread, the resonance lines broadened or shifted to higher or lower fields, according to whether the mass extremum was a maximum or minimum. Except near $\alpha = 0$, these shifts were almost linear for mass maxima and less so for mass minima; the sensitivity to tipping decreased as the mass spread increased. Smith (1967) concluded that the line shape modifications, due to field tipping for the extremal mass cases arose mainly from changes in the electric field distribution and not from a shift in

the resonant orbit.

For larger tip angles (sin $\alpha > \delta/L$), electrons near limiting points on the free electron model spiral out of the zero tip skin depth before being scattered, causing an increase in the effective skin depth. Smith (1967) shows that for $\dot{J}_{rf} || \dot{H}$, this increase in the effective skin depth causes the resonances to invert, with the inverted peaks lying close to the positions of the zero tip normal AKCR resonance. Similar inversions have been reported for $\dot{J}_{rf} \perp \dot{H}$ (Koch, Stradling, and Kip, 1964, and Figure III-16 of this thesis). For $\dot{J}_{rf} \perp \dot{H}$, the width of the band of contributing orbits decreases as α increases and, consequently, the resonance weakens and normally disappears. Also, for $\dot{J}_{rf} \perp \dot{H}$, the bands contributing to resonances from mass extrema become narrower and the resonance disappears as α increases.

Resonances may still be observed at large tip angles for $\vec{J}_{rf} ||\vec{R}$, however, via the "current sheet" mechanism of Grimes et al (1963) and Spong and Kip (1965). This mechanism arises from electrons near Fermi surface limiting points which have the same cyclotron masses, essentially the same drift velocities, and spiral along \vec{H} on very open helices of constant pitch but with varying radii. Such electrons are excited in the skin depth, then drift out of the skin depth, but are refocused each cyclotron period at
depths, $d_n = nT_c v_p \sin \alpha$ (where n is an integer), into the sample. At these depths d_n, they are again moving parallel to the sample surface and hence produce sheets of r.f. currents deep inside the sample. Electrons from the opposite side of the Fermi surface drift through these current sheets toward the sample surface and, if $2\omega = m\omega_{c}$ where m is an integer, interact in phase with all of these current sheets. Grimes et al (1963) and more recently Smith (1967) show that these interactions alter the surface impedance, and lead to inverted resonance series having twice the usual number of resonance peaks (since $2\omega = m\omega_{c}$). The calculation of Smith (1967) is significant in that it shows (in contrast to the situation for aluminum (Grimes et al, 1963, and Spong and Kip, 1965) where $ec{v}_{_{\mathrm{D}}}$ is essentially constant for many holes on a spherical cap) that such doubling can also be expected to occur for the free electron model where the limiting point electrons dominate even though the current sheets might be expected to be badly smeared due to a continuous distribution of drift velocities near the limiting point.

Smith (1967) also shows that, at very large tip angles (\sim 10° to 80°), the condition for stationary phase, which determines the band of contributing electrons, is $\partial^2 A/\partial k_{\rm H}^2 = 0$. This, as was suggested by Koch, Stradling, and Kip (1964), explains the observation of apparently normal (not inverted or doubled) resonances corresponding to nearly cylindrical Fermi surface sections at very large tip angles (up to 70°). For such sections, a large number of quasi-stationary electrons return many times to the skin depth region and contribute in phase to the resonance signal.

D. De Haas-van Alphen Theory

For free electrons of mass, m, in a field, \vec{H} , along the z direction, the Hamiltonian is

$$H = \frac{1}{2m} \left(\vec{p} - \frac{e\vec{A}}{c} \right)^{2} = \frac{1}{2m} \left(p_{x}^{2} + p_{z}^{2} + (p_{y} - m\omega_{c}x)^{2} \right) (I-18)$$

where \vec{A} is the vector potential $\vec{A}(0, Hx, 0)$ whose curl gives the field \vec{H} in the z direction. If $x_0 = p_y / m\omega_c$, Equation I-18 can be written as

$$H = \frac{1}{2m} \left[p_{x}^{2} + m^{2} \omega_{c}^{2} (x - x_{0})^{2} \right] + \frac{\tilde{n}^{2} k_{z}^{2}}{2m}$$
(I-19)

which is the Hamiltonian for a harmonic oscillator of frequency ω_c centred at x_0 , plus an energy term varying with k_z along H. The eigenvalues are therefore $\frac{\pi^2 k}{2}$

$$E_{n} = (n + \frac{1}{2}) \tilde{n} \omega_{c} + \frac{n^{2} k_{z}^{2}}{2m}$$
 (I-20)

where n is an integer. In physical terms, the field has split the zero field parabolic band structure, $E(k)=n^2k^2/2m$, into a series of oscillator levels, called Landau levels. By box normalization (with x_0 remaining inside the box), the degeneracy of these levels is found (Ziman, 1964) to be $\frac{eH}{4\pi^2 \hbar c} \Delta k_z$, for states of each spin, per Landau level, for a slice of thickness Δk_z of \vec{k} space perpendicular to \vec{H} , when the crystal has unit volume.

Orbits differing in energy by $\hbar\omega_c$ are separated in area by $\Delta A = (dA/dE)\hbar\omega_c$ so that, via Equation I-5, the energy quantization in Equation I-20 shows that the cross sectional areas enclosed by the \vec{k} space orbits are quantized in units of $2\pi eH/\hbar c$. More generally, the \vec{k} space quantization may be obtained from the Bohr-Sommerfeld quantization rule (Onsager, 1952)

$$\oint \vec{p} \cdot d\vec{r} = (n + \gamma) 2\pi \vec{n}, \quad (I-21)$$

where $\vec{p} = \vec{n}\vec{k} + \frac{e\vec{A}}{c}$ is the conjugate momentum, γ is a phase factor ($\frac{1}{2}$ for free electrons) and \vec{r} and \vec{k} specify the position of the electron in real and reciprocal space respectively. By Stokes theorem,

$$\frac{e}{c} \oint A \cdot dr = \frac{e}{c} \int \nabla \times \vec{A} \cdot d\vec{a} = \frac{e}{c} \Phi$$

where Φ is the product of H and the projected area of the real space orbit on a plane normal to \vec{H} . Also, from Equation I-1 (integrated),

$$\frac{e}{c} \oint \vec{n} \vec{k} \cdot d\vec{r} = -\frac{e}{c} \oint \vec{r} \times \vec{H} \cdot d\vec{r} = -\frac{eH}{c} \oint \vec{r} \cdot d\vec{r} = \frac{2e}{c} \Phi.$$

Hence

and

$$\Phi = \frac{2\pi\hbar c}{e} (n + \gamma)$$

$$A_{N} = \frac{2\pi eH}{\hbar c} (n + \gamma) \qquad (I-22)$$

since the real and reciprocal space orbit dimensions are scaled by a factor eH/hc.

Equation I-22 is independent of the component of \vec{k} along \vec{H} . Thus the effect of the applied field is to constrain the allowed states in \vec{k} space to lie on a series of tubes, called Landau levels, whose cross sectional areas are constant along \vec{H} , but whose sections may vary. Generally these tubes need not be parallel to \vec{H} (Gold, 1968, for example).

For a given slice of momentum space normal to \vec{H} , the Landau levels expand with increasing \vec{H} , and states which attain energies greater than the Fermi energy, E_F , are depopulated and redistributed to lower levels whose degeneracies increase with field. This redistribution gives rise to changes in the free energy which are periodic in reciprocal magnetic field. The contributions to the free energy from different slices vary in phase, however, and are additive only for regions of the Fermi surface for which the Fermi surface cross sectional areas normal to \vec{H} are stationary as a function of the component k_H of \vec{k} along \vec{H} . Experimental observables, like the dHvA effect (oscillations in torque or magnetization which arise from the oscillations in the free energy) therefore depend only on the extremal areas of the Fermi surface.

The free energy for a system of electrons in sharp Landau levels at finite temperatures has been evaluated by Lifshitz and Kosevich (1956). For unit volume, the free energy, FE, for a Fermi-Dirac assembly is given by

$$FE = NE_{F} - kT \sum_{i} ln(l + exp\{(E_{F} - E_{i})/kT\})$$
(I-23)
where E_F is a fixed Fermi energy and N is the number of
electrons. The energy E_i for state i is expressed as

 $E_{i} = E(n + \gamma, k_{z}) = (n + \gamma)\hbar\omega_{c} + f(k_{z}) \qquad (I-24)$ where n is the orbital quantum number specifying a particular Landau level. This is a generalization of Equation I-20. When expressed as a sum over Landau levels and an integration over k_{z} , Equation II-23 becomes

$$FE = NE_{F} - kT \int_{0}^{\infty} \left(\frac{eH}{2\pi^{2}\pi c} \sum_{n=0}^{\infty} ln(l+exp\{(E_{F}-E(n+\gamma,k_{z})/kT\})\} dk_{z} \right) dk_{z}$$
(I-25)

where the degeneracy of the Landau levels has been introduced and spin effects are ignored. Ziman (1964) shows that the integrand in Equation I-25 may be evaluated for each of its fourier components separately using the Poisson summation formula. His result, for oscillatory terms only, is

$$FE_{osc} = 2kT \sum_{s=1}^{\infty} \int_{-\infty}^{\infty} \frac{eH(-1)^{s}}{4\pi^{2}\pi cs} \frac{\cos(sc\hbar (E_{F},k_{z})/eH)}{\sinh(2\pi^{2}skT/\hbar\omega_{c}(E_{F},k_{z}))} dk_{z} (I-26)$$

where the fourier components are represented by s and $A(E_F,k_z)$ is the cross sectional area of the Fermi surface at the slice k_z . For ordinary magnetic fields, the phase $c\hbar A(E_F,k_z)/eH$ is very large and the cosine term in Equation I-26 oscillates rapidly with changing k_z . Thus, since Equation I-26 is a Fresnel-type integral, the major contributions to the oscillatory part of the free energy arise from Fermi surface cross sectional extrema $(\partial A(E_F,k_z)/\partial k_z = 0)$ where the phase is stationary. Then, expanding about an extremal area, A_0 , at $k_z = k_0$, the cross sectional area may be represented by

$$A(E_{F},k_{z}) = A_{0} \pm \frac{1}{2}A_{0}"(k_{z}' - k_{0})^{2}$$

and Equation II-26 yields

$$FE_{osc} = 2kT \sum_{s=1}^{\infty} (-1)^{s} \left(\frac{eH}{2\pi sch}\right)^{\frac{3}{2}} (A_{0}^{"})^{-\frac{1}{2}} \frac{\cos\left(\frac{schA_{0}}{eH} \pm \frac{\pi}{4}\right)}{\sinh\left(\frac{2\pi^{2}skT}{\hbar\omega_{c}}\right)} (I-27)$$

where A_0 " is the local curvature of the Fermi surface about k_0 and the plus and minus signs refer to minimum and maximum cross sectional areas respectively. The high harmonics (s > 1) are normally rapidly damped by the sinh term, so that generally the fundamental (s = 1) term dominates. Then the oscillations in free energy with changing field H are periodic in reciprocal magnetic field and have a dHvA frequency, F,

$$F = \frac{chA_0}{2\pi e}$$
 (I-28)

Dingle (1952b) and Williamson et al (1964) have shown that the effects of broadening of the Landau levels

(cont. T)

due to scattering may be accounted for by multiplying each term in Equation I-27 by $\exp(-2\pi^2 \mathrm{skX}_D/\hbar\omega_c)$, where X_D is the Dingle temperature, $X_D = \frac{\hbar}{2\pi k} \left\langle \frac{1}{\tau} \right\rangle$, and $\left\langle \frac{1}{\tau} \right\rangle$ is the reciprocal of the relaxation time, τ , averaged around the orbit.

The effect of spin is to split the Landau levels into two levels, equally spaced above and below the unsplit level. This can be incorporated (Dingle, 1952a, and Cohen and Blount, 1960) into Equation I-27 by multiplying each term by $\cos(\pi \text{sgm}_c/\text{m}_e)$, where g is the effective g-factor for the conduction electrons, and m_e is the free electron mass.

A complete expression for the oscillatory part of the free energy, per unit volume, is

$$FE_{OSC} = 2kT \left[\frac{eH}{2\pi ch} \right]^{\frac{3}{2}} \left[\frac{\partial^2 A}{\partial k_z^2} \right]^{-\frac{1}{2}} \times \left[\frac{(-1)^S}{(s)^{3/2}} \frac{exp(-2\pi^2 skX_D/\hbar\omega_c)}{\sinh(2\pi^2 skT/\hbar\omega_c)} \right]$$
(I-29)
$$\cos\left(\frac{\pi sgm_c}{2m_e} \right) \cos\left(\frac{2\pi sF}{H} \pm \frac{\pi}{4} \right)$$

where the curvature $\partial^2 A/\partial k_z^2$ and m_c are evaluated at the Fermi surface extremal A_0 .

The oscillatory parts of the magnetization, M_{osc} , or of the torque, T_{osc} , on the sample may be found by differentiating Equation I-29 with respect to H or θ from the expressions

$$M_{osc} = -\frac{\partial F_{osc}}{\partial H}|_{H,V}$$
, $T_{osc} = \frac{\partial F_{osc}}{\partial \theta}|_{H}$. (I-30)

In either case the phase of the oscillatory term is not changed. An expression for M_{OSC} is given in Equation V-2. The amplitude of T_{OSC} contains a factor $\partial F/\partial \theta$; hence the torque becomes zero when the dHvA frequency is stationary with respect to rotations θ .

Unlike the AKCR or magnetoacoustic attenuation effect phases, the phase $2\pi F/H$ for the dHvA effect is large $(\sim 10^2 \text{ to } 10^4)$. This means that only sections of the Fermi surface normal to $\dot{\vec{H}}$ which are very close to the extremal section contribute appreciably to the oscillations and it is a well defined extremal which is measured. Also when the field \vec{H} is rotated, the dHvA frequency F(θ) changes and the torque or magnetization, at constant \vec{H} , oscillate once per unit change in phase. Small changes in area can therefore be measured to accuracies of the order of 1 part in 10⁴, for example, if the phase is 10⁴. By studying the temperature and field dependence of the dHvA amplitudes, both the effective mass and the relaxation time may be determined. Measurements of τ , however, may be complicated by field dependencies due to other causes such as crystal imperfection or field inhomogeneity (Shoenberg, 1969) or magnetic breakdown.

Finally, the dHvA effect may be regarded as arising from changes in the effective density of states at the Fermi surface, as the field \hat{H} changes. These changes give rise

to similar oscillatory effects in the electrical and thermal conductivities and in the microwave surface impedance, for example. The dHvA frequencies of all such oscillations are related to extremal cross sectional areas of the Fermi surface via Equation I-28.

CHAPTER II

THE FERMI SURFACE OF MERCURY

Mercury is a divalent metal which crystallizes at approximately 223°K into a rhombohedral lattice at atmospheric pressure (Barrett, 1957) with one atom per unit cell. The Brillouin zone (Jones, 1960 and Keeton and Loucks, 1966) and certain lattice parameters for the rhombohedral phase of mercury are shown in Figure II-1. We adopt the notation that (lmn) represents a direction in reciprocal space, or a plane perpendicular to the (lmn) direction. Similarly, real space directions, and planes normal to them, are represented by [lmn]. Sets of equivalent directions will be represented by {lmn} in reciprocal space and by <lmn> in real space. It should be noted that the three orthogonal real space directions [111], [112], and [110], specify the same directions as the corresponding reciprocal lattice directions (111), $11\overline{2}$), and (110). The trigonal-bisectrix {110} planes which bisect the Brillouin zone along the line X-U-L-U-T are the mirror planes for mercury, and there is threefold symmetry about the (111) direction, and twofold symmetry about the (110) direction.



Symbol	Value	Definition
a ₀ a	2.9863 Å	Real space lattice vector at 5°K
a	70°44.6'	Real space rhombohedral angle at 5°K
goa	2.3002 $Å^{-1}$	Reciprocal space lattice vector at 5°K
βä	104°21.7'	Reciprocal space rhombohedral angle at 5°K
k _F b	1.3707 Å-1	Free electron Fermi radius
TU b	0.6254 Å ⁻¹	
xu b	0.6345 Å^{-1}	
LU b	1.0337 Å ⁻¹	
XK b	0.3611 Å ⁻¹	
TW b	0.7220 Å^{-1}	

^a Source C.S. Barrett, Acta Cryst. <u>10</u>, 58 (1957).

^b Bogle et al, Phys. Rev. <u>177</u>, 1122 (1969).

Figure II-1. The Brillouin zone for mercury, and certain lattice parameters.

Both theory and experiment (Keeton and Loucks, 1966, Brandt and Rayne, 1966, Datars and Dixon, 1967, Dishman and Rayne, 1968, and Bogle et al, 1969) predict that, like the free-electron sphere which has a volume equal to that of the Brillouin zone, the Fermi surface of mercury cuts through the L faces to yield lens-shaped second-zone electron discs centred at L on the {100} faces of the Brillouin zone, and a multiply-connected first-zone hole surface. Unlike the free-electron sphere, however, the Fermi surface also contacts the X and T faces to produce openings, centred at X and T in the hole surface, as illustrated in Figure II-2. In what follows, the pair of arms formed on either side of the breakthrough region in the X face, in Figure II-2, will be called the " β arms", and the two breakthrough regions at X and T will be called the X and T face openings.

