

## Some New Aspects of Cyclotron Resonance in Copper\*

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(Received 17 June 1963)

The development of improved techniques has made possible a new series of cyclotron resonance experiments on Cu which gives more accurate and extended information on electron cyclotron masses. New orbits which have been observed and measured include a limiting point orbit, orbits extending through three and four Brillouin zones, orbits whose centers are neither at the center nor edge of the zone, and orbits observed with the magnetic field tipped at large angles (up to 80°) with respect to the crystal surface. The neck orbit has been observed and measured with the field along the [111] direction. The cyclotron mass ratio for this orbit is 0.46. Certain discrepancies and puzzling aspects of earlier data have been clarified and the present data are in excellent accord with de Haas-van Alphen and magnetoacoustic data on the geometry of the Fermi surface. The principal cause of difficulties in the former experiments has been shown to be some perturbations of the absorption spectra of cyclotron resonance when the applied magnetic field is tipped as little as a fraction of a degree with respect to the metal surface. These effects are responsible for the phase shifts in the cyclotron resonance series reported earlier, as well as for the rapid changes in measured cyclotron masses which occurred in certain regions of field orientation. Although these effects are not understood in all details, their principal causes are fairly clear, and the effects can be avoided by maintaining the field accurately parallel to the sample surface.

### I. INTRODUCTION

WE are reporting a new series of experiments on cyclotron resonance in a single crystal of Cu. In view of the earlier extensive study of cyclotron resonance in the same metal by Kip, Langenberg, and Moore<sup>1</sup> we feel compelled to explain our repetition and extension of these experiments. The present work is the result of the realization of the importance of the effect of tipping the magnetic field relative to the sample surface. Improvements in the experimental technique and the use of higher experimental frequency in the present work have allowed the removal of some troubling aspects of the earlier study. In the first place, KLM report the observation of an unexplained "phase shift," i.e., a nonzero intercept of the straight line plots of  $1/H$  versus number of subharmonic. This was observed for practically all orientations of  $H$  in the (110) sample surface used, and in some directions had a value as large as 0.3. The theory of Azbel<sup>2</sup> and Kaner<sup>3</sup> fails to predict such unusually large phase shifts and there has not been a satisfactory explanation for their occurrence. A further difficulty with the earlier work concerns the detailed values of cyclotron masses observed. Gross aspects of the data were reasonably consistent with the general features of the model of the Fermi surface suggested by Pippard.<sup>3</sup> However, an attempt by Roaf<sup>4</sup>

to fit the effective mass anisotropy data reported by KLM to the detailed model of the Fermi surface derived from Shoenberg's de Haas-van Alphen measurements<sup>5</sup> proved impossible. The sharp breaks and kinks in the KLM experimental curves are in sharp contrast to the expectations of the model. In addition, the KLM data do not fit the neck dimensions suggested by the dH-vA results or the acoustic attenuation experiments in Cu.<sup>6</sup>

The possibility that both phase shifts and errors in the effective mass measurements might be caused by experimental difficulties, was first suggested by some work on the dependence of cyclotron resonance signals in Cu on the angle of inclination of the dc magnetic field to the sample surface.<sup>7</sup> It was found that such tipping to even a relatively small fraction of one degree could cause drastic changes in both the measured cyclotron mass and the phase shift. In particular, the phase shift was observed to vanish when the field was accurately parallel to the sample surface. This observation prompted us to re-examine the KLM data and repeat effective mass measurements for an adequately flat Cu sample with accurate control of field tipping. (Drastic effects on cyclotron mass and phase were not detected in the earlier studies at lower frequency, probably because of the surface roughness of the sample used.)

A number of modifications in the experimental arrangement were required for this investigation. These included (1) a new cavity and sample geometry which made possible accurate alignment of the field to the sample surface, (2) new treatment of the sample surface to improve its flatness, and (3) increase in the experimental frequency from 24 kMc/sec to 67 kMc/sec. This increase in the microwave frequency had three advan-

\* This work was supported in part by the U. S. Office of Naval Research and by Grant AF-AFSOR-290-63 from the U. S. Air Force Office of Scientific Research

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<sup>1</sup> A. F. Kip, D. Langenberg, and T. W. Moore, *Phys. Rev.* **124**, 359 (1961).

<sup>2</sup> M. Ya. Azbel' and E. A. Kaner, *J. Phys. Chem. Solids* **6**, 113 (1958).

<sup>3</sup> A. B. Pippard, *Phil. Trans. Roy. Soc. (London)* **A250**, 325 (1957).

<sup>4</sup> D. J. Roaf, *Phil. Trans. Roy. Soc. (London)* **A255**, 135 (1962).

<sup>5</sup> D. Shoenberg, *Phil. Trans. Roy. Soc. (London)* **A255**, 85 (1962).

<sup>6</sup> H. V. Bohm and V. J. Easterling, *Phys. Rev.* **128**, 1021 (1962).

<sup>7</sup> J. F. Koch, Ph.D. thesis, University of California, Berkeley, 1962 (unpublished).

tages: the effects of field tipping are less serious at higher frequencies, the smaller cavity used at the higher frequency exposed only the central part of the Cu crystal to the microwave field, thus improving the effective flatness of the sample, and the increased  $\omega\tau$  made for better resolution of the resonance signals. As a result of these changes we have been able to greatly extend and improve the effective mass data. The phase shifts and errors in the cyclotron masses because of tipping have been removed, and the cyclotron mass data now seem completely consistent with Roaf's<sup>4</sup> analysis of the dH-vA results. The considerably increased  $\omega\tau$  has allowed the identification and measurement of effective masses due to orbits in a range of angles where KLM were unable to interpret their data. Extended orbits through 3 and 4 zones have been observed, as well as orbits not centered at the center of the Brillouin zone or at a zone face, i.e.,  $k_H \neq 0$  orbits. We have observed a limiting point resonance with the field along the [100] axis. The measurement of the neck orbit has been extended over a wider range of angles. The neck mass measured is considerably different from the extrapolated value suggested by the previous work, the discrepancy probably being due to the poor resolution of the 24 kMc/sec data. In addition, we have found that in most orientations in Cu it proves possible to observe cyclotron resonance with  $H$  at arbitrarily large angles to the sample surface. Such measurements give additional detailed geometrical information about the Fermi surface.

The following section will deal with the experimental arrangement used for the present work as well as with the preparation of the sample surface. Section III will be concerned with some detailed measurement of the effects of field tipping. The interpretation of the effective mass data in the fourth section follows in spirit the orbit identification scheme proposed by KLM,<sup>1</sup> but has been extended to include orbits through 3 or 4 zones, as well as some  $k_H \neq 0$  orbits. We have used the Pippard model of the Fermi surface with the dimensions and shape derived from Roaf's analysis<sup>4</sup> of the dH-vA data.

## II. EXPERIMENTAL ASPECTS

The measurements reported in this work were carried out at liquid He temperatures and at a frequency of 67 kMc/sec, using a standard reflection spectrometer. To allow frequency scaling of certain experimental results we have also made measurements at 35 kMc/sec. The sample forms the end wall of a cylindrical cavity and the system is stabilized to the frequency of this cavity. The magnetic field is modulated at a low audio frequency and the signal recorded is the derivative of the power absorption. We shall restrict the discussion here to the cavity arrangement used in the present work and the preparation of samples.

The previously used experimental arrangement in which the cavity was mounted at the end of the wave guide made difficult the exact alignment of the field to

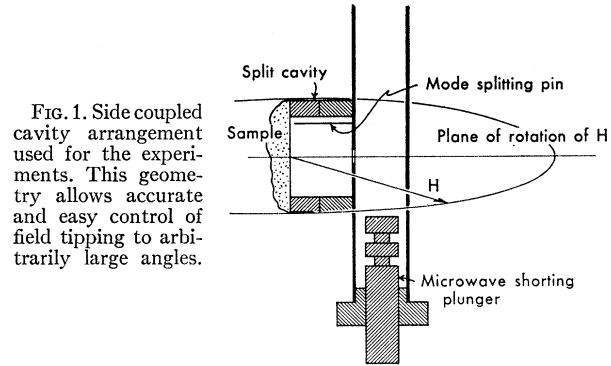


FIG. 1. Side coupled cavity arrangement used for the experiments. This geometry allows accurate and easy control of field tipping to arbitrarily large angles.

the plane of the sample surface. In addition, it did not allow tipping to large angles. In order to permit accurate and easy control of field tipping to arbitrarily large angles, we have used a cavity coupled somewhat off-center on the broad face of the rectangular wave guide as in Fig. 1. The cavity is designed to operate in a  $TE_{111}$  mode and is split in the center as indicated in the figure. The top half is rigidly fixed to the wave guide. The sample is lightly clamped to the bottom half over a choke joint groove (not shown in the figure). A geared drive mechanism rotates the lower half of the cavity about the cylindrical axis. In effect, the sample can be rotated without change in cavity coupling, relative to a fixed combination of dc magnetic field and rf current directions.

