It is relevant at this juncture to point out that while the surface we have been discussing is the actual Fermi surface of thallium (defined as the boundary between occupied and unoccupied regions of reciprocal space), for the purposes of transport theory it is more properly called the zero-magnetic-field Fermi surface. For sufficiently strong magnetic fields, breakdown between the third and fourth, and fifth and sixth, zone sheets is possible at any point in the AHL plane. Thus, in the limit of field strengths greater than the largest breakdown fields occurring on the Fermi surface (approximately 40 kg), the surface we have described should be rearranged into the double zone representation, with the AHL sections of the third and fourth, and fifth and sixth, sheets being contiguous. For intermediate field strengths both representations are in some sense valid, there being a finite probability of an electron successfully passing the AHL plane barrier and disappearing into some other zone.

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Relativistic Band Structure and Fermi Surface of Thallium. II. Comparison with Experiment*

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The results of a band-structure calculation for thallium reported previously by the author are compared with the currently available experimental information. The purpose of the comparison is to test the validity of the theoretical model of the Fermi surface and to further refine that model. We also discuss in some detail the effects of the magnetic breakdown phenomenon on the properties of thallium. The experimental information considered in the paper deals with the galvanomagnetic, magnetoacoustic, and magnetic-susceptibility (de Haas–van Alphen effect) properties of thallium. It is found that the theoretical model is consistent with the experimental data.

I. INTRODUCTION

THIS paper reports the results of a detailed comparison between the available experimental information on the properties of thallium and the results of a relativistic orthogonalized-plane-wave calculation of the band structure and the Fermi surface of the metal. The calculation itself is discussed in detail elsewhere.[1]

We conclude in I that the band structure and Fermi surface of thallium bear a close resemblance to the "free-electron" model. The departures from this model are due to the usual effects of the finite crystal potential plus effects due to spin-orbit coupling. The relativistic effects were shown to be particularly large in thallium. This follows from the high atomic number ($Z=81$) of the material. As discussed in more detail in I and below, the major effect of the spin-orbit coupling is to change the topology of the Fermi surface.

The available experimental data are of three kinds: magnetoresistance measurements,[2,3] which yield qualitative information about the topology of the Fermi surface and about the energy gaps in the band structure, and the de Hass–Van Alphen[4] and magnetoacoustic attenuation[5] periods, which yield quantitative measurements of the Fermi surface. The experimental information will be considered from two points of view. One, it will be used, of course, as a test of the validity of the results of I. Two, we will attempt to employ it as an adjunct to the results of I, with the aim of resolving an ambiguity remaining in the theoretical calculation. This concerns the connectivity of the fourth zone sheet of

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[1] P. Soven, preceding paper, Phys. Rev. **137**, A1706 (1965). We will refer to this paper as I.

[2] A. Alekseevskii and Yu. P. Gadiukov, Zh. Eksperim. i Teor. Fiz. **43**, 2094 (1962) [English transl.: Soviet Phys.—JETP **16**, 1481 (1963)].

[3] A. R. Mackintosh, L. E. Spanel, and R. C. Young, Phys. Rev. Letters **10**, 434 (1963).

[4] M. G. Priestley (to be published). We are indebted to Dr. Priestley for allowing us the use of his results prior to publication.

[5] J. A. Rayne, Phys. Rev. **131**, 653 (1963).

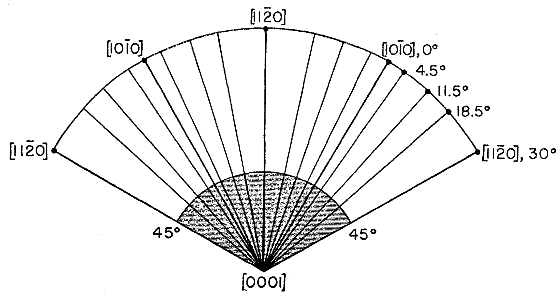

FIG. 1. The stereogram presented in Ref. 2 showing the magnetic field directions for which open orbits appear to exist in thallium.

the Fermi surface in the [0001] direction. We shall find, however, that the experimental information is also ambiguous on this point, and will suggest the appropriate experimental situation capable of resolving the question.

We will discuss in turn the available data on the galvanomagnetic properties, the de Haas–van Alphen effect, and the geometric resonances in the ultrasonic attenuation in thallium, with emphasis on the consistency of the experiments with the theoretical model and on the role played by magnetic breakdown effects in determining the properties of the metal.

II. GALVANOMAGNETIC PROPERTIES

The theory of the high-field galvanomagnetic properties of metals has been discussed in the work of Chambers,[6] Lifshitz, Azbel, and Kaganov,[7] Lifshitz and Peschanskii,[8] and others. The experimental information on the galvanomagnetic properties of thallium deals only with the transverse magnetoresistance, i.e., with the resistivity in directions perpendicular to the external magnetic field. We will therefore mention only those results of the above authors that deal with this case. They show that the power law of the field dependence of the leading term in the high-field transverse magnetoresistance may be related to the topological properties of the Fermi surface. The work of Onsager[9] showed that in the presence of an external magnetic field the carriers on the Fermi surface may be viewed as traversing the constant energy orbits formed by the intersection of planes perpendicular to the magnetic field and the Fermi surface. If the surface consists entirely of closed sheets, all orbits on them will also be closed. There are then two possible field dependencies for the resistivity. If the volume enclosed by orbits having electron character (i.e., orbits traversed in the right-hand screw sense about the magnetic field direction) is not equal to the volume enclosed by orbits of hole character, then the resisitivity saturates as the field strength increases. When this situation prevails one is dealing with a so-called uncompensated metal. The situation where the two volumes are equal, the so-called compensated case, results in a quadratic dependence of the resistivity on field strength, i.e., the resisitivity increases without limit with increasing magnetic field.