Since mercury has been shown to be a compensated metal by magnetoresistance (Datars and Dixon, 1967), the volumes of the hole and electron surfaces are equal.

Figure II-3 shows that, while the theoretical models for the mercury Fermi surface are topologically similar, they differ significantly in detail, particularly for the sizes and shapes of the breakthrough regions about L, X, and T. With the exception of the dimension k_{in}^{XU} in this figure, there have been no certain direct measurements of the sizes of the X and T face openings, or of the outside of the β arm near K. Some closed cyclotron orbits predicted by the hole Figure II-2.

A nearly-free electron model (after Dixon and Datars, 1968) of the firstzone hole Fermi surface of mercury with modifications to illustrate breakthrough at X and T. The outside of the solid lines at X and T represent the breakthrough region sizes predicted by Keeton and Loucks (1966), while the dotted lines represent those predicted by the MAG V model of Dishman and Rayne (1968); there is no breakthrough at X and T in the nearly-free electron model. Black areas on the model are regions where the surface is joined to other parts of the extended zone surface. In the actual hole surface, the sharp corners are expected to be more round, and the X face section is expected to be narrower, than in this model. For the nearly-free electron model, the cyclotron effective mass for an orbit is the total angle through which the electron moves divided by 2π , since the effective mass of an electron going around a sphere without Bragg reflections is unity (Harrison, 1960).



Figure II-3.

Relative sizes of the contact regions for several models of the first-zone hole Fermi surface (after Dishman and Rayne (a) and Bogle et al (b)). In the table, a dimension, k_{in}^{PQ} , represents the distance from P to the hole surface in the direction PQ, measured in $^{a-1}$. RAPW is a relativistic-augmented-plane-wave model, MAG V is an empirical 4-parameter model based on magnetoresistance data, and the remainder are plane wave pseudo-potential calculations. Detailed descriptions and the limitations of each of the models are given in sources (a) and (b); none of these models can simultaneously accurately fit the hole surface, the electron surface, and the magnetoresistance data.



Model	kin	k ^{XU} in	k ^{TU} in	k ^{TW} in	k _{in}
RAPW a	0.948	0.117	0.117†	0.117+	0.115
HAA ^a	0.831	0.354	0.382†	0.382†	0.347
MAG V a	0.860	0.260	0.198†	0.198+	0.209
8PW a	0.874	0.177	0.071+	0.071+	0.209
dHvA 3PW a	0.897	0.299	0.324+	0.324 +	0.182
4PW b	0.870	0.300	0.270	0.340	0.185
Experiment b	0.90	0.30			

^a For a summary of these models see J.M. Dishman and J.A. Rayne, Phys. Rev. <u>166</u>, 728 (1968).
^b See T.E. Bogle et al, Phys. Rev. <u>177</u>, 1122 (1969).

* Circular approximation.

surface model (Figure II-2) are shown in Figure II-4, and others are illustrated in Figure 8 of Bogle et al (1969); approximate ranges over which some of these orbits are expected to be observed are as follows:

The γ and τ orbits have previously been observed in de Haas-van Alphen (dHvA) experiments (Brandt and Rayne, 1966, and Moss, 1968), while γ , μ , and τ have been reported in Azbel'-Kaner cyclotron resonance (AKCR) experiments (Dixon and Datars, 1968), and more recently in magnetoacoustic attenuation experiments (Bogle et al, 1969). The minimum extremal cross sectional area (or effective mass) for τ will be at $\vec{H} \mid | (1\overline{1}0)$, for γ at \vec{H} 25±10 degrees from (112) toward (110) in the (110) plane, and for μ at \vec{H} near (112). In the (111) plane, the ranges of all three are limited by the sizes of the X and T face openings and for γ by the shape of the β arm near K, and are small. γ and μ have considerably larger ranges in the (110) plane; γ is cut off by the β arms beyond (112) and (110), and μ is cut off before (110) and (001), also by the β arms. τ has a wide angular range, centred at (110) in the (112) plane, and may be possible for a limited range near (001) for \hat{H} close to, but probably not in, the (110) plane. These ranges, particularly the latter, are very sensitive to the size of the X and T face openings, and to the shape of the β arm near K.





Figure II-4. Some possible hole orbits on the firstzone Fermi surface.

The κ , $\mu\gamma$, and γ_2 orbits shown in Figure II-4 are reported to have been observed for the first time in this study. These orbits, and the v orbit which has not yet been observed, are all expected to have limited ranges. The κ orbit cannot occur for field directions in either of the (110) or (112) planes, but is expected to occur for a very limited range 10 ± 5 degrees from $(11\overline{2})$ in the (111) plane. From this point, it is expected to have ranges of a few degrees toward (110), and 15±5 degrees toward (001) with its minimum a few degrees toward (001); the cutoffs depend sensitively on the regions of the β arms near K. Similarly, the v orbit is expected to have a very limited range near (110) which depends entirely on the β arm shape near K. Both these orbits are extremal, but are expected to be difficult to observe because of the relatively narrow bands of electrons which may contribute in phase to the dHvA or AKCR effects. The uv orbit is expected to have a range of perhaps ± 10 degrees centred about 15 degrees from $(11\overline{2})$ in the (111) plane, with its minimum about 30 degrees from this centre toward (110). This orbit cannot be observed in the $(1\overline{1}0)$ plane, but might be observed for a limited range approximately 20 ± 5 degrees from $(1\overline{1}0)$ in the $(11\overline{2})$ plane. The γ_2 orbit is expected to be observed only in, or very near, the $(1\overline{1}0)$ plane from a sharp cutoff near $(11\overline{2})$

toward (110); this cutoff depends sensitively on the size of the T face opening and on the β arm shape again near K. The orbit cannot exist at (112) if the T face opening is much larger than 0.2 Å⁻¹.

The hole surface also supports β orbits (Brandt and Rayne, 1966) shown in Figure II-4, around the β arms in the X face, and a non-extremal bow-tie orbit μ_2 (Bogle et al, 1969) around the X face on either side of the X face opening. There are also electron neck orbits, δ and ε_1 , about X and L (Bogle et al, 1969) respectively, and a similar orbit around T is expected; these orbits have small angular ranges for \vec{H} near (110), (100), and (111) respectively. A hexagonal-shaped hole orbit η about the outside of the T face for \vec{H} near (111), and an even larger hole orbit ε_2 which threads through T face openings in the (100) plane on the outside of the hole surface, have recently been observed by magnetoacoustic attentuation experiments (Bogle et al, 1969).

The electron lenses support α orbits which exist for all directions with maximum dHvA frequencies (and effective masses) for \overrightarrow{H} near {100} and minima near the {100} plane.

The first-zone Fermi surface supports three principal open orbits along the (100), (110), and (110) directions (Datars and Dixon, 1967). Signals due to these open orbits

have been used extensively in this investigation to orient the mercury crystals within the experimental apparatus.

When this study began, the AKCR results of Dixon and Datars (1968) for samples with curved surfaces, the magnetoresistance results of Datars and Dixon (1967), the dHvA results and the 3PW model of Brandt and Rayne (1966), and the Keeton and Loucks RAPW model (1966), were available, if not published. The magnetoresistance results and several models by Dishman and Rayne (1968) and the dHvA measurements of Moss (1968) have since appeared. Recently Bogle et al (1969) reported their magnetoacoustic attenuation experiments and 4PW model.

The present study has been concerned primarily with extending experimentally the results available by AKCR and dHvA for the mercury Fermi surface.

Much of this work has been made possible by the successful growth of large, very flat, crystals for cyclotron resonance studies. As shown in Chapter III, these samples permitted more accurate determinations of effective masses, the observation of several new orbits, and a study of the effects of tipping the magnetic field out of the sample surface. These flat samples were invaluable for the experiments reported in Chapter IV, in which variations of the cyclotron effective mass with temperature due to

changes in the electron-phonon mass enhancement were observed for the first time. These samples also permitted measurements of the temperature dependence of the relaxation time τ ; as reported in Chapter IV, the phonon part of τ^{-1} was found to vary faster than the expected T^3 dependence, due to Umklapp processes involving the α orbit studied.

The dHvA apparatus of Moss (1968) was improved and, with extensive use of fourier analysis of the dHvA spectra, previous results have been extended; the η and γ_2 orbits were observed for the first time by dHvA. These results are discussed in Chapter V.

The experimental results and conclusions are summarized in Chapter VI.

CHAPTER III

AZBEL'-KANER CYCLOTRON RESONANCE IN MERCURY

A. Experimental Apparatus

The cyclotron resonance, quantum oscillations, and open orbit data were taken using a Varian model V-4503 EPR microwave reflection spectrometer operated at approximately 35.0 GHz. The spectrometer was operated in its "Absorption Operate" mode generally with 40 Hz. magnetic field modulation and phase sensitive detection, and with the klystron frequency locked to that of a TE_{113} resonant cavity. The magnetic field modulation was parallel to the magnetic field, \vec{H} , applied to the sample, and the resulting signal was the field derivative dR/dH of the sample surface resistance.

Flat mercury single crystals formed the end wall of a horizontal, cylindrical TE₁₁₃ resonant cavity which was side coupled to the broad face of an 8 mm. rectangular wave guide suspended vertically between the pole pieces of a 0 to 20 k0e electromagnet. A tunable shorting plunger terminated the wave guide below the cavity, and allowed the cavity coupling and hence the spectrometer sensitivity to be adjusted during an experiment. The microwave currents were polarized vertically in the sample surface by a copper

stirrup positioned horizontally across the iris hole of the cavity. The polarization $(\vec{J} \mid \vec{H})$ was shown to be better than 90% by measuring the Electron Spin Resonance (ESR) signals corresponding to a uniform dispersion of diphenyl picryl hydrazyl (DPPH) on the surface of a copper test sample first with $\vec{H} \mid \vec{H}_{rf}$, and then with $\vec{H} \mid |\vec{H}_{rf}$, where \vec{H}_{rf} was the microwave \vec{H} field. ESR signals from a small piece of DPPH mounted in the cavity served as a convenient field calibration point during the experiments.

The cavity was split in the centre so that the half containing the sample could be rotated by a gear drive mechanism about the axis of the cavity. By rotating the magnet and the cavity, the tip angle between the applied field \vec{H} and any direction in the sample surface could be varied, and measurements could be made in any crystallographic direction. Large tip angles were measured to an accuracy of ± 0.2 degrees using a precision potentiometer attached to the magnet base. An optical lever arrangement, using the image of a fine crosshair projected via a mirror mounted on the magnet (and two fixed mirrors to fold the optical path) onto a recorder chart on a nearby wall, was used for small tip angle measurements with a resolution of $\pm .005$ degrees. For cyclotron resonance studies, the field was aligned to within ± 0.01 degrees of the sample surface by setting it at the position about which the small angle field tipping effects to be discussed in section D of this chapter were symmetrical.

The field derivative, dR/dH, of the surface resistance was generally recorded as a function of field or of magnet rotation angle on a strip chart recorder. Field marks were put on the strip chart recordings automatically every 100 (or 1000) gauss by a null-detecting circuit driven by a Rawson (Series 820, 0.1% accuracy) rotating coil gaussmeter calibrated with an NMR gaussmeter. Marks were also put on the recorder traces every 1 or 10 degrees during field rotation experiments with a circuit driven by the voltage across the precision potentiometer turned by the rotating magnet.

Since the cyclotron resonance signals for mercury were found to improve considerably from 2.1 to 1.1°K (see Figure IV-2, for example) measurements were carried out with the cavity and sample immersed directly in the helium bath. Temperatures as low as 1.1 degrees Kelvin were obtained by reducing the helium vapour pressure with an Edwards 50 liter/ sec. vacuum pump. The temperature was measured by monitoring the vapour pressure directly above the helium bath with a precision pressure gauge. Changes in the spectrometer sensitivity due to helium level changes in the wave guide were eliminated by placing a cellulose tape seal over the

end of a wave guide flange just above the cavity, and evacuating the wave guide above it. This flange also allowed the cavity assembly containing the sample to be removed for storage in liquid nitrogen.

The samples used in these studies were flat single crystal discs approximately 0.6 inches in diameter and 0.2 inches thick, and were grown from 99.999% pure mercury supplied by the United Mineral and Chemical Corporation of New York. The crystal growing apparatus is shown in Figure III-1. Chlorotrifluoethylene (Kel-F) was used for the molds, since its stated thermal expansion nearly matches that of mercury below - 20°C, and it is easily machined. The mold and microscope slides were carefully cleaned in heptane, washed in nitric acid, and finally washed in methyl hydrate. To facilitate easy removal after crystal growth, paraffin coatings, $\sim l\mu$ thick, were applied to the microscope slides before assembly of the mold by drawing them slowly and uniformly out of a very dilute, clean solution of paraffin in heptane. The molds were filled with mercury with a clean hypodermic syringe reaching to the bottom of the growth tube to avoid bubbles in the growth tube and on the sample surfaces. In a typical crystal growth, the temperatures of the cold pedestal and the hot can were stabilized at -10°C, the mold placed on the pedestal, and the pedestal slowly cooled.



Figure III-1. The mercury crystal growing apparatus

A layer of acetone on the pedestal led to better thermal contact between the stainless steel foot of the mold and the pedestal. Crystals with comparable $\omega\tau$'s could be grown with cooling times ranging from 30 minutes to several hours. Numerous attempts to seed crystals accurately oriented in crystallographic planes were unsuccessful in this apparatus, although a few attempts indicated some seeding was possible. The crystals were stored in liquid nitrogen, complete with microscope slides to protect their surfaces.

The tubes on either end of the sample were carefully cut away with a fine jeweller's saw, the glass slides were removed, and the samples fixed to the cavity, in liquid nitrogen. Because of the very humid conditions in the laboratory, this was done in a specially constructed dry room to reduce accumulation of frost inside the cavity, on the sample surface, and between the sample and the cavity.

The crystals were held firmly against a blunt edge on the end of the cylindrical cavity using a stiff phosphor bronze spring. A backing plate was placed between the crystal and the spring to minimize damage to the crystal. Another spring held the two halves of the cavity together. Care was taken to load these springs as tightly as possible to reduce vibrations arising from the interaction of the applied field \vec{H} and eddy currents induced by the field modulation in the sample; noise due to such vibrations could sometimes be reduced by wedging the wave guide assembly

against one side of the dewar.

B. CYCLOTRON MASS MEASUREMENTS

B.1. Sample Orientations

Two samples oriented, as shown in Figure III-2, within 12 and 20 degrees of the (110) plane were studied by AKCR. For these samples, directions in the sample surface will be specified relative to the P(lmn) projections, or by the angles, θ_c , which are defined in Figure III-2.

The samples were oriented within the experimental apparatus to better than ± 0.5 degrees using open orbit signals obtained in field rotation traces, and using the symmetry of masses corresponding to the electron α orbits. These methods of crystal orientation will be described in sections B.4 and C.1 of this chapter.

B.2. Method of Analysis

A typical Azbel'-Kaner cyclotron resonance trace is shown in Figure III-3 for sample #2 with \vec{H} in the sample surface and $\theta_s = 90.0$ degrees. Six distinct resonance series with up to 20 subharmonic resonances are well resolved in this trace. Orbits responsible for these traces will be discussed later. The trace also shows a signal due to DPPH at 12.22 kOe, and a sharp signal at 0.35 kOe due to the superconducting transition which saturated the spectrometer. Figure III-2.

Stereographic projections on the (111) plane of the orientations of Samples #1 and #2. The figures show the normals, N, to the sample surfaces, the great circles corresponding to these surfaces, and the positions (represented by thin lines between (lmn) directions and the sample surface) of the projections, P(lmn), of several (lmn) directions onto the sample surface. Field directions in the sample surface are specified by the angles, θ_s , measured relative to $P(\overline{112})$ in the senses shown by the arrows near $P(\overline{112})$. The coordinates (θ, ϕ) for N are (69.7°, 59.9°) for Sample # 1, and (82.1°, 51.5°) for Sample #2.



Figure III-3.

A typical Azbel'-Kaner cyclotron resonance trace for mercury. The region from 12 to 17 k0e is inset at the top right hand corner. The trace shows resonance series with fundamental resonances at fields of 8.47, 8.84, 9.50, 13.30, 14.50, and 16.30 k0e. Subharmonic sets for each of these series are indicated by sets of vertical lines below the trace. In each set the fundamental resonance is indicated by an arrow, and peaks obscured by larger ones are represented by dashed lines. The numbers under the lowest of these sets are the subharmonic numbers, N, corresponding to the resonance peaks in that set.



With a time constant of 0.1 seconds, noise on the original trace was less than the width of the pen used in this reproduction. The fundamental resonances at 13.3 and 14.5 kOe for two resonance series are not clearly evident in the reproduction, but were clearly visible in the original trace. The trace in this figure was taken at about 1.1°K with a microwave frequency of 34.285 GHz. Similar traces were taken at 2.5 degree intervals of θ_s , with frequent checks of small angle tipping effects to ensure accurate alignment of the field to the sample surface.

Effective masses were determined from the experimental data using a computer program to least-squares fit a line to a plot of the reciprocal magnetic field values, $1/H_N$, at which the peaks in dR/dH occurred, versus the subharmonic numbers, N, corresponding to these peaks. The reduced effective masses, m*, normalized to the free electron mass, m_e , were

$$m^* = \frac{e}{c \ \omega \ m_e \ \Delta(1/H)}$$
(III-1)

where ω is the microwave frequency and $\Delta(1/H)$ is the slope of the straight line fitted to the data. A phase shift, ΔN , defined as the intercept of the straight line on the N axis, was also calculated. This phase shift is the negative of the phase shift defined by Kip, Langenberg, and Moore (1961).

