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Abstract

The absorption of longitudinal ultrasonic waves has been studied as a function of transverse magnetic field in pure single crystals of lead at 1.2°K. The results were found to be generally consistent with the Fermi surface of lead suggested by Gold. In particular a detailed study of the magnetoacoustic oscillations as a function of sample orientation and field direction allowed the determination of some of the dimensions of the hole surface in the second Brillouin zone and revealed a number of new electronic orbits on the multiply connected surface in the third zone. The absolute magnitude of the attenuation and its dependence on propagation direction and on magnetic field was studied and the results interpreted in terms of the general theory of ultrasonic absorption.

ULTRASONIC ATTENUATION IN LEAD

A. R. Mackintosh*

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1. Introduction

Bömmel (1955) first observed experimentally that the absorption of high frequency longitudinal ultrasonic waves in pure tin, at liquid helium temperatures, exhibited an oscillatory variation when a transverse magnetic field was applied. It was suggested by Pippard (1957) that these oscillations correspond to a variation in the relative sizes of the ultrasonic wavelength and the extension of the closed electron orbits in the magnetic field. He showed that the oscillations should be periodic in reciprocal field and suggested the relation

$$\Delta\left(\frac{1}{H}\right) = \frac{e\lambda}{ch k_e} \quad (1)$$

for the period of the magnetoacoustic oscillations, where λ is the ultrasonic wavelength and k_e is the extremal extension of the Fermi surface in $\underline{k}$ space, normal both to the direction of propagation and the magnetic field.

This result was verified for the free electron gas by the detailed calculations of Kjeldaas and Holstein (1959) and Cohen, Harrison and Harrison (1960). In particular, they found that magnetoacoustic oscillations only

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appear when the product of the acoustic wave number q , and the electronic mean free path l is greater than about 3, and further that the high field oscillations do not become much more distinct on increasing ql beyond about 15, although more oscillations appear at lower fields.

Pippard (1960) examined the problem of calculating the ultrasonic attenuation in real metals in some detail and found that, under certain conditions, equation (1) indeed determines the period of the magnetoacoustic oscillations, although in general it must be applied with caution.

Although quite detailed studies of magnetoacoustic oscillations have been made in a number of metals (see "The Fermi Surface", John Wiley and Sons, Inc., 1960; for a number of examples) no investigation of a large variety of crystal orientations and magnetic field directions in one metal has yet been reported. A study of this kind is clearly required to make a detailed comparison between theory and experiment and to obtain the maximum amount of information about the electronic structure. The investigation reported in this paper was undertaken in an attempt to carry out such a study on a metal whose electronic structure was sufficiently well understood to allow a fairly detailed analysis in terms of a simple theory of the interaction between electrons and acoustic waves. Lead was chosen for these experiments, both because of the knowledge of its Fermi surface, which was available from the work of Gold (1958), and because of its availability in a sufficiently pure form to make the observation of magnetoacoustic oscillations feasible.

A brief description of this work has already been published (Mackintosh 1960 (a)).

2. Experimental Method

2. (a) The Apparatus and Experimental Technique

The absorption of longitudinal ultrasonic waves in cylindrical specimens of pure lead was measured by the "pulse echo" technique. A schematic diagram of the apparatus used is given in figure 1. The ultrasonic waves were generated by applying a high frequency voltage pulse across a quartz transducer and transmitted to the specimen through a thin film of high-viscosity silicone oil. Successive ultrasonic reflections from the free end of the specimen were reconverted by the transducer into electrical signals which were amplified, demodulated, and displayed on a cathode-ray oscilloscope. The attenuation was measured by comparison with a pulse from a high frequency oscillator which was fed into the receiver through a calibrated piston attenuator, thus eliminating the effect of non-linearity of the receiver.

The transducers used in these experiments were X-cut quartz discs with a fundamental resonant frequency of 15.6 Mc/s, and they were normally excited on their fifth harmonic by one microsecond high-voltage electromagnetic pulses. The choice of this frequency was determined by the electronic equipment, the purity of the specimens, and the available magnetic field.

The specimen was immersed in liquid helium in a conventional nitrogen-shielded cryostat and the temperature could be further reduced to about 1.2°K, the normal working temperature, by pumping on the helium bath. A Weiss electromagnet produced a transverse magnetic field of up to 14 kilogauss with a pole gap of 6.4 centimetres.

A series of measurements was made of the ultrasonic absorption as a function of magnetic field for each specimen. In order to cover the maximum range of relative orientations of the magnetic field and crystal axes, the specimens were rotated about their axes and their angle of rotation measured on a scale at the top of the cryostat. Readings were normally taken every ten degrees around the zone, although in regions of rapid variation of attenuation with angle, five degree intervals were used. The signal received in a magnetic field was compared to that in the superconducting state, and since the electronic attenuation in superconducting lead at 1.2°K is negligible, this gave an absolute value of the ultrasonic absorption by the electrons.