The situation is more complicated when the Fermi surface under consideration is multiply connected. For those field directions in which open orbits actually exist the resistivity is given by

$$\rho = A + BH^2 \cos^2\alpha\,,$$

where A and B are constants and α is the angle between the open orbit direction in reciprocal space and the current direction. It is seen that saturation occurs only when the sample axis is accurately perpendicular to the open orbit direction.

The variety of types of orbits that can exist on a given Fermi surface is often greatly increased by the existence of magnetic breakdown. This phenomenon, first described by Cohen and Falicov,[10] and later by Blount[11] and Pippard,[12] involves the tunneling of an electron between different sheets of the Fermi surface. In the absence of an external magnetic field an electron traversing an orbit on the Fermi suface is Bragg reflected at the Brillouin zone faces owing to the presence of a finite crystal potential. If, however, the crystal field were removed and an external magnetic field applied to the electrons, the quasiclassical orbits would form circles. In a real metal placed within a magnetic field, there is a finite probability of either of these two types of orbits existing.

Blount[11] and Pippard[12] have shown that the probability that an electron will tunnel between adjacent bands is $e^{-H_0/H}$ where H_0, the "breakdown field," is defined by the equation

$$E_g \simeq (\hbar\omega_c E_f)^{1/2}\,,$$

and E_g is the band gap in the neighborhood of the point of tunneling, E_f the Fermi energy, and ω_c the cyclotron frequency of the electrons. We have mentioned that it is the topology of the Fermi surface that determines the field dependence of the resisitivity in strong magnetic fields. In view of the fact that the effect of magnetic breakdown is to change the effective topology of the Fermi surface, we expect that under the proper conditions the effects of breakdown will be visible in the magnetoresistance properties of a metal.[13] It will

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[7] I. M. Lifshitz, M. Ya. Azbel, and M. I. Kaganov, Zh. Eksperim. i Teor. Fiz. **30**, 220 (1955) [English transl.: Soviet Phys.—JETP **3**, 143 (1956); **4**, 41 (1957)].

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[10] M. L. Cohen and L. M. Falicov, Phys. Rev. Letters **7**, 231 (1961).

[11] E. I. Blount, Phys. Rev. **126**, 1636 (1962).

[12] A. B. Pippard, Proc. Roy. Soc. (London) **A270**, 1 (1962).

[13] L. M. Falicov and P. R. Sievert, Phys. Rev. Letters **12**, 558 (1964), and to be published.

be seen presently that the phenomenon of magnetic breakdown plays an important role in the galvanomagnetic properties of thallium.

Alekseevskii and Gaidukov,[2] and Mackintosh, Spanel, and Young[3] have reported the results of detailed measurements of the transverse magnetoresistance in thallium. Their results show that there are a variety of magnetic field directions for which open orbits appear to exist. They also show in a particularly clear manner that magnetic breakdown effects are present in thallium.

A stereogram of the magnetic field directions for which open orbits appear to exist in thallium is shown in Fig. 1. The stereogram shows that there is a two-dimensional region centered about the [0001] axis and existing for an angular range of approximately 45° adjacent to this direction. The [0001] direction itself is excluded from this region. For field directions outside the two-dimensional region there exist only isolated one-dimensional regions extending into the basal ((0001)) plane. While Alekseevskii *et al.* and Mackintosh *et al.* infer the existence of a discrete set of open orbits from the presence of minima in the angular dependence of the magnetoresistance, they also report saturation of the magnetoresistance for field directions adjacent to the one-dimensional regions shown on the stereogram. The saturation value, of course, is a function of the field direction, being a local minimum in the "open-orbit" directions.

Alekseevskii and Gaidukov report that the magnetoresistance does not saturate when both the magnetic field direction and the sample axis are in the basal plane. At first sight this fact would appear to resolve the question discussed in I concerning the contact of the fourth zone posts with the ΓMK plane. In the absence of magnetic breakdown this would be true, for in that case the third and fourth zone sheets would be isolated from each other. Contact of the posts with the ΓMK plane would then imply that open orbits could exist on them with a general direction parallel to [0001]. The existence of such open orbits would cause a saturation of the resistivity when the field direction and the sample axis were in the basal plane, contrary to the observed results. We might add that the third zone sheet cannot support open orbits parallel to the [0001] direction.

When we include the possibility of breakdown, this argument must be modified. In the limit of field strengths very much greater than the relevant breakdown fields, the third and fourth zone surfaces effectively coalesce into one continuous surface, and the open orbits implied by contact of the posts with the ΓMK plane would have degenerated into large closed orbits having no significant effect on the transport properties. The nonsaturation of the resistivity in this configuration would then presumably be caused by the presence of the same open (or greatly-elongated closed) orbits that are responsible for the saturation when the field is in the basal plane but the sample axis is along [0001]. We note that the existence, for at least some values of the magnetic field strength, of these latter orbits is independent of whether or not the post actually make contact with the ΓMK plane.

The actual experimental situation lies between these extremes. Our calculation indicates that the ratio of the breakdown field at the base of the posts to the experimentally used fields is approximately 2. This implies a small but by no means negligible breakdown probability ($e^{-2}\simeq 0.125$). Model calculations by Falicov and Sievert[13] indicate that the open orbits on the post (if they exist) will have effectively vanished even with this low value of the breakdown probability. Thus the nonsaturation of the resistivity when the field direction and the sample axis are in the basal plane gives no clue regarding the question of contact.