Equation III-1 is accurate in the limit that the

relaxation time, τ , tends to infinity and the field is accurately aligned in the sample surface (Kip, Langenberg, and Moore, 1961, and Koch, Stradling, and Kip, 1964). As τ decreases the resonance peak positions shift to higher positions and, because the shifts for low N are proportionately larger than those for large N, the plot of $1/H_N$ versus N is no longer linear. Masses predicted from Equation III-1 under such conditions are high, and the error decreases with the number of subharmonics included in the least-squares fit.

Changes of the peak positions and mass errors computed from Equation I-15 for electrons with common ω_c are shown in Figure III-4. In Chapter IV, $\omega \tau$ will be shown to be 20 for α orbits at 1.15°K; errors in m* due to $\omega \tau$ may be expected to be lower than \sim 1.5%, and to depend on the number of subharmonics available. In studies of field tipping effects (Section III-D), the fundamental resonances were found to shift less than 10% per degree for $\vec{J}_{rf} \perp \vec{H}$. Thus, since samples #1 and #2 were found to be flat to approximately 12 and 3 minutes of arc respectively, errors due to field tipping effects and sample curvature are expected to be less than ±1% for both samples particularly when several subharmonics are available. Errors due to $\omega \tau$ are expected to be characterized by positive phase shifts ΔN .

Figure III-4.

Changes in the resonance peak positions and errors in the effective masses due to $\omega\tau$ for electrons with common ω_c . The error in the mass m*, computed for a series of N subharmonics, is less than the shift of the position of the Nth subharmonic. The values of $\omega\tau$ shown here are approximately those found for the α orbit at temperatures of 1.15 and 2.1°K in Table IV-1.


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B.3. Effective Masses and Orbit Assignments

Effective mass curves for samples #1 and #2 are shown in Figures III-5 and III-6. Points denoted by open circles in these figures were obtained from fundamental resonances only, and were included only because they appeared to be extensions of well defined mass curves. A number of low amplitude peaks for which no subharmonic series could be found were disregarded because of the possibility that they might not be due to AKCR. The phase shifts, ΔN for both samples were generally below ± 0.03 and were not systematically positive. These phase shifts are attributed to scatter in assigning field values to the dR/dH peaks. From the phase shifts and the data in Figures III-5 and III-6, the errors in m* are estimated to be generally less than ± 2 %.

For convenience in subsequent discussion, the notation X_z^y will be used to identify the mass curve X, and to convey the information that this branch corresponds to sample #y and is attributed to the Fermi surface orbit z.

Mass curves S_{α}^{1} and D_{α}^{2} correspond to the dominant signals, for their respective samples, with over ten subharmonics visible for every field direction. Signals corresponding to T_{α}^{1} , V_{α}^{1} , F_{α}^{2} , and G_{α}^{2} were considerably weaker, so that typically only four or five subharmonics could be observed. These six mass curves are attributed



Figure III-5. Effective masses for sample #1.



Figure III-6. Effective masses for sample #2.

to α orbits on the electron lenses because they could be observed through 180 degrees of field rotation, indicating closed surfaces, and because each was at a maximum near the angle of closest approach of \vec{H} to a {100} axis. S_{α}^{1} and D_{α}^{2} , and indeed all of the α orbit mass curves, reach their maxima for field directions several degrees from P{100} toward P{111}. These shifts indicate the electron lens mass maxima are tipped approximately 3 degrees from (100) toward (111); this will be confirmed later in this chapter using an empirical interpolation scheme fitted to the α orbit data for both samples.

The mass curves U_{γ}^{1} and H_{γ}^{2} were both characterized by four or five subharmonics. These mass curves are attributed to the γ orbits approximately normal to (110) in Figure III-2; the positions of the minima for these curves and their relatively large ranges agree qualitatively with the predictions in Chapter II for the γ orbit. These mass curves show that the minimum γ orbit mass is $18\pm2^{\circ}$ from (11 $\overline{2}$) toward (110) in the (1 $\overline{10}$) plane, with a value m* = 0.65. Note that at P(11 $\overline{2}$), the masses for both U_{γ}^{1} and H_{γ}^{2} are approximately 0.73; this confirms the assignment by Dixon and Datars (1968) of the mass curves D, E, and F of their Figure 6 to the γ orbit.

Mass curves $P_{\gamma_2}^1$, Q_{μ}^1 , $R_{\epsilon_1}^1$, $A_{\gamma_2}^2$, $B_{\mu\gamma}^2$, and C_{μ}^2 corresponded to weaker signals for which, typically, only five

subharmonics could be observed. Only two or three subharmonics corresponding to E_{K}^{2} were observed. Assignments of orbits to these mass curves were made with the aid of Figure III-7, which shows several views of the model of Figure III-2 for orientations corresponding to samples #1 and #2.

Figures III-7 (d), (e), and (f) are for orientations near the minima of mass curves $A_{\gamma_2}^2$, $B_{\mu\gamma}^2$, and C_{μ}^2 respectively. A_{γ_2} is assigned to a γ_2 orbit because this orbit may exist for Figure III-7 (d), but probably does not exist for Figure III-7 (e), and cannot exist for Figure III-7 (f). Similarly, mass curve $B_{\mu\nu}^{2}$ is assigned to a $\mu\gamma$ orbit because this orbit appears most likely to exist for Figure III-7 (e), and should exist for Figure III-7 (d). Both this $\mu\gamma$ orbit, and the γ orbit corresponding to mass curve H_{γ}^2 , exist for the orientation of Figure III-7 (f) thus indicating that the β arms do not extend beyond the Brillouin zone point K, in agreement with the theories illustrated in Figure I-3. Mass curve $C_{i_1}^2$ is assigned to a μ orbit, which can exist for Figures III-7 (e) and (f) if the X face openings are just slightly larger than the size predicted by Dishman and Rayne (1968). Mass curve E_{μ} is assigned to a κ orbit, which cannot exist for Figures III-7 (d) and (e), but should exist for a small angular range to the right of $P(11\overline{2})$ in Figure III-6 and in Figure III-7 (f).

Figure III-7.

Orbits on the hole surface for several orientations corresponding to samples # 1 and #2. For each orientation, the sample surface, represented by the line S-S, is perpendicular to the plane of the drawing, and the applied field \vec{H} is in the sample surface and parallel to S-S; the Fermi surface orbits are in planes perpendicular to \vec{H} . The angles θ_s , which specify the orientations of \vec{H} relative to P($\vec{112}$), are approximately -22, 0, and +15 degrees in (a), (b) and (c) respectively (for sample #1), and -25, -15, and +10 degrees in (d), (e), and (f) respectively (for sample #2).



SAMPLE I



SAMPLE 2

This orbit is expected to cut off, as does mass curve E_{κ} , near P(112) because as the field is rotated toward P(110) the orbit can no longer pass between the two β arms near the point W in Figure III-7 (f). It is fortuitous that the orientation of sample #2 allowed the observation of all these orbits for fields in the sample surface.

Figures III-7 (a), (b), and (c) are for orientations near the minima of mass curves $P_{\gamma_2}^1$, Q_{μ}^2 , and $R_{\varepsilon_1}^1$ respectively. Mass curve $P_{\gamma_2}^1$ is assigned to a γ_2 orbit since this orbit can exist for Figure III-7 (a), is less likely to exist for Figure III-7 (b), and cannot exist for Figure III-7 (c). Similarly, mass curve Q_{μ}^1 is assigned to a μ orbit, which may just exist for Figure III-7 (a), reaches its minimum for Figure III-7 (b), and can still exist for Figure III-7 (c). Mass curve $R_{\varepsilon_1}^1$ is tentatively assigned to the ε_1 orbit expected for \vec{H} near (001); it is surprising that this orbit could be observed when \vec{H} passed only within 8.2 degrees of (001), that the orbit is centred approximately 5 degrees from P(001) toward P($\vec{112}$), and that it could be observed over a range of 15 degrees. However, there appears to be no other explanation for this mass curve.

The assignments of γ_2 , $\mu\gamma$, μ , and κ orbits near P($\overline{112}$) are strengthened by the observation that, as the orientation changes from that of sample #2 to that of sample #1, the minimum masses for the γ_2 and μ orbits are

expected to decrease and signals for the $\mu\gamma$ and κ orbits are expected to disappear, as observed. Also, for the nearly-free electron model, the mass corresponding to γ_2 is expected to be more than double that of γ for \vec{H} near ($\vec{112}$) as observed for sample #1.

Only a few subharmonics could be observed for mass curve L_{τ}^{2} because this series was partially obscured by subharmonics corresponding to D_{α}^{2} ; three or four subharmonics were typically observed for W_{τ}^{1} . Both these mass curves are assigned to the τ orbits shown in Figures III-7 (f) and (c). The cutoff points for these orbits toward P($\overline{112}$) depend critically on the size of the T and X face openings and on the sample orientations, but could not be determined accurately for either sample because of the variety of other resonances observed in this region.

Mass curves $X_{\mu_2}^1$ and $K_{\mu_2}^2$ were characterized by only two or three resolved subharmonics. These curves have been assigned to μ_2 orbits since they are centred near P[001] and P[010] respectively. Mass curve $J_{\mu_2}^2$ is tentatively assigned to a similar orbit centred near P[001]. The signals which give rise to these mass curves probably do not correspond to mass extrema. Apparently, the phases of the contributions to the surface impedance of the μ_2 orbits vary sufficiently slowly with k_{μ} over the region for which these orbits are possible that the Fresnel-type integral representing the sum of these contributions yields an amplitude large enough to be observed. Bogle et al (1969) report observing this orbit by magnetoacoustic attenuation experiments.

Two or three subharmonics were typically observed for mass curve Z^{1} but no orbit has been found to explain these signals.

B.4. Crystal Orientation Using the α Orbit Masses

The α orbit data of Figures III-5 and III-6 were superimposed, using symmetry, on one quarter of an electron lens, as shown on {100} projections in Figure III-8 (a) and (b).

The α orbit data cross over six times for each sample in Figures III-8 (a) and (b), and therefore were used to orient both of the samples by adjusting the orientation of each so as to minimize the sum of the squares of the mass differences at the crossover points. This was done by computer with a program that located the crossovers, smoothed the mass curves and interpolated for the masses at these crossovers, obtained the required sum for a particular orientation, and then adjusted the orientation until this sum was a minimum. The orientation was specified by θ and ϕ (see Figure III-2), the coordinates of the normal

Figure III-8.

The data planes for, and some predictions of, a ten parameter fit to the α orbit masses. In (a) and (b), for samples #1 and #2 respectively, field directions in the sample surface are determined relative to the three distinct sets of electron lenses centred on (100), (010), and (001) and superimposed by symmetry on a single {100} stereographic projection. The great circles representing the sample surface are reflected at the {110} mirror planes and, because the data have inversion symmetry, the ends of a great circle are equivalent points. Each of the great circles is labelled by the mass curve corresponding to it, and the bar and arrow represent the positions of $P(\overline{11}2)$ and the sense of the magnetic field rotation respectively. The stereographic projection (c) shows contours of constant mass obtained from a ten parameter interpolation expression (Equation III-2), which included terms to fourth order in the direction cosines relative to the orthogonal X, Y, and Z axes, simultaneously least-squares fitted to the data of Figures III-5 and III-6. Masses predicted by this interpolation scheme for the (100) plane are given in the polar plot (d).

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to the sample surface, and by a shift $\Delta \theta_s$ in θ_s ; since mass curves D_{α}^2 and S_{α}^1 vary rapidly with θ_s , this procedure was particularly sensitive to $\Delta \theta_s$. The accuracy was limited by the accuracy of the mass data; θ and ϕ were found to agree, to within ±1.0 degrees, with the corresponding values obtained from open orbit data (see Chapter III-C.1). Because the latter values are believed to be more accurate, and because there were unknown shifts of \vec{H} in the sample surface between the open orbit and AKCR experiments, the orientations for samples #1 and #2 have been obtained by fixing θ and ϕ at the open orbit values, and using the method based on α orbit data to determine $\Delta \theta_s$. The orientations are accurate to ±0.5° and are given in Figure III-2.

B.5. Interpolation scheme for the α Orbit Masses

Because the α orbit data sample wide ranges of the stereographic projections in Figures III-8 (a) and (b), these data form a good basis for fitting a three-dimensional empirical interpolation scheme to the masses corresponding to the electron lens. The data were fitted simultaneously to the expression

$$\left(\frac{1}{m^{*} (\alpha\beta\gamma)}\right)^{2} = A_{1}\alpha^{2} + A_{2}\beta^{2} + A_{3}\gamma^{2} + A_{4}\alpha\gamma + A_{5}\alpha^{2}\beta^{2} + A_{6}\alpha^{2}\gamma^{2} + A_{7}\beta^{2}\gamma^{2} + A_{8}\alpha\beta^{2}\gamma + A_{9}\alpha^{3}\gamma + A_{10}\alpha\gamma^{3} (III-3)$$

where α , β , and γ are direction cosines relative to the orthogonal X, Y, and Z axes in Figure III-8 (c). The axes Y and Z are parallel to (010) and (100) respectively. Equation III-2 includes all terms to fourth order in the direction cosines which satisfy the mirror symmetry $m^*(Y) = m^*(Y)$ about the (110) plane in Figure III-8 (c), and the inversion symmetry $m^*(\alpha\beta\gamma) = m^*(\alpha\beta\gamma)$ inherent in the mass data. The second order terms in Equation III-2 are those in the Shockley expression for the effective mass of an ellipsoidal Fermi surface tipped an angle 0.5 $atan(A_4/(A_3-A_1))$ from (100) toward (111). These terms themselves yield a fair approximation to the electron lens masses. Coefficients and the rms deviation for the fourth order ten parameter fit are given in Table III-1, and the fit to the data is shown in Figure III-9. Contours of constant mass corresponding to this fit for masses in the (100) plane are given in the {100} stereographic projection of Figure III-8 (c), while a polar plot for masses in the (100) plane is given in Figure III-8 (d). Masses predicted by the fourth order fit for a variety of planes are shown in Figure III-10.



FIGURE III-9 FOURTH ORDER FIT TO Q-ORBIT MASSES

Table III-1.

Coefficient Value Coefficient Value 2.289 -0.019 A₁ AG 2.652 -0.266 A₇ A2 -9.405 0.320 A₃ A₈ 9.635 A9 -9.915 A4 -0.793 -9.835 A10 A_5 RMS deviation in $m^* = 0.015 \sim 1.5$ %.

Coefficients for a fit of $(1/m^*)^2$ to Equation III-2.

As illustrated in Figure III-10, the fourth order fit confirms that the position of the maximum mass is tipped 3.0 degrees from (001) toward (111) in the (110) plane. The fit also shows that the broad mass minimum for \vec{H} in the (110) plane is also shifted 3.0 degrees in the same sense. Maximum and minimum masses in this (110) plane are 1.78 and 0.66 respectively for the fourth order fit.

The fourth order fit also predicts, as shown in Figures III-8 (d) and III-10, that in the (100) plane the mass has a minimum value of .614 for \vec{H} along (OII), and increases 9% to a maximum value of .669 near [00I] and [100]. This is clear evidence that the electron lens is not a surface of revolution about the line FL in Figure II-1. Because mass values in the (100) plane

Figure III-10.

Predicted α orbit mass curves for several symmetry planes. The predictions are based on the fourth order fit. Each curve is labelled by the plane to which it corresponds and by the crystallographic direction at $\theta = 0$ degrees; except for the (110) plane, the curves are symmetric about the labelled direction. The curve for the (110) plane is shifted 3.0 degrees from (001) toward (111) indicating that the lens is tipped out of the (001) plane in this sense.



are roughly proportional to the radius of the lens measured in the (100) plane and perpendicular to \vec{H} , Figure III-8 (d) shows that the electron lens has a minimum radius in the LU direction, and is pulled out approximately 9% in the LK direction. Note that Brandt and Rayne (1966) measure, and Keeton and Loucks (1966) predict, 6 and 10% increases in the dHvA frequencies between (011) and [001] or [100]. These results are in good agreement with the predictions of the fourth order interpolation scheme presented here. This agreement, the small deviations between the fit and the data in Figure III-9, and the wide ranges of the stereographic projections sampled by the measured planes in Figures III-8 (a) and (b), show that the fourth order interpolation scheme is reliable over 4π steradians. It is therefore useful for calculations of the mass in any desired plane or direction.

The systematic deviations in Figure III-9, are, however, slightly larger than expected from errors in m* and the sample orientations. Note specifically that the 6% deviation of the fit from the data at the top of the mass curve, S, would correspond to an apparent three degree misorientation of the sample toward the (110) plane. This is well beyond the 0.5 degree error bar for the orientation and therefore very unlikely. Also, the deviation appears not to be due to tipping effects, even though sample #1 was flat only to 12 minutes of arc, since the phase shifts ΔN

for these four points were less than ±0.03 and not systematically positive. These deviations suggest that the fourth order model may not be sharp enough about the (100) plane, and that better interpolation schemes may be obtained from higher order fits. Such fits might best be carried out with orthogonal basis functions, composed of spherical harmonics, chosen to satisfy the symmetry of the mass data. Such a basis has the advantage that, because the functions are orthogonal, one can estimate the effects of errors in the data and of taking measurements in a limited number of planes, by checking for changes in the coefficients corresponding to lower order terms when higher order terms are included in the leastsquares fits. Preliminary tests of this suggestion indicate that significantly better fits cannot be obtained for the present data; this is because once a fit equivalent to the present one is achieved, the lower order coefficients begin to readjust, indicating that the data are then being "overfitted". It is apparent that data in more planes, and even more carefully oriented samples are required to significantly improve the present fit.

