Experimental and Theoretical Study of Magnetic Breakdown in Magnesium*

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It is found that the energy-band structure of magnesium in the symmetry plane Γ*KHA* of the Brillouin zone is such that regions of intersecting or nearly intersecting energy surfaces give rise to magnetic breakdown. A theoretical investigation of the profiles of the energy surfaces is carried out by means of an interpolation scheme based on values of the energy at the symmetry points taken from an existing orthogonalized-plane-wave (OPW) calculation. The energy gaps due to spin-orbit effects are computed using a three-OPW approximation.

Experimental measurements of the de Haas–van Alphen effect for magnetic fields close to the *c* axis are reported. A giant orbit of area larger than the cross section of the Brillouin zone is found. The experimental parameters for this orbit are in excellent agreement with the theoretical estimates.

1. INTRODUCTION

THE energy-band structure of magnesium metal has recently been investigated from both the experimental[1–3] and the theoretical[4–7] viewpoints. The good agreement generally found between these theoretical predictions and experimental data gives confidence that the theoretical calculations can be pushed far enough to explain some of the fine details observed experimentally. In particular, magnesium is the first metal in which magnetic breakdown[8–10] effects have been found experimentally.[2,3] Because of the small gaps between the second and third bands of the energy structure, the motion of the electrons in the presence of high magnetic fields close to the *c* axis of the crystal is such that they make multiple transitions between the two bands. This results in a qualitative change in the experimental picture, e.g., the appearance of saturation in the transverse magnetoresistance[2] and new periods in the de Haas–van Alphen oscillations.[3]

In the first parts of this paper we give more detailed theoretical considerations than those which led to the prediction of the effect,[8] compute those geometrical features of the energy surfaces that are relevant to the so-called "giant orbit," and include the spin-orbit effects. We also report the experimental data obtained from de Haas–van Alphen measurements at high magnetic fields by means of the pulsed-field technique.[11] Finally, the results of these experiments are compared with the theoretical values.

Sections 2 and 3 are devoted to the theoretical investigation. In Sec. 2 the energy surfaces in the Γ*KHA* plane of the Brillouin zone (Fig. 1), where the breakdown takes place, are determined by means of an interpolation scheme based on a nine orthogonalized-plane-wave (OPW) approximation. The energy parameters are determined from an existing band structure calculation[5]; the symmetry properties of the Γ*KHA* plane are used to factorize the resulting secular equation.

In Sec. 3 we compute the spin-orbit splitting of the accidentally degenerate bands, using a representation of the wave functions in terms of 3 OPW's whose parameters are obtained from Ref. 5.

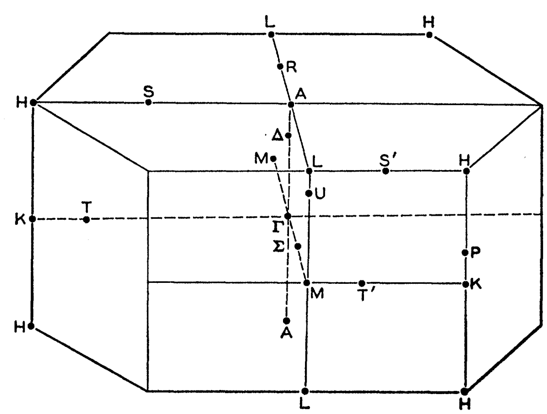

FIG. 1. The Brillouin zone in the hexagonal-close-packed structure showing points and lines of symmetry.

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1 W. L. Gordon, A. S. Joseph, and T. G. Eck, in *The Fermi Surface*, edited by W. A. Harrison and M. B. Webb (John Wiley & Sons, Inc., New York, 1960), p. 84.

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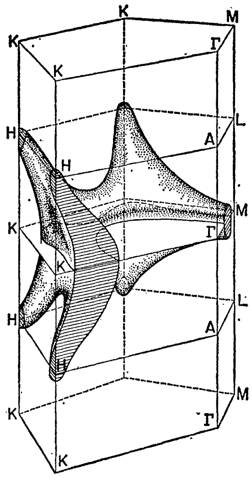


FIG. 2. The Fermi surface of magnesium in the first and second zones for the "without spin" case. Sections of the surface with the ΓKHA and ΓMLA planes are shown.

The experimental details and data from the de Haas–van Alphen measurements of the giant orbit are given in Sec. 4. Section 5 is devoted to the comparison of theory with experiment and discussion of the results.