A pin is used to split the degeneracy of the cavity and allow choice of two perpendicular current modes. This pin is set so as to make the current lines respectively parallel or perpendicular to the component of the magnetic field in the surface. The coupling to either of the modes can be conveniently adjusted by means of a shorting plunger in the wave guide termination. The current lines for a  $TE_{111}$  mode are not strictly linear over the end wall. We have determined the ratio of rf power in the two perpendicular components by measuring the absorption signal amplitude from a DPPH sample distributed evenly over the end wall. This ratio was about 1/20; our polarization was therefore 95% pure.

The dc magnetic field can be rotated in the plane perpendicular to the wave guide to make possible a three-dimensional search for cyclotron resonance signals. The magnet position could be set by means of a dial indicator to an accuracy of better than  $10''$  of arc, allowing accurate and detailed tipping measurements.

In this work we have used the (110) Cu sample prepared by KLM for their earlier work. This disk-shaped sample had been acid-string cut from a single crystal boule so that the surface of the sample was approximately a (110) plane of the crystal. There is an error of one degree in the orientation of the surface with respect to this plane. The resistivity ratio (between room temperature and 4°K) is in excess of 5000. The sample surface has been relapped and repolished in an attempt to

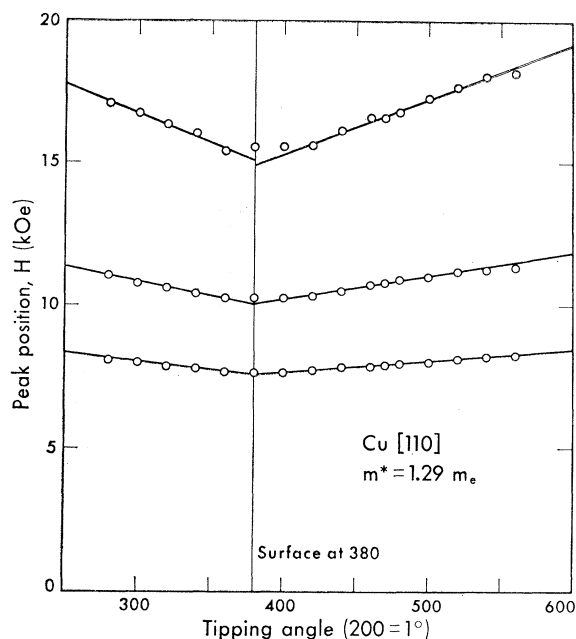


FIG. 2. Plot of the derivative peak positions in Cu versus the angle of field tipping for the second, third and fourth subharmonics of the  $m^* = 1.29m_e$  resonance at the  $[110]$  axis.

produce a more nearly flat surface. A 60-40 phosphoric acid-distilled water electropolishing solution was used. The sample was mounted with its flat surface upward and accurately horizontal, and the current density was adjusted empirically to give the best possible mirror surface. The optimum current density was about 50 ma/cm<sup>2</sup>. Effects due to the over-all rounding of the surface caused by the extensive electropolishing were minimized by exposing only a small central portion of  $\frac{1}{4}$ -in. diam to the microwaves. The diameter of the entire sample was nearly  $\frac{3}{4}$  in.

In order to understand better the possible effects of the remaining departure from perfect flatness of the Cu sample, we have also made measurements on optically flat oriented Sn single crystals. These samples were grown between polished quartz plates, and the surfaces were not treated. The resistivity ratio for such Sn samples was on the order of 100 000.

### III. EFFECTS OF FIELD TIPPING

The results of our experiments on the effects of tipping the magnetic field with respect to the sample surface readily fall into two categories; the regime of small angles of tip (a small fraction of a degree to about one or two degrees from the surface) and the regime of arbitrarily large angles. We discuss these two regimes separately below.

#### A. Small-Angle Tipping

The effects of small-angle tipping of the field with respect to the sample surface have been studied for a

number of orientations in the  $(110)$  plane of Cu. We show in Fig. 2 the effect of tipping on the peak positions of a subharmonic series found with the field in the  $[110]$  direction. This series exhibits many of the characteristic features of the tipping experiments in Cu: the peaks shift rapidly to higher fields as the tip angle is increased from zero, the shift is linear with angle except for the first 10' of arc, and beyond about 2° the signals broaden and decrease in amplitude to such an extent that measurements are no longer possible. The higher order subharmonics are found to be successively less shifted by field tipping. In Fig. 2, the fractional increase of the resonance field over that parallel to the surface ( $\Delta H/H$ ) is about 18% per degree for the second harmonic. The two succeeding subharmonics shift by 11 and 7% per degree of tipping, respectively. The amplitude decrease and broadening with increasing tip angle is less pronounced for the higher subharmonics. In contrast to these results at the  $[110]$  axis, we have also observed resonances for which the peak position shifts are to lower rather than higher fields. Some other series exhibit no appreciable shift with tipping angle. Most of the peak positions shifts that we have observed for the fundamental resonance are of the order of 10 or 20% per degree of tipping. (We have observed an even more pronounced shift of 40% per degree for the second harmonic of a limiting point resonance<sup>8</sup> at the  $[001]$  axis in Sn. This measurement was made at 35 kMc/sec and the series of subharmonics for this resonance could be observed only over the first 15' of tipping angle.)

The symmetry in the observed shifts of the derivative peaks for positive and negative tipping angles allows accurate alignment of the magnetic field to the sample surface. When the field is accurately parallel to the surface, the derivative peak positions are strictly periodic in  $1/H$ , and the plot of  $1/H$  versus the index number of the subharmonic forms a straight line through the origin. Thus, for the accurately parallel geometry there are no appreciable phase shifts, although for small  $\omega\tau$  values the calculations of KLM predict small phase shifts. As we believe that the  $\omega\tau$  values for our samples are much greater than 30, phase shifts from this cause should be negligible. In fact, we could detect no phase shift greater than 0.02, and even this figure was not reproducible and was probably caused by the scatter in experimental points. (In contrast to this, KLM reported phase shifts of up to 0.3.) This is not generally the case for measurements made with tipped fields. When the field is tipped, the relative displacements in field for the various subharmonics results in a relationship which is not strictly periodic in  $1/H$ , and it is impossible to fit the data points perfectly to a straight line. As in Fig. 2 the effect of tipping is to shift the reso-

<sup>8</sup>The limiting point, in the terminology of Azbel' and Kaner (see Ref. 2), refers to points on the Fermi surface where the tangent plane is normal to the applied field direction. The orbit at this point is vanishingly small and only orbits close to this point contribute to limiting point cyclotron resonance.

nance field for the fundamental by the greatest fractional amount and to shift the fields for the corresponding higher order subharmonics by successively smaller amounts. The fields for the very highest order subharmonics observable are found to shift negligibly. As a consequence of these shifts a plot of the data points ( $1/H$  versus index number of the subharmonics) when the field is tipped will show the point for the fundamental shifted by the greatest fractional amount from the straight line formed when  $H$  is parallel to the surface. The data points for successively higher order subharmonics approach the straight line asymptotically. For any straight line fitted to these data points there will be a considerable and consistent deviation of the points from the line.

The observed shifts cannot be accounted for by a mere change in the cyclotron mass. A simple change in cyclotron mass would require that the fractional changes in the peak positions of all harmonics be identical. Any attempt by the least-mean squares method to fit a straight line to the tipped field data will usually result in a somewhat different slope than in the parallel field case, and the line will fail to go through the origin, i.e., a phase shift will result. The cyclotron mass and the phase shift determined from the slope and intercept of the straight line, respectively, will depend sensitively on the number of harmonics available for determining the straight line.

