Magnetic Breakdown Effects in Helicon Propagation in White Tin*†

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The propagation of helicon waves through white tin has been studied at low temperatures and related to topological properties of the Fermi surface. The large-amplitude quantum oscillations observed at 1.2°K as the field was varied arise from magnetic breakdown. They provide a sensitive means of investigating some features of the Fermi surface. Comparisons are made with the results of a recent band-structure calculation by Weisz.

I. INTRODUCTION

PLASMA effects in solids have been the subject of many experimental and theoretical investigations in recent years.[1] In contrast to the well-known skin effect, propagation of audiofrequency electromagnetic waves through a metal can occur when a sufficiently strong magnetic field is applied. The work described here arose out of an interest in the influence of the band structure on such propagation.

The propagation in single crystals of white tin at 4.2°K can be related to some dimensions of the Fermi surface that have recently been calculated by Weisz.[2] At 1.2°K the propagation characteristics were found[3] to depend in an oscillatory way on the magnitude of the magnetic field, just as in the de Haas–van Alphen and Shubnikov–de Haas effects. Further studies have enabled the oscillations to be attributed to oscillations in the magnetoresistance and Hall coefficient arising from magnetic breakdown between the sheets of the Fermi surface in the third and fourth Brillouin zones. The period of the predominant oscillation disagrees with the value deduced by Weisz. In the present case, the identification of the extremal cross section associated with this particular oscillation is clarified by the orientation dependence of the quantum oscillation amplitude, and it appears that the oscillation with period 5.7×10^{-7} G^{-1} was correctly attributed by Gold and Priestley[4] to the δ orbit of the third Brillouin zone sheet of the Fermi surface.

Most of the work described here was carried out in fields less than 15 kG. We were, however, able to study some aspects of the quantum oscillations up to 150 kG at the National Magnet Laboratory, M.I.T.

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[1] *Proceedings of the Seventh International Conference on the Physics of Semiconductors, Paris, 1964*, edited by J. Bok (Academic Press Inc., New York, 1965).
[2] G. Weisz, Phys. Rev. **149**, 504 (1966).
[3] D. A. Hays and W. L. McLean, Phys. Letters **17**, 215 (1965).
[4] A. V. Gold and M. G. Priestley, Phil. Mag. **5**, 1089 (1960).

II. HELICON THEORY

In an isotropic metal in a magnetic field, normal modes of electromagnetic wave propagation along the direction of the field are circularly polarized. The response of the medium to electromagnetic waves is described here in terms of a complex effective conductivity σ. When the mean free path l of the charge carriers and their mean time between collisions τ are, respectively, much less than the wavelength $2\pi/q$ and the period $2\pi/\omega$ of the electromagnetic wave, the relation between current density and electric field is local and the effective conductivity is related to the direct-current resistivity ρ and Hall coefficient A in the presence of the magnetic field H by

$$\sigma_{\pm}=1/(\rho\pm iAH),$$

where the + and − refer to the right and left circularly polarized components, respectively. As far as reliable estimates of l and τ can be made, in the present experiments the condition $\omega\tau\ll1$ is certainly satisfied but ql may be as large as 0.5 at the highest frequencies that were used.

A particular configuration that is of interest from the experimental point of view is that of an infinite sheet of metal with its faces normal to a steady magnetic field (along which is chosen the z axis) and with a much smaller, oscillating magnetic field applied parallel to the faces (along the x axis). The propagation of circularly polarized waves along the steady field direction can then be detected from the presence of oscillating flux in the direction mutually perpendicular to the two applied magnetic fields (i.e., parallel to the y axis).

The solution to Maxwell's equations with appropriate boundary conditions in such a case involves a number of difficulties, some of which have been discussed by Legéndy.[5] In most of the work described here, we have used the solution of Chambers and Jones[6] which, although not exact, agrees well with the experiments in many respects. According to their solution the spatial average of h_y, the instantaneous complex in-

[5] C. R. Legéndy, Phys. Rev. **135**, A1713 (1964).
[6] R. G. Chambers and B. K. Jones, Proc. Roy. Soc. (London) **A270**, 417 (1962).

duction along y, is given by

$$\bar{h}_y=\mu h_{0x}=\frac{4}{(\pi^2)}h_{0x}\sum_n \frac{u}{n^2[1+iQ(\omega/\omega_n-\omega_n/\omega)]}, \quad (1)$$

where h_{0x} is the complex applied oscillating field (along x), $u=AH/\rho$, $Q=\frac{1}{2}(1+u^2)^{1/2}$, $\omega_n=(n^2\pi/4t^2)\times(\rho^2+A^2H^2)^{1/2}$, t is the thickness of the sheet, and the summation runs over all odd positive integers. For ω near ω_1, the transverse permeability μ is sufficiently accurately represented by just the first term of the sum, i.e.,

$$\mu=\frac{4u/\pi^2}{1+iQ(\omega/\omega_1-\omega_1/\omega)}. \quad (2)$$

In an anisotropic solid, the electrical conduction can no longer be characterized by a scalar resistivity and Hall coefficient as above: Instead, the anisotropy must be taken into account by using the full resistivity tensor. Real metals with multiply connected Fermi surfaces are highly anisotropic in their conduction on account of the open cyclotron orbits that can exist for certain orientations of the magnetic field. The effects of the open orbits in a simple model of the Fermi surface on the propagation of electromagnetic waves have been discussed by Buchsbaum and Wolff.[7] Strong damping of helicon waves is expected to occur in an uncompensated metal (density of electrons not equal to density of holes) with open orbits, as has been confirmed by the experiments of Grimes, Adams, and Schmidt[8] on silver. In a metal such as white tin, which is compensated, the effects are more complicated.[7] When the magnetic field is oriented so that firstly, all the cyclotron orbits are closed, and secondly, each sheet of the Fermi surface has either only hole or only electron orbits, the propagating electromagnetic waves in a very pure sample are Alfvén waves and not helicon waves. However, when the magnetic field is along [001] in white tin, although the first condition is satisfied, the second is not and there is a finite Hall coefficient owing to geometric discompensation.[9] Thus in high-purity white tin at low temperatures, helicon propagation can occur with small damping.