2. THE ENERGY SURFACES IN THE REGION OF MAGNETIC BREAKDOWN

Magnetic breakdown refers to interband transitions which occur across sufficiently small energy gaps in sufficiently high magnetic fields. These transitions between bands occur for fields high enough to satisfy the approximate condition[8,9]

$$\hbar\omega_c E_F > E_g^2, \tag{1}$$

where E_g is the energy gap, ω_c the cyclotron frequency

$$\omega_c = eH/m^*c, \tag{2}$$

and E_F the Fermi energy. Consequently, it is necessary to look for small gaps, less than 0.1 eV, say, in order to make such an effect experimentally observable.

In general, gaps of these orders of magnitude arise only from the removal by spin-orbit effects of degeneracies of the energy-band levels. Such an example has been discussed[6,7] in the case of the AHL plane of the Brillouin zone of the hexagonal close-packed metals, where the degeneracies are due to the symmetry of the lattice (sticking together of the bands). In our case, however, we shall be interested in the removal of accidental degeneracies[12] which appear in the ΓKHA plane.[8]

In order to understand the existence of these small gaps and their location in the Brillouin zone, we must first describe the Fermi surface of magnesium as found both theoretically and experimentally.

If for the time being we neglect spin-orbit effects, the first and second, and also the third and fourth bands, stick together, due to the degeneracy of the levels on the AHL plane. The Fermi surface in the first and second zone (Fig. 2) is a single, multiply connected sheet usually known as the "monster" or the "region of holes." In the third and fourth zone (Fig. 3) the surface consists of several closed, singly connected sheets enclosing electrons:

(a) A surface centered on Γ, resembling an oblate spheroid.

(b) Two surfaces centered on K, and forming cigars of triangular cross section which run along the KH line.

(c) Six small surfaces about L.

We shall be mainly concerned with the "monster" and the "cigars," and we shall show that in fact they touch each other at one point of accidental degeneracy on the ΓKHA plane. In addition, the profiles of the two surfaces in that plane run parallel and very close to each other. This is due to the following properties of the band structure[5]:

(a) The existence of a line of double levels P_3 along the KH line.

(b) The crossing of the P_3 line with the P_1 line which is due to the ordering of the K_1, K_5, H_1, and H_2 levels, and the compatibility relations between them.

(c) The existence of an accidental degeneracy in the ΓK line, where, because of the compatibility relations, the T_1 and T_4 levels must cross each other.

(d) The existence of a saddle-point of the $E(k)$ curve in the second band in the ΓKHA plane, where K_5 and A_1 are points of local minima, and H_2 as well as a point on the ΓK line are points of local maxima.

To find the relevant energy profiles in the ΓKHA plane, a pseudopotential interpolation scheme was used. Only the first 9 plane waves such that their k vectors were smallest at K were taken into account. In the approximation used, the matrix elements of the Hamiltonian were assumed to be

$$\langle k|\mathcal{H}|k'\rangle = Pk^2\delta_{kk'} + U(k-k'), \tag{3}$$

where P included an average constant effective mass correction and $U(G)$ were the Fourier coefficients of

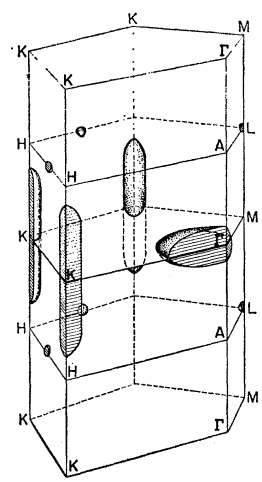


FIG. 3. The Fermi surface of magnesium in the third and fourth zones for the "without spin" case.

[12] C. Herring, Phys. Rev. 52, 365 (1937).

TABLE I. Single-group representations for the general point of the ΓKHA plane.[a]

Representations	G	U
$\{\epsilon\|0\}$	1	1
$\{\rho_2''\|\tau\}$	$e^{ik\tau}$	$-e^{ik\tau}$

[a] $\tau=\frac{1}{2}t_1+\frac{1}{3}t_2+\frac{2}{3}t_3$. Γ_1^+, K_1, Δ_1, T_1, $P_1\rightarrow G$; Γ_3^+, Γ_4^-, Δ_2, $T_4\rightarrow U$; A_1, K_5, H_1, H_2, H_3, P_3, $S_1\rightarrow G+U$.

the pseudopotential for reciprocal lattice vectors G. The pseudopotential was assumed to be independent of k.