In view of the fact that the  $1/H$  plot versus harmonic numbers for the tipped field departs considerably from a straight line and the consequent difficulties in the interpretation in terms of cyclotron mass, we have chosen to discuss the effects of tipping in terms of peak position shifts. Khaikin<sup>9</sup> has reported a study of the dependence of the cyclotron resonance in Sn on the angle of tipping, and does interpret his data in terms of a shift in  $m^*$  without regard for the fact that the various subharmonics are not strictly periodic in  $1/H$  when the field is tipped. We have repeated his experiments in Sn and find it impossible to fit these tipping data to straight lines.

For the small-angle tip measurements the flatness of the sample surface is of great importance. The measurements are meaningful only at such angles of tip that exceed some average angle due to a possibly wavy or rounded surface that results from electropolishing. Even though a major improvement has been made in the flatness of the Cu crystal surface, there remains an appreciable curvature of the surface. Some measure of the influence of this curvature on our results has been obtained by comparison of our measurements on Cu with those on optically flat Sn samples where peak position shifts depart from linearity only at angles below about  $1'$  or  $2'$  of arc from the surface. Even this remaining departure could be explained as due to experimental factors other than surface flatness. In contrast to these

<sup>9</sup> M. S. Khaikin, Zh. Eksperim. i Teor. Fiz. 42, 27 (1962) [translation: Soviet Phys.—JETP 15, 18 (1962)].

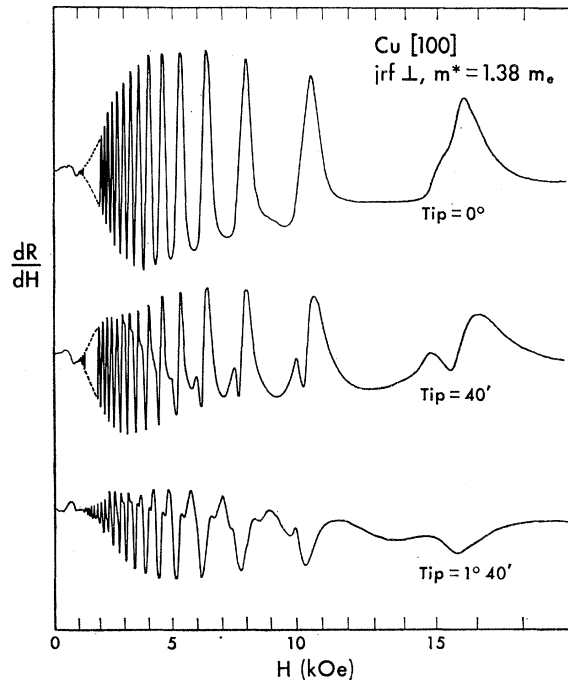


FIG. 3. Cyclotron resonance signals at the [100] axis in Cu for three angles of field tipping. With increased tipping the various subharmonics split and sharp derivative minima appear. The high-order subharmonics are less affected by tipping the magnetic field, than the fundamental or first few subharmonics.

observations in Sn, the peak-position shift data in Cu depart from linearity below a full  $10'$  of arc. We attribute this difference to the residual curvature of the Cu sample surface. Comparison of the field values for peaks measured at zero angle of tip with a straight line extrapolation of values measured at finite tip angles shows that the error due to remaining surface curvature can be 2 or 3% for the second harmonic in the most tip-sensitive orientations. This difference decreases considerably with increase in the number of the subharmonic, so that the cyclotron mass determined from the periodicity of some 6 to 10 harmonics should not be in error by more than 1% because of the residual curvature of the Cu surface.

In addition to the harmonic series plotted in Fig. 2 and discussed at length above, we observe another series with somewhat lower cyclotron mass in the same [110] direction. Subharmonics of the latter series show relatively little sensitivity to field tipping. Over the same range of angles as in Fig. 2 peaks are shifted by at most 2 or 3%.

The effect of field tipping when the field is along the [100] axis is more complicated. In Fig. 3, the resonance signal is shown for three angles of field tipping. As the field is tipped from the surface, the harmonic peaks split into two series with one series moving to lower fields and the other to higher fields. The behavior of the peaks of each series is similar in character to that observed along the (110) axis. In particular, the shifts are linear with angle of tip, the peaks broaden with increasing angle,

and the higher order harmonics are less affected by tipping. Additional features evident in Fig. 3 are the development of sharp derivative minima as the peaks split, and the presence of a small hump on the low-field side of the second harmonic when the field is as parallel to the surface as is possible with the present Cu sample. It is not clear whether or not this hump would disappear with a strictly flat sample surface. The data may be complicated by the presence of signals from rosette orbits or from a  $k_H \neq 0$  mass extremum, and a complete explanation of all the details has not been possible. However, the splitting and broadening of peaks can be interpreted in terms of a simple Doppler shift theory as outlined in a latter part of this section.

With the field along the [111] direction the data in Cu are practically insensitive to field tipping. There are no appreciable peak position shifts due to tipping for either the belly orbit or the neck orbit.

We have studied the frequency dependence of the peak position shifts in Cu and Sn by comparing results at 35 and 67 kMc/sec. We find that with increased frequency, the shifts are less pronounced. The slope of the fractional change in the field of a given subharmonic plotted against tipping angle is found to decrease with increased frequency approximately as the two-thirds power of the frequency. The measurements to date are not wholly reliable and are complicated by the fact that the resolution of harmonics changes with frequency, as well as by the fact that the slopes are sensitive to the orientation of  $H$  in the surface and that this may have differed by a few degrees for these scaling experiments. It is not impossible that various orbits will give somewhat different scaling results.

The variety of experimental observations on the effects of field tipping is difficult to fit under one theoretical roof. We should expect that with  $H$  tipped relative to the surface, carriers with a finite average velocity parallel to  $H$  (i.e.,  $\bar{v}_H \neq 0$ ) will contribute differently to the resonance than will stationary orbits. One reason for this is that such carriers, which one successive traversals of the surface region appear at different depths, will see an effectively Doppler shifted frequency<sup>10,7</sup> because the rf field changes both in amplitude and phase with depth into the metal. For an electron with a given value of  $\bar{v}_H$ , this mechanism would predict both a shift and line broadening that are linear with angle of tip. Using the expression for the rf field in the normal skin effect regime, the expected shift in magnetic field for small angles of tip  $\theta$  is given by

$$\frac{\Delta H}{H} \approx \frac{\bar{v}_H \theta}{\delta \omega} \quad (1)$$

where  $\delta$  is the skin depth for penetration of rf fields,  $\omega$  the experimental frequency. This equation is equivalent to one derived by Miller and Haering<sup>10</sup>. The spectrum of frequencies seen by the drifting electron will be a Lorentzian centered about the Doppler shifted fre-

quency. This mechanism would serve well to explain the linear shift and broadening observed for the limiting point resonance in Sn where  $\bar{v}_H$  is essentially constant for all electrons contributing to the resonance. However, when dealing with orbits about an external cross-sectional area of the Fermi surface, there is a distribution of orbits having increasing values of  $|\bar{v}_H|$  about the central  $\bar{v}_H = 0$  orbit. In this case the simple Doppler shift theory would predict a line broadening but not a shift in peak position. However, particular sections of certain Fermi surfaces may produce line splitting and linear peak shifts. These sections must have stationary ( $\bar{v}_H = 0$ ) orbits away from the extremal  $k_H = 0$  region and slowly varying mass functions in the intermediate  $k_H \neq 0$  regions. A plot of  $\bar{v}_H$  against  $k_H$  for such a surface will have  $\bar{v}_H = 0$  at  $k_H = 0$  and, also, at two values of  $k_H$  symmetrically placed on either side of  $k_H = 0$ . Close to  $k_H = 0$  the plot should be linear. In the intermediate regions  $\bar{v}_H$  must have a maximum value on one side of  $k_H = 0$  and a minimum value on the other side. When the field is tipped, similar maximum and minimum values will be shown in a plot of Doppler shifted resonance fields [given by Eq. (1)] against  $k_H$  provided that the mass variation is small. In this case the strongest absorption should occur at fields close to the maximum and minimum values as this is where the most electrons are at resonance. Thus, two peaks should occur, and these peaks should move linearly with angle of tip as predicted by Eq. (1). In fact, we have observed very striking examples of this behavior with resonance series arising from sections of Fermi surface which should fulfill the required conditions. These sections are the fourth-zone hole surface in Sn (Khaikin's<sup>11</sup> "barrel-shaped" surface) which is observed with the field along the [001] axis, and the [100] belly orbit section in Cu (see Fig. 3 and earlier paragraphs).