Magnetic breakdown in metals[10] can be expected to modify the above considerations in two different ways. Firstly, there are the nonoscillatory effects which reflect the dependence on field of the probability of an electron tunneling from one sheet of the Fermi surface to another. The probability p at a magnetic field H is given by

$$p=\exp(-H_0/H),$$

where H_0, the breakdown field, is related to the Fermi energy E_F and the band gap Δ (associated with the separation of the two sheets of the Fermi surface in the region of k space where tunneling occurs) by

$$\hbar(eH_0/mc)E_F\sim\Delta^2.$$

The tunneling can cause a change in the nature of the electron orbits—for example, from open to closed—thus changing the high-field behavior of the magnetoresistance.[11] In a simple case, breakdown should be easily detectable by its rapid increase at fields near H_0. If, however, the band gap varies considerably over the region of k space where breakdown can occur, the sharpness of the onset will be less clear. The second type of breakdown effect is the oscillatory variation with field of the elements of the conductivity tensor. This arises from electron wave interference effects on relatively small orbits coupled to others by tunneling.[12] It is the oscillatory kind of breakdown effect which is important in understanding the quantum oscillations that were the aspect of main interest in the present experiments.

At high frequencies, when $ql\gg1$, strong damping can occur because of the Doppler-shifted cyclotron resonance effect if the condition $\mathbf{q}\cdot\langle\mathbf{v}_F\rangle=\omega_c$ holds for any electrons at the Fermi surface, where ω_c is the cyclotron frequency and $\langle\mathbf{v}_F\rangle$ the average of the component of the Fermi velocity along the propagation direction. The feasibility of using this effect to determine features of the Fermi surface topology has been discussed by a number of authors.[13]

III. EXPERIMENTAL PROCEDURE

A. Apparatus

Helicon waves were produced by the oscillating field of a primary coil wound with its axis parallel to the large faces of the plate sample. Flux perpendicular to this field was detected by a secondary coil wrapped around the sample with its axis perpendicular to the axis of the primary coil. The secondary voltage was preamplified and then measured with either a vacuum tube voltmeter or a phase-sensitive detector. The primary and secondary coils consisted of 200 and 850 turns of #32 and #40 copper wire, respectively. The two coils and sample were set in varnish and attached to a phenolic block. The rigid assembly was fastened to one of a pair of meshing right-angle aluminum gears the other of which was connected to a vertical thin stainless-steel tube that could be turned from the top of the cryostat, thus rotating the sample and coils about a horizontal axis. Rotation of the sample about a vertical axis was accomplished by turning the cryostat cap within the helium Dewar. Thus the orientation of

[7] S. J. Buchsbaum and P. A. Wolff, Phys. Rev. Letters **15**, 406 (1965).
[8] C. C. Grimes, G. Adams, and P. H. Schmidt, Phys. Rev. Letters **15**, 409 (1965).
[9] E. Fawcett, Advan. Phys. **13**, 139 (1964).
[10] M. H. Cohen and L. M. Falicov, Phys. Rev. Letters **7**, 231 (1961); E. I. Blount, Phys. Rev. **126**, 1636 (1962).
[11] L. M. Falicov and P. R. Sievert, Phys. Rev. Letters **12**, 558 (1964); Phys. Rev. **138**, A88 (1965).
[12] A. B. Pippard, Proc. Roy. Soc. (London) **A287**, 165 (1965).
[13] J. L. Stanford and E. A. Stern, Phys. Rev. **144**, A534 (1966).

the crystal axes of the sample with respect to the strong magnetic field (in a horizontal direction) could be changed during the experiment.

It was found necessary to take care over mounting the sample so that it was not strained by differential contraction on cooling. In an early experiment, after the sample had been more firmly attached to the coil assembly than previously, no quantum oscillations were detectable. After removing the sample, annealing it and then mounting it with adhesive along one edge only, the oscillations reappeared. Mounting the sample sufficiently firmly that it will not vibrate but so that it is not strained is one of the main practical problems of this type of experiment.

B. Sample Preparation

The samples were prepared from 99.9999% pure Cominco tin. A single crystal was selected from the ingot and oriented using back-reflection x-ray Laue photographs. Plate samples were cut from the single crystal with a Servomet spark-cutter. The plates were approximately $1\times10\times10$ mm^3.

The large faces of the plate were made sufficiently close to parallel by use of a spark-planer. The samples were etched for a few minutes in concentrated hydrochloric acid to remove any surface layers of strained material. X-ray photographs were again taken after handling to ensure that the samples had not been damaged.

The residual resistance ratio, determined by the eddy current decay method of Bean, DeBlois, and Nesbitt,[14] was found to be about 10^4. However, the resistivity at 4.2°K can be found by measuring $\omega_1=(\pi/4t^2)(\rho^2+A^2H^2)^{1/2}$ at small fields. The residual resistance ratio, for current flow in the plane normal to [001], obtained in this way was as 2×10^4.