In the context of magnetoresistance measurements the question can probably be resolved by careful study in very pure samples of the field strength dependence of the resistivity when the field direction and sample axis are both in the basal plane. Examination of the Fermi surface presented in I and of the Harrison free-electron surface,[14] modified to include spin, shows that the two alternatives will produce very different field dependencies. If there is contact, then, as the field strength increases, open orbits parallel to [0001] that tend to produce saturation when both field direction and sample axis are in the basal plane gradually break down into closed orbits, while simultaneously closed orbits in different planes perpendicular to the field are transforming into open orbits parallel to the current direction. The net result should be a change in curvature in the resistivity versus field-strength curves as the dependence changes from an approach to saturation to a quadratic behavior. If there is no contact of the posts with the ΓMK plane, no change of curvature should be observed. For in this case one has compensation in the low-field limit, and open orbits parallel to the sample axis in the high-field limit. Both of these produce a quadratic field dependence (for $\omega_c\tau\gg 1$, where ω_c is the cyclotron frequency and τ the appropriate relaxation time) and hence, no break in the curve should be discernible.

We consider now the types of orbits that can exist on our model of the Fermi surface. The fifth and sixth zone sheets are simply-connected closed surfaces. and can support only electron-like closed orbits for all directions of magnetic field. The third and fourth zone sheets are of a more complicated nature; depending upon the direction and strength of the magnetic field, closed electron- and hole-like orbits, and open orbits may exist on them. For the purpose of elucidating the topology we will first consider the types of orbits that exist in the absence of magnetic breakdown ($H\rightarrow 0$), and afterwards discuss the modifications introduced by the presence of a finite magnetic field.

[14] W. A. Harrison, Phys. Rev. 118, 1182 (1960).

In the absence of breakdown the third zone can support only closed hole-like orbits. There is actually an exception to this rule: open orbits are possible in exactly the [10$\bar{1}$0] direction, where the vanishing of the spin gap along the AL line permits propagation of orbits passing successively through the third and fourth zones. A set of orbits of such small measure cannot contribute appreciably to the transport properties. The fourth zone sheet is more complicated and exhibits a greater variety of orbits. We denote by θ the angle between the field direction and [0001]. It is convenient in describing the types of orbits existing on the fourth zone sheet to divide the totality of field directions into four regions:

(1) $\theta=0°$. For this field direction the fourth zone supports only closed orbits. These are hole like in planes near the midsection of the hexagonal mesh and electron like in planes intersecting the posts. This is a singular-field direction for which the Fermi surface exhibits geometric decompensation, i.e., because of the fact that there are closed hole-like orbits on the nominally electron-like sheet the metal acts like an uncompensated material having only closed orbits.

(2) $0<\theta\lesssim 24°$. The hole- and electron-like orbits existing for $\theta=0°$ are also present in this region. Owing to the tilt of the magnetic field, there are planes in which both types of closed orbits exist simultaneously, implying the existence of aperiodic open orbits on the boundary between them.[8] These aperiodic orbits can exist since neither the electron nor the hole-like orbits are surrounded by orbits of opposite character. They propagate with an average direction in the basal plane. The upper boundary of the region in which the aperiodic open orbits can exist is given by the smallest angle at which either the electron-like or hole-like orbits vanish. Owing to the smaller ratio of height to diameter of the hexagonal mesh as compared to the posts, it is the hole-like orbits which limit the region. Strictly speaking, the maximum value of θ for this region is a function of the azimuth of the field direction, the 24° being only the approximate value for the bounding angle.

(3) $24°\lesssim\theta\lesssim 45°$. In this region one finds electron-like closed orbits around the posts and the arms of the hexagonal mesh. The upper limit of the region is defined as the angle at which the orbits on the posts cease to exist. The aperiodic open orbits of region (2) are replaced by open orbits which run in unique directions along the inside of one of the segments of the mesh and then around one of the arms, reappearing in the adjacent segment. These open orbits can propagate in the [10$\bar{1}$0] and [11$\bar{2}$0] directions. There must of necessity also be extended closed orbits existing for field directions adjacent to those producing the open orbits. In relatively impure samples the extended orbits produce a resistivity field dependence characteristic of true open orbits. For pure samples, however, our model appears to predict the existence of anisotropy in the magnetoresistance for field directions in this region. As discussed below, this anisotropy will be either reduced or destroyed completely by magnetic breakdown effects. The orbits propagating in the [11$\bar{2}$0] direction disappear at approximately 45°. The equality of this angle with the angle at which the closed orbits on the posts disappear is fortuitous and only approximate.

(4) $45°\lesssim\theta\leqslant 90°$. In this region, there are electron-like closed orbits on the arms of the hexagonal mesh together with the open orbits propagating in the [10$\bar{1}$0] direction already mentioned in connection with region (3). These latter orbits disappear at an angle somewhat smaller than 90°. There is also the vanishing small set of open orbits in the same direction which exist by virtue of the degeneracy of the AL line and which have already been mentioned in connection with the third zone sheets. For all field directions in this region other than those producing the open orbits in [1010] directions the metal is effectively compensated.

This is the picture that we have before invoking magnetic breakdown. In the presence of a finite magnetic field the situation is as follows. Region (1) is unchanged by breakdown, implying that the magnetoresistance should saturate for all axes in the basal plane, in agreement with the experimental results. Regions (2) and (3) effectively coalesce into one. With increasing field strength the open orbits described above as existing in region (3) gradually disappear, while simultaneously aperiodic open orbits of the type already present in region (2) gradually come into existence. Because of the fact that these aperiodic open orbits have a field-dependent amplitude [a point which will be discussed in more detail in connection with region (4)] it is probable that some of the anisotropy in the resistivity that would exist in the absence of breakdown will remain even in the presence of breakdown. In any event, open orbits will exist for all field directions within an angle of approximately 45° from the [0001] axis. This is in accord with the experimental results.