In general, the Doppler-shift mechanism does predict a frequency scaling such that the fractional changes in peak positions decrease as the two-thirds power of the experimental frequency. This is in good agreement with the results of our frequency scaling experiments.

The Doppler shift ideas outlined above cannot readily be applied to explain the fact that higher order harmonics are less sensitive to field tipping, nor does it account for the observation of peak shifts unaccompanied by splitting as seen with the field along the [110] direction in Cu. In order to reconcile theory with all the experimental data a number of additional factors may have to be considered.

One possibility is that the application of a magnetic field may alter the electric field distribution within the metal with the result that the skin depth  $\delta$  in Eq. (1) need no longer be constant. This has been demonstrated by the theoretical work of Azbel<sup>12</sup> and the experimental

<sup>11</sup> M. S. Khaikin, Zh. Eksperim. i Teor. Fiz. 41, 1773 (1961); *ibid.* 43, 59 (1962) [translations: Soviet Phys.—JETP 14, 1260 (1962); 16, 42 (1963)].

<sup>12</sup> M. Ya. Azbel', Zh. Eksperim. i Teor. Fiz. 39, 400 (1960) [translation: Soviet Phys.—JETP 12, 283 (1961)].

<sup>10</sup> P. B. Miller and R. R. Haering, Phys. Rev. 128, 126 (1962).

studies of Gantmakher<sup>13</sup> which have shown that a magnetic field applied parallel to the surface may cause orbiting electrons to transport current into the bulk of the metal. Under certain conditions a strong electric field may appear at a depth greater by two orders of magnitude than the normal skin depth. A further complication arises when the magnetic field is tipped, as electrons for which  $\bar{v}_H \neq 0$  will now transport current into the bulk by their drifting motion. Grimes<sup>14</sup> has argued that this could affect the surface impedance in such a way as to cause the splitting and inversion of peaks which is observed in Na and K<sup>15</sup>. When the field is applied perpendicularly to the surface, Miller and Haering<sup>10</sup> have shown that the skin depth may also be increased drastically.

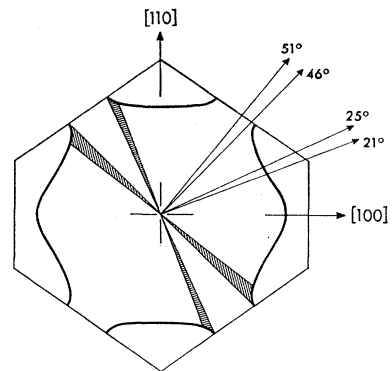
A final consideration is that the contribution to the resonance from carriers moving in the field direction may be diminished over that from stationary orbits when the field is tipped. This is because such carriers are eventually removed from the surface and, therefore, may have a lower lifetime and  $\omega\tau$  than electrons in stationary orbits.

Although these several factors affecting the position and shapes of cyclotron resonance lines have not yet been applied successfully to explain the details of our observations on the effects of field tipping in the small-angle regime, they do give a qualitative picture of at least some aspects. Our experimental observations on the effects of field tipping have prompted us to make the necessary changes and improvements in the experimental techniques so that field tipping can be avoided, and reliable and meaningful data can be obtained. On the basis of our new experiments we are in a position to estimate the accuracy of our results as well as to explain the origin of phase shifts and errors in the effective mass data of the earlier work in Cu. In particular it is now clear that the phase shifts resulted from fitting a straight line to data points that are not always strictly periodic in  $1/H$  because of tipping effects. We also believe that some of the kinks in the KLM mass anisotropy curves resulted from a change in the number of harmonics available to determine the straight line. As was discussed earlier in this section, both the slope and intercept of the straight line fitted to the data when the field is tipped are strongly sensitive to the number of data points used. This would also serve to explain the strong correlation of kinks in the phase and mass anisotropy curves in the earlier work.

### B. Cyclotron Resonance with $H$ at Large Angles to the Surface

The new experimental arrangement of sample and microwave cavity has allowed us to search for cyclotron resonance signals with the magnetic field tipped to arbitrarily large angles with respect to the crystal sur-

FIG. 4. Ranges of angles and corresponding orbits in Cu for which the signal amplitude of large-angle cyclotron resonance is unusually large. These angles are contained in the (110) plane normal to the (110) sample surface.



face. We have found reasonably strong, well-resolved series of subharmonics over large ranges of solid angle. Signal amplitudes in some orientations are comparable to those for signals observed with  $H$  parallel to the surface. Usually such resonances at large angles are strictly periodic in  $1/H$  and do not exhibit large phase shifts. We have observed two-zone extended orbits, as well as orbits contained in a single zone. The cyclotron masses determined in the large angle geometry are generally different by some 5% or so from those measured in the surface in an equivalent crystallographic direction. The observation of cyclotron resonance perpendicular to, or at large angles to, the surface has previously been reported by Koch and Kip<sup>16</sup> in Sn and by Everett<sup>17</sup> in Bi.

The discrepancy between cyclotron masses obtained with  $H$  in the sample surface and those measured with large angles of tip is not understood. At large angles it might be expected that the electron orbits contributing to the resonance would be limited to a very narrow range of  $k_H$  about the value of  $k_H$  for a stationary orbit. This measurement should then give most accurately the mass of the orbit about the extremal cross-sectional area. In contrast to the large-angle cyclotron resonance, the signals parallel to the surface could be due to orbits over a much larger range of  $k_H$ , and the measured cyclotron mass therefore an average value of masses near the extremal value. One should then expect the mass anisotropy curves of the large angle measurements to fit Roaf's analysis even better than the parallel field results. Contrary to this expectation the effective mass curves for tipped fields show rapid breaks in the slope and extra kinks that would not fit the model.

The large angle measurements can, however, give detailed geometrical information about the Fermi surface. The amplitude of the signals in the (110) plane that is normal to the (110) sample surface becomes unusually large over the range of angles indicated in Fig. 4. The amplitude is increased sharply by a factor of 4 or 5 over that for neighboring directions. The stationary orbits corresponding to these angles are those immediately adjacent to the neck. Since the amplitude of the large

<sup>13</sup> V. F. Gantmakher, Zh. Eksperim. i Teor. Fiz. 43, 345 (1962) [translation: Soviet Phys.—JETP 16, 247 (1963)].

<sup>14</sup> C. C. Grimes (private communication).

<sup>15</sup> C. C. Grimes and A. F. Kip, Bull. Am. Phys. Soc. 8, 247 (1963).

<sup>16</sup> J. F. Koch and A. F. Kip, Phys. Rev. Letters 8, 473 (1962).

<sup>17</sup> G. E. Everett, Phys. Rev. 128, 2564 (1962).