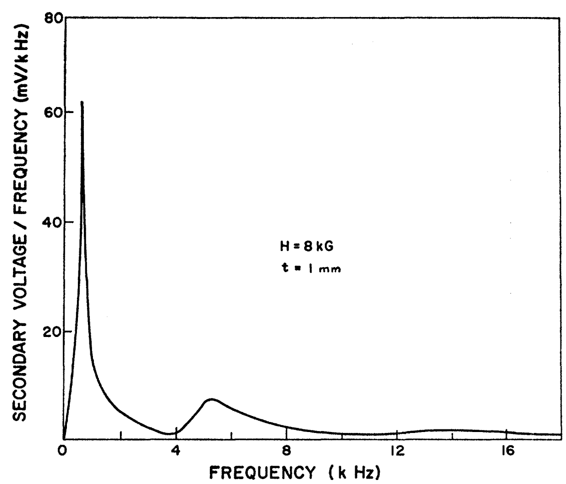

FIG. 1. The fundamental, second, and third helicon standing-wave resonances at a fixed field in a slab of tin—as observed in the effective transverse permeability. The latter was found from measurement of the amplitude of the secondary voltage at constant primary current amplitude in a crossed coil arrangement. **H** was along [001].

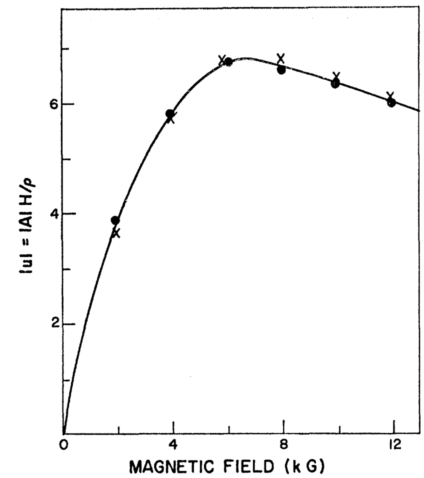

FIG. 2. Field dependence of the tangent of the Hall angle for **H** along [001]. ●—from width of fundamental resonance, ×—from its height.

IV. EXPERIMENTAL RESULTS

We shall first describe in Sec. IV A the results obtained at 4.2°K, where the quantum oscillations were not observable, and then in Sec. IV B we will deal with the oscillatory aspects.

A. Helicon Propagation

1. H *along [001]*

Figure 1 shows a plot as a function of frequency of a quantity proportional to $|\mu|$, obtained by dividing the amplitude of the voltage across the secondary coil by the frequency of the primary current. The magnetic field was fixed at 8 kG and directed along [001], which was nearly parallel (within a few degrees) to the plate normal. The maxima correspond to the standing-wave resonances that occur when an odd number of half-wavelengths equals the sample thickness.

Figure 2 shows $|u|=|A|H/\rho$ as a function of H. $|u|$ was found from the width at half-height of the standing-wave resonance by using Eq. (2). The crosses on the same graph were found from the height of the resonance, which is proportional to $|u|$.

The fundamental resonant frequency (corresponding to the helicon wavelength being twice the plate thickness) is given by

$$f_1=\omega_1/2\pi=(|A|H/8t^2)(1+1/u^2)^{1/2}. \quad (3)$$

For $u\gg1$, $f_1=|A|H/8t^2$. The experiment showed that for fields up to 14 kG df_1/dH was constant and equal to (0.076 ± 0.001) (G sec)$^{-1}$. The thickness at liquid-helium temperatures was estimated from room-

[14] C. P. Bean, R. W. DeBlois, and L. B. Nesbitt, J. Appl. Phys. **30**, 1976 (1959).

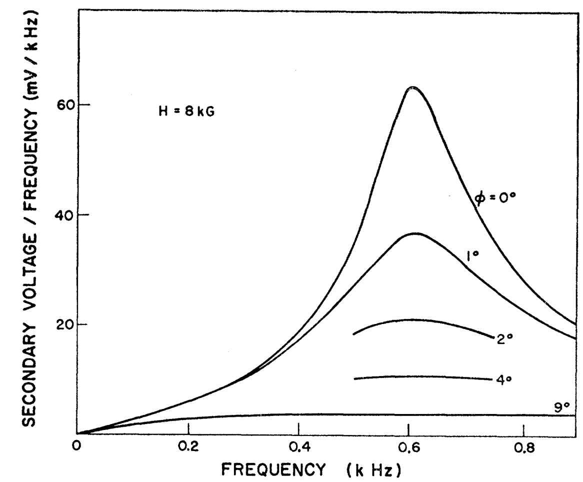

FIG. 3. The fundamental resonance ($n=1$) at a fixed magnitude of field for different orientations of the field. ϕ is the angle between **H** and [001].

temperature measurements and a correction of 0.7% for thermal contraction[15] to be (1.00±0.01) mm. The formula of Chambers and Jones[6] requires correction since the boundary conditions were not properly taken into account in its derivation. The solution of Legéndy[5] indicates that in the present case the value of $|A|$ obtained from (3) is too large by 8%. Thus we find $|A|=(5.5\pm0.3)\times10^{-3}$ emu. This should be compared with the result obtained by Kachinskii,[16] using the conventional direct-current method,

$$A=-(4.8\pm0.2)\times10^{-3}\text{ emu}.$$

The independence of $|A|$ on field and the decrease in $|u|$ for $7<H<14$ kG shown in Fig. 2 imply that the magnetoresistance was still increasing as H^2. Preliminary measurements with a 25-kG superconducting solenoid have shown that beyond 14 kG the magnetoresistance was beginning to saturate while the Hall coefficient no longer remained constant but had begun to decrease.