It is in region (4) that the most interesting magnetic breakdown occurs. Mackintosh *et al.*[3] explain the existence of minima in the angular dependence of the resistivity in terms of the free-electron model, assuming that the posts contact the ΓMK plane and that the observed open orbit directions are those for which orbits may propagate through the third and fourth zone while simultaneously avoiding the posts. Inasmuch as we believe that the preliminary de Haas–van Alphen data indicate that the posts do not in fact make contact with this plane, we offer an alternative explanation based upon magnetic breakdown considerations and upon the assumption that the posts do not touch the ΓMK plane.

Open orbits may propagate in any direction in the basal plane. These pass successively through the third and fourth zones, with the existence of the spin-induced gap between these zones implying that these orbits have a field-dependent amplitude. The necessity of tunneling between the zones implies that at a given field strength those orbits will be most prominent which maximize the number of times that they pass between the zones near regions of small breakdown field for every passage near a region of relatively large breakdown field.

In Fig. 2 we reproduce curves given by Alekseevskii and Gaidukov showing the field dependence of the magnetoresistance for a configuration having the sample axis parallel to [0001] and the field in the basal plane. The curves marked 11.5°, 4.5°, and 15.5° show an initial roughly quadratic rise interrupted by a change in curvature and then an apparent approach to saturation. Our assumption concerning the posts implies that the metal would be exactly compensated (except for **H**||[11$\bar{2}$0]) if breakdown did not occur. This is the cause of the initial quadratic rise in resistivity as the field strength increases from zero. For sufficiently large fields open orbits appear, thus causing the approach to saturation. These ideas are illustrated in Fig. 3, which is a schematic representation of the third and fourth zone surfaces in the AHL plane.

Passage between the third zone (unshaded) and the fourth zone (shaded) is possible at any point of contact, but it is most probable at points equivalent to (a) and least probable at points equivalent to (b). The orbits marked 0° and 30° always tunnel near regions of small (approximately zero) breakdown field. Consequently, when the magnetic field is perpendicular to directions equivalent to the direction of these orbits, no change in character (compensated to open) should be observed. This is in accordance with the experimental results. The next most probable orbits are those that pass through relatively high breakdown field regions only once for every passage through regions of low breakdown field. These are marked 11° in Fig. 3, and comparison with Fig. 2 shows the expected behavior. The process can be continued to predict the field dependence of the resistivity for any magnetic field direction. We note that

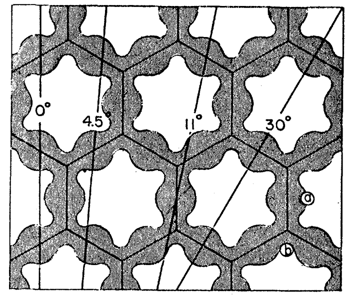

FIG. 3. The third (unshaded) and fourth zone surfaces in the AHL plane. The drawing is somewhat schematic in the sense that the two surfaces do not actually make contact in [11$\bar{2}$0] directions. The heavy lines indicate the open orbit directions corresponding to the field directions of Fig. 2.

these ideas imply eventual saturation for any field direction in the basal plane, in accordance with the reported experimental results. The curve marked 15.5° in Fig. 2 is an example of the resistivity versus field strength dependence when the field is in a non "open-orbit" direction. It is seen that the difference between this curve and one taken for the field in an "open-orbit" direction is simply one of degree and not of kind. The fact that saturation occurs at different values depending upon the field direction has been explained by Falicov and Sievert[13] as being due to the addition of incoherent scattering processes associated with the breakdown phenomenon.

III. DE HAAS–VAN ALPHEN PERIODS

We have compared the results of measurements of the de Haas–van Alphen (dH–vA) periods in the magnetic susceptibility in thallium as reported by Michael Priestley[4] with the predictions of our model. As is well known, these periods arise from extremal areas of the Fermi surface. The period P in $1/H$ is given in terms of the area A of an extremal cross section of the surface by the relation $P=(2\pi e)/(hcA)$.[15] For comparison with the experimental data we note that for P in G^{-1} and A in atomic units the relationship becomes $P=(0.2673)10^{-8}/A$. The Fermi level quoted in I was positioned by requiring that reasonable agreement be

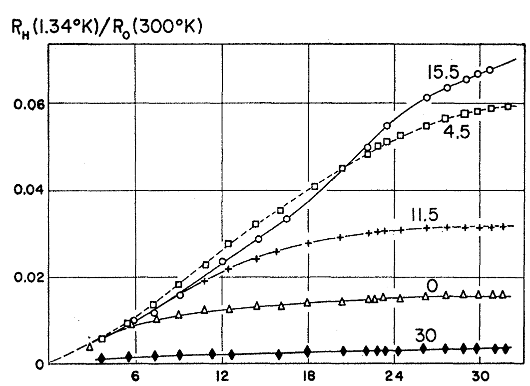

FIG. 2. Resistivity versus field strength for a thallium sample having its axis parallel to [0001] and the field in various directions in the basal plane. The angles are those between the magnetic field direction and the [10$\bar{1}$0] axis. (Taken from Ref. 2.)

TABLE I. Summary of de Haas–van Alphen[a] periods in thallium.