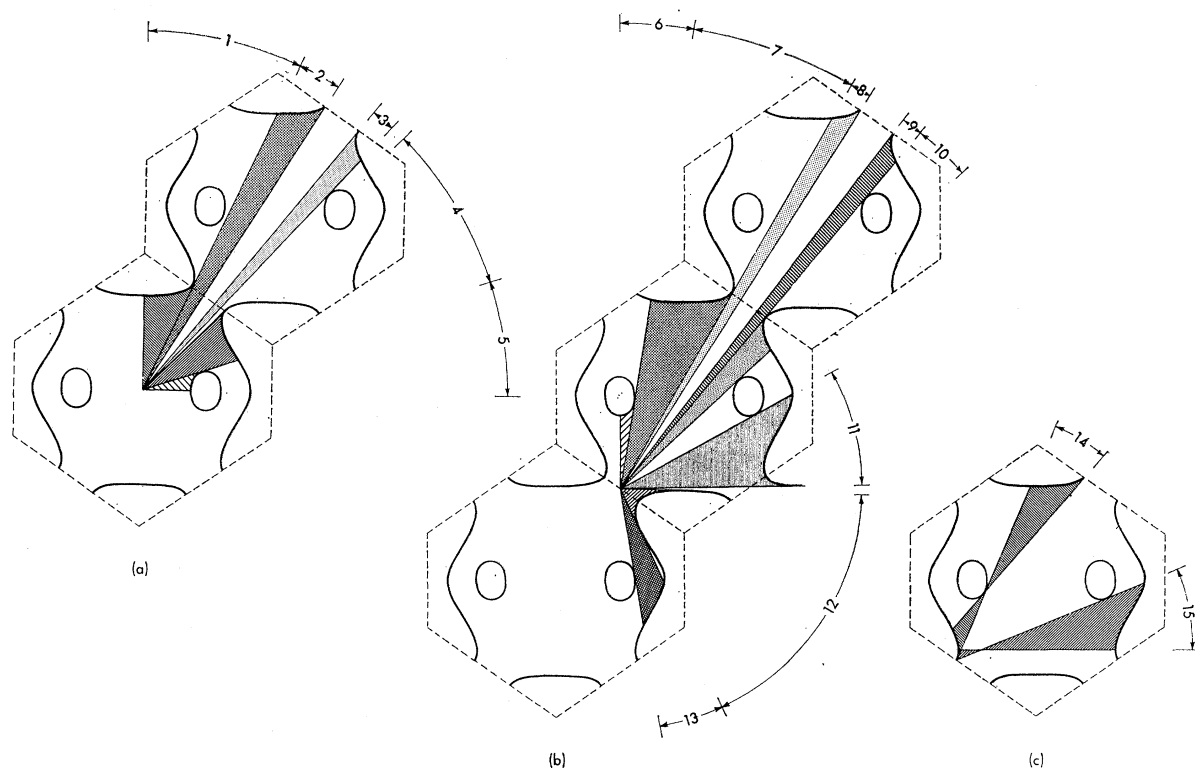


FIG. 5. Projections of the Fermi surface of Cu on the (110) plane showing regions of cyclotron orbits with extremal mass. The lines are the projections of the orbits at the limiting angles, and the shading shows the range of allowed angles for each type of orbit. When an electron is performing a particular orbit, the steady magnetic field is normal to the plane of the orbit. For the sake of clarity only half of each orbit is shown in Figs. (a) and (b). Figure (c) shows orbits whose centers are neither at the zone center nor at the zone edge.

angle signals should be directly proportional to the number of effectively stationary electron orbits, we interpret the signal enhancement as due to an increase in the number of stationary electrons because of the perturbation caused by the necks on the Fermi surface. Equivalently the requirement for large signal amplitude at large angles is that the cross-sectional area remain reasonably constant for some range of  $k_H$  about the extremum in the cross-sectional area, or that  $d^2A/dk_H^2$  be small at the extremum. The necks on the Cu Fermi surface do in fact achieve this. The limiting angles for the large amplitude regions have proven to be a sensitive measure of the neck diameter. From the values of these limiting angles noted in Fig. 4, we conclude that the angle subtended by the neck at the center of the Brillouin zone is less than or equal to  $21^\circ$ .

#### IV. CYCLOTRON MASS RESULTS AND THEIR INTERPRETATION

In this section we will summarize the results of cyclotron mass data arising from the many different types of orbits on the Cu Fermi surface. A number of these orbits have already been observed and discussed by KLM. However, in addition to allowing a more accurate determination of the effective masses, our improved experimental resolution and technique have

permitted the discovery of a number of new types of orbits. The planes of all the orbits which have been positively identified are shown in Fig. 5. The Fermi surface shown in this diagram is that derived by Roaf<sup>4</sup> to fit Shoenberg's<sup>5</sup> dH-vA data on Cu. The range of magnetic field directions where the different cyclotron orbits could be observed is in excellent agreement with those predicted by this surface.

The nomenclature developed by KLM and also by Shoenberg<sup>5</sup> has been used to describe the different types of orbits shown in Fig. 5. Groups 1 and 4 are "belly" orbits, whereas groups 7, 10, and 11 are "mass two" extended orbits, all of which were observed by KLM. In addition to these mass-two series, orbits which extend through more zones have been discovered during the recent experiments. In Fig. 5 these are groups 2 and 3 which extend through three complete zones, and groups 8 and 9 which pass through four zones. Orbits which pass through even more zones, and also open orbits which pass through an infinite number of zones, should exist when the steady magnetic field is close to  $35^\circ$  from the [100] axis. However, none of these orbits were observed, either because their intensities were so weak as to make the series undetectable, or because of a slight misorientation of the sample surface. Groups 5 and 6 are negative mass orbits, 5 being the dog's bone and 6, the four-

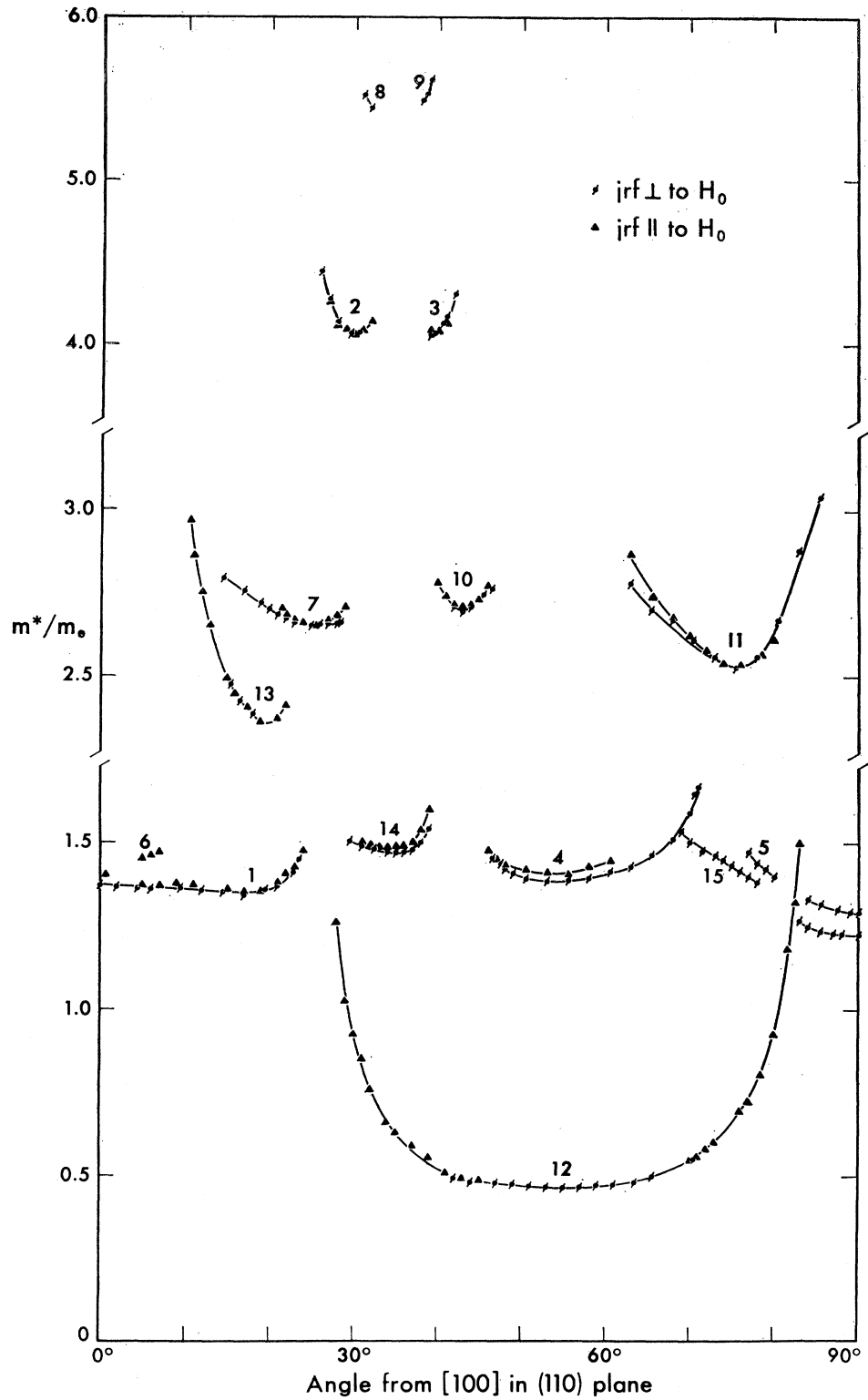


FIG. 6. Experimental data of the cyclotron effective mass in Cu with the steady magnetic field in the (110) plane.

cornered rosette. Group 12 is composed of neck orbits, whereas mass-two orbits make up group 13 which may be considered as an extension of the group-12 neck orbits.