C. OPEN ORBIT PEAKS AND DHVA-LIKE OSCILLATIONS

The experimental traces shown in Figure III-11 show the sharp peaks and oscillations of interest in this section. The peaks will be shown to correspond to open

Figure III-11.

Examples of open orbit peaks and dHvA-like quantum oscillations in dR/dH. In (a), the field is rotated very nearly in the (111) plane and types of open orbit directions corresponding to these peaks are labelled. Quantum oscillations are shown in the field rotation trace (b) for \vec{H} passing close to a β arm dHvA frequency minimum ($\vec{H} \mid |<100>$) at $\theta_{\rm H} = 315^{\circ}$; the peaks are open orbit peaks, and signals due to a second β arm are seen near $\theta_{\rm H} = 280^{\circ}$. Quantum oscillations are also shown in (c) for an orientation near the point A in Figure III-ll; here oscillations due to two β arms are beating, and the large signal at 12.4 kOe is due to DPPH.



orbits, and the oscillations to de Haas-van Alphen like signals arising from the β arms of the mercury Fermi surface. Both the peaks and the oscillations will be shown to arise, in part, from apparent changes in the microwave absorption in the cavity due to torques on the mercury sample.

C.l. Open Orbit Peaks

The positions of the sharp peaks observed for samples #1, #2, and #3 are summarized in Figure III-12. Since all these points lie on the known open orbit pattern for mercury (Datars and Dixon, 1967), it is clear that the observed peaks are associated with the open orbits. In what follows, it will be shown that, even though the crystal and cavity were tightly clamped together, torques similar to those which form the basis of the open orbit torque experiments of Moss (Moss and Datars, 1967) and Cook (Cook and Datars, 1970) were responsible for the apparent open orbit peaks in R and dR/dH observed in this experiment. Such torques result from the interaction of eddy currents induced in the sample by turning (or modulating) the magnetic field, with the applied field. Peaks in the torque occur, because the torque is small except for field directions perpendicular to the open orbit where the torque is determined by the open orbit conductivity, and

Figure III-12.

(111) stereographic projection of the positions of the measured open orbit peaks for samples #1, #2, and #3. The orientations P(Q,P) of the poles corresponding to these samples are P1(-69.7,89.9), P2(82.1,21.4), and P3(-82.2,9.6) degrees respectively. These orientations were obtained by least-squares fitting the {100} open orbit data to the {100} planes according to the known open orbit pattern for mercury (Datars and Dixon, 1967).



becomes much larger. Because these torques slightly change the contact between the sample and the cavity, they change the cavity Q (and hence the reflected microwave signal) and are therefore observed.

Perhaps the best indication that a torque mechanism was responsible for the peaks in dR/dH was the observation that these peaks varied widely in shape, amplitude, and even sign from trace to trace for the same crystal orientation. Cyclotron resonance traces taken at the same sample orientation, but with \vec{H} parallel to the sample surface, did not change appreciably. Changes in the peaks were particularly apparent when the apparatus was tapped lightly with a hammer. The system was also unstable in that the peak shape and indeed its baseline sometimes changed when the magnet was rotated through a peak in opposite senses. Also, the amplitudes and instability of the peaks was generally found to increase as the sample was loosened and vice versa. Finally, these properties remained essentially unchanged after the surface of a test sample was uniformly damaged (by scratching it with a cotton swab) so that all traces of previously strong cyclotron resonance signals were removed. The latter observation indicates that the peaks were due to effects, such as torque, in the volume of the sample rather than to changes in the surface im-

pedance of the crystal.

Peaks in the apparent absorption R corresponding to the open orbits were also observed by measuring the dc signal at the microwave crystal detector. These peaks were small and significantly sharper than the peaks in dR/dH. With rare exceptions, they were of the same sign, and changed signs when the sense of the magnet was reversed. The amplitudes of these peaks increased with both the applied field \vec{H} and the magnet rotation speed ω_{m} , and dropped to zero if the field rotation was stopped inside the angular range of the peak. These peaks did not vary with amplitude of the field modulation. However, the dR/dH peaks increased with \vec{H} , were independent of ω_m , and did not change appreciably when the rotation was stopped at a peak position. All these properties of the apparent peaks in R and dR/dH are consistent with induced torque mechanisms; those in R being due to torques induced by the field rotation, and those in dR/dH being due to torques induced by the field modulation. Assignment of these peaks to large induced torgues corresponding to open orbits therefore seems quite certain.

Beyond understanding their origin, studies of the open orbit peaks are of little interest because of the indirect and often unstable relationship between the torque

and the resulting signal. Plots such as Figure III-12 were, however, very useful for orienting the cyclotron resonance samples "in situ" within the cyclotron resonance experimental apparatus. Such orientations were accurate and unambiguous and avoided problems, special to mercury, of keeping the cyclotron resonance samples cold and their surfaces free of frost and surface damage during X-ray orientations (Dixon, 1966). This method of orientation had the additional advantage that before starting a detailed resonance experiment the sample could be shown to be a single crystal throughout its volume by the absence of spurious peaks. A computer program, in which the orientation was adjusted to obtain the best least-squares fit of {100} open orbit data to the {100} planes, was used to refine the orientation to better than ±0.5 degrees. This accuracy was limited by errors in specifying field directions corresponding to the open orbit peaks for each field rotation trace, and in specifying the angular rotation of the cavity corresponding to that trace. The orientations shown in Figure III-12 were obtained by this method.

C.2. De Haas-van Alphen Like Oscillations

Oscillations like those illustrated in Figures III-11 (b) and (c) were observed over wide ranges of sample orientation and tip angle between the field \vec{H} and the sample

surface for several samples. The oscillations were periodic in reciprocal field, and yielded dHvA frequencies corresponding to β arms of the mercury Fermi surface (Dixon, 1966, and Poulsen, Dixon, and Datars, 1967). Measurements of the dHvA frequencies, F, corresponding to these oscillations were made in twenty planes for sample #3 to an accuracy of 1.0%. This large amount of data will be summarized here in the form of a least-squares fit of (1/F)² to the expression

 $\left(\frac{1}{F}\right)^{2} = A_{1}\alpha^{2} + A_{2}\beta^{2} + A_{3}\gamma^{2} + A_{4}\alpha\gamma + A_{5}\alpha^{2}\beta^{2} + A_{6}\alpha^{2}\gamma^{2} + A_{7}\beta^{2}\gamma^{2} + A_{8}\alpha\beta^{2}\gamma + A_{9}\alpha^{3}\gamma + A_{10}\alpha\gamma^{3}$ (III-3)

for all the frequencies measured. Such a fit is possible since, as for the electron lenses, there are distinct β arm groups centred on <100> directions in the trigonal-bisectrix planes, and the β arm frequency data can be superimposed by symmetry in a single quarter zone.

The coordinate system chosen and coefficients determined for both second and fourth order fits are shown in Figure III-13. Agreement between the fourth order fit and the measured dHvA frequencies for a typical non-symmetry plane is shown in Figure III-14 and is within the experimental error for the determination of the orientation of that plane and of the frequencies. This fit will be compared later, in Chapter V, with β arm frequencies measured by the dHvA effect.



Coefficients for fits of $(1/F)^2$

to Equation III-3.

Coefficient	Value (units of 10^{-4} Tesla ⁻²)	
	Second order	Fourth order
^A 1	-0.055	0.010
A2	-0.311	-0.297
A3	1.823	-1.828
A4	-0.125	3.224
A ₅	0.0	0.160
A ₆	0.0	-0.136
A7	0.0	-0.045
A8	0.0	-3.169
A ₉	0.0	-3.295
A10	0.0	-3.390
RMS Deviatio	n: 1.51 Tesla	1.46 Tesla

FIGURE III-13

The coordinates and coefficients for, and a 115 Tesla Frequency Contour from, a fit of Equation III-3 to the β -arm data



Figure III-14. dHvA β arm frequencies for a sample oriented as shown in the stereogram.

The anisotropy of frequencies predicted by the fourth order fit is illustrated in the 115 Tesla countour in Figure III-13, and in Figure III-15, which shows frequency spectra in the (110) and (110) planes compared with frequencies for a cylinder with the same minimum frequency. These figures show that the β arm deviates significantly from a hyperboloid of revolution. In Figure III-15, the curve for the (110) plane approximates that for the cylindrical fit but is shifted two degrees from [001] toward (001). This shows the β arm is centred about a direction which is two degrees from [001] toward (001), and that the (110) cross section of the β arm flares out very slowly at the ends where it joins the rest of the Fermi surface. However, as $|\theta|$ increases, the β frequencies for the (110) plane increase more rapidly than for the cylindrical fit. Thus the (110) cross section of the β arm flares out significantly as it joins the rest of the Fermi surface, as expected, since this section includes the shape of the X face opening.

Oscillations similar to those reported here have been observed at microwave frequencies in aluminum and tin (Spong and Kip, 1965). As for aluminum, the amplitudes of the mercury quantum oscillations were found to depend only weakly on the angle of inclination of the magnetic field from the sample surface. For mercury, the amplitudes generally increased as the angle, between the field \vec{H} and the β arm dHvA frequency minimum near <100>, decreased.





Quantum oscillations arise from the same variations with field of the effective density of states at the Fermi level which give rise to the dHvA effect, and have been considered theoretically by Azbel' (1958). According to this theory, only electrons which have real space orbits small enough to lie within the skin depth region, contribute appreciably to the measured microwave surface impedance at fields above the AKCR fundamental. Also, only electrons which are very nearly stationary, and hence not readily lost from the skin layer because of drift along the magnetic field, are expected to contribute. The β arms of mercury satisfy these conditions well; not only is the radius of the real space orbit sufficiently small (\sim 9 \times 10⁻⁶ cm. at 20 kOe), but also the β arms are very nearly cylindrical, as shown in Figure III-15, so that a wide band of electrons can be expected to be nearly stationary and to contribute in phase to yield a relatively strong signal.

The signals observed in the present experiment cannot, however, be attributed only to quantum oscillations in the microwave surface impedance; they were apparently also due in part to the effects of the strong torques on the sample arising from the dHvA effect for the β arms. Evidence for the existence of such torque effects includes the observation that the amplitudes increased considerably when the sample was mounted less tightly in the cavity, and

vice versa. Even when the sample was clamped tightly in the cavity, the amplitudes were very sensitive to tapping of the dewar, and often could not be reproduced from trace to trace. Also, amplitudes for one sample changed little after the sample surface was very severely damaged with a cotton swab; true quantum oscillations, being sensitive to scattering in the skin depth, would have been expected to disappear.

Signals were, however, observed for fields near the β arm minima, where the torque effects must disappear. This suggests that the signals were also due in part to true quantum oscillations in the microwave surface impedance. Because of problems of separating the torque effects from these oscillations, it was not possible to study the properties of the true quantum oscillations in the microwave surface impedance.

D. Magnetic Field Tipping Effects

A brief description of the effects of tipping the magnetic field, \vec{H} , out of the sample surface is presented in this section. This is done primarily to demonstrate the usefulness of these effects in aligning \vec{H} parallel to the sample surface and in estimating surface flatness, and to discuss the tip sensitivity of several orbits. Some interesting features of the large angle tipping effects will be shown, however, to illustrate areas which warrant further study.
A variety of these tipping effects is illustrated for sample #2 in Figures III-16, III-17, and III-18. In these figures, and in subsequent discussion, the resonances are labelled by the mass curves corresponding to them in Figure III-6, the orientations are specified by the angle θ_{s2} in Figure III-6, and positive tip angles are directed toward (110) in Figure III-2. The relative amplitudes among the experimental traces shown are not absolute because the cellulose tape which excluded helium from the wave guide was cracked (and could not readily be replaced) so that the spectrometer sensitivity varied with liquid helium level changes.

Figures III-17, III-18, and III-19 illustrate the rapidity at which the line shapes for several resonances changed with increasing tip angle, α , and the symmetry of the small angle tipping effects for positive and negative α . Note, specifically, that in Figure III-17 the line shape for the fundamental of the D_{α}^2 resonance changed significantly for $-0.035 < \alpha < +0.035$ degrees thus indicating that sample #2 was flat to better than \pm 0.035 degrees along the field direction specified by $\theta_{s2} = 120$ degrees. Similar measurements indicated sample #1 was flat to better than \pm 0.1 degrees. Both of these estimates are conservative in the sense that the flatness is specified by tip angles for which the line shapes had very obviously changed and



Figure III-16. Large angle field tipping effects for $\theta_{s2} = 120$ degrees.



for $\theta_{s2} = 120$ degrees.





Figure III-19. Field positions of several resonances as functions of the tip angle for $\theta_{s2} = 120$ degrees.

the samples may be somewhat flatter.

The magnetic field was aligned parallel to the sample surface using the symmetry (for positive and negative α) of the small angle tipping effects as follows. First, the field magnitude was set just slightly on the low field side of one of the more tip sensitive, strong, fundamental resonances, at the orientation, θ_{c} , of interest. Then the magnet was rotated and, for a resonance which shifted to higher fields as α increased, the signal amplitude decreased as $|\alpha|$ increased on either side of the apparent zero tip position. This zero tip position was most accurately determined by measuring the tip angles corresponding to positions of equal signal amplitudes slightly on either side of the position of maximum amplitude; the mean of these two tip angles was chosen as the position of zero tip. The field magnitude was then reset and the procedure repeated several times to refine the previous determinations. Using this method, the field could be aligned to within ±0.01 degrees of the average sample surface; this accuracy is demonstrated in Figure III-17, where the traces are very nearly symmetrical about the zero tip position determined by the procedure outlined above. The method depended on tip sensitive resonances; fortunately strong tip sensitive resonances could be found to permit such an alignment procedure at all orientations for both samples.

The resonances D_{α}^2 , F_{α}^2 , and G_{α}^2 in Figures III-16, III-17, and III-18 all correspond to nearly ellipsoidal electron lenses with low mass spread but exhibit very different sensitivity to tipping effects depending on the orientation of the lens with respect to both \vec{H} and the sample surface. For $\theta_{g2} = 120$ degrees (Figures III-16 and III-17), for example, the lens (normal to (001) in Figure III-2) corresponding to D^2_{α} was viewed almost edge on by \dot{H} and a wide band of essentially stationary electrons contributed to the resonance with a very favourable geometrical factor so that strong, and eventually inverted, resonances were observed over a wide range of tip angles. As illustrated in Figure III-19, the first peak on the high field side of the split shifted 9.5% per degree for the fundamental but only 5.5% per degree for the second harmonic, while the position of the split remained virtually unchanged. However, for $\theta_{s2} = 40$ degrees (Figure III-18), the same lens was viewed nearly along (001) so that only a very narrow band of stationary electrons contributed to the D_{α}^2 resonance and the amplitudes observed with $\hat{J}_{rf} \downarrow \hat{H}$ dropped rapidly with increasing α as the non-stationary electrons drifted out of the skin depth. For this orientation, the D_{γ}^2 resonance peaks did not appear to shift in field for several measurements between $-0.3 < \alpha < +0.3$ degrees but rather only decreased in amplitude. The F_{α}^2 and G_{α}^2 resonances corresponded to

electron lenses which were tipped with respect to the sample surface. While these resonances showed slight signs of splitting, the major effects of field tipping were to cause a shift in the position of the dominant peak to higher fields and to cause the peak to broaden. Such shifts were found to be less than 10% for the fundamental resonances with $\vec{J}_{rf} \perp \vec{H}$ for F_{α}^2 and G_{α}^2 at all orientations θ_{s2} .

In Figure III-17 for $\theta_{s2} = 120$ degrees and in an experiment at $\theta_{s2} = -27$ degrees near the minimum of the γ orbit resonance, H_{γ}^2 was found to be relatively insensitive both in amplitude and field position to field tipping effects. In both cases, the resonance peaks shifted continuously to lower fields at ~ 0.5 % per degree as the tip angle changed from -1.0 to +1.0 degrees. This shift is interpreted as being due to changes in the field orientation relative to the Fermi surface rather than to field tipping effects, since as \hat{H} approaches the (110) plane the γ orbit mass decreases.

At $\theta_{s2} = -27$ degrees, the third subharmonics of both $A_{\gamma_2}^2$ and $B_{\mu\gamma}^2$ resonances shifted approximately 3% per degree of tip to higher fields indicating that these resonances were relatively tip sensitive.

The preceeding results show that the fundamental resonance shifts for $\vec{j}_{rf} \perp \vec{H}$ for several typical orbits were less than 10% per degree of tip and that the field \vec{H} could be aligned parallel to the mean sample surface to an

accuracy well inside the sample flatness. Hence, even for sample #1 which was flat to better than ±0.1 degrees, errors due to field tipping for these orbits are believed to be less than ±1% if only the fundamental was observed, and considerably less when several subharmonics were observed. However, for samples with surfaces flat to \sim ±0.8 degrees such as those studied by Dixon (Dixon, 1966, and Dixon and Datars, 1968) errors, due to field tipping effects, of several per cent or more might be expected for $\dot{J}_{rf} \mid \dot{\vec{H}}$ for the α orbit resonances, particularly when only the fundamental resonance was measured. Such errors depend sensitively on the orientation of the Fermi surface relative to the sample surface and on variations of the curvature of the sample surface. Where splitting of the peaks occurred, Dixon could have achieved greater accuracy by using the position of the split rather than the position of the dominant maximum, to determine the α orbit masses. Aside from reducing the errors due to field tipping, the flat samples used in the present experiment permitted the observation of several orbits which were obscured by field tipping effects in Dixon's experiments, and measurements of the temperature dependence of the effective mass and relaxation time for an α orbit as reported in Chapter IV.