The small group of k for points in the ΓKHA plane contains two essentially different symmetry operations, i.e., the identity $\{\epsilon|0\}$ and a glide-plane $\{\rho_2''|\tau\}$.[13] Consequently, the group has two irreducible representations which we call G and U, whose characters are given in Table I. The compatibility relations in this plane are also given.

Because of the existence of these two representations, the original 9×9 secular equation can be easily factorized into a 4×4 G equation and a 5×5 U equation. The 5 pseudopotential coefficients appearing in them as well as the effective mass parameter P were considered adjustable parameters and were chosen so as to fit the energy levels along the KH and ΓK lines as given in Ref. 5. The values used in the calculation are given in Table II. It was found that the fitting could not be accomplished in a perfect way for all the points on the boundaries of the ΓKHA rectangle, but the errors in this approximation were of the order of, but less than, 0.02 Ry in the region of interest, which is less than the quoted errors of the energies in Ref. 5. This fact points out very clearly that the pseudopotential must necessarily be k-dependent, although for the present purposes a k-independent pseudopotential gives fairly satisfactory answers.

The energy profiles, as computed numerically, are given in Fig. 4. The locus of points of accidental degeneracy is shown in Fig. 4(d); these points exist for values of the energy between 0.676 and 0.6835 Ry. The Fermi energy is estimated to be 0.680 Ry from Ref. 5. It is interesting to note that this accidental degeneracy exists only in a range of about 0.008 Ry

TABLE II. Parameters of the pseudopotential interpolation scheme.[a]

G	$[G]$ (Ry)
$\pm G_2$; $\pm G_3$; $\pm G_4$	$[10\bar{1}0]=0.210$
$2G_1$	$[0002]=0.060$
$\pm G_2+G_1$; $\pm G_3\pm G_1$; $\pm G_4\pm G_1$	$[10\bar{1}1]=0.225$
$\pm G_2\pm 2G_1$; $\pm G_3\pm 2G_1$; $\pm G_4\pm 2G_1$	$[10\bar{1}2]=0.060$
0	$[0000]=0.040$

[a] $[G]=U(G)/S(G)$. $S(G)=\frac{1}{2}+\frac{1}{2}\exp(iG\tau)$. Effective mass parameter $P=m^{*-1}=1.56$.

[13] C. Herring, J. Franklin Inst. **233**, 525 (1942).

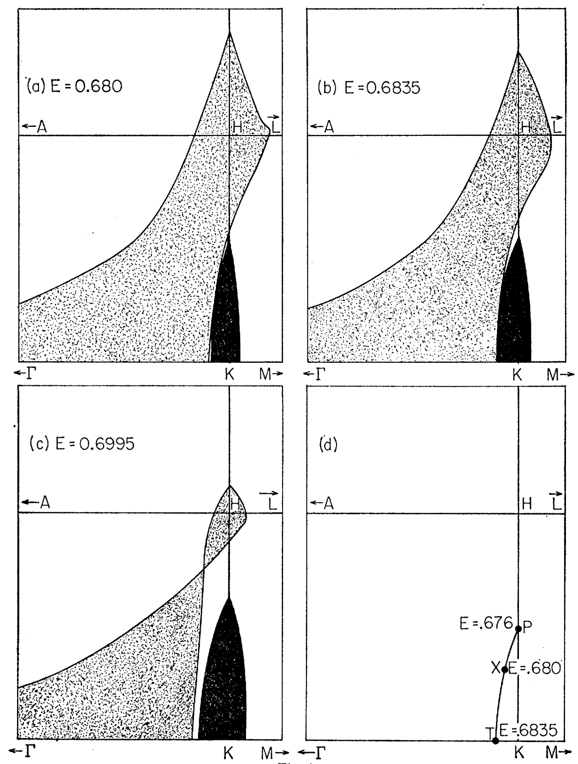


Fig.4

FIG. 4. The profiles of the energy surfaces in the ΓKHA plane for the "without spin" case at various energies: (a) $E=0.68$ Ry, the estimated Fermi energy; (b) $E=0.6835$ Ry, energy at which the degeneracy occurs on the ΓK line; (c) $E=0.6955$ Ry, energy of the saddle point; (d) The locus of points of accidental degeneracy as a function of energy.

and, consequently, the fact that magnetic breakdown is observed experimentally defines sharply the relative position of the Fermi level in the neighborhood of the point K. We may also point out that the existence and location of the line of accidental degeneracy are rather insensitive to the choice of the pseudopotential parameters, as long as the ordering of the levels at the symmetry points is conserved. In this context, the values of the energy quoted above can be considered meaningful, even when the errors of the initial band structure calculations[5] were estimated to be of about 0.03 Ry.