Period	Field directions in which observed[b] θ	φ	Period in 10^{-8} G^{-1} for field in central symmetry direction	Area in atomic units
P_{1a}	0°	···	0.460	0.583
P_{1b}	0°	···	0.480	0.558
P_2	70°–90°	0– 9°	0.88	0.301
P_3	60°–90°	0– 9°	1.06	0.253
P_4	90°	12°–30°	1.02	0.262
P_5	62°–90°	0–30°	3.66	0.073
P_6	60°–75°	0°	2.5	0.107
P_7	90°	0–30°	19.75	0.0135

[a] Taken from Ref. 4.
[b] θ is the angle between H and [0001], while φ is the angle between the plane of H and [0001] and the (10$\bar{1}$0) plane.

[15] See e.g., I. M. Lifshitz and M. I. Kaganov, Usp. Fiz. Nauk **78**, 411 (1962) [English transl.: Soviet Phys.—Usp. **5**, 878 (1963)].

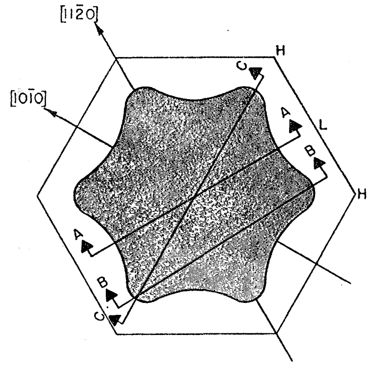

FIG. 4. The third zone sheet in the AHL plane. The section lines refer to extremal areas relevant to the discussion of the de Haas–van Alphen periods in thallium.

obtained between one of the measured areas and the corresponding extremum of our model of the Fermi surface. The fact that we were able to obtain good agreement for nearly all of the experimentally determined areas using only one adjustable parameter (i.e., the position of the Fermi level) attests to the at least internal consistency of the model. In Table I we summarize part of Priestley's results.

Periods (1a) and (1b) have been grouped together since their size and approximate equality indicates that they arise from the $AHLA$ sections of the Fermi surface in the third and fourth zones. In the absence of spin-orbit coupling these periods would coalesce, and hence the separation between the two is a measure of the influence of relativistic effects on the band structure. The theoretical areas (in atomic units) are 0.548 for the third zone sheet and 0.582 for the fourth. These are to be compared with the measured areas of 0.558 and 0.583.

Periods P_2, P_3, and P_4 are of such a size that they can only be attributed to orbits in nearly vertical planes in the third and fourth zone. There are several possibilities for these extremal areas. It would appear at first that because of its small curvature the section of the third zone sheet by the ΓMLA plane, Sec. AA of Fig. 4, would contribute a strong dH–vA period, even though magnetic breakdown also allows open orbits to exist in this and neighboring planes. However, our theoretical area for this section is 20% smaller than any of the reported experimentally determined areas. We believe that because of the approximately zero-breakdown field in planes neighboring this one (it is exactly zero in the ΓMLA plane) that the zero-field closed orbits have a negligible probability of existing in the 10^5 G pulsed fields of the experiment. Thus, the theoretical area should correspond to no observable period. Support for this view comes form the fact that if the third zone extrema in the ΓMLA plane produced an observable period so would the section of the arms of the hexagonal mesh labeled (AA) in Fig. 5. Aside form the fact that none of the experimental areas corresponds to the theoretical area for this section either, there is the fact that such a period would be readily identifiable by its angular dependence, i.e., its ellipsoidal-like shape in the neighborhood of the extrema would cause the period to decrease less quickly than the cosine of the angle between H and [11$\bar{2}$0] as the field was moved away from [11$\bar{2}$0]. No such angular dependence has been observed. We therefore conclude that the closed orbits in the ΓMLA plane of both the third and fourth zone surfaces are so completely destroyed by magnetic breakdown that they are able to produce no observable dH–vA period.

We believe that period P_3 arises from the section of the third zone surface labeled (BB) in Fig. 4. Our calculated area of 0.275 is somewhat bigger than Priestley's measured 0.253. It is possible that this discrepancy is due to the difficulty in accurately determining the area of an extremum not lying in a plane of symmetry. For example, a relatively slight change in the shape of the "bumps" on the upper and lower parts of the surface is sufficient to bring the theoretical area into close agreement with the experimentally determined area. The shape of the surface is such that as the field is rotated in the basal plane away from [11$\bar{2}$0] the extermal area will first decrease slightly and then increase again. The data are not yet complete, but this appears to be in agreement with the experimental angular dependence. When the field is parallel to [10$\bar{1}$0] the section of the surface labeled (CC) in Fig. 4 is the extremum. The theoretical area is 0.273 which is to be compared to the experimentally determined area of 0.262.

The period P_2 is more of a problem. There are only two nonequivalent extrema on the third zone surface, both of which we believe have been accounted for. Thus, P_2 must come from the fourth zone surface. We believe that it arises from the section labeled (BB) in Fig. 5. It is extremely difficult to calculate the area of this and neighboring sections. However, it appears to be a legitimate maximum, since any motion of the cutting plane parallel to itself causes the orbits to depart from the posts on the top and bottom of the surface. We estimate the area of this section to be in the range 0.25–0.35, which is, at least, consistent with Priestley's 0.301. The angular dependence offers more convincing proof of the

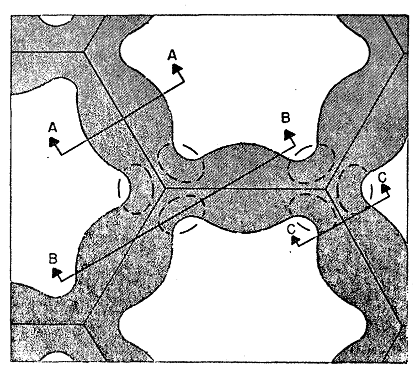

FIG. 5. The fourth zone sheet in the AHL plane. The section lines refer to extremal areas relevant to the discussion of the de Haas–van Alphen periods in thallium.