As illustrated in Figure III-16 for $\theta_{s2} = 120$ degrees, the D_{α}^2 resonance was inverted at large tip angles roughly as

expected for a Fermi surface section, like the electron lens, with low mass spread. The masses corresponding to the inverted peaks were found to increase $\sim 2\%$ for $-10 < \alpha < +10$ degrees, the change being consistent with changes in the Fermi surface geometry as predicted from the ten parameter interpolation scheme of section B.5 of this chapter. Similarly, for a constant tip of $\alpha = -4.75$ degrees, the masses for the inverted peaks were consistent (within the $\sim 1\%$ scatter in the data) with the zero tip masses (and the predictions of the interpolation scheme) for the range of orientations $70 < \theta_{s2} < 160$ degrees beyond which the resonances broadened and decreased in amplitude to such an extent that analysis was no longer possible.

The results in the preceeding paragraph are interesting in view of the observation that, over the same angular ranges, plots of $1/H_N$ versus N (N=1, 2, 3) for the features A-B-C in the $\alpha = 4$ degree trace of Figure III-16 were essentially linear and had nearly the same slopes as similar plots for the inverted D_{α}^2 resonances. Thus these extra features A-B-C, appear to have been associated with the electron lens responsible for mass curve D_{α}^2 . For the indices N=1,2,3,..., the phase shifts, ΔN , for $1/H_N$ versus N plots for A-B-C were grouped about $\Delta N = -0.5\pm0.1$. However, with N=3,5,7,..., $\Delta N = 0.0\pm0.1$ for A-B-C with a corresponding decrease in the slope of the $1/H_N$ versus N plots by a factor

of 2. The latter observations suggest the features A-B-C might have been due to mass doubling, current sheet effects; however, since these features were observed with $\vec{j}_{rf} \perp \vec{H}$, and since for this orientation, \vec{H} was along the sharp edge of the ellipse where there are relatively few limiting point electrons, this mechanism appears unlikely.

The line shapes of the D_{α}^2 fundamental in Figure III-16 differ in several respects from line shapes previously reported for large angle tipping effects. First, as shown also in Figure III-19, there are two, not one, peaks on the high field side of the split. Second, for the $\alpha = 2.0$ degree trace on Figure III-16 for example, there are slight oscillations apparent in the region of fields just above the split. Finally at larger tip angles, the inverted peak is itself apparently split into two parts. It may be that these features, and the A-B-C structure can be explained as arising from the specific shape and orientation of the electron lens in a theory such as that of Smith (1967) (see section I-C.2) if a difficult, detailed calculation, which includes the Fermi surface shape of the lens and does not assume the equivalent of $\mathtt{A}_{_{\rm UV}}$ (Equation I-13) to be diagonal, could be carried out. These extra features might also be associated with the propagation of plasma-like waves into the sample near Azbel'-Kaner cyclotron resonance (Kaner and Skobov, 1964, Walsh and Platzman, 1965, Walsh, 1968, and Kaner and Skobov, 1968). This possi-

bility might best be investigated using the interference methods of Walsh (1965,1968) with very thin, plane parallel, samples.

CHAPTER IV

TEMPERATURE DEPENDENCE OF THE EFFECTIVE MASS AND RELAXATION TIME IN MERCURY

A. Introduction

The effective masses measured by Azbel'-Kaner cyclotron resonance and the de Haas-van Alphen effect are expected to differ, at low temperatures and cyclotron frequencies, from band masses predicted in the one-electron model because of mass enhancement due to many-body electronelectron and electron-phonon interactions. Mass enhancements due to electron-electron interactions are small (less than 10%) and temperature independent (Rice, 1965). Much larger enhancements (up to 150%) are expected from electronphonon interactions at low temperatures and frequencies for metals with strong electron-phonon interactions, like aluminum and lead (Ashcroft and Wilkins, 1965) and mercury (Grimvall, 1969).

Experimental evidence for such mass enhancements has so far been limited to the noting of differences between the measured masses and calculated band masses (Walsh, 1968) and to evidence from superconducting tunnelling experiments which will be discussed later. The purpose

of this section is to report measurements of the temperature dependence of an effective mass for mercury which provide new experimental evidence for the mass enhancement due to the electron-phonon interaction. The temperature dependence of the relaxation time, τ , was measured to aid in the analysis of the mass data; τ^{-1} was found to vary faster than the expected T^3 dependence with temperature (Azbel' and Kaner, 1958), and evidence will be presented to indicate that this deviation is due to Umklapp processes.

The electron-phonon interaction distorts the band structure, E(k) versus k, relative to the single particle band structure (Heine, 1969, for example) in a very small region, of energy $\hbar\omega_{\rm D}$ where $\omega_{\rm D}$ is the Debye frequency, about the Fermi energy, $E_{\rm F}$. Because $\hbar\omega_{\rm D}$ << $E_{\rm F}$, changes in the Fermi surface dimensions are expected to be small; and indeed, no deviations between observed and calculated single particle Fermi surfaces have, thus far, been conclusively attributed to electron-phonon many-body effects. In this context, it is important to note that techniques like the dHvA effect, which in the single particle model yield information about the Fermi surface dimensions, continue to do so in the presence of many-body electron-phonon effects (Nakajima and Watabe, 1963). It is because $\hbar\omega_c$, the cyclotron energy $\hbar eH/m_c$, is typically very much less than $\hbar\omega_{\rm p}$ that techniques, which measure the cyclotron effective

mass ($m_c \propto \partial A/\partial E$, where A is the Fermi surface cross sectional area at the mass extremal), sample the distorted region and observe masses which deviate from the single particle masses. Such deviations are expected to be both frequency and temperature dependent since, as ω increases, a wider range of the distorted region is sampled while, as T increases, the distortion of E(k) versus k changes.

Nakajima and Watabe (1963) have shown that, at low temperatures and low cyclotron frequencies, the electronphonon interaction enhances the effective mass, and

$$m^* = m_{\rm b} (1 + \lambda) , \qquad (IV-1)$$

where m_b is the single particle band mass, and λ , which will be defined more specifically in Equations IV-2 and IV-3, is a positive, dimensionless, number which characterizes the electron-phonon interaction strength.

Scher and Holstein (1966) find, for an isotropic electron-phonon gas at T = 0°K, that the mass enhancement rises 6% from its low frequency value to a maximum near $\omega_c = \omega_D$, and then drops to unity for $\omega_c >> \omega_D$. The electronphonon part, $\omega_c \tau_p$, of the product of the cyclotron frequency and the phonon part of the relaxation time was found to decrease with increasing ω_c even at T = 0°K because, as ω_c increases, the electron-hole pairs, which are energized by an external electromagnetic field, sample a greater range (up to the Debye cutoff) of phonon modes in their real

collisions with the phonons. Thus far, the high frequency cyclotron resonance experiment suggested by Scher and Holstein (1966) to observe variations of the mass enhancement with $\boldsymbol{\omega}_{c}$ has not been successfully applied, because these variations are very small over the frequency range provided by presently available cyclotron resonance apparatus. This experiment may become feasible with the advent of submillimeter cyclotron resonance apparatus, but will require high magnetic fields and sensitive apparatus to observe signals for $\omega_{c} \sim \omega_{p}$, since these signals will be damped because of decreased relaxation times arising from changes in the imaginary part of the electron self-energy. The suggestion, by Fowler and Prange (1965), that the mass enhancement might be determined from the field dependence of the dHvA amplitude has also not been successfully applied because significant changes in the mass enhancement occur only at very high fields. Interpretation of the results of such an experiment may also be complicated by amplitude changes resulting from changes in the relaxation time, which cannot easily be separated from amplitude changes arising from variations in the mass enhancement.

Wilkins (1968) has studied the temperature dependence of the mass enhancement and shown, for a Debye model, that λ (T) rises a few per cent from λ (T=0) to a peak at approximately T/T_D \sim 0.3, where T_D is the Debye temperature,

and then drops slowly to zero for $T >> T_D$. As the temperature increases, the phonon part of the relaxation time, τ_p , decreases due to increased scattering of the electrons by real phonons; thus the method (Fowler and Prange, 1965) of determining changes in the mass enhancement via the temperature dependence of the dHvA amplitude may be complicated by changes in the relaxation time with temperature. In the following few paragraphs, equations from Wilkins (1968) and theoretical estimates of λ (T) from Grimvall (1969) will be presented to illustrate rather than explain the theory, and to demonstrate why the temperature dependence of λ (T) might best be observed by low frequency Azbel'-Kaner cyclotron resonance for mercury. The material to be presented here will later provide a basis for understanding the results of the present experiment.

The essential information concerning both the electron-phonon interaction and the phonon spectrum is contained in a function, $\alpha^2 F(\omega')$, which can roughly be viewed as the product of the square of a frequency dependent electron-phonon coupling constant, $\alpha(\omega')$, and the phonon density of states, $F(\omega')$, for phonons of frequency ω' (Scher and Holstein, 1966, and Wilkins, 1968). A formal definition of $\alpha^2 F(\omega')$ is given in Equation IV-8.

In terms of $\alpha^2 F(\omega')$, (Wilkins, 1968), $\lambda(0) = 2 \int d\omega' \frac{\alpha^2 F(\omega')}{\omega'}$ (IV-2) at T = 0°K, and at finite temperatures,

$$\lambda(\mathbf{T}) = 2 \int d\mathbf{E} \left(-\frac{\partial f(\mathbf{E})}{\partial \mathbf{E}} \right) \int d(\mathbf{\tilde{n}}\omega') \frac{\alpha^2 F(\mathbf{h}\omega')}{\mathbf{\tilde{n}}\omega' + \mathbf{E}}, \qquad (IV-3)$$

where f(E) is the Fermi distribution function and the energy E is measured relative to the Fermi energy. For $\omega_{c} << \omega_{D}$, the electron-phonon contribution to the scattering rate is (Wilkins, 1968)

$$\tau_p^{-1} = 4\pi \int d\omega' \alpha^2 F(\omega') \left(\frac{1}{e^{\beta\omega'} + 1} + \frac{1}{e^{\beta\omega'} - 1}\right)$$
 (IV-4)
with $\beta = \hbar/k_BT$. At low temperatures, τ_p^{-1} depends primarily
on the low ω' edge of a sharply rising $\alpha^2 F(\omega')$ function,
since as ω' increases $\beta\omega'$ rises beyond unity and $\alpha^2 F(\omega')$
is strongly damped by the exponential terms in Equation IV-4.

 $\alpha^2 F$ has not yet been computed directly for mercury since the phonon spectra are not known, but has been determined by McMillan and Rowell (1969) by inversion of the superconducting tunnelling data of Bermon and Ginsberg (1964). This spectrum, and an estimate due to Grimvall (1969) of the temperature dependence of $\lambda(T)$ corresponding to it, are shown in Figure IV-1. The $\alpha^2 F$ spectrum for mercury is exceptional in that the value, $\lambda(T=0) \sim 1.6$, corresponding to it is one of the largest known and the strong peak is at an energy corresponding to an "effective" Debye temperature of only 20°K. This compares with values of T_D of from 68 to



 $\lambda(T)/\lambda(0)$ (after Grimvall) for mercury.

55°K obtained between 1 and 2°K by specific heat measurements in the normal (not superconducting) state (van der Hoeven and Keeson, 1964).

The large value of λ and the low effective Debye temperature for mercury give rise to the relatively large (~2.1%) change in effective mass predicted in Grimvall's calculation between 1.15 and 2.1°K. This shift is expected to be sufficiently large to be observable by Azbel'-Kaner cyclotron resonance for flat crystals. As shown in Figure III-4, corrections to the masses due to $\omega\tau$ can be estimated via Equation I-15, and are expected to be only weakly temperature dependent. Changes in $\omega\tau$ with temperature are therefore not expected to seriously complicate the experiment, provided $\omega\tau$ is sufficiently large ($\omega\tau \sim 10$) so that, say, 10 subharmonics can be observed.

In a cyclotron resonance experiment, even a small angle deflection (of order δ/L where δ is the anomalous skin depth and L is the mean free path) of a resonant electron caused by an electron-phonon scattering event will be sufficient to displace the electron into an orbit which either strikes the sample surface or does not pass through the skin depth. Such an electron is rendered ineffective, in the sense that it no longer contributes in phase to the resonance signal, and the AKCR signal amplitude is reduced; because of this, the electron-phonon contribution, τ_p^{-1} , to the scattering rate is normally expected to vary as the number of phonons, that is, as T³ at liquid helium temperatures. This was first suggested by Azbel' and Kaner (1958), has since been confirmed for copper (Haussler and Welles, 1966) and gallium (Moore, 1966), and has become generally accepted rule (Chambers, 1969). Two mechanisms will now be discussed which can, however, lead to variations of τ_p^{-1} with temperature which are faster than T³.

For a spherical Fermi surface and Debye-model phonons, the scattering angle for normal (not Umklapp) processes is approximately T/T_p , so that, if $T/T_p << \delta/L$, the scattered electrons will not be rendered totally ineffective. Under such conditions, the scattering rate will depend not only on the number of phonons but also on the size of the scattering angle, thus causing τ_p^{-1} to vary more rapidly with temperature than T^3 . This mechanism has been invoked to explain a $T^{3.3}$ dependence of τ_p^{-1} in tin (Gantmakher and Sharvin, 1965) for radio frequency size effect studies where, as for AKCR, scattering angles of the order of δ/L are sufficient to render electrons ineffective.

The scattering angles resulting from electron-phonon Umklapp processes are large, and the number of phonons having the energy, C, which is required for a particular Umklapp process may be expected to vary approximately as exp(-C/kT), according to the Boltzman factor at low tempera-

tures (T<C/k). Thus, if Umklapp processes contribute significantly to the scattering rate, the component τ_p^{-1} of τ^{-1} will vary roughly exponentially, and therefore τ_p^{-1} will vary faster than T³ with temperature. The possibility of such deviations from T³ is included in Equation IV-4, since contributions from Umklapp processes are included in $\alpha^2 F$. The fact that the $\alpha^2 F$ spectrum for mercury, in Figure IV-1, rises abruptly at ~ 0.5 mev. indicates (McMillan and Rowell, 1965) that Umklapp processes play a significant role in determining $\alpha^2 F$, and are therefore expected to give rise to a τ_p^{-1} variation with temperature which is faster than T³ from Equation IV-4.

B. Experimental Apparatus and Crystal Orientation

Measurements of the temperature dependence of m^* and τ were carried out in the cyclotron resonance apparatus described in Chapter III. The sample was immersed directly in the helium bath, the temperature was varied between 1.1 and 2.2 degrees Kelvin by throttling the pumping line to the dewar, and the temperature was measured by monitoring the helium vapour pressure with a precision pressure gauge. The wave guide above the cavity was evacuated to eliminate changes in sensitivity due to helium level changes. Because the signal amplitude decreased rapidly with increasing temperature, no attempt was made to design and build apparatus for measurements above the lambda point of He⁴. For each measurement, the field was aligned to within ±0.01 degrees of the sample surface by locating the magnet position about which the field tipping effects for the fundamental resonance were symmetrical.

Because $\omega \tau < 50$, the method of Chambers (1965) was not applicable, and the method of Haussler and Welles (1966) was used to measure $\omega \tau$. Accurate measurements of dR/dH versus H were made at various temperatures using low amplitude 10 Hz. field modulation, with an external oscillator to drive the Varian spectrometer, to avoid attenuating the higher subharmonics by over-modulation or insufficient penetration of the modulation field into the sample. Care was taken to operate in the square law region of the detector and to use short time constants and slow field sweeps which were approximately linear in 1/H.

Changes in the effective mass were studied, with a 20 Hz. modulation frequency, by measuring the positions of the resonance peaks with an NMR gaussmeter. In order to locate each peak accurately, the measured fields for two points of equal amplitudes, slightly on either side of the peak, were averaged and the peak position taken as the average of several such measurements. The amplitudes used for these determinations were greater than 90% of the peak

amplitude, except for the ninth and tenth subharmonics at high temperatures where the measurements were made further down the sides of the peaks to reduce errors due to low signal to noise ratios.

Sample #2 was used for the studies reported here; because it was flat to approximately 0.05 degrees, errors in m* and $\omega\tau$ arising from small angle tipping effects due to surface curvature were expected to be small. The measurements were made at the field orientation \vec{H} shown in Figure IV-4 for which $\theta_{s_2} = 173^{\circ}$. A wide band of contributing electrons and a very favourable geometrical factor for the α orbit on the electron lens normal to (001) led to signals, for that orbit, which were ten times stronger than all others, thus permitting unambiguous data analysis.