3. SPIN-ORBIT EFFECTS

The accidental degeneracy between the "monster" and the "cigar" discussed in the last section is removed by spin-orbit effects. To estimate the critical field for magnetic breakdown given by (1), it is necessary to know the value of the spin-orbit splittings. In the "without spin" case the two degenerate states are those of G and U symmetry which merge at K to form the K_5 level. Since the accidental degeneracy occurs close to K, for the consideration of spin-orbit effects the wave functions of the pertinent states can be fairly well represented by linear combinations of three OPW's.

TABLE III. Double-group representations for the general point of the ΓKHA plane.[a]

Representations	C_1	C_2
$\{\epsilon\|0\}$	1	1
$\{\bar{\epsilon}\|0\}$	-1	-1
$\{\rho_2''\|\tau\}$	$ie^{ik\tau}$	$-ie^{ik\tau}$
$\{\bar{\rho}_2''\|\tau\}$	$-ie^{ik\tau}$	$ie^{ik\tau}$

[a] C_1 and C_2 are degenerate because of time-reversal symmetry. Γ_7^+, Γ_8^+, Γ_8^-, A_6, K_7, K_8, K_9, H_8, H_9, Δ_7, Δ_8, P_6, $T_5 \rightarrow C_1+C_2$; H_5, H_6, S_3, $S_5 \rightarrow C_1$; H_4, H_7, S_2, $S_4 \rightarrow C_2$.

The spin-orbit splitting can then be computed in the fashion of Ref. 7, once the coefficients of the OPW's are known. These, in fact, were also obtained from Ref. 5.

When spin-orbit terms are taken into account, the symmetry properties of the wave functions for a given k vector are given by the irreducible representations of the so-called double group.[14] For the points under consideration there are two irreducible representations, here called C_1 and C_2, which are degenerate because of time-reversal symmetry.[15] The characters of these representations are given in Table III.

In order to choose a suitable direction for the spin quantization, we define a right-handed system of orthogonal coordinates x, y, z, such that x is in the ΓK direction, z is in the ΓA direction. We call spin-up states $|\uparrow\rangle$ those in which the spin lies parallel to the y axis. With this convention it is easy to prove that the wave function with G symmetry and spin up $|G\uparrow\rangle$, and the function with U symmetry and spin down $|U\downarrow\rangle$ belong to the C_1 representation, and $|G\downarrow\rangle$ and $|U\uparrow\rangle$ belong to the C_2 representation. For any point in the plane, both G functions are degenerate; the same applies to the U functions. At the point of accidental degeneracy the spinless part of the Hamiltonian $\mathcal{H}_0$ has for these four wave functions the representation

$$\begin{bmatrix} E & 0 & 0 & 0 \\ 0 & E & 0 & 0 \\ 0 & 0 & E & 0 \\ 0 & 0 & 0 & E \end{bmatrix},$$

and the spin-orbit term because of the symmetry must be of the form

$$\begin{bmatrix} 0 & F & 0 & 0 \\ F^* & 0 & 0 & 0 \\ 0 & 0 & 0 & F^* \\ 0 & 0 & F & 0 \end{bmatrix},$$

where the off-diagonal matrix element F connects states of the same symmetry, C_1 or C_2. The spin splitting ΔE_{spin} is then equal to

$$\Delta E_{\text{spin}} = 2|F|. \tag{4}$$

The spin Hamiltonian is equal to[14]

$$\mathcal{H}_s = \frac{\hbar}{4m^2c^2}(\nabla V \times \mathbf{p} \cdot \boldsymbol{\sigma}), \tag{5}$$

where V is the lattice potential, $\mathbf{p}$ the momentum operator, and $\boldsymbol{\sigma}$ the Pauli spin operator. The wave functions to be considered, $|G\rangle$ and $|U\rangle$, are linear combinations of three and two OPW's, respectively; their coefficients and core parts were taken from Ref. 5.