correctness of the association of the experimental period with this section of the Fermi surface. Examination of the surface shows that there is only a limited range of magnetic field directions for which one can expect to observe periods arising from this and neighboring sections. As the field is rotated in the basal plane the orbit will vanish entirely as soon as the cutting plane touches the adjacent corner of the surface. We calculate that this will happen when the angle between the field and $[11\bar{2}0]$ is approximately 9°. The data is sparse in this region, but the experimental period does vanish at an angle between 6° and 12°. As the field is rotated toward [0001] in the $(10\bar{1}0)$ plane, the orbit will cease to be an extremum when it can no longer remain on the posts. We calculate that this occurs at roughly 20° above the basal plane, which is to be compared with the measured 22°. We think that the evidence from the angular dependence is reasonably conclusive, and sufficient to identify P_2 with the fourth zone surface in the manner indicated.

If this is the correct identification it implies that the fourth zone posts do not make contact with the ΓMK plane. Perhaps at this point we should mention that if the posts do make contact with this plane, then the open orbits existing $\|$[0001] for small magnetic fields will have broken down in the fields used for dH–vA measurements into very large closed orbits capable of producing an observable period. Such a period would be a factor of 2–3 smaller than those arising from orbits contained entirely within a single zone. No such period has as yet been observed.

Period P_5 requires a hyperboloid-like surface to account for the fact that it decreases faster than $\cos\varphi$ as as the field is moved away from $[11\bar{2}0]$. The only possibility is the section labeled (CC) in Fig. 5. This is certainly a minimum area, since motion of the cutting plane parallel to itself in one direction causes the area of the orbits to increase by virtue of the hyperboloidal shape of the surface, while motion in the other direction causes the orbits to go over the posts. The calculated area is 0.069 as opposed to the experimental area of 0.073. As noted above, the angular dependence of the period as the field is moved away from the symmetry direction is in at least qualitative agreement with experiment, although it is impossible to say without further calculation whether all details are reproduced with the same accuracy.

Period P_6 has not as yet been identified with an extremal section of the model. P_7 is very nearly isotropic; this and its large size suggest that it arises from orbits around one or both of the fifth- and sixth-zone electron surfaces. For instance, the sixth-zone surface yields an extremal area of approximately 0.01, of the same order of magnitude as the experimental 0.013. In general, however, the data on the extremely slow periods is very fragmentary, and any positive confirmation of the existence and size of the electron pockets must await more detailed experiments.

TABLE II. Experimental[a] and theoretical calipers of the thallium Fermi surface.

Propagation direction	Field direction	dc experimental	dc theoretical	Zone
[0001]	$[10\bar{1}0]$	0.846	0.96	3
		0.34	0.33	4
		0.074	0.10/0.16	$\frac{5}{6}$
	$[11\bar{2}0]$	0.772	0.76/0.83	3
		0.276	0.22–0.34	4
		0.074	0.10/0.16	$\frac{5}{6}$
$[1\bar{2}10]$	$[10\bar{1}0]$	0.328	0.28–0.40/0.34	$\frac{3}{4}$
	[0001]	0.772	0.76/0.83	$\frac{3}{4}$
$[10\bar{1}0]$	$[11\bar{2}0]$	0.328	0.28–0.40/0.34	$\frac{3}{4}$
	[0001]	0.846[b]	0.96/1.01	$\frac{3}{4}$

[a] Taken from Ref. 5.
[b] Extrapolated value.

IV. MAGNETOACOUSTIC ATTENUATION

We consider now the experiments of Rayne[5] measuring the attenuation of longitudinal ultrasonic waves in a transverse external magnetic field. We reproduce in Table II some of his principal results, together with the calipers of our model that we believe correspond in some sense to the observed periods in the field dependence of the attenuation.

There is considerable ambiguity in the interpretation of magnetoacoustic attenuation experiments when the Fermi surface under consideration assumes the partially concave and multiply-connected form of the surface in thallium. The usual interpretation[16] of the observed periods is based on the idea that the contribution of each orbit to the attenuation is dominated by segments of the orbit near its external calipers in a direction perpendicular to the sound wave vector and the magnetic field.

The total attenuation is in turn dominated by the extrema of the individual orbit calipers. When both the field and propagation directions lie in symmetry planes this doubly extremal caliper will also be an extremal caliper of the Fermi surface as a whole; this is not, however, necessarily true for other experimental configurations. The relationship between the extremal caliper K_e and the period in $1/H$ of the oscillations is

$$K_e = \frac{2e}{hc}\frac{\lambda}{\Delta(1/H}\,,$$

where $\Delta(/1H)$ is the period, λ the sound wavelength, and the other symbols have their usual meaning. That this interpretation is often valid has been confirmed theoretically by the work of Cohen, Harrison, and Harrison[17] for a spherical Fermi surface and by the results of may experiments.

It is also known, however, that there are situations in which the interpretation of the observed attenuation

[16] A. B. Pippard, Proc. Roy. Soc. (London) **A257**, 165 (1960).
[17] M. H. Cohen, M. J. Harrison, and W. A. Harrison, Phys. Rev. **117**, 937 (1960).