C. Experimental Results and Analysis

Typical traces for the $\omega\tau(T)$ measurements are shown in Figure IV-2 to illustrate the dominant resonance due to the α orbit studied, and the substantial changes in envelope shape, resonance line widths, and the envelope amplitudes, B_N , between 1.15 and 2.1 degrees Kelvin. The traces reproduced in this figure were noisy and of low amplitude relative to the trace in Figure III-3, since both the time constants and the modulation amplitudes were small to minimize errors in the $\omega\tau$ measurements. The m*(T)



measurements were carried out with higher time constants and modulation amplitudes, and hence with better signal to noise ratios.

Using the Haussler-Welles method, $\omega \tau$ was determined from the relation $\omega \tau = -2\pi/S$, where S was the slope of a least-squares fit of a line to plots of $\ln(B_N^{}/N^2)$ versus N for $2\pi N > \omega \tau$, and $B_N^{}$ was the envelope amplitude (illustrated in Figure IV-2) of the Nth subharmonic of the dR/dH versus H experimental trace. Two such plots, and the $\omega \tau$ values corresponding to them, are shown in Figure IV-3 for traces similar to those of Figure IV-2. The linearity of these plots confirms the applicability of the Haussler-Welles method for the orbit studied. Also, the observation that the points do not fall below the fitted lines, at high N, indicates that the frequency and amplitude of the field modulation were sufficiently low to avoid errors due to attenuation of the higher subharmonics.

Measured values of $(\omega \tau)^{-1}$ are shown plotted against T^3 in Figure IV-4 to illustrate both the temperature dependence of $\omega \tau$ and the scatter in the data due to noise on the experimental traces and variations in the tip angle between \vec{H} and the sample surface from trace to trace. These data were least-squares fitted equally well (with rms errors of ~ 2 %) to the equations

$$(\omega \tau(T))^{-1} = 0.047 + 0.00136 T^{4.8}$$
 (IV-5)



Figure IV-3. Typical Haussler-Welles plots for mercury

Figure IV-4.

The temperature dependence of $\omega \tau$. The data correspond to an α orbit on the electron lens normal to (001) for the field direction H shown in the stereographic projection. Least-squares fits to the data are given in Equations IV-5 and IV-6. The upward curvature of the data shows the deviation of the temperature dependent part of $(\omega \tau)^{-1}$ from a T³ variation with temperature.



and

$$(\omega \tau (T))^{-1} = 0.049 + 4.68 \exp(-9.8/T)$$
 (IV-6)

which are also plotted in Figure IV-4. The data could not be satisfactorily fitted by a power law, for $(\omega\tau(T)^{-1})$, with a constant plus quadratic and cubic temperature terms. At 1.15°K, $\omega\tau$ dropped 8% for a tip angle of 23 minutes and 10% when the modulation frequency was increased from 10 to 80 Hz.

Field positions corresponding to the first ten subharmonics and $\omega\tau$ values are summarized in Table IV-1 for temperatures between 1.15 and 2.1°K. The points for 1.15 and 2.1°K are each the average of three independent measurements with mean deviations of 0.1%, while those for the remaining temperatures are for one measurement each and are consequently less accurate. The changes in peak positions with temperature in Table IV-1 will now be shown to be due, not only to changes in $\omega\tau$ like those illustrated in Figure III-4, but also to changes in the effective mass, m*, with temperature.

First consider only the resonances at 1.15 and 2.1°K. Changes in the peak positions between these two temperatures are shown in Figure IV-5 for the first ten subharmonics. The solid curve is the $\omega\tau$ contribution to these shifts estimated from Equation I-15, with $\omega\tau$ changing from 10.8 to 20.1, as shown previously on Figure

Te mperature (°K)	2.08	2.01	1.83	1.64	1.46	1.15
ωτ	10.9	11.8	14.0	16.4	18.2	20.2
N = 1	8194	8177	8116	8069	8032	7983
2	4102	4095	4070	4046	4035	4006
3	2731	2728	2718	2703	2698	2679
4	2047	2045	2039	2030	2027	2013
5	1636	1635	1635	1624	16 21	1612
6	1363	1360	1357	1353	1349	1342
7	1164	1165	1162	1159	1155	1150
8	1019	1018	1016	1013	1008	1005
9	904	904	901	899	894	892
10	814	813	810	808	803	802

Table IV-1. Resonance Fields as a Function of Temperature.*

* The microwave frequency, ω , was 34.285 GHz; the magnetic field field values are in Oersteds.

Figure IV-5.

Shifts of the resonance peak positions resulting from a temperature change from 1.15 to 2.1°K. The solid curve represents shifts due only to changes in $\omega\tau$, and the dashed curve is a least-squares fit, to the data, of the sum of the solid curve and a constant shift, $\Delta m^*/m^*$, attributed to changes in the effective mass.



III-4. The dashed curve is the result of a least-squares fit, to the measured shifts, of the solid curve plus a constant, Am*/m*, which represents the shift due to changes in the effective mass m*. The observed shifts are consistent with this interpretation, within experimental error, and $\Delta m^*/m^*$ is found by this method to be (1.03 ± 0.2) %. In this analysis, the ten harmonics and the wt and Am* contributions to the resonance shifts are uniformly weighted. The good fit which results confirms that the estimates of the wr contributions to the peak shifts via Equation I-15 were reasonable and lends credence to the value determined for Am*/m* between 1.15 and 2.1°K. Note that Equation I-15 is expected to be applicable since the electron lens is nearly ellipsoidal and the resonance is dominant. Alternatively, the masses corresponding to the 1.15 and 2.1°K data in Table IV-1 were calculated in the standard manner and then corrected for changes in wt based on Equation I-15. This approach yielded mass values of 0.6505 and 0.6580 respectively, and a shift of $\Delta m^*/m^* = (1.16 \pm 0.2)$ between 1.15 and 2.1°K.

Effective masses computed in the standard manner from the data in Table IV-1 are shown in Figure IV-6, together with masses corrected for the effects of finite $\omega\tau$. Note that, as in Figure III-4, the corrections for ten subharmonics with $\omega\tau$ between 10.8 and 20.1 were approximately 0.8%



cyclotron mass.

as determined from Equation I-15, and only very weakly temperature (or $\omega\tau$) dependent. A least-squares fit to the corrected masses is also shown in Figure IV-6; this fit yielded an rms error of ±0.15% and a value of (1.3±0.3)% for $\Delta m^*/m^*$ between 1.15 and 2.1°K.

Figure IV-6 and the analysis summarized in Figure IV-5 show that a temperature variation in m*(T) has in fact been observed.

D. Discussion

The effective masses in Chapter III and in this chapter are in good agreement with those used by Dixon (1968) to show that the mass enhancement relative to the SOPW model band mass is $(1 + \lambda) = 1.72$. Also, estimates based on the minimum α orbit mass of 0.615 for \vec{H} along (1 $\vec{1}$ 0) in Figure III-8 (d) yield more reliable $(1 + \lambda)$ values of approximately 2.3 and 2.2 relative to the RAPW model of Keeton and Loucks (1966) and to the model of Bogle et al (1969). These $(1 + \lambda)$ values are lower than the McMillan and Rowell value of $(1 + \lambda) \sim 2.6$, indicating possible discrepancies in the determinations of λ from the measured masses and band structure calculations and/or in the determination of the McMillan and Rowell α^2_F spectrum and its use, via Equation IV-2, to determine λ . Also, since the λ obtained from the McMillan and Rowell spectrum represents
an average over the Fermi surface while the RAPW estimate, for example, corresponds to a particular orbit, there is the possibility that the discrepancies may be due, in part, to anisotropy of the mass enhancement. The value of $(1 + \lambda)$ is thus not yet well determined but is important since λ is one of the Landau-type Fermi liquid parameters for the electronphonon interaction (Heine et al, 1966).

The mass enhancement is large relative to the less than ten per cent electron-electron mass enhancements predicted by Rice (1965) and therefore is attributed primarily to the electron-phonon interaction. Since the enhancements due to the electron-electron interaction are temperature independent, the variation of m*(T) with temperature seen in this experiment is attributed to the temperature dependence of the electron-phonon mass enhancement. This is the first reported measurement of this temperature dependence, and provides new experimental evidence for the electron-phonon mass enhancement.

To first order, the change, $\Delta m^*/m^*$, in mass between two temperatures is related to the change $\Delta(\lambda(T)/\lambda(0))$ in $\lambda(T)$ for the same temperature range by

$$\frac{\Delta m^*}{m^*} = \frac{\lambda(0)}{1 + \lambda(0)} \Delta(\frac{\lambda(T)}{\lambda(0)}), \qquad (IV-7)$$

since $m^* = m_b(1 + \lambda(T))$ and $\lambda(0) \sim \lambda(T)$. With $\lambda = 1.6$

and $\Delta(\lambda(T)/\lambda(0) \sim 0.034$ for a change in T from 1.15 to 2.1°K, Grimvall's theory in Figure IV-1 predicts a change $\Delta m^*/m^* = 0.021$ or 2.1%. A corresponding change of (1.3 ± 0.3) % was measured in this experiment. Thus the McMillan and Rowell $\alpha^2 F$ function used by Grimvall leads to an estimate for the mass shift which is significantly higher than the experimental result.

The discrepancy between the measured and predicted values of $\Delta m^*/m^*$ is probably due to the use of the McMillan and Rowell average $\alpha^2 F$ spectrum for the estimates instead of a "directional" $\alpha^2 F$ spectrum appropriate to the narrow band of electrons on the α orbit studied. This point is illustrated by Equation IV-8 where, for the directional $\alpha^2 F$ function for the α orbit, the average over states k must be confined only to states corresponding to the orbit studied. The expression for the average $\alpha^2 F$ spectrum is (Wilkins, 1968)

$$\alpha^{2}F = \frac{\int \frac{dS_{k}}{\vec{v}_{F}(k)} \int \frac{dS_{k}}{\vec{v}_{F}(k')} \frac{|g_{k-k'}|^{2}}{(2\pi)^{3}} \delta(\omega' - \omega'_{k-k'})}{\int \frac{dS_{k}}{\vec{v}_{F}(k)}}$$
(IV-8)

where the integrals are over the Fermi surface S, $\vec{v}_F(k)$ is the carrier velocity from Equation I-2, and $|g_{k-k'}|^2$ is the electron-phonon transition probability for both normal and Umklapp transitions between k and k'. Reliable calculations of the directional $\alpha^2 F$ function for the α orbit have not yet been made in view of the lack of experimental phonon spectra, and are expected to be complicated by difficulties in calculating the transition probabilities for both normal and Unklapp processes near the zone boundary, and in determining the width of the band of electrons which contributes effectively to the resonance signal.

Like the average $\alpha^2 F$ function, the directional $\alpha^2 F$ function for the α orbit studied is expected to be sharply peaked at low w' due to Umklapp transitions across the broad face of the electron lens as illustrated in Figure IV-7. From Bogle et al (1969), the longest q vector, q., across the broad face of the lens is $\sim 0.35 \text{ Å}^{-1}$, and the longitudinal velocity of sound, s, is typically 2×10⁵ cm/sec. A simple Debye model estimate, $\hbar\omega'_{1} = \hbar sq_{1} \approx 4.6$ mev therefore shows that the peak in $\alpha^2 F$ corresponding to Umklapp processes across the lens and involving low energy transverse phonon modes may occur at energies well below 4 mev. Estimates based on the temperature dependence of wt, later in this section, indicate that the leading edge of the $\alpha^2 F$ function for the α orbit is at approximately 1.0 mev. As in Figure IV-1, this $\alpha^2 F$ spectrum will include a high energy tail arising from normal processes on the lens.

Using the measured value of $\Delta m^*/m^* = 1.3\pm0.3$ and Grimvall's value of $\Delta(\lambda(T)/\lambda(0)) \sim 0.034$, a value

Figure IV-7.

A (110) cross section of the mercury Fermi surface. The thin lines represent the free electron surface and the heavy lines represent the surface generated by the 4PW calculation of Bogle et al (1969). The latter illustrates the τ and α (electron lens) cross sections. An Umklapp process from states k to k' across the broad face of the electron lens and the angular range of the η orbit are illustrated. The lens is 0.35 Å⁻¹ thick.



of $1 + \lambda \sim 1.67 \pm 0.25$ is obtained from Equation IV-7, in near agreement with estimates based on the nearlyfree electron (SOPW) model. This is only an estimate, however, in the sense that, because the $\alpha^2 F$ spectrum for the α orbit differs from the average McMillan and Rowell spectrum, Grimvall's value of $\Delta(\lambda(T)/\lambda(0))$ is not strictly valid for the α orbit unless the difference between the two $\alpha^2 F$ spectra is one only of normalization by a factor 0.67/1.6.

Finally, it may be well to note that, if Equation IV-7 is satisfied for a measured value of $\Delta m^*/m^*$ and values of λ and $\Delta(\lambda(T)/\lambda(0))$ corresponding to a calculated directional $\alpha^2 F$ spectrum for the same orbit, that $\alpha^2 F$ spectrum and the λ corresponding to it can be considered reliable. Such measurements and calculations for several orbits could then, in principle, lead to experimentally verified determinations of the anisotropy of the mass enhancement and the electron-phonon interaction, which would complement estimates based on comparisons of measured masses with calculated band masses.

Now consider $\omega\tau(T)$ measurements. We first note that both of the fits, in Equation IV-5 and IV-6, yield $(\omega\tau(0))^{-1} \sim 0.048$. This value is due to scattering from static imperfections, and corresponds to a Dingle temperature, $T_D = \hbar/2\pi k\tau(0)$, of 0.012° K, thus indicating that

the sample was very pure and relatively free of crystal imperfections. This estimate is, in fact, high because $(\omega\tau(0))^{-1}$, as measured by the Haussler-Welles method, also contains contributions from surface roughness and the effects of mass spread near the mass extremal studied, as shown in Equation I-17.

It is interesting to note that the value for the Dingle temperature for mercury determined here is significantly lower than the values of 2°K reported by Moss (1968) and \sim 1.1°K reported by Palin (Shoenberg, 1969) which were determined by studies of the field dependence of the dHvA amplitude. This is yet another example of the generally anomalously large values of Dingle temperatures determined by dHvA relative to those determined from AKCR or the resistivity. Possible reasons for such discrepancies have been discussed recently by Shoenberg (1969); it appears that these discrepancies are not yet well understood and that systematic studies of the effect of crystal orientation, type of orbit, and the influence of known mosaic structure strains, and dislocation densities would be well worth while.

The upward curvature of the $(\omega\tau(T))^{-1}$ versus T^3 plot in Figure IV-4 and the least-squares fit to these data show that $(\omega\tau_p)^{-1}$ varied approximately as $T^{4\cdot7}$ for the α orbit studied. This clear deviation from a T^3 dependence shows that the general conclusion (Chambers, 1969) that $(\omega \tau_p)^{-1}$ varies as T^3 in cyclotron resonance experiments as predicted by Azbel' and Kaner (1958), is not necessarily valid.

For mercury, $\delta \sim 10^{-5}$ cm., $L \sim 4 \times 10^{-2}$ cm., and $T_D \sim 20^{\circ}$ K using the α^2 F estimate for T_D . Thus, T/T_D $\sim 200 \ \delta/L$ at 1°K, which shows the observed $T^{4.7}$ dependence of $(\omega \tau)^{-1}$ cannot be due to scattering angles which are insufficiently large to render the electrons ineffective. Electron-electron interactions contribute a very small T^2 component to $(\omega \tau)^{-1}$ (Moore, 1966). No such component was observed in the present experiment. It is apparent therefore that contributions from the electron-phonon scattering, or from other, as yet unthought of, scattering processes, must account for the observed temperature dependence of $(\omega \tau)^{-1}$.

Because the low energy peak in the average $\alpha^2 F$ spectrum in Figure IV-1 provides evidence of very low energy Unklapp processes and the electron lens is rich in Umklapp processes across its broad face (Figure IV-7), the observed $T^{4.7}$ dependence of $(\omega \tau)^{-1}$ is attributed to electron-phonon Umklapp processes involving electrons on the α orbit studied and low energy transverse phonon modes. This conclusion is supported by the observation that the temperature dependent part of the measured $(\omega \tau)^{-1}$ could be well fitted by the exponential term 4.68 exp(-9.8/T) in Equation IV-6. From this fit, the Umklapp processes which determined $(\omega \tau_p)^{-1}$ are seen to have been characterized by a temperature of approximately 9.8°K or, equivalently, an energy of approximately 0.9 mev, in the exponential term exp(-C/kT). This energy should correspond approximately to the leading edge of the $\alpha^2 F$ spectrum for the α orbit since $(\omega \tau_p)^{-1}$, in Equation IV-4, depends principally on the leading edge of a sharply rising $\alpha^2 F$ spectrum.

The McMillan and Rowell $\alpha^2 F$ spectrum (Figure IV-1), when used in Equation IV-4, yields a value $(\omega \tau_p)^{-1} \sim 1$ at T = 1.8°K, compared with a measured value of $(\omega\tau)^{-1}$ \sim 0.068. Since contributions to ($\omega\tau$)⁻¹ are additive, this spectrum is not consistent with the present experimental results. It is apparent that the leading edge of the $\alpha^2 F$ spectrum appropriate to the α orbit studied here must be at a slightly higher energy that that of the McMillan and Rowell spectrum, thereby reducing $(\omega \tau_p)^{-1}$ via the damping terms in Equation IV-4. A shift of 0.6 mev is sufficient to yield agreement between the measured and calculated values of $(\omega \tau_n)^{-1} \sim 0.037$ at 2°K, assuming the shape of the McMillan and Rowell spectrum. This suggests the leading edge of the α orbit $\alpha^2 F$ function should be at approximately 1.1 mev, in approximate agreement with the value of 0.9 mev determined previously from the exponential term

in Equation IV-6. When shifted 0.6 mev, the McMillan and Rowell spectrum leads to a temperature dependence of $(\omega \tau_p)^{-1}$ of $T^{7.7}$ for $1 < T < 2^{\circ}K$, indicating that the directional $\alpha^2 F$ for the α orbit studied may rise slightly less rapidly than the McMillan and Rowell spectrum.