2. H *Inclined to [001]*

If the magnetic field is inclined to [001], the singular field direction,[9] some of the cyclotron orbits are open and the magnetoresistance becomes much larger. Strong damping of the helicon propagation may thus occur even in fields of a few kilogauss, as was found by Grimes *et al.*[8]

Figure 3 shows the fundamental resonance for different orientations of the magnetic field, with its magnitude fixed at 8 kG. The direction of the magnetic field with respect to the crystal axes is specified by the angles (θ,ψ,ϕ) where θ is the angle between [001] and the rotation plane of the magnetic field, ψ is the angle between [100] and the line of intersection of the plane of rotation of the magnetic field and the plane containing [100] and [010], and ϕ is the angle between the magnetic field and the projection of [001] on the rotation plane. The oscillating magnetic field produced by the primary current was always in the rotation plane.

As the magnetic field was tilted away from [001], the helicon amplitude dropped sharply. A resonance was barely detectable beyond about 6° away from [001]. Over the range for which the fundamental resonance was observable the resonant frequency f_1 was independent of orientation thus implying that the Hall coefficient is independent of orientation for the magnetic field near [001].

By obtaining curves similar to Fig. 3 for different magnetic fields, the magnetoresistance can be determined for different orientations of the magnetic field. For all orientations near [001], the magnetoresistance up to 14 kG was found to vary as H^2.

Figure 4 shows the dependence of the helicon amplitude on ϕ with $(\theta,\psi)=(0, 22.5°)$ for a magnetic field of 10 kG and frequency of 760 Hz. The strong damping is due to aperiodic open orbits. The maxima at $\phi\sim\pm37°$ occurred near the end of the range of angles for which there are open orbits. Beyond this angle there are no open orbits on the hole sheet of the Fermi surface in the fourth zone, according to the interpretation by Alekseevksi and Gaidukov[17] of their magnetoresistance measurements in terms of a multiply connected surface which is topologically similar to the one proposed by Gold and Priestley.[4] A similar rotation curve was obtained for a plate sample in which [001] was approximately 15° from the plate normal instead of parallel to it, as was the case with the sample above, thus

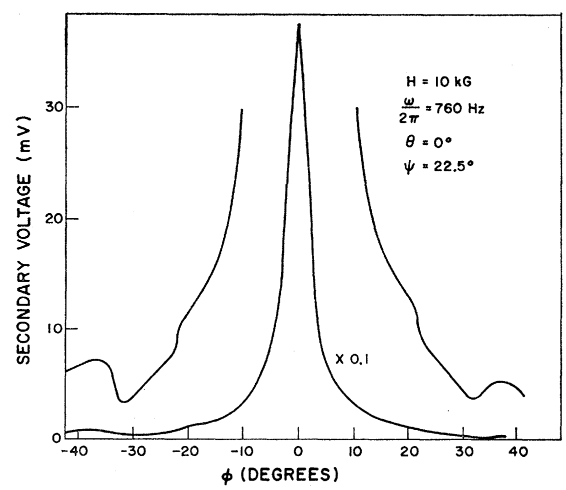

FIG. 4. Rotation curve: the helicon signal (secondary voltage) at fixed field and frequency plotted against the angle between **H** and [001]. θ and ψ define the plane of rotation of **H**.

[15] H. D. Erfling, Ann. Physik **34**, 136 (1939).

[16] V. N. Kachinskii, Zh. Eksperim. i Teor. Fiz. **43**, 1158 (1962) [English transl.: Soviet Phys.—JETP **16**, 818 (1963)].

[17] N. E. Alekseevskii and Yu P. Gaidukov, Zh. Eksperim. i Teor. Fiz. **41**, 1079 (1961) [English transl.: Soviet Phys.—JETP **14**, 770 (1962)].

demonstrating that the bumps were not associated with the arrangement of the faces of the plate sample with respect to the crystal axes.

B. Quantum Oscillations

The results discussed so far were obtained at about 4.2°K. At 1.25°K, it was found that when the magnetic field was varied (keeping the frequency fixed) large amplitude oscillations occurred in the amplitude of the secondary voltage. These oscillations were superimposed on the smoother variation with field that was observed at 4.2°K. According to the analysis presented above, the helicon propagation depends on the magnetic induction H, the Hall coefficient A, and the magnetoresistance ρ. Oscillations in any of these quantities could therefore be responsible for the observed effects. The possibility that the observed oscillations could be the "giant quantum oscillations" predicted by Miller[18] and Quinn[19] is ruled out since only the third of the three conditions for the mechanism of such oscillations to operate—$ql \gg 1$, $qv_F \gg |\omega_c|$, and $\hbar|\omega_c| \gg kT$, where T is the absolute temperature—was satisfied in the present case. We therefore continue the discussion in terms of the analysis that should be valid if nonlocal and retardation effects are negligibly small.

Figure 5 shows a pen-recorder trace of the part of the secondary voltage proportional to the real part of μ (proportional to the part of $\bar{h}_y$ in phase with h_x) taken as the magnetic field was varied from 10.5 to 14.5 kG, while directed along [001]. The temperature was 1.25°K and the helicon frequency 1060 Hz. The oscillatory part of the response has a period in $1/H$ of $(5.70 \pm 0.05) \times 10^{-7}$ G^{-1}. Such a period was found in the de Haas–van Alphen measurements of Gold and Priestley[4] who associated it with the δ orbit of the hole sheet in the third Brillouin zone of the nearly-free-electron Fermi surface. The temperature dependence of the quantum oscillation amplitude in the present

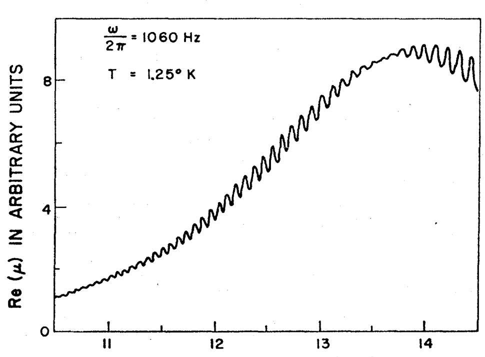

FIG. 5. Part of the helicon signal (secondary voltage) in phase with the exciting field (or primary current), at a fixed frequency, plotted against field. **H** was along [001].