The matrix elements F were evaluated with the potential given in Ref. 7 and using the techniques there described. The values of the splitting at the three points T, X, P shown in Fig. 4(d), as well as the splitting at K_5 are given in Table IV. They are roughly equal to 2×10^{-4} Ry, which corresponds to breakdown fields of about 100 G.

This value is for the minimum gap, precisely at the point where the accidental degeneracy has been removed. In general, for those points of the ΓKHA plane in which the "monster" and the "cigar" are nearly

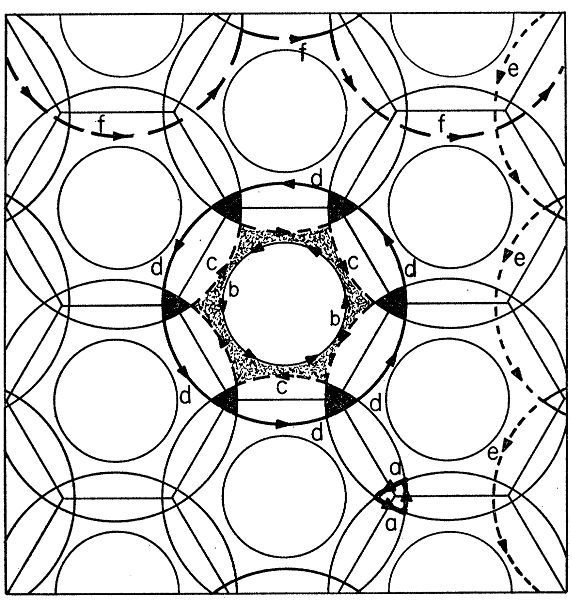


FIG. 5. The cross section of the "cigars" and the "monster" with the basal plane ΓKM (repeated zone scheme), showing the possible orbits before and after magnetic breakdown is complete: (a) orbit around the "cigar"; (b) orbit around the outer waist of the "monster"; (c) orbit around the inner waist of the "monster"; (d) the giant orbit; (e) open orbits for fields tilted off the c axis towards [11$\bar{2}$0]; (f) open orbits for fields tilted off the c axis towards [10$\bar{1}$0].

TABLE IV. The spin-orbit splitting at the points of degeneracy.

Point	k_x (a.u.)	k_z (a.u.)	Splitting (Ry)
K	0.695	0	2.8×10^{-4}
T	0.664	0	2.3×10^{-4}
X	0.677	0.102	1.0×10^{-4}
P	0.695	0.160	1.7×10^{-4}

[14] R. J. Elliott, Phys. Rev. 96, 280 (1954).
[15] C. Herring, Phys. Rev. 52, 361 (1937).

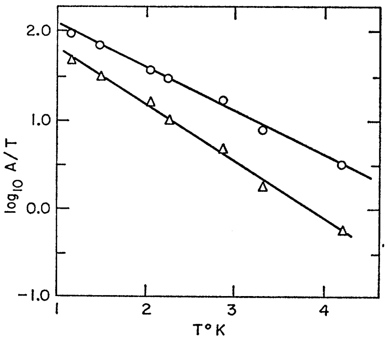


FIG. 6. The $\log_{10}(A/T)$ versus T for the de Haas–van Alphen oscillations, $P=1.61\times10^{-9}$ G^{-1}. The amplitude A is measured in arbitrary units. The open circles are for $H=174$ kG and the triangles for $H=136$ kG.

degenerate, the gap between the second and third band is due not only to spin-orbit splitting but has also a small contribution from the crystal potential. Nonetheless, the value of the energy gap turns out to be small, of the order of 10^{-3} Ry, corresponding to breakdown fields of the order of a few kG.

The trajectories of the electrons in k space in the presence of magnetic fields are given semiclassically by the intersection of the surfaces of constant energy with planes perpendicular to the magnetic field. When the field is exactly parallel to the c axis, the orbits to be expected from the "cigars" and the "monster" are those indicated by (a), (b), and (c) in Fig. 5. However, when the fields are larger than the critical value for magnetic breakdown, transitions between the various sheets of the Fermi surface give rise to new kinds of orbits. In particular, the one indicated by (d) in Fig. 5 corresponds very closely to a maximum circle of the free electron sphere, whose area, from Ref. 5, is equal to

$$\mathcal{A}_0 = 1.662 \text{ a.u.} = 5.94 \text{ Å}^{-2}.$$

In addition, when the fields are tilted off the c axis and the orbits do not pass through all the regions where breakdown is possible, open orbits of the kind indicated by (e) and (f) in Fig. 5 appear. These, as well as orbit (d), are responsible for the saturation of the magnetoresistance found experimentally.[2]