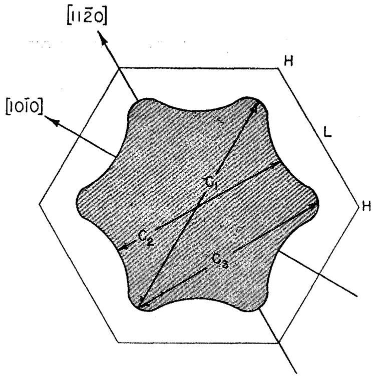

FIG. 6. The third zone sheet in the *AHL* plane. The dimension lines refer to the extremal calipers of the surface.

pattern is not quite as straightforward as the scheme described above. We mention the case of zinc, since the situation there is quite analogous to one we shall encounter in thallium.[18] Gibbons and Falicov[19] have measured the geometric resonance oscillations in the ultrasonic attenuation in zinc. Their results seem to indicate that the outer "waist" of the second-zone region of holes in zinc (the "monster" in the usual terminology) has an extremal caliper in the [11$\bar{2}$0] direction of approximately 2.9×10^8 cm^{-1}. The experiments of Stark[20] on magnetic breakdown effects on the galvanomagnetic properties of zinc definitely imply that the surface actually approaches very closely to the corner of the hexagonal Brillouin zone. This would imply an extremal caliper of 3.15×10^8 cm^{-1}. We conclude from this that for some reason the geometric resonances or the usual interpretation of the beat pattern tend to underestimate the Fermi surface dimension in this case. One can speculate on the possible cause of this discrepancy. The region of the Fermi surface in zinc that should produce the extremal caliper is one of rather small radius of curvature. Pippard[21] has emphasized that the relevant phase in the ultrasonic attenuation process is rather low (at most $20\text{–}30\times2\pi$); this perhaps makes it less likely that the true extremal will completely dominate the attenuation process in situations where the Fermi surface dimensions are rapidly varying. It is also possible that the difficulty lies in the analysis of the complicated beat patterns resulting form the mixing of several periods of greatly differing amplitudes. In any event, we shall keep in mind the situation existing in zinc when we consider some of the experimental results for thallium.

It is also reported[5] that open orbit resonances[22,23] exist in the ultrasonic attenuation in thallium. These occur when the magnetic field propagation directions are such that the trajectory of the open electron orbit in real space is on the average parallel to the propagation direction. The resonances appear in the form of extremely sharp peaks in the attenuation whenever the spatial period of the orbit in real space is equal to the sound wavelength. The real space periodicity may be related through the relation $\hbar\mathbf{k}=(e/c)\mathbf{v}\times\mathbf{H}$ to the period in reciprocal space, which can only be a low multiple of a Brillouin zone dimension.

We will discuss in detail only a part of the experimental results. In a configuration having the magnetic field $\mathbf{H}\|[0001]$ and the propagation vector $\mathbf{q}\|[11\bar{2}0]$ the published data indicate the existence of one extremal caliper of magnitude 0.772. In our model of the Fermi surface a caliper of this size can only refer to a dimension in the *AHL* section of the third and/or fourth zone sheets. Inspection of Fig. 6 shows that the calipers C_2 and C_3 of the third zone surface can contribute a period. In addition, the fourth zone sheet will contribute a caliper identical to C_2. Our theoretical estimates for calipers C_2 and C_3 are 0.760 and 0.830, respectively. Because of its small curvature, caliper C_2 will dominate, leading to good agreement between our model and the experimental results.

With the field and propagation directions interchanged, i.e., $\mathbf{H}\|[11\bar{2}0]$ and $\mathbf{q}\|[0001]$, the experimental results show no change. While similar experimentally, from a theoretical point of view the two configurations are very different. For one, in the second configuration there are no fourth zone orbits spanning a caliper of size near 0.772. For another, the orbits in the third zone that in principle would span C_2 of Fig. 6 are now in planes near the ΓALM plane, and are thus limited in amplitude by the possibility of magnetic breakdown. We note that we have argued above that the third-zone orbits in these planes do not contribute a high field de Haas–van Alphen period. We conclude that either the observed period arises in connection with caliper C_3 of Fig. 6 or that because of the small magnetic fields employed in these experiments ($H\lesssim10^3$ G) the orbits in the third zone have sufficient amplitude to produce an observable period. The exact equality within experimental error of the results in the two configurations makes the second of these seem likely. If, however, it is actually caliper C_3 of Fig. 4 that is contributing most strongly to the observed period, then the lack of agreement experiment and theory could perhaps be explained by the "phase effect" discussed above.

When the propagation direction is parallel to [0001] and the field to [10$\bar{1}$0] a period corresponding to a caliper of 0.846 is observed. This can only correspond to the third-zone caliper labeled C_1 in Fig. 6. However, the calculated size of the caliper is 0.96. In view of the good agreement between theory and experiment on the size of the de Haas–van Alphen period arising from the *AHL* section of the third-zone sheet, and also on the size of the caliper labeled C_2 in Fig. 6, we believe this

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[18] See e.g., *The Fermi Surface*, edited by W. A. Harrison and M. B. Webb (John Wiley & Sons, Inc., New York, 1960).

[19] D. F. Gibbons and L. M. Falicov, Phil. Mag. **8**, 177 (1963).

[20] R. W. Stark, Phys. Rev. Letters **9**, 482 (1962).

[21] Page 230 of Ref. 18.

[22] E. A. Kaner, V. G. Peschanskii, and I. A. Privorotskii, Zh. Eksperim. i Teor. Fiz. **40**, 214 (1961) [English transl.: Soviet Phys.—JETP **13**, 147 (1961)].

[23] J. D. Gavenda and B. C. Deaton, Phys. Rev. Letters **8**, 208 (1962).

discrepancy to reflect the difficulties in interpretation discussed above. We might add that it appears to be impossible to construct a reasonably shaped surface that is consistant with the Brillouin zone symmetry and the apparent attenuation data that will also predict the correct de Haas–van Alphen period.