[18] P. B. Miller, Phys. Rev. Letters **11**, 537 (1963).
[19] J. J. Quinn, Phys. Letters **7**, 235 (1963).

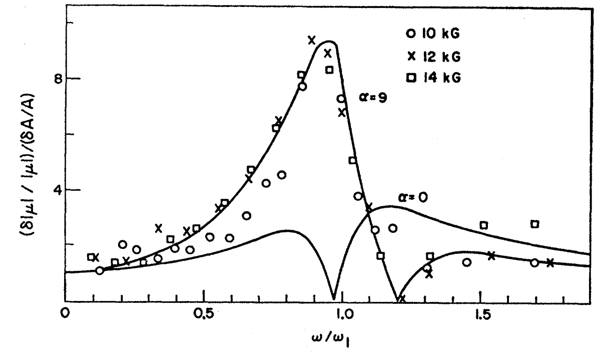

FIG. 6. Dependence on frequency of the amplitude of the quantum oscillations. The solid lines are theoretical estimates for two different values of $\alpha = (\delta\rho/\rho)/(\delta A/A)$, the ratio of the fractional quantum oscillation amplitude in the magnetoresistance to the fractional amplitude in the Hall coefficient.

experiments gives the same cyclotron effective mass of the δ orbit, $(0.10 \pm 0.01) \times$ (free-electron mass), as was found by Gold and Priestley. As can be seen from Fig. 5, the amplitude of the oscillations varies considerably with field having a maximum value of about 15% of the mean response averaged over a range of fields spanning a few cycles of the oscillations. Such a large amplitude was difficult to understand when one examined the magnitude of quantum oscillations in A, ρ, and H determined by other investigators from direct measurements of each of these quantities. As far as is known, there has been no report of quantum oscillations in A in white tin, although A itself has been studied extensively[16]—implying that such oscillations are not large. Alers[20] observed quantum oscillations in the direct-current magnetoresistance which were just detectable with a 25-kG magnetic field along [001]. The de Haas–van Alphen effect study by Gold and Priestley[4] with pulsed magnetic fields of 80 kG showed, at 1.2°K and with the magnetic field along [001], a differential susceptibility of 3×10^{-3} associated with the quantum oscillation of the same period as the one which is dominant in the present experiment. When account is taken of the large demagnetization effects in our experiments it seems unlikely that the oscillations could be due to the de Haas–van Alphen effect. The absence in Fig. 5 of another period of quantum oscillation due to electrons on the μ orbit of the fifth Brillouin zone supports this conclusion since it was found by Gold and Priestley that the μ orbit oscillation had a larger amplitude than the δ orbit oscillation. Henceforth we shall assume the induction to be equal to the externally applied magnetic field.

The oscillations in the detected signal, which is proportional to the modulus of the transverse permeability μ, can be related to the oscillations in ρ and A from Eq. (2). It is of interest to see if these contributions can be disentangled so that the amplitudes of the oscillations in ρ and A can be found separately for

[20] P. B. Alers, Phys. Rev. **107**, 959 (1957).

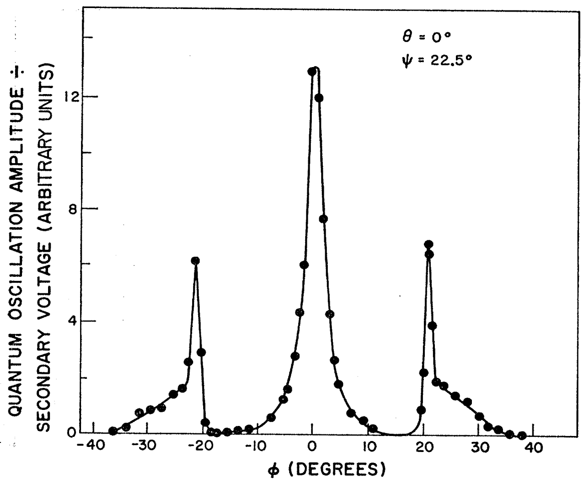

FIG. 7. Rotation curve of the amplitude of the quantum oscillations. Amplitude of quantum oscillation of the helicon signal (secondary voltage) divided by the steadily varying part of the signal.

comparison with theory. Differentiating $|\mu|$ obtained from Eq. (2) with respect to ρ and A independently we find that for small changes $\delta|\mu|$ caused by small changes $\delta\rho$ and δA,

$$\frac{\delta|\mu|}{|\mu|}=\frac{\delta A}{A}K(\omega,H)+\frac{\delta\rho}{\rho}L(\omega,H)\,, \tag{4}$$

where

$$K=1+\frac{\pi^4}{32}|\mu|^2\left(1-\frac{\omega_1{}^2}{\omega^2}\right),$$

$$-L=1+\frac{\pi^4}{32}|\mu|^2\left(1-\frac{\omega^2}{\omega_1{}^2}\frac{u^2+1}{2u^2}-\frac{\omega_1{}^2}{\omega^2}\frac{u^2-1}{2u^2}\right).$$

By appropriately choosing ω, the relative importance of changes in ρ and A can be varied. The experimental procedure used here was to observe at each of a number of different values of ω a few cycles of the quantum oscillations in the secondary voltage by sweeping the magnitude of the strong field over a small range centered on H. The amplitude of the quantum oscillations was thus obtained as a function of H and $\omega/2\pi$.