In conclusion, the present experiment has provided new evidence for the mass enhancement due to many-body electron-phonon interactions, since only these interactions are expected to lead to a temperature dependence of the effective mass. The measured change in mass with temperature agrees only approximately with the predictions of Grimvall (1969), and it is apparent that a "directional" α^2 F function, appropriate to the α orbit electrons studied, is required for a more quantitative comparison. The temperature dependent part of $(\omega\tau)^{-1}$ was found to vary more rapidly than the expected T³ dependence, and the deviation attributed to a significant fraction of Umklapp scattering processes involving the electrons studied. Finally, there is evidence that the directional $\alpha^2 F$ function for the α orbit studied should rise abruptly at approximately 1 mev due to these Umklapp processes.

CHAPTER V

DE HAAS VAN-ALPHEN EFFECT IN MERCURY

The results of studies of the de Haas-van Alphen (dHvA) effect for mercury will be presented in this chapter for the trigonal-binary and trigonal-bisectrix planes. Two new orbits have been observed, and the results of Moss (1968) and Brandt and Rayne (1966) extended and clarified.

A. Theory of the Large Amplitude Field Modulation Technique

The theory of the large amplitude field modulation (LAFM) technique used here to detect dHvA signals has recently been reviewed (Stark and Windmiller, 1968), and will be only briefly summarized here.

Equations I-29 and I-30 may be generalized (Gold, 1968, for example) to show that the oscillatory changes in the magnetization arising from the dHvA effect have the form

$$\vec{M}(H,\theta,\phi,T) = \sum_{i} \vec{M}_{i}(H,\theta,\phi,T) \qquad (V-1)$$
where
$$\vec{M}_{i}(H,\theta,\phi,T) \propto kT \sum_{s=1}^{\infty} C_{i}\hat{M}_{i}(\theta,\phi) H^{-\frac{1}{2}}s^{-\frac{1}{2}} \times exp\left[-\frac{2\pi^{2}kcsX_{D}m_{i}(\theta,\phi)}{\hbar eH}\right] \times \left[\sinh\frac{2\pi^{2}kcsTm_{i}(\theta,\phi)}{\hbar eH}\right]^{-1} \times \cos\left[\frac{\pi g_{i}s m_{i}(\theta,\phi)}{2m_{e}}\right] \times sin\left[\frac{2\pi sF_{i}(\theta,\phi)}{H} + \beta_{i}(\theta,\phi,s)\right], \qquad (V-2)$$

and the sum over i represents contributions from various Fermi surface extrema. The symbols are defined in the next few paragraphs.

In this equation, $2\pi F/H>>1$ for ordinary fields, and the sine term varies much more rapidly with H than the other terms. Thus the ith extremal cross sectional area of the Fermi surface gives rise to quantum oscillations in the magnetic susceptibility of fundamental frequency $F_i(\theta,\phi)$ and phase $\beta_i(\theta,\phi)$ at temperature T for a field \vec{H} in the (θ,ϕ) direction relative to the crystal axes. The frequency F_i is related to the ith extremal cross sectional area A_i normal to \vec{H} by the Onsager relation (Equation I-28)

$$F_{i}(\theta,\phi) = \frac{\hbar c}{2\pi e} A_{i}(\theta,\phi) \qquad (V-3)$$

where $\hbar c/2\pi e = 1.047 \times 10^4$ Tesla/Å⁻². It is via Equation V-3 that the dHvA effect is a powerful method with which to determine Fermi surface geometries.

The effective mass m_i is the full effective mass enhanced by electron-phonon and electron-electron interactions, as is the Dingle temperature (Dingle, 1952b), $X_D = \hbar/2\pi k\tau$. The arguments in the exponential and sinh terms of Equation V-2 are much greater than unity; oscillations can only be seen at low temperatures and high magnetic fields, and often only the fundamental (s=1) of the dHvA oscillation is important. Effective masses, m_i , and the Dingle temperature, X_D , may be measured by studies of the temperature dependence of the dHvA amplitude. When m_i varies over a large range, as it does for the electron lens, the amplitude of the dHvA fundamental signal may go to zero as $\cos(\pi g_i m_i / 2m_e)$, where g_i is the g-factor of the conduction electrons, and m_e is the electron mass. Such "spin-splitting zeros" yield the possibility of determining the effective orbital g-factor g_i , and its anisotropy (Ketterson and Windmiller, 1968).

 $C_{i} \stackrel{\circ}{M}_{i}$ is determined by the Fermi surface geometry near the ith extremal; it is proportional to the area A_{i} and inversely proportional to the square root of the curvature $\partial^{2}A_{i}(\theta,\phi)/\partial k_{z}^{2}$ (where k_{z} is parallel to \dot{H}), and is in the direction

$$\hat{\mathbf{M}}_{i} = \frac{\dot{\mathbf{M}}_{i}}{|\vec{\mathbf{M}}_{i}|} = \frac{\nabla_{\vec{\mathbf{H}}} \psi_{i}}{|\nabla_{\vec{\mathbf{H}}} \psi_{i}|}$$

$$= -\hat{\mathbf{u}}_{\mathbf{H}} + \frac{1}{\mathbf{F}_{i}(\theta,\phi)} \frac{\partial \mathbf{F}_{i}(\theta,\phi)}{\partial \theta} \hat{\mathbf{u}}_{\theta}$$

$$+ \frac{1}{\mathbf{F}_{i}(\theta,\phi)} \frac{\partial \mathbf{F}_{i}(\theta,\phi)}{\sin \theta} \frac{\partial \mathbf{F}_{i}(\theta,\phi)}{\partial \phi} \hat{\mathbf{u}}_{\phi} \qquad (V-4)$$

where $\psi_{i} = 2\pi F_{i}(\theta, \phi)/H$ and \hat{u}_{H} is a unit vector along \vec{H} . The direction \hat{M}_{i} can often be determined by inspection, since it is often nearly parallel to the direction of minimum frequency.

For complete rigor, \vec{H} should be replaced by

 $\vec{B} = \vec{H} + 4\pi \vec{M}$ in Equation V-2; this is one source of sum and difference frequencies in the dHvA spectra.

For convenience in the following discussion, \tilde{M}_{i} will be approximated by the fundamental as

$$\vec{M}_{i} = K_{i} (H, \theta, \phi, T) \hat{M}_{i} \sin(\psi_{i} + \beta_{i}), \qquad (V-5)$$

where K_{i} varies slowly with H, θ , and φ .

In the LAFM technique for measuring the dHvA effect, the sample is placed at low temperatures in a modulated magnetic field \vec{H} + hhcos ω t, and the voltage induced by the time rate of change of \vec{M}_i (H, θ , ϕ ,T) in a pickup coil around the sample is measured. When the modulation amplitude, h, is large enough to modulate over several dHvA oscillations, the non-linear nature of \vec{M}_i causes harmonics of the modulation frequency to be generated. The voltage induced in the pickup coil due to the ith extremal is (Stark and Windmiller, 1968)

$$V_{i}(t) \propto \sum_{n=1}^{\infty} n\omega J_{n}(\delta \psi_{i}) K_{i}(H, \theta, \phi, T)(\hat{M}_{i} \cdot \hat{u})$$
$$\times \sin \left(\psi_{i} + \beta_{i} + \frac{n\pi}{2} \right) \sin (n\omega t) \qquad (V-6)$$

where \hat{u} is a unit vector along the axis of the pickup coil, $J_n(\delta \psi_i)$ is the Bessel function of the first kind of order n, and

$$2\pi \leq \delta \psi_{i} = \nabla_{\hat{H}} \psi_{i} \cdot h \hat{h} \propto \hat{M}_{i} \cdot \hat{h} . \qquad (\nabla -7)$$

The total voltage at the output of a phase-sensitive detector tuned to the nth harmonic nw is, therefore,

$$V_n(H,\theta,\phi,T) \propto \sum_i n\omega J_n(\delta \psi_i) K_i(H,\theta,\phi,T) (\hat{M}_i \cdot \hat{u})$$

 $\times \sin \left[\psi_i + \beta_i + \frac{n\pi}{2} \right].$ (V-8)
By choosing the directions \hat{h} and \hat{u} such that

$$\hat{M}_{i} \cdot \hat{h} = \hat{M}_{j} \cdot \hat{h} = \hat{M}_{k} \cdot \hat{u} = \hat{M}_{l} \cdot \hat{u} = 0,$$
 (V-9)

the signals corresponding to the i, j, k, and 1 Fermi surface extrema can simultaneously be eliminated. Further selectivity results from the amplitude factor $J_n(\delta \psi_i)$, which can be set on maxima (or minima) to maximize (or eliminate) the signal corresponding to a fifth extremum. Also, for $\delta \psi_i \leq 1$, $J_n(\delta \psi_i) \sim \delta \psi_i^n$, so that for higher harmonics signals from Fermi surface extrema with large frequencies $F_i(\theta,\phi)$ can be greatly enhanced relative to smaller ones since $\delta \psi_i$ is proportional to F_i . Additional selectivity can be gained by sweeping 1/H linearly in time, so that the dHvA signals are periodic in time and specific frequencies Finally, many dHvA frequencies can be resolved by fourier analysing complicated dHvA spectra.

In the present experiment, $\hat{h} | |\hat{u}| |\hat{H}$ so that only the $J_n(\delta \psi_i)$ selectivity could be widely used, and the signals were reduced by $\hat{M}_i \cdot \hat{h}$. For this geometry, $\delta \psi_i = \nabla_{\hat{H}} \psi_i \cdot \hat{hh} = \frac{2\pi F_i(\theta, \phi)h}{\mu^2}$, (V-10) and $\delta \psi_i$ could be held constant by varying h as H². Electronic filtering and fourier analysis were also used.

B. Experimental LAFM Apparatus

The Westinghouse superconducting magnet system used in this work has been described by Moss (1968). It provides fields up to 55 k0e that are uniform to 1 part in 10⁴ in a 0.5 inch sphere. The field was scanned approximately linearly in H (or 1/H) at variable rates. Field readings were taken from a stable magnet current shunt calibrated by NMR. At the sweep rates used in these experiments, the field-current relationship was linear after the field was swept several k0e. A Magnion niobium-tin superconducting solenoid was used at 110 k0e for one experiment. For this experiment, field measurements were made by measuring the magnetoresistance of a copper coil (Scott et al, 1968), wound outside the inner dewar, after calibration against a Rawson rotating coil gaussmeter calibrated by NMR.

The LAFM apparatus used in this experiment is shown schematically in Figures V-1 and V-2.

The apparatus (Figure V-1) which rotated the crystal and supported the removable modulation and pickup coil assembly was constructed by Moss (1968) principally from Kel-F with a nylon belt. The belt tightener was added



Figure V-1. The modulation coil assembly and sample goniometer for the LAFM apparatus.



Figure V-2. Block diagram for the LAFM apparatus

for this experiment to reduce noise due to vibrations of the lower drum relative to the pickup coil; the belt was tightened at liquid nitrogen temperatures, and did not loosen significantly due to differential thermal contraction when cooled to liquid helium temperatures. The pickup coils were 4500 turns each, wound in opposite senses and balanced to 1 part in 10⁴ in air; the modulation coil was 700 turns. Vibrations within the coils were eliminated by gluing them together, layer by layer, with General Electric Glyptal glue; the wedge at the top of the modulation coil assembly reduced noise due to vibrations of the coils relative to the sample holder.

The sample goniometer shown in Figure V-1 was constructed for these experiments so that crystals could be conveniently and accurately oriented for measurements in specific crystallographic planes. Samples were grown inside the inner race of this goniometer; a thin starting tube used to initiate single crystal growth was carefully cut off with a jeweller's saw. The goniometer was adjusted to $\pm 1^{\circ}$ in a series of torque magnetometer experiments by using a combination of β arm dHvA oscillations and open orbit peaks to determine the orientation. To orient a crystal for measurements in the trigonal-bisectrix plane, for example, the three open orbits crossing at [001] in Figure III-12 and the beats between dHvA signals arising from the [100] and [010] β arms at A were used as sensitive indicators of crystal orientation.

The apparatus in Figure V-1 was designed to allow working space in which to orient the sample. Because of this, the pickup coils were large, and therefore weakly coupled to the sample and particularly sensitive to noise arising from vibrations of the coil assembly as a whole. Such noise could be reduced by wedging the coil assembly against the dewar wall, but often remained the limiting noise in this system. When inserted in the dewar, the pickup coils became somewhat unbalanced, apparently due to distortion of the modulation field arising from nonuniform eddy currents in the dewar. This unbalance led to noticeably reduced signal-to-noise ratios arising from harmonic content in the modulation field. Twin-T filters, between the oscillator and amplifier and tuned to 2w and 3w, considerably reduced this noise in early experiments using a Hewlett-Packard 200 CDR oscillator with a McIntosh 75 amplifier. These filters were unnecessary with a Krohn-Hite 4100R oscillator; however a Krohn-Hite UF101AR amplifier used with it produced considerable harmonic distortion unless the modulation coil was made resistive by tuning it with a parallel capacitor. A modulation frequency of 517 cps. was used, and the modulation amplitude, h, was limited to less than 75.0 gauss, above which

the magnet was driven normal.

The pickup coils were impedance matched to a twin-T filter tuned to remove the fundamental frequency ω , and the resulting signals were phase sensitively detected at harmonics n ω with a Princeton Applied Research (PAR) HR-8 lock-in amplifier operated in the "select external" mode. Optional band pass filters tuned to the harmonic of interest were used occasionally between the twin-T and the PAR. Care was taken to reduce noise pickup by tightly twisting the pickup coil leads together, but since the signal-to-noise ratio was limited by vibrations of the sample and pickup coil assembly, no attempt was made to place a matching transformer shielded in superconducting lead inside the inner dewar (Stark and Windmiller, 1968).

Reference signals were provided to the PAR via a non-linear circuit and tuned amplifier. The output from the PAR was filtered by a Krohn-Hite 3322R band pass filter, principally to remove background drift and, occasionally, high frequency noise. The filtered signals were recorded both on a strip chart recorder, and on magnetic tape together with magnetic field voltages using the data acquisition system described by Moss (1968) for subsequent fourier analysis. Usually the field was scanned linearly in 1/H, and typically 10 to 20 pairs of field and dHvA voltages were recorded per dHvA oscillation on magnetic tape, for over 100 oscillations. Harmonics from the second to seventh were used, low harmonics to obtain a general view of the dHvA spectra, and the seventh to enhance low level high dHvA frequency signals relative to strong low frequency signals.

The experiment relied heavily on the resolution provided by fourier analysis of the dHvA signals. Jones (1968) and Moss (1968) but principally Dunsworth (unpublished) have contributed to the computer programs now used in this laboratory for data handling and the fourier analysis. In the version used in these experiments, the data were first pre-smoothed by subtracting from them a least-squares parabola fitted to the data, and then interpolating (using the IBM subroutines ATSM and ALI) for signal values spaced equally in reciprocal field. The IBM subroutine RHARM was then used to compute fourier coefficients from which the fourier amplitudes were calculated for the dHvA frequency range of interest. Subroutine ATSM requires a monotonic data table; it was found necessary to pre-smooth the field values locally for traces in which many points were recorded at low sweep rates, when noise on the field voltage could destroy the monotonicity. This pre-smoothing was applied sparingly, as was low pass filtering of the field voltage.

The fourier amplitudes were plotted versus dHvA frequency on a Benson-Lehner plotter, except during experimental runs, when considerably faster computer turnover times could be gained by displaying the data graphically with the line printer. The square root of the fourier amplitude was generally plotted to suppress the dominant peaks and improve the visibility of the low amplitude peaks. A sample spectrum, Figure V-5, will be discussed in the next section.

C. Experimental Results and Discussion

Figures V-3 and V-4 show the dHvA frequency plots for a sample oriented in the trigonal-bisectrix (110) plane with the goniometer shown in Figure V-1. The data in Figure V-3 correspond to the β arms; the frequencies corresponding to [100] and [010] are not resolved because the sample is oriented very nearly in the trigonal-bisectrix plane. The measured curves agree closely with the solid curve based on the fourth order fit to the β arm quantum oscillations described in Chapter III; this lends weight to that analysis, and shows that the field calibrations and sample orientations are consistent in the two experiments.

Signals corresponding to the γ branch in Figure V-4 were stronger than all others; they are assigned to the γ orbit because the position of the minimum and the







Figure V-4. dHvA frequencies for the (110) plane.

angular range for this branch are those expected for the γ orbit. As for the cyclotron resonance results, the γ orbit minimum was approximately 20 degrees from (112) toward (110) and signals were observed for a range of 76°. The minimum frequency was 1900±10 Tesla and the frequencies at (110) and (112) were 2080±10 and 2040±10 Tesla respectively.