In the configuration having **q**||[1210], **H**||[10$\bar{1}$0] Rayne reports the existence of pronounced attenuation peaks suggestive of open orbit resonances. He interprets these in terms of the "free-electron" model as arising from open orbits propagating on the fourth zone posts in an average direction parallel to [0001]. If this interpretation is correct it would resolve the ambiguity concerning the possible contact of the fourth zone posts with the ΓMK plane. Contact would definitely occur, for the other possibility would prohibit the existence of open orbits. We believe, however, that it is possible that this interpretation is not the correct one. Firstly, as discussed above, it appears to be difficult if not impossible to explain the de Haas–van Alphen data under the assumption that the surface is multiply connected in the [0001] direction. Secondly, there is as yet no firm theoretical basis for distinguishing between situations involving open orbits perpendicular to the field and propagation directions, and situations involving closed orbits having an extension perpendicular to **q** and **H** of essentially the suggested open orbit period and very much greater than the extension in the plane of **q** and **H**. For these reasons we feel that the reported experimental results must be taken as being perhaps suggestive but by no means conclusive evidence of the existence of open orbits parallel to [0001].

Rayne reports the existence of several intermediate periods corresponding to calipers of order 0.2–0.4 atomic units. Again there are difficulties in comparing the experimental results with the predictions of our model. For the most part these difficulties are due to the fact that for several configurations of magnetic field and propagation direction only one period of the relevant size is reported, whereas our model (and, we might add, the "free-electron" model as well) indicates that several should exist.

With **q**||[0001] and **H**||[11$\bar{2}$0] and [10$\bar{1}$0], Rayne reports the existence of periods corresponding to calipers of 0.276 and 0.340, respectively. These do not appear when the field and propagation directions are interchanged, indicating that they arise from the arms of the fourth zone surface.

We believe that the 0.34 caliper can be identified with the extremal dimension of the arms of the fourth-zone sheet in the [11$\bar{2}$0] direction. The theoretical value is approximately 0.33. We note that if this interpretation is correct then the ultrasonic attenuation period is again measuring an orbit for which no dH–vA period has as yet been observed. The remarks made above concerning magnetic breakdown are applicable here also. We are not able to identify the 0.276 calipers with a single dimension of the Fermi surface. Examination of the surface shows that the model contains several extremal calipers for **H**||[11$\bar{2}$0], **q**||[0001], the configuration being considered. The range of values is given in Table II. Without more data it is fruitless to speculate further on the exact assignment of the experimental calipers.

A similar situation occurs in configurations having both the field and the propagation direction in the basal plane. For these configurations, with measuring dimensions parallel to [0001], Rayne reports a single extremal caliper of 0.328, whereas our model predicts a range of calipers from 0.28 to 0.40.

Finally, Rayne reports the existence of a roughly isotropic long period corresponding to a basal-plane caliper of 0.074. This probably arises from orbits on the electron pockets in the fifth and/or sixth zones. Our theoretical values for such calipers are 0.10 and 0.16. In view of the small size of these sheets we do not consider the lack of agreement to reflect on the accuracy of the results of I.

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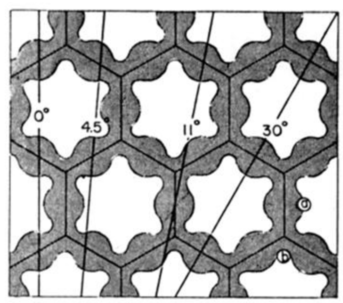

FIG. 3. The third (unshaded) and fourth zone surfaces in the *AHL* plane. The drawing is somewhat schematic in the sense that the two surfaces do not actually make contact in [11$\bar{2}$0] directions. The heavy lines indicate the open orbit directions corresponding to the field directions of Fig. 2.

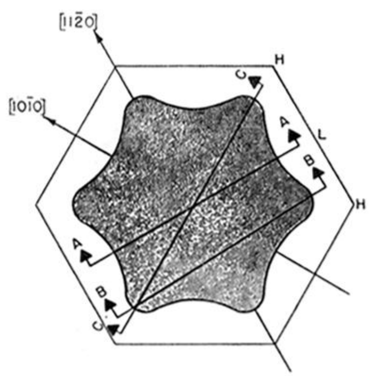

Fig. 4. The third zone sheet in the AHL plane. The section lines refer to extremal areas relevant to the discussion of the de Haas–van Alphen periods in thallium.

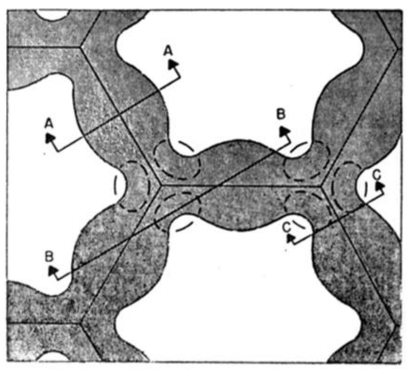

FIG. 5. The fourth zone sheet in the *AHL* plane. The section lines refer to extremal areas relevant to the discussion of the de Haas–van Alphen periods in thallium.

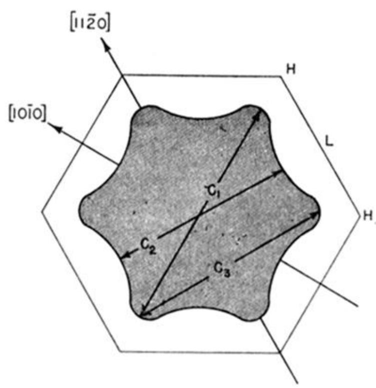

FIG. 6. The third zone sheet in the *AHL* plane. The dimension lines refer to the extremal calipers of the surface.