The quantum oscillation amplitude passes through zero at an angular frequency ω_0, when $K+\alpha L=0$, where $\alpha=(\delta\rho/\rho)/(\delta A/A)$. This condition reduces to

$$\frac{\omega_0}{\omega_1}=\left(\frac{u^2-1+2\alpha}{u^2+1}\right)^{1/2},$$

so that α can be determined from u, ω_0, and ω_1. We thus find $\alpha=+9$, the positive sign indicating that the oscillations in ρ and in A are in phase. This can be inferred from the origin of the oscillations discussed later. A comparison between the experimental results and Eq. (4) is given in Fig. 6.

In spite of difficulties with vibrations, the quantum oscillations were observed up to 150 kG with the field parallel to [001]. There was no evidence of any breakdown effects other than the oscillations that had been observed at lower fields. However, further effects could have been obscured to some extent by vibrations. A very careful examination was made to see if there were any oscillations of smaller period superimposed upon the main oscillations. Such oscillations would have been near the limit of detectability and were not in fact observed. Small steps at intervals corresponding roughly to the expected period were attributed to the sliding contact moving from turn to turn in the current-control rheostat.

So far, results of quantum oscillations in the helicon amplitude have been presented and discussed for the magnetic field along [001]. Figure 7 shows the variation with ϕ of a quantity proportional to the amplitude of the oscillations in $|\mu|$ divided by the steadily varying part of $|\mu|$ for $(\theta,\psi)=(0, 22.5°)$. These data were taken by modulating the magnetic field at an amplitude of 5 G and a frequency of 22 Hz. The modulated secondary voltage was detected with a lock-in amplifier. The causes of the variations in the amplitude are discussed below.

V. DISCUSSION

The comparison of the present results with models of the Fermi surface can best be carried out with reference to the model of Gold and Priestley[4] shown in Fig. 8. This surface was arrived at by combining information from de Haas–van Alphen experiments with a version of the nearly free-electron model in which all small-scale features were smoothed out. It has been shown by Alekseevskii and Gaidukov[17] to be consistent with the magnetoresistance measurements of Alekseevskii *et al.*[21] The band-structure calculation of Weisz,[2] fitted mainly to the caliper measurements of Gantmakher (see Weisz[2]) predicts a surface which differs topologically in some respects as well as quantitatively in others from Fig. 8.

Leaving aside for the moment the effects of magnetic breakdown, a knowledge of the Hall coefficient A with the magnetic field along the singular field direction [001] allows a determination of Δk_z, the thickness measured parallel to [001] of the region of the hole surface of the fourth Brillouin zone on which the orbits are electron orbits.[9] Although this dimension is not readily obtainable from the data given by Weisz,[2] it can be seen that it is predicted to lie between $0.91k_0$ and $1.05k_0$, where $k_0=2\pi/a=1.08\times10^8$ cm^{-1}. Using the value of A obtained here for $H<14$ kG, we find $\Delta k_z=(1.10\pm0.07)k_0$.

It may be noted that the Hall coefficient found here differs by more than the experimental error from the

[21] N. E. Alekseevskii, Yu P. Gaidukov, I. M. Lifshitz, and V. G. Peschanskii, Zh. Eksperim. i Teor. Fiz. 39, 1201 (1960) [English transl.: Soviet Phys.—JETP 12, 837 (1961)].

value obtained by Kachinskii[16] using conventional methods. Although the possibility of systematic errors in the conventional technique has been discussed frequently, care must be taken in the helicon method to correct Eq. (2), properly allowing for the boundary conditions. If the main concern is an accurate determination of A, it is best to use a well-defined sample geometry, as was done by Harding and Thoneman.[22] Alternatively, if no additional damping mechanisms arise at frequencies sufficiently high that the helicon wavelength is much less than the thickness of the plate, the interference fringe method[13,23] can be used.

The preliminary results obtained between 14 and 25 kG showed that $|A|$ was decreasing as is to be expected[11] from the magnetic breakdown effects discussed later, where there is a transition from the independent ζ and δ orbits at low fields to a closed hole orbit, comprising the parts of one ζ and two δ orbits, linked by magnetic breakdown.

As was mentioned in the previous section, with **H** along [001] the magnetoresistance in our experiments appeared to be still increasing as H^2 between 7 and 14 kG, rather than reaching saturation as was found by Alekseevskii *et al.*[21] Their experiments were on samples of residual resistance ratio similar to ours but extended up to 23 kG. The saturating resistivity was found to be 27 times the very-low-field value. This is probably why no deviation from the quadratic increase was detectable in the present experiments up to 14 kG. It is possible that between the second x raying (which revealed no signs of damage nor of mosaic structure) and the experiment, our samples were slightly strained—by thermal stress, for instance—but the strength of the quantum oscillations makes this seem unlikely. The preliminary results obtained between 14 and 25 kG have shown a tendency toward saturation. It is worth noting that Grimes *et al.*[8] observed a rapid increase in the helicon damping at fields greater than 15 kG which they interpreted as possibly arising from magnetic breakdown. A change in the Hall coefficient in the direction described above could also produce an increase in the helicon damping.