Groups 14 and 15 are orbits whose centers are neither at the zone center nor at the zone boundary. As orbits of this type have not been identified previously in metals, although similar orbits have been observed in semi-



TABLE I. Cyclotron masses in Cu.

Orbit	Angle of $H$ from [100] axis (deg)	Cyclotron mass $m^*/m_e$	
		$j_{II}$	$j_{II}$
(A) Masses derived from cyclotron resonance experiments <sup>a</sup>			
1. Belly	0	1.37 ± 0.005	...
	18	1.34 ± 0.005	1.35 ± 0.005
2. 3 zone extended	30	4.07 ± 0.01	4.06 ± 0.01
3. 3 zone extended	39½	4.07 ± 0.01	4.08 ± 0.01
4. Belly	55	1.385 ± 0.005	1.41 ± 0.005
5. Dog's bone	90	1.29 ± 0.005	...
5. $k_H \neq 0$ belly		1.225 ± 0.005	...
6. Rosette	5	...	1.45 ± 0.005
7. 2 zone extended	25	2.65 ± 0.01	2.65 ± 0.01
8. 4 zone extended	31½	5.44 ± 0.03	...
9. 4 zone extended	38	5.48 ± 0.03	...
10. 2 zone extended	43	2.69 ± 0.01	2.71 ± 0.01
11. 2 zone extended	76	2.53 ± 0.01	2.54 ± 0.01
12. Neck	55	0.46 ± 0.02	...
13. 2 zone extended	19	...	2.36 ± 0.01
14. $k_H \neq 0$ belly	35	1.47 ± 0.005	1.485 ± 0.005
limiting point	0	...	0.48 ± 0.05
(B) Theoretical masses obtained by Segall <sup>b</sup> $m^*/m_e$			
1. Belly	0	1.1 ± 0.1	
5. Dog's bone	90	1.12 ± 0.06	
12. Neck	55	0.41 ± 0.02	
(C) Masses derived from de Haas-van Alphen <sup>c</sup> measurements			
1. Belly	0	1.28 ± 0.07	(1.1–2.0°K)
4. Belly	55	1.18	(1.1–1.7°K)
4. Belly	55	1.48	(1.8–2.4°K)

<sup>a</sup> The errors quoted for the experimentally determined cyclotron masses are derived from the spread in experimental points, and do not include any systematic errors arising from tip effects caused by the residual surface roughness of the sample. Such errors do not exceed 1%. Also given with the de Haas-van Alphen masses is the temperature range over which the particular experiment was performed.

<sup>b</sup> See Ref. 19.

<sup>c</sup> See Ref. 5.

conductors,<sup>18</sup> it is instructive to consider the reasons for their appearance as distinct series in the cyclotron resonance spectrum. If the mass of orbits in the relevant region of the Fermi surface is plotted against  $k_H$  for a particular direction of the magnetic field, then the mass becomes infinite for the two values of  $k_H$  which have orbits whose planes touch the neck. In between these values of  $k_H$  the mass must have a minimum value, which implies that a considerable number of electrons will have approximately the same mass and will be capable of producing a mass series. The mass becomes infinite when an orbit plane touches a neck because at this point the electron velocity has no component along the orbit path (since  $v = 1/\hbar \text{grad}_k \epsilon$ ), and the electron then takes an infinite time to complete the orbit. In general, this will happen whenever the plane of the orbit is a tangent plane to the Fermi surface, the only exception being for a limiting point resonance. In the latter case the cyclotron period remains finite as both the path length and the orbital velocity approach zero together. In addition to the groups of resonances shown in Fig. 5,

<sup>18</sup> D. M. S. Bagguley and R. A. Stradling, Proc. Phys. Soc. (London) **78**, 1078 (1961).

we believe that we have observed a limiting point resonance when the magnetic field was close to the [100] direction.

The experimentally observed masses are shown in Fig. 6 as a function of the magnetic field orientation in the (110) plane. Table I gives the cyclotron masses obtained with the magnetic field along certain directions together with the masses derived theoretically<sup>19</sup> and from dH-vA measurements.<sup>5</sup> In Fig. 6 different symbols are used for masses obtained with different modes of polarization. In general it is seen that as the magnetic field orientation is varied, the cyclotron mass of each group goes through a minimum value, and is increasing when the series disappears. From the argument outlined in the previous paragraph it is obvious that the cyclotron mass should be infinite at the limiting angle. However the series becomes too weak to be detectable before this occurs. It, therefore, follows that the range of angles over which any mass series is observed experimentally will be slightly less than the range allowed by the Fermi surface. Table II lists the experimentally determined range of angles together with those predicted by the Shoenberg-Roaf Fermi surface. Of particular importance in determining these angles is the size of the neck region. Also given in Table II are the limits on the angular size of the neck that can be deduced from each group of measurements. The angle quoted is that subtended at the center of the zone by the neck which is assumed to be circular. From our results it appears that this angle is between 19° and 21°. This conclusion agrees well with the angles deduced from other types of experiment. A neck size of 21° was obtained from dH-vA<sup>4</sup> measurements, whereas magnetoacoustic experiments have suggested angles of<sup>20</sup> 17½° and 20°. In general the

TABLE II. Range of angles over which different types of cyclotron orbit in Cu may be observed.

Orbit	Experimental angles (deg)	Angles from Shoenberg-Roaf surface (deg)	Information about neck size from cyclotron resonance measurements
1. Belly	0–24	0–25	<23°
2. 3 zone extended	26–32	25–32	>19°, <21°
3. 3 zone extended	39–42	38½–43	<24°
4. Belly	46–71	45½–71½	<22°
5. Dog's bone	77–90	71½–90	...
6. Rosette	5–7	0–10½	...
7. 2 zone extended	14½–29	10½–30	<25°
8. 4 zone extended	31–31½	30–32½	>17°, <26°
9. 4 zone extended	38–39	37½–40	>17°, <25°
10. 2 zone extended	40–46½	40–48	<21°
11. 2 zone extended	63–85½	62–88	<25°
12. Neck	28–83	26–87	...
13. 2 zone extended	10½–22	10½–26	<21°
14. $k_H \neq 0$ belly	29½–39	25–41	...
15. $k_H \neq 0$ belly	70–90	68–90	...

<sup>19</sup> B. Segall, Phys. Rev. **125**, 109 (1962).

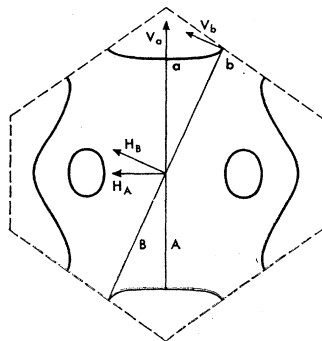
<sup>20</sup> R. W. Morse, in *The Fermi Surface*, edited by W. A. Harrison and M. B. Webb (John Wiley & Sons, Inc., New York, 1960), p. 214.

agreement between our results and the predictions of the Shoenberg-Roaf surface is excellent.

Further confirmation of the limiting angles was provided by the large angle cyclotron resonance experiments discussed in Sec. III. This can be a particularly useful method of observing resonances close to the limiting angles in copper because the signal intensity may increase suddenly in this region. For example, in the conventional geometry, the signals from the belly orbits (1 and 4) are weak close to the limiting angles and are dominated by other stronger series; whereas in the tipped geometry, these signals become very large when the magnetic field is between 21° and 25° and between 46° and 51° from the [100] axis in a (110) plane. It is thought that between these angles the perturbation of the neck regions on the Fermi surface produces a large number of electrons with little or no net drift velocity along the magnetic field, thus, ensuring that most of the electrons in this region of the Fermi surface return to the skin depth after completing several cyclotron orbits.