Because curves α_1 and α_2 extend through 180 degrees, they are assigned to α orbits on the electron lenses normal to (100) and (010). These two branches were just resolved, by an amount that indicates that the crystal was rotated about one degree out of the trigonal-bisectrix plane about an axis at approximately the -40 degree position in Figure V-4. The α_3 branch is assigned to an α orbit on the electron lens normal to the (001) axis. The amplitudes of the signals corresponding to the α branches all decreased rapidly with increasing frequency, and α_3 did not reappear near (001) at higher frequencies, even at 100 k0e.

Estimates based on an ellipsoid of revolution of 3 to 1 semiaxis ratio (for the electron lens $k_{LU}/k_{LT} =$ 0.538/0.176 = 3.05, from Bogle et al, 1969), scaled to a minimum frequency of 3250 Tesla, showed that the α amplitudes decreased significantly faster than expected

from the effects of Fermi surface curvature, $\hat{M} \cdot \hat{u}$, and changes in the effective mass m; in Equation V-2. The observed decreases in amplitude therefore appear to be due in part to smearing of the phase $\psi = 2\pi F/H$, due to field inhomogeneity, or more probably, to strains introduced in the crystal when the starting tube was cut from the crystal. Phase smearing due to mosaic spread in the crystal apparently can be ruled out because near (001), where $\partial F/\partial \theta \rightarrow 0$, α_3 did not reappear. Spin-splitting zeros may also have contributed to the rapid decrease in amplitude, since for 0.61 < $m_i * < 1.8$ and $g \geq 2$, $\cos(\pi g m_i * / 2)$ will go through one or more zeros. Little can be said about this contribution, however, because the signals did not reappear at high frequencies, and attempts based on the ellipsoidal model to use the amplitude terms in Equation V-2 to determine effective g values (2n + 1)m,* were inconclusive due either to appreciable phase smearing or to inadequacies of the model. Finally, note that with the ellipsoidal model (or from estimates based on the dimensions ${\bf k}_{\rm LU}$ and ${\bf k}_{\rm LW}$ in Bogle et al, 1969) a maximum frequency in excess of 9500 Tesla is expected for the α_3 branch near (100). The points at 6800 Tesla previously attributed to the α_3 branch (Moss, 1968 and Brandt and Rayne, 1966) were shown in this experiment to correspond to second harmonic and sum frequencies, due

to α_1 and α_2 , which could be seen throughout this plane except between (111) and (110) where α_1 and α_2 signals were very weak.

The frequency branch η in Figure V-4 has not previously been observed by dHvA, and was observed only with the 116 k0e magnet. This branch is assigned to the hexagonal shaped hole orbit, n, centred at T and completely enclosing the T face, which was first reported by Bogle et al (1969) in their magnetoacoustic attenuation studies. The orbit was observed by dHvA for a range of 12 degrees toward (100), where it was cut off sharply by the L face opening , and 18.4 degrees toward (110), where it was cut off sharply by the X face opening; the total range of 30.4 degrees is in close agreement with the value of 30 degrees subtended at T by the hole section about U in Figure 7 of Bogle et al (1969). This angle is illustrated in Figure IV-7. Bogle et al (1969) report a caliper dimension, $k_{\text{out}}^{\text{TW}} \sim 0.90 \ \text{\AA}^{-1}$, from T through W to the outside of the hole surface. The frequency corresponding to a hexagon with this dimension is 22,100 Tesla, and compares favourably with the minimum value of 21,900 Tesla observed in this experiment.

Figure V-5 shows the host of dHvA harmonic and sum frequencies observed near (112). The γ_2 branch shown

Figure V-5.

Sample fourier amplitude and dHvA frequency plots near (112) for fields in the (110) plane. The fourier amplitude plot corresponds to an experimental trace for which \vec{H} was along (112), and the seventh harmonic was detected with $\delta \psi$ small to suppress signals due to the γ branch. The frequency plot shows the variety of harmonic and sum frequencies observed, and demonstrates the need for fourier analysis in resolving the spectra. The γ_2 frequency branch is seen to correspond to a relatively strong signal in the fourier amplitude plot.



there was observed for the first time in this experiment; signals corresponding to this orbit were strong, particularly just before a sharp cutoff near (112). Because of this cutoff and the increase in frequency toward (110), this orbit has been assigned to the γ_2 orbit, illustrated in Figure II-4 and believed also to have been observed by AKCR in Figures III-5 and III-6. The cutoff depends critically on the size of the T face hole and on the amount by which the β arms pass inside the X face at K. Assuming, for example, that the orbit passes through K as it does in the MAG V model of Figure I-3, one finds that the radius kmm of the T face opening must be $\stackrel{\sim}{\sim}$ 0.2 \mathring{A}^{-1} at the cutoff, in agreement with the value of 0.198 Å⁻¹ estimated by Dishman and Rayne (1968) from their MAG V model based on magnetoresistance open orbit studies. The alternate explanation that the γ_2 branch could be a μ orbit cut off near $(11\overline{2})$ by postulating a large T face hole opening of radius \sim 0.5 ${\rm \AA}^{-1}$ can be ruled out by the appearance of mass curves γ_2 , μ , and $\mu\gamma$ in Figures III-5 and III-6, as well as by the magnetoresistance open orbit studies, all of which indicate that the T face hole cannot be so large. Finally, the break in the points labelled γ in Figure 2 of the Bogle et al (1969) paper, if real, may indicate that not only γ but also γ_2 orbits were observed. For \dot{H} near (112) and the stated geometry, the Fermi surface calipers along (111) corresponding to γ and γ_2 orbits are expected to be nearly equal.

The branch ascribed to the X face,electron neck, δ orbit by Moss (1968) was shown to correspond to the sum frequency $\gamma + \alpha_3$. No signals corresponding to this orbit, δ , or to the T face electron neck were observed, although the dHvA frequency region between 50 to 5000 Tesla was carefully studied both near (110) and near (111). This is disconcerting, but perhaps due to narrow bands of contributing electrons or to a high sensitivity of these orbits, which are in the zone boundary, to strain-induced phase smearing. Moss (1968) reports weak signals for \vec{H} along (111) at 1950 Tesla which were tentatively assigned to the T face, electron neck, orbit. A very careful search in this region was unsuccessful in the present study; it may be the present sample was more strained than the one studied by Moss (1968).

The branch ascribed to the μ orbit by Moss (1968) was shown to correspond to the sum frequencies $\alpha_1+\gamma$ and $\alpha_2+\gamma$ illustrated in Figure V-5. No signals could be found for the μ orbit at 55 k0e and, unfortunately, an experiment in the 116 k0e magnet had to be terminated, due to difficulties with the magnet power supply, before the region between (110) and (001) could be studied. The μ orbit therefore has not yet been observed by dHvA although evidence that it exists has been presented by Bogle et al (1969) by the magnetoacoustic effect, and by the cyclotron mass curves assigned to it in Figures III-5 and III-6 of this report. It is possible that this μ orbit is obscured by the variety of signals near (112) in Figure V-5, or that it has a low "spin splitting factor" for dHvA.

DHvA frequency plots for a sample oriented near the trigonal-binary ($\overline{2}$ ll) plane are shown in Figures V-6 and V-7. The growth tube for this sample was oriented by magnetoresistance, and the sample mounted in the apparatus of Figure V-1 in a cylinder machined so that the trigonalbinary ($\overline{2}$ ll) plane would be measured. The actual orientation was obtained with a computer program which searched for the orientation yielding the best least-squares fit of the measured β arm frequencies to the model of section III.C.2. The best fit and the orientation corresponding to it are shown in Figure V-6; the normal P(Q,P) to the measured plane has angular coordinates P(86.5,9.0) in the stereographic projection.

The frequency branches α in Figure V-7 are due to α orbits on the electron lenses; again the amplitudes corresponding to these branches dropped more rapidly than expected. The branch τ has been assigned to a τ orbit since its minimum is near P(0Ī1) as expected. Finally the γ branches on either side of P(0Ī1) correspond to γ orbits, and are not symmetrical about P(0Ī1) because they are very sensitive to rotations about the trigonal axis.





The minimum frequency for τ was 1600 ± 10 Tesla for $\vec{H} | |\vec{P}(0\vec{1}1)$, where P($0\vec{1}1$) was 9.0 degrees from ($0\vec{1}1$); this value compares favourably with the value of 1580 Tesla obtained by Brandt and Rayne (1966) at ($0\vec{1}1$), since the frequency increases slightly as \vec{H} is rotated from ($0\vec{1}1$) in the (111) plane. The observed angular range of 47 degrees for the τ orbit means that the sum, $k_{in}^{TW} + k_{in}^{XK}$, is at least, and probably greater than, $0.22 \ \text{Å}^{-1}$, since the measured frequency is not rising rapidly at the ends of the τ frequency branch. When compared with the corresponding values in Figure II-3, the above estimate shows that the RAPW model underestimates the sizes of the X and T face openings.

The field \vec{H} passed within 3.5 degrees of the (111) direction in the ($\vec{2}$ 11) plane. However, as for the ($\vec{110}$) plane sample, the T face electron neck orbit was not observed. Also a $\mu\gamma$ orbit which was expected to be just geometrically possible at the 63 ± 5 degree position in Figure V-7 was not observed. The fact that these orbits were not observed does not, however, rule out the possibility that they do exist.

A planned dHvA experiment for the (111) plane was not carried out, to allow the temperature dependence experiments in Chapter IV to be done. Such a dHvA
experiment would have been useful, since the γ_2 , $\mu\gamma$, ν , and κ orbits are all expected in the (lll) plane and, if observable, would have yielded new information on the details of the Fermi surface geometry via both their positions and angular ranges. Also, observations of the $\mu\gamma$ and κ orbits would have yielded supporting evidence for the assignment of cyclotron resonance mass curves to these orbits in Figures III-5 and III-6. With fourier analysis and sensitive detection, the shapes and cutoffs of the γ and τ dHvA frequency curves may be specified more accurately than by Brandt and Rayne (1966), thereby yielding further new information concerning the sizes of the X and T face openings and the shape of the β arm near the Brillouin zone point K.

CHAPTER VI CONCLUSIONS

Azbel'-Kaner cyclotron resonance studies have been carried out for two, very flat, mercury single crystals having Dingle temperatures of 0.015°K. Because the crystals were flat, possible errors in the effective masses arising from field tipping effects were much smaller than for the corresponding results of Dixon and Datars (1968), and several new orbits, ($\mu\gamma$, γ_2 , κ , μ_2 , and ϵ_1) were observed for the first time by AKCR. Measurements in nonsymmetry planes and the use of crystal symmetry permitted the development of a three dimensional interpolation scheme for the α orbit masses. This interpolation scheme showed that the second-zone electron lens is tipped three degrees out of the (100) plane toward the (111) plane, and that there is a 9% anisotropy of the mass in the (100) plane. A similar interpolation scheme for the β arm quantum oscillations showed that the β arms deviate significantly from hyperboloids of revolution and are centred about directions which are two degrees from <100> toward {100}.

The AKCR orbit assignments reported here are in agreement, for the α , γ , and τ orbits, with those of Dixon

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and Datars (1968). However, their assignment of some very weak signals (G in their Figure 9) to the μ orbit seems to be incorrect since, for the stated orientation, the suggested μ orbit is cut off by the [100] β arm in their stereogram and the μ orbit mass is expected to be considerably larger, relative to the t orbit mass, than the value reported. These points G are more likely to be due to a μ_2 orbit (about the (110) Brillouin zone face in their stereogram) for which the mass is a minimum at the same orientation. For the orientation corresponding to Figure 10 of Dixon and Datars (1968), β arm signals are expected to be centred near the 65 and 135 degree positions but not at the 170 position corresponding to curve D. The assignment of their mass curve D to a β orbit is therefore doubtful, though no more definite interpretation presents itself.

For a dominant α orbit resonance series, the effective mass was found to increase 1.3%, and the relaxation time was found to decrease by a factor of two, between 1.15 and 2.1°K. The observed change in mass with temperature provides new evidence for the mass enhancement due to many-body electron-phonon interactions, since only these interactions are expected to lead to a temperature dependence of the effective mass. The measured change agreed only approximately with the predictions of Grimvall (1969) and a "directional" $\alpha^2 F$ function, appropriate to the α orbit studied, is required for more qualitative comparison. The temperature dependent part of the scattering rate, τ^{-1} , was found to vary more rapidly with temperature than T^3 and the deviation attributed to a significant fraction of Umklapp electron-phonon scattering processes involving the α orbit electrons studied. Also, there is evidence that the directional $\alpha^2 F$ function for the α orbit should rise abruptly at approximately one mev. due to these Umklapp processes.

The dHvA results of Brandt and Rayne (1966) and Moss (1968) were extended and clarified, and two new orbits (n and γ_2) were observed for the first time by dHvA. DHvA frequency curves assigned by Moss (1968) to δ and μ orbits were shown to be due to sum frequencies, while points at 6800 to 7000 Tesla near {100}, which had previously been assigned to the maximum frequency corresponding to the electron lens, were shown to be sum and harmonic frequencies. DHvA frequencies and AKCR cyclotron masses are summarized, in Table VI-1, for a variety of orbits. The minimum frequencies determined here for the β , τ , γ , and α orbits confirm the values of Brandt and Rayne (1966) and Moss (1968) within the combined frequency and orientational accuracies. However, the value (3780 Tesla) for the α orbit frequency, with \vec{H} along (111), found here is considerably larger than

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Orbit	Field Direction	dHvA Frequency [†] (Tesla)	AKCR Reduced Mass
β	2° from [001] toward (001) ^a	73.9±0.5	
Υ	19° from (112) toward (110) ^a	1900 ± 10	≲ 0.65 ^b
	(110)	2080 ± 10	0.73±0.01
	(112)	2040 ± 10	0.73±0.01
τ	(011) ^a	≲ 1600 ± 10 ^C	< 0.71
η	(111) ^a	21,900 ± 300	
μ	(112) ^a		1.24±0.01
ad	(111)	3780 ± 10	0.733 ^e
	[110]	3410 ± 10	0.661 ^e
	(110)	3630 ± 10	0.700 ^e
	(1Ī0) ^a	≲ 3250 ± 10 [℃]	0.614 ^e

Table VI-1. Some dHvA Frequencies and AKCR Effective Masses.

[†]The corresponding cross sectional areas $({\rm \AA}^{-2})$ are 0.955 × 10⁻⁴ times the dHvA frequencies (Tesla).

^aDirections of minimum dHvA frequency and AKCR mass for the corresponding orbit.

^bMisoriented 7 degrees from the (110) plane; from Figure III-5.

^CMisoriented 9 degrees toward {121}; from Figure V-7.

^dThe α orbit values correspond to the lens normal to (001).

 e_{From} the ten parameter α -mass interpolation scheme.

the value (3450 Tesla) reported by Brandt and Rayne (1966). For measurements in the (110) plane, the observed angular range of the η orbit was in good agreement with the range predicted by the 4PW model of Bogle et al (1969), while the sharp cutoff of the γ_2 orbit was consistent with the MAG V model of Dishman and Rayne (1968). These angular ranges indicate that the dimensions k_{in}^{LU} , k_{in}^{XU} , and k_{in}^{TU} for the hole surface are approximately 0.87, 0.30, and 0.20 Å⁻¹ respectively.

Apparent peaks in dR/dH found during field rotation (dR/dH versus θ) experiments were shown to be due to torques induced in the sample for magnetic field directions which support open orbits. These peaks were primarily important in that they could be used to accurately orient the AKCR crystals "in situ" within the experimental apparatus. An alternate method based on the symmetry of the measured α orbit masses was also developed to orient AKCR crystals. DHvA samples were conveniently oriented in a specially constructed goniometer using open orbit torque and β arm dHvA signals, measured with a torque magnetometer, as sensitive indicators of the crystal orientation.

The observed AKCR cyclotron mass and dHvA frequency curves (including those for the $\mu\gamma$, γ_2 , and κ orbits which were suggested in this study) have been shown to be in qualitative agreement with the topographical features of

previously proposed mercury Fermi surface models having electron lenses in the second-zone and a multiply-connected first-zone hole surface with breakthrough regions at all the Brillouin zone faces. However, as for previous studies, the results reported here are not accurately fitted by the existing band structure calculations and there remains a need for further detailed band structure calculations to resolve apparent discrepancies between existing experimental results and calculations. Because mercury is a heavy element (atomic number 80, $\dots 5d^{10}6s^2$) and the 5d band is close to the 6s conduction band (Keeton and Loucks, 1966), it appears (Dishman and Rayne, 1968) that future calculations should simultaneously include spin-orbit coupling effects and a non-local pseudopotential formulation while at the same time adjusting the Fermi level to maintain carrier compensation (ie.equal numbers of electrons and holes) during fits to the experimental data. In addition to providing more accurate data for the α , τ , and γ orbits, the present dHvA results provide new data for the η and γ_{2} orbits which will be useful for such band structure calculations.

When more precise band structure calculations are available, the cyclotron effective masses reported here can be used to make more reliable estimates of the electronphonon mass enhancement factor and its anisotropy for various

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orbits. Also, when both accurate band structure calculations and experimentally determined phonon spectra for mercury become available, calculations of directional $\alpha^2 F$ spectra may be carried out to provide estimates of the mass enhancement factor, and the temperature dependence of both the electron-phonon mass enhancement and the relaxation time, for specific orbits. Calculations for the α orbit studied in Chapter IV are of particular interest for quantitative comparison with the measured temperature dependences of the effective mass and relaxation time corresponding to that orbit.

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