4. THE EXPERIMENTAL STUDY OF THE GIANT ORBIT

In the course of a detailed study of the de Haas–van Alphen effect in magnesium,[3] oscillations which appeared to contradict the crystal symmetry were found when the magnetic field was along [0001]. The period was $(1.61\pm0.01)\times10^{-9}$ G^{-1}, which corresponds[16] to an area of 1.66 a.u., i.e., larger than the corresponding cross section of the Brillouin zone (1.255 a.u.).

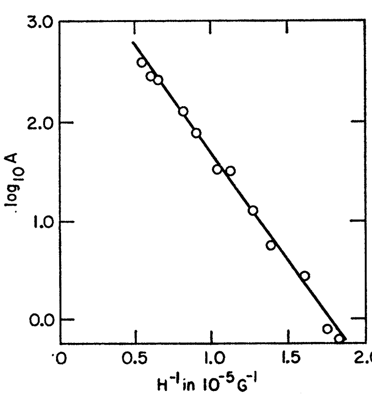
First we had to eliminate the possibility the oscillations were the second harmonic of some period whose fundamental had low amplitude because of the spin-splitting factor[17] which modifies the amplitude.[18] This factor is $\cos(ym^*g/2)$, where y is the order of the harmonic, m^* is the effective cyclotron mass in units of the electronic mass, and g is the spectroscopic splitting factor. For magnesium we expect g to be close to 2 so the fundamental amplitude should be zero for $m^*=n+\frac{1}{2}$, where n is an integer. The temperature dependence of the amplitude of the oscillations (Fig. 6) has been used to measure the effective mass, giving a value 1.37 ± 0.05. The effective mass of the postulated fundamental would, thus, be 0.69, which would not account for its absence. We conclude that the observed oscillations are not the second harmonic of a missing fundamental. We interpret the giant orbit, therefore, in terms of magnetic breakdown in the ΓKHA plane, between the third zone "cigars" and the second zone "monster."[8]

The field dependence of the amplitude of this giant orbit was measured in an attempt to find the transition region. The results are given in Fig. 7 and the observation of the expected linear relation between the logarithm of the amplitude and the reciprocal of H shows that the transition region is well below 53 kG, the lowest field at which the oscillations could be seen. This is in agreement with recent measurements of the Hall effect for H along [0001],[19] which shows that the transition region occurs below a few kG.

The collision broadening temperature,[20] deduced from the slope of Fig. 7, is 1.3°K which is a factor of 2 higher than that found for the longer periods in similar samples. It is not known to what extent this difference can be ascribed to breakdown effects.

The absolute amplitude of the oscillations at $H=185$ kG was compared with that calculated from the theoretical expression,[18] using the above values of the effective mass and collision relaxation time. The comparisons are made in terms of the value $4\pi|dM/dH|$ where M is the magnetic moment per unit volume. The observed amplitude at resonance was 150 mV which gives a measured value of $4\pi|dM/dH|=27$. The calculated value at 185 kG and 1.2°K was 0.45, assuming $|\partial^2\mathcal{A}/\partial k_z^2|^{-1/2}=(2\pi)^{-1/2}$, the value for a spherical surface. The errors in these estimates are considerable,

FIG. 7. The field dependence of the amplitude of the de Haas–van Alphen oscillations, $P=1.61\times10^{-9}$ G^{-1}. The amplitude is measured in arbitrary units, $T=1.2$°K.



[16] L. Onsager, Phil. Mag. **43**, 1006 (1952).
[17] R. B. Dingle, Proc. Roy. Soc. (London) **A211**, 500 (1952).
[18] I. M. Lifshitz and A. M. Kosevich, Zh. Eksperim. i Teor. Fiz. **29**, 730 (1955) [translation: Soviet Phys.—JETP **2**, 636 (1956)].
[19] R. W. Stark (private communication).
[20] R. B. Dingle, Proc. Roy. Soc. (London) **A211**, 517 (1952).