It is also worth mentioning that in the conventional geometry the parallel mode of polarization of the rf electric field (see Sec. II) often produces the strongest signals when the magnetic field is close to a limiting angle. This is because the maximum signal amplitude occurs when the electric field is parallel to the velocity of the electron when it is in the skin.<sup>21</sup> If the point of the orbit which is within the skin depth in real space is near a neck region in momentum space, then most of the electron's velocity will be along the magnetic field, and hence, the signal amplitude will be greater in the parallel mode. This conclusion follows even though the electrons concerned have no net drift velocity along the magnetic field. An example of this is given in Fig. 7 which shows belly orbits when the magnetic field is along the [100] direction (orbit A), and along the limiting angle (orbit B). Points *a* and *b* are the points on the orbits where the electron is within the skin depth with a (110) sample surface. It can be seen that at point *a* the electron velocity has no component along the field, whereas at point *b* all the velocity is parallel to the field. Experimentally, when the mag-

FIG. 7. Projection of Fermi surface in Cu on (110) plane showing belly orbits when the magnetic field is along the (100) direction (orbit A) and along the limiting angle (orbit B). The figure gives the direction of the electron velocity when it is within the skin depth.



<sup>21</sup> J. F. Koch and A. F. Kip, Bull. Am. Phys. Soc. 7, 477 (1962).

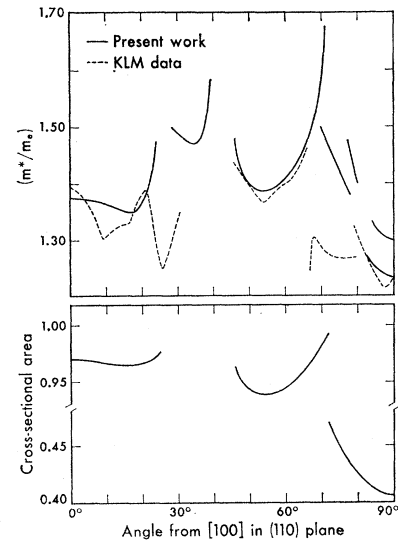


FIG. 8. Comparison of Cu cyclotron-mass values obtained in the present work with those measured by KLM. The resonance series concerned are those with mass ratios between 1.2 and 1.7. Also shown, are the cross-sectional areas of the copper Fermi surface derived from de Haas-van Alphen measurements of Shoenberg.

netic field was along a [100] direction, the signal amplitude in the parallel mode was only a few percent of that in the perpendicular mode, and even this fraction could be attributed to the lack of perfectly linear rf polarization (see Sec. II). However, close to the limiting angle the parallel mode signal was stronger and persisted over a wider range of angles, in agreement with the above argument.

It is interesting to note that there are differences of the order of a percent between the masses measured with the two modes of polarization. This is very obvious for groups 4 and 14 in Fig. 6. As these particular masses are insensitive to the effects of magnetic field tipping, it is unlikely that the differences arise from the residual roughness of the sample surface (see Sec. II). We believe that the discrepancies occur because every cyclotron resonance series contains contributions from electrons which have different values of mass and  $k_H$ , and also different velocity components within the skin. The cyclotron mass observed has some average value for this group of electrons. Each polarization will weight the contributions from the individual electrons according to their velocity components in the skin, emphasizing those with the greatest velocity along the rf electric field. Therefore, it is not surprising that the observed masses differ slightly. Studies of these differences coupled with a knowledge of the geometrical shape of the Fermi surface may show whether a mass extremum is a maximum or a minimum.

The majority of the data obtained from the present work has already been presented in Fig. 6 and Tables I and II. The remainder of the discussion will be confined to points of interest concerning individual mass series. A comparison of our work with that of KLM demonstrates the importance of preparing a sample with a flat surface, and of having the magnetic field carefully aligned with this surface. Figure 8 illustrates

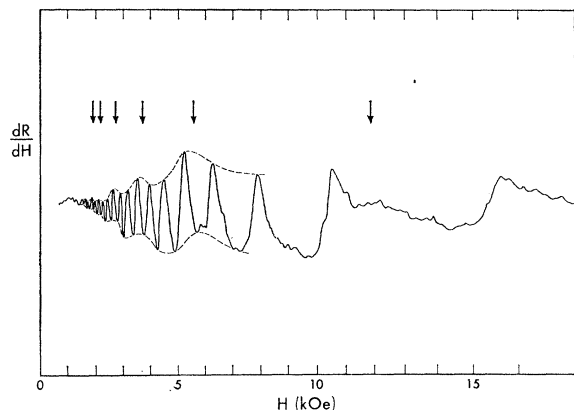


FIG. 9. Experimental recording of derivative absorption in Cu with  $H$  along the (100) axis and with  $j_{rf}$  parallel to  $H$  showing how the limiting-point-resonance series (identified by arrows) modulates the envelope of the belly-orbit series.

this point. Here our results for series with mass ratios between 1.2 and 1.7 are compared with those of KLM. In addition this figure shows the cross-sectional areas calculated for the belly and dog's bone orbits by Roaf.<sup>4</sup> It is interesting that the mass variations for both belly-orbit regions have a similar appearance to the cross-sectional area variations. The masses change more rapidly with magnetic field orientation, however. This is expected as the masses should become infinite at the limiting angles, whereas the areas should remain finite.

The belly orbit series 1 shows the most striking discrepancies compared to the results of KLM as is seen in Fig. 8. It is uncertain why KLM observed belly masses beyond the limiting angle of  $25^\circ$ , but it is possible that a  $k_H \neq 0$  series (orbit 14) was identified as the belly series 1. The best agreement between the two sets of data is for the belly orbit 4. This is to be expected as this series was very insensitive to tipping effects. Close to the [110] axis comparisons are difficult as we were able to resolve another mass series in this region.

The smoothness of our experimental mass variation and the lack of any phase shift in a plot of harmonic peak positions (see Sec. III) leads us to believe that any errors in our masses introduced by the surface roughness of the sample are small, probably of the order of a percent in the most tip-sensitive region. In such a region close to the [100] axis the first few harmonics of the belly series had structure on the low-field side of the resonance lines (see Fig. 3). On tipping the magnetic field, the structure became resolved into a separate series as discussed in Sec. III. We are uncertain whether this structure is due to the residual roughness of the surface, or whether it is a real mass series which would be observed with an optically flat surface. In the latter case the series probably arises from  $k_H \neq 0$  belly orbits with an extremal mass. The position on the low field side of the main series would be consistent with this hypothesis, as it is reasonable to think of a plot of  $m^*/m_e$  against  $k_H$  having a minimum value somewhere away

from  $k_H \neq 0$  before increasing to infinity at the value of  $k_H$  where the orbit runs into the neck. It seems unlikely that such a plot would have a region of sensibly constant mass above the  $k_H = 0$  value. It was this argument which caused us to assign mass series 6 to the four-cornered rosette, as the masses observed are larger than the belly masses. Series 6 was observed over only a small range of angles close to the [100] direction, and only with the parallel mode of polarization. Inspection of Fig. 5 shows that an electron performing a rosette orbit away from the symmetry direction has a considerable component of its velocity along the magnetic field when it is within the skin depth, and hence, should be observed with parallel polarization.