The strong damping of the helicon waves by open orbits when the field is tilted away from [001], first reported by Grimes *et al.*,[8] was described here in the previous section in a little more detail. Although we have not attempted a quantitative comparison between the results shown in Fig. 4 and what can be calculated from particular models of the Fermi surface, some of the structure in the vicinity of $\phi=20°$ to $30°$ can be seen to follow from the explanation below of the more striking variation of the amplitude of the quantum oscillations with ϕ. The difficulty of carrying out a quantitative analysis arises from the complexity of the

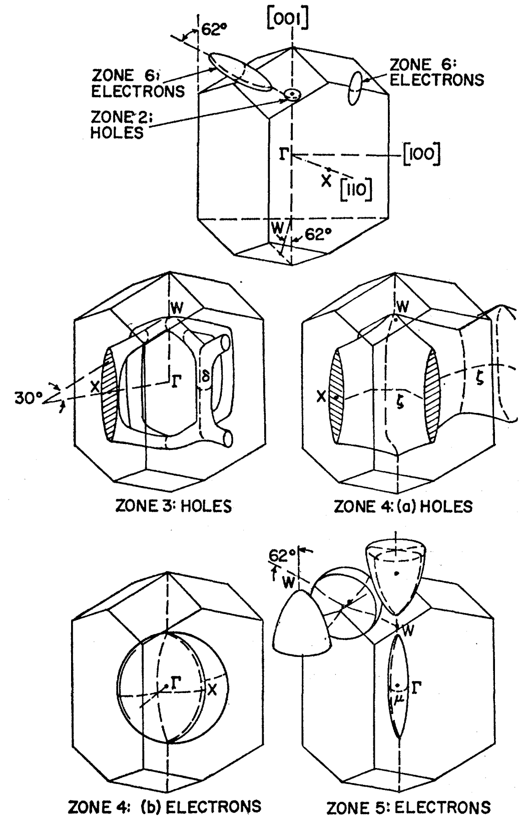

FIG. 8. The nearly free-electron Fermi surface of Gold and Priestley. The observed quantum oscillations arise from magnetic breakdown between the δ and ζ orbits at X.

wave equation. When the wave vector and the field lie along [001], the propagation can be described in terms of two parameters ρ and A. In general, however, the propagation vector is not along the field and more than two elements of the resistivity tensor are needed. Also, the difficulties mentioned in connection with Eq. (2) in dealing with accurate solutions for finite specimens are even more formidable.

We turn now to discuss the quantum oscillations. Values of $\delta A/A$ and $\delta\rho/\rho$ at various values of H can be obtained by fitting the curves of Fig. 6 to Eq. (4). At 14 kG, the oscillation amplitude of ρ is approximately 12% ρ while for A it is approximately 2% A. The resistivity amplitude should be contrasted with the much smaller value found by Alers.[20] We have already mentioned that strains can easily cause the oscillations to disappear. Recently, Young[24] has repeated his direct measurements of the magnetoresistance[25] of tin by conventional methods at 1.2°K instead of at 4.2°K and did indeed see large quantum oscillations. He has also been able to explain their origin in some detail in terms of a relatively simple extension of Pippard's[12] analysis of magnetic breakdown. The results of Young were ob-

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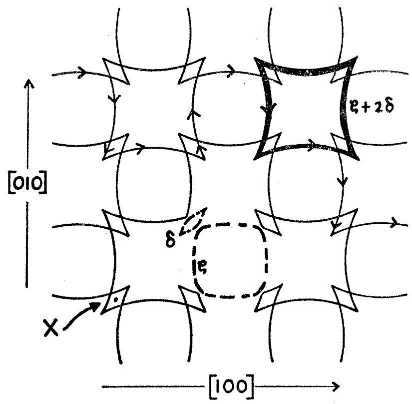

FIG. 9. Two-dimensional network of coupled orbits in (001) plane passing through origin of k space of tin, with **H** along [001]. The dashed lines in the lower middle indicate in an exaggerated way the δ and ζ orbits in low fields (before breakdown). The thick curve in the upper right, a typical orbit at high fields (complete breakdown). One of the many possible paths of an electron at intermediate fields is indicated by the series of arrows. X is at the point shown by the single arrow.

tained with the magnetic field tilted at 5° with respect to [001] where the magnetoresistance is much higher than when the field is along [001].

The most interesting aspect of the present results is the marked variation of the amplitude of the quantum oscillations when the field is tilted away from [001]—as shown in Fig. 7—and its connection with the topology of the Fermi surface.

The period of the quantum oscillations observed in the present experiments with the field parallel to [001] was $(5.70\pm0.05)\times10^{-7}$ G^{-1}, close to the de Haas–van Alphen period attributed by Gold and Priestley to the δ orbit. This result should be contrasted with the value 8×10^{-7} G^{-1} calculated by Weisz.[2]

The variation of the period with the angle ϕ between [001] and the magnetic field was found in our experiments to be proportional to $\cos\phi$ for ϕ lying between 0 and 37°—as would be expected if the orbit were on a cylindrical sheet. The sudden change of the amplitude of the quantum oscillations at $\phi\sim20°$ discussed below also supports the identification of the 5.7×10^{-7} G^{-1} oscillation with the δ orbit.

When the magnetic field is parallel to [001], i.e., when $\phi=0$, $\theta=0$, $\psi=0$, the effect of magnetic breakdown is to couple the otherwise closed ζ and δ orbits so that a two-dimensional network of coupled orbits arises in the plane normal to [001] and passing near X, as shown in Fig. 9. The variation with field of the conductivity of a similar network has been investigated by Falicov *et al.*[26] and found to oscillate owing to phase coherence effects on the smaller orbits—such as the δ orbits in this case. If the field is tilted a few degrees away from [001] with $\theta=\psi=0$, the only extended orbits passing through the breakdown region run on a course parallel to [010], since the band gap at a point in k space near X increases and therefore the probability of breakdown falls as the point moves away from X. The two-dimensional network thus breaks up into a set of essentially linear chains. If the field is tilted a few degrees away from [001] but with $(\theta,\psi)=(0, 22.5°)$ as was the case in Fig. 7, then not even a linear chain remains and the only orbits passing through the breakdown region are closed. The rapid fall in the quantum oscillation amplitude shown in Fig. 7 as ϕ increased from 0° to about 15° is due mainly to the breaking up of the two-dimensional network of coupled orbits—which has a conductivity oscillatory in field—and partly to the appearance of aperiodic open orbits, which reduce the importance of the oscillatory part of the conductivity.