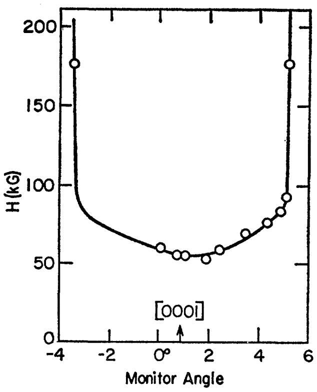


FIG. 8. The field at which the giant orbit amplitude reaches 100 μV as a function of monitor angle.

but probably less than a factor of 2, so there is a large discrepancy between the measured and calculated amplitudes. Part of this can be ascribed to the effective amplification of the resonant circuit discussed by Shoenberg,[21] but this seems unlikely to account for a difference of a factor of 50.

Shoenberg also predicts an anomalously large harmonic content when $4\pi|dM/dH|\sim 1$ but this was not observed in these measurements.

An attempt was made to measure the field dependence of the angular range over which the giant orbit could be seen. The only practicable way to do this was to measure the amplitude as a function of field for a given angle between H and [0001] and then to plot against this angle the lowest field at which the oscillations could be seen. In fact, the field at which the amplitude reached 100 μV was plotted against the monitor angle. This voltage was just above the noise level and represented the smallest signal that could be reliably detected. The precision of the relative angular measurements is of the order of 0.1°.

The results of these measurements are shown in Fig. 8 for a plane which passes through [0001] and intersects the basal plane 14° from [10$\bar{1}$0]. The main feature of the graph is that above 100 kG the angular range of the giant orbit is independent of field to 0.1° which suggests that at high fields the disappearance of the giant orbit as the field is tipped away from [0001] is governed by some geometrical feature of the Fermi surface.

At 177 kG the maximum observed angular range is $8.4\pm 0.2°$, and it should be noted that this value is not at all sensitive to the criterion used to determine the disappearance of the oscillations.

When breakdown is complete the orbit around the "cigar" [the one called (a) in Fig. 5] for H along [0001] should disappear. The "cigar" orbit was observed at fields up to 35 kG, but an attempt to measure it at much higher fields was not successful because of some experimental difficulties.

For the sake of completeness we may mention that the orbits around the outer and inner waist of the monster [(b) and (c) in Fig. 5] were not found experimentally.

5. CONCLUSIONS

From the discussion in the last sections it is evident that the giant orbit found experimentally can be explained only in terms of magnetic breakdown. Any electron trajectory in k space which stays only in one band cannot give an area larger than the cross section of the Brillouin zone, provided the field is along [0001]. In addition, we have shown that the effect cannot be due to spin splitting of the Landau levels.

The explanation of this orbit in terms of magnetic breakdown is supported by several experimental data. The area, as obtained from the period of the oscillation, agrees to three significant figures with the theoretical value for the free electron sphere.

The angular dependence is also explained in terms of the model. The geometrical factor most likely to limit the angular range of the giant orbit can be explained as follows: As the field is tipped, the orbit will run off the horizontal arms of the "monster" which lie parallel to [11$\bar{2}$0]. Since we know the minimum cross-sectional area of these arms,[3,2] we can calculate the angular range, assuming a circular cross section. This gives a calculated range of 8.5° in the plane studied, in surprisingly good agreement with the measured value, since the cross section of the arms is not really expected to be circular. If this interpretation of the extent of the giant orbit is correct, we can place a lower limit of 0.106 a.u. in the k_z direction for the region in which breakdown takes place. If the range of the giant orbit is determined by breakdown effects, which seems unlikely because of the constancy of the observed angular range for H greater than 100 kG, the 0.106 a.u. is then the k_z range of the breakdown region. The k_z range over which the giant orbit exists has also been determined by Stark[19] from Hall effect measurements for H along [0001] and the result agrees quantitatively with the above value.

The existence of the breakdown region in the ΓKHA plane opens the possibility of many different kinds of new orbits for fields tilted off the c axis. In addition to the open orbits (e) and (f) of Fig. 5, which have been found experimentally by means of magnetoresistance measurements[2] and which agree very well with the theoretical predictions, it is also possible to find various closed orbits which run between the "tentacles" of the "monster" and the "cigars." Some of these have been found by means of the de Haas–van Alphen effect.[3]

We are grateful to Professor M. H. Cohen and Dr. D. Shoenberg for very stimulating discussions and to Dr. R. W. Stark for communicating the result of his experiments prior to publication.

[21] D. Shoenberg, Phil. Trans. Roy. Soc. London A255, 85 (1962).