In addition to the series discussed above, when the field was within  $5^\circ$  of the [100] axis, we have observed a weak and broad resonance series with the parallel mode of polarization which we believe to be due to a limiting point resonance. In this type of orbit the electron is moving with nearly all its velocity along the magnetic field, and hence, should be observed only with parallel polarization. A typical experimental recording taken with this mode of polarization and with the field along the [100] direction is shown in Fig. 9. The strong series in the recording arises from belly orbits which probably appear because the rf electric field has a small component perpendicular to the steady magnetic field. The fundamental of the limiting point series appears as a broad peak at about 11.5 kOe on the high-field side of the third harmonic of the belly series. The other harmonics do not appear as separate peaks because of their weakness and breadth, but modulate the envelope of the belly-orbit series. Some six harmonics of the limiting point series are seen. The effective mass ratio obtained from this series is  $0.48 \pm 0.05$ . However, this mass may be too large by up to 10% because of the surface roughness of the sample which has the effect of tipping the magnetic field with respect to much of the surface. Because of the large values of the velocity along the magnetic field, the limiting point resonance should be extremely sensitive to small angles of tip. In support of this idea, the limiting point resonance, in fact, disappeared when the magnetic field was tipped only  $\frac{1}{3}^\circ$  from the surface position. In a Sn sample with an optically flat surface the limiting point resonance observed when the magnetic field was close to the [001] axis was much more affected than any other series by tip effects, with peaks moving about 10% when the field was tipped  $\frac{1}{4}^\circ$  (see Sec. III). Similarly, in Cu we believe that the peaks of the limiting points series are appreciably shifted and broadened even though the other series are relatively unaffected by the residual roughness of our surface.

Limiting point masses are of particular interest as they give information about small regions of the Fermi surface; whereas larger orbits have masses which are less sensitive to local variations in shape of the surface. In order to determine whether the low mass observed was reasonable for the limiting point resonance ob-

served in Cu, attempts were made to estimate the mass using the expression derived by Shockley<sup>22</sup>

$$m^* = \frac{\hbar^2}{2\pi} \oint \rho \frac{d\varphi}{\partial\epsilon/\partial\rho}$$

where  $(\rho, \varphi, k_H)$  are the cylindrical polar coordinates of a point on the electron orbit, and  $\epsilon$  is the electron energy. Unfortunately, Roaf's<sup>4</sup> analytical expression merely yields the  $k$  vectors which make up the Fermi surface, and is not a true  $\epsilon$  versus  $k$  band structure dispersion relation. It was impossible, therefore, to calculate absolute mass values, but the ratio of limiting point mass to central orbit mass was estimated by calculating the two values of  $\oint \rho d\varphi / (\partial\epsilon/\partial\rho)$  on the assumption that Roaf's coefficients do not depend on  $p$ . Such a calculation yielded a value of 0.3 for the ratio of the two masses which is in approximate agreement with the experimental results indicating that the above assumption may be valid. Estimates of  $\rho / (\partial\epsilon/\partial\rho)$  from Segall's published curves<sup>19</sup> support this ratio. It would appear that the low mass of the limiting point resonance arises from the small radius of curvature of the Fermi surface along the [100] direction. This produces a larger component of the Fermi velocity along the orbital path while leaving the magnitude of the velocity relatively unaffected, thereby decreasing the cyclotron period.

Another mass of interest is that of the orbits around the neck region. KLM failed to obtain the mass value with the field along the [111] direction where the mass should be a minimum as the electrons are performing their most circular orbits; and they had to rely on an extrapolation of their results obtained with the field between 65 and 80° from the [100] direction. During the present experiments, signals from neck orbits have been observed over the complete range of angles from 28 to 83° from the [100] direction. The two sets of data coincide at the angle of 80°, but progressively diverge as the field is moved towards the [111] direction. The present work yields a value of  $0.46 \pm 0.02$  for the neck mass ratio compared with the extrapolated value of 0.6 obtained by KLM and the theoretical value of 0.41 calculated by Segall.<sup>19</sup> The figure of 0.46 was confirmed by a number of additional measurements at 36 kMc/sec, removing the possibility that the mass observed might depend on the experimental frequency. Such a frequency dependence could arise from a decrease with increasing frequency of any electron-phonon contribution to the effective mass.<sup>23,24</sup> The discrepancy probably arose from the poor resolution of the KLM experiments, and is not due to the greater sensitivity of their experiments to the effects of magnetic field tipping as the neck series was found to be tip insensitive.

<sup>22</sup> W. Shockley, Phys. Rev. **79**, 191 (1950).

<sup>23</sup> J. J. Quinn, in *The Fermi Surface*, edited by W. A. Harrison and M. B. Webb (John Wiley & Sons, Inc., New York, 1960), p. 58.

<sup>24</sup> S. Nakajima and M. Warabe, Progr. Theoret. Phys. **29**, 341 (1963).

In general, the signal intensities from the neck orbits were weak, and many harmonics were obscured by stronger series. Often only one or two high-field harmonics could be resolved. However, because of the small mass involved, the neck series usually persisted to low fields where other series had died out; and four or five low-field harmonics would be observed. This was despite the fact, that fewer harmonics were observed for the neck series than for other series. Typically, the highest order harmonic observed at 67 kMc/sec would be the 13th, whereas many other series showed over 30 harmonics. This observation might suggest that the collision time for neck orbits is considerably less than for other orbits, as was predicted by Ziman<sup>25</sup> for certain types of impurity and phonon scattering. However, a study of the width of the fundamental resonance peaks indicates that the collision time for the neck orbits is probably of the same order as that round the belly orbits. The deterioration in quality of the higher order harmonics may be due to strong mass spread effects<sup>1</sup> which can be thought of as destructive interference between the resonance series of electrons performing orbits with different periods. There was no change in appearance of the resonance spectrum on going from 4.2 to 2.5°K indicating that the collision times are determined by impurity scattering.

As the magnetic field is rotated towards the [110] direction, a  $k_H \neq 0$  belly series appears at an angle of 70° and the dog's bone at 77°. However, it was impossible to follow these series continuously through to the [110] direction because of the presence of the neck series and a strong two-zone mass series. We, therefore, were unable to determine which of the masses observed with the field along the [110] direction corresponded to the  $k_H \neq 0$  series and which to the dog's bone series.

In addition to the series discussed in the previous paragraphs, a number of isolated peaks or groups of peaks were observed in the present work, but these could not be identified as belonging to any series. A particularly strong group was seen close to the [111] direction which may arise from six-cornered rosette<sup>5</sup> orbits.

## V. CONCLUSION

It has been shown that the cyclotron resonance absorption spectrum has the form predicted by the Azbel-Kaner theory<sup>2</sup> only when the magnetic field is aligned accurately parallel to the surface of the sample. When the magnetic field is tipped only a small fraction of a degree away from the surface position the absorption peaks may be broadened and shifted in field. Any sample which does not have a very flat surface may show these tip effects, as any surface roughness, such as may be produced by some electropolishing techniques, will tip the magnetic field with respect to certain regions of the surface. The tip effects require further interpretation, but they may arise from a Doppler-shift mechanism.

<sup>25</sup> J. M. Ziman, Phys. Rev. **121**, 1320 (1961).

The magnitude of the shifts in peak position decrease as the experimental frequency is increased, as would be expected from a simple Doppler-shift theory. When the magnetic field is not along the surface of the sample, a plot of reciprocal fields of the absorption peaks against harmonic number is no longer a straight line, and does not extrapolate through the origin. This behavior produced the phase shifts reported in the work of KLM on cyclotron resonance in Cu. The effective masses in their experiments showed strong correlations with the phase shifts observed, and both were very dependent on the magnetic-field orientation.

In the present experiments tip effects have been much reduced by using a higher microwave frequency and improving the electropolish on the sample surface, and by more accurate alignment of the magnetic field to the surface. No phase shifts were detected when the magnetic field was parallel to the surface, and the effective masses obtained were smoothly varying functions of the magnetic field orientation. In addition, the resultant increase in resolution has permitted the identification of signals from many more groups of electrons. These

observations include a limiting-point resonance, orbits which extend through three and four Brillouin zones, and orbits whose centers were neither at the center nor at the edge of the Brillouin zone ( $k_H \neq 0$  orbits). The neck-orbit series was observed for the first time with the magnetic field along the [111] axis, and the mass ratio obtained was  $0.46 \pm 0.02$ , which was considerably lower than the value estimated from an extrapolation of the results obtained previously when the magnetic field was more than  $10^\circ$  away from the [111] direction.

The range of angles over which the individual resonance series were observed is in excellent agreement with those predicted by the Fermi surface computed by Roaf from the dH-vA experiments of Shoenberg. Furthermore, the mass variation and the limiting point mass obtained in the present work are consistent with the Roaf surface, although no connection is established between the absolute mass values obtained from dH-vA and from cyclotron resonance experiments. It appears that the same Fermi surface can be used to describe the results of two very different experiments on a metal.