The sudden increase in the quantum oscillation amplitude at $\phi\sim\pm21°$ coincides in the nearly free-electron model with where open orbits, which would exist even in the absence of breakdown, begin to pass through the breakdown region, i.e., near X. At such angles, the open orbits dominate the conductive behavior and as soon as the threshold angle is reached and their breakdown can occur, a large oscillatory dependence of the conductivity is to be expected, in addition to a change in the steadily varying part of the resistivity as was observed at 4.2°K by Young.[25] It does not seem possible that the return to a smaller amplitude at $\phi\sim\pm22°$ can be attributed to any topological properties of the Fermi surface—for instance, either a crater near W, of the sort suggested by Weisz,[2] or a set of sharp peaks near W, of the type that occur in the only infinitesimally perturbed free-electron model (as opposed to the smoothed version due to Gold and Priestley). The sharp peak at $\phi\sim\pm21°$ is probably caused by a small angular displacement of the sample which varies periodically with field in phase with the quantum oscillations. Such a displacement could be caused by the oscillatory torque of the de Haas–van Alphen effect. As can be seen from Fig. 4, the variation of helicon wave amplitude near $\phi\sim\pm21°$ is very sensitive to the angle ϕ. Beyond $\phi\sim\pm22°$, where the relative amplitude of the quantum oscillations varies linearly with ϕ, the vibration effect should be small and the change in the quantum oscillation amplitude caused mainly by the diminishing number of open orbits that pass near X.

The disappearance of quantum oscillations for $|\phi|>37°$ coincides with the disappearance of open orbits. Even when breakdown occurs the result is merely the formation of a large closed orbit.

The absence of any oscillations with a period corresponding to what can be estimated for the ζ orbit shows that there was no phase coherence around these larger orbits. This might be expected from the fact that the oscillations observed were sinusoidal, and not of the Fabry-Perot fringe type,[24] indicating appreciable scattering.

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VI. CONCLUSION

Although the interpretation of the characteristics of the helicon propagation is far more complicated than the interpretation of direct resistance and Hall effect measurements by conventional techniques, the helicon technique is more suitable for low magnetoresistance studies. As the results that we have discussed show, useful information about the topology of the Fermi surface can be obtained from the oscillations arising from magnetic breakdown.

A more detailed study of the quantum oscillations should enable further quantitative checks to be made, not only of the band-structure calculations but also of the theory of magnetic breakdown.

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Splitting of Conduction-Electron Spin Resonance in Potassium

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The 0.5-G splitting of the conduction-electron spin resonance in potassium (at 4200 G) observed by Walsh, Rupp, and Schmidt can be quantitatively explained providing the conduction electrons are in a charge-density-wave (CDW) ground state. The Fermi-surface distortion caused by the CDW energy gap leads to an anisotropic conduction-electron g factor depending on the angle between **H** and the wave vector **Q** of the CDW. The extremal values of g, corresponding to $\mathbf{Q}\perp\mathbf{H}$ and $\mathbf{Q}\parallel\mathbf{H}$, differ by $(3V/8E_F)\Delta g$, where $\Delta g=-0.0025$ is the observed g shift. V is the observed threshold energy of the Mayer–El Naby optical-absorption anomaly, and E_F is the Fermi energy. The predicted maximum splitting is 0.56 G. Interpretation of the data requires the sample to have a macroscopic domain structure, caused by thermal stress and plastic flow when the potassium-Parafilm sandwich is cooled to He temperature. The orientation of **Q** in stress-free regions should be parallel to **H**. In regions of high stress, **Q** is presumed perpendicular to the surface, and therefore approximately perpendicular to **H**.

I. INTRODUCTION

TWO years ago, Walsh, Rupp, and Schmidt[1] observed conduction-electron spin resonance (CESR) in extremely pure potassium. Linewidths as narrow as 0.13 G were obtained at a resonance field of 4200 G and a temperature of 1.3°K. The observed g shift, caused by spin-orbit coupling, was $\Delta g=-0.025(4)$, and represents an experimental shift of 5.3 G. A very puzzling feature of their result was that the CESR signal split into two well-resolved components as the magnetic field was tilted away from an initial orientation parallel to the surface of the potassium. The splitting reached a maximum value of about 0.5 G at a tilt angle of 9°. At larger tilt angles the signal intensity diminished, and one component vanished rapidly. The splitting has been observed in a number of specimens[2] having sufficient purity to yield comparably narrow lines.

The possibility that the splitting is caused by an artifact arising from excitation of both faces of a thin sample has been ruled out by Lampe and Platzman,[3] who computed the surface impedance spectra for a variety of configurations. They found line-shape variations but no splitting. The possibility that it is caused by new collective modes (paramagnetic spin waves), resulting from conduction-electron exchange interactions, was ruled out by Platzman and Wolff.[4] They indeed found that such modes exist theoretically and provide a quantitative explanation of the sidebands reported by Schultz and Dunifer.[5] The 0.5-G splitting, however, remained a mystery.[4]

An important characteristic of CESR is that individual electrons undergo several thousand quantum transitions among levels at the Fermi energy during a spin relaxation time T_2. Consequently the resonance field is determined by the average g factor (the average over all levels that contribute to the paramagnetism). Since an averaging process necessarily results in a unique number, a single resonance must always be

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