2. (b) Preparation of Specimens

The specimens used in the experiments were single crystals of high purity lead in the form of cylinders, one centimetre in diameter and one centimetre long. These were made by casting Associated Lead Manufacturers' high purity (99.999%) lead into a graphite mould in vacuo. Great care was taken to keep impurities out of the molten material. The crucible was made

of pure graphite and was outgassed before use by heating to over 1000°C in a high vacuum. The material was thoroughly cleaned before casting, by etching with concentrated hydrochloric acid and washing with distilled water.

In order to produce crystals of suitable pre-determined orientations, a series of seed crystals in the form of rods, 2 millimetres in diameter and about 10 centimetres long, was grown in a horizontal furnace. Each was then placed in a narrow hole in the graphite mould and partially melted with the material for the crystal, the bottom end being prevented from melting by water cooling. On cooling, a one centimetre cylindrical crystal of the same orientation as the seed was formed.

The single crystal cylinder was carefully removed from the crucible and the ends very gently turned off in a lathe until it was about 10.5 millimetres long. It was then waxed into a jig, which had been prepared with faces plane parallel to better than 10^{-3} millimetres, and the crystal faces were ground flat and parallel on a glass plate, with emery powder of decreasing roughness, and finally with rouge powder. This treatment left a thin polycrystalline layer, about one tenth of a millimetre thick, on the end faces of the specimen, which caused acoustic scattering and some loss of power. It was found possible to remove this layer, however, by etching in strong acid for some hours, and the etch was sufficiently even to leave a flat monocrystalline surface. The specimens were allowed to anneal at room temperature for some days before use.

The specimens were oriented on an optical goniometer, by etching in concentrated hydrochloric acid and observing etch pit reflections from prominent faces ($[100]$ and $[111]$). For this purpose, a fine scratch was made on the curved side of the crystal parallel to the axis and this, together with the direction of the axis served to define the crystal orientation completely. The orientations of the ten specimens used in the investigation are shown in figure 2a.

The electronic free path in the lead crystals was estimated by measuring the resistance ratio $R_{300^\circ\text{K}}/R_{1.2^\circ\text{K}}$, and using the ratio of conductivity to mean free path deduced by Chambers (1952), from anomalous skin effect measurements. Superconductivity was destroyed, at low temperatures, by means of a longitudinal magnetic field, and the effects of magnetoresistance taken into account by extrapolating the resistance curve to zero field. The resistance ratios of filaments carefully cut from three of the crystals were measured and were all found to lie between 2.0×10^4 and 2.5×10^4 . From Chambers' value of the ratio of conductivity to free path for lead, and the mean ultrasonic velocity, ql for these crystals was found to be between 20 and 25 at 80 Mc/s and 1.2°K .

3. The Experimental Results

3. (a) The Magnetoacoustic Oscillations

Because of the relatively high value of ql , well defined magnetoacoustic oscillations are observed for almost all field directions. Although as many

as six maxima can be distinguished in some orientations, above the superconducting critical field, these cannot, in general, be analyzed unambiguously into contributions periodic in reciprocal field. This is readily explained by the fact that there are, for any magnetic field direction, many sections of Fermi surface contributing oscillations, and these interfere in a complex manner. In some cases, however, oscillations periodic in reciprocal field are observed, and these probably indicate that the contribution from one section of the Fermi surface is dominating that from the others.

The difficulty of resolving the oscillations into periodic contributions renders the use of equation (1) for determining the Fermi surface impracticable in most cases. It is found, however, that the final peak in a train of oscillations, which occurs at the highest magnetic field and which therefore corresponds to the smallest electronic orbit, is much larger and better defined than the other maxima. This feature is also present in the calculations for the free electron model, and it can be used to make an estimate of the sizes of the electron orbits which contribute to the oscillations. To make this estimate, a parameter Δk is used, defined by the equation

$$\Delta k = \frac{8e}{\hbar c q} H_m \quad (2)$$

where H_m is the magnetic field at which the prominent final maximum of a train of oscillations occurs. For the free electron gas Δk is just the diameter of the Fermi surface, as can be readily verified from the calculations of

Kjeldaas and Holstein (1959). Δk is therefore related to the extension of the Fermi surface normal to the direction of propagation and the magnetic field. The electron orbits are not in general circular, however, and the phase of the oscillations can be altered by effects associated with deformation of the Fermi surface, as discussed by Pippard (1960), so it is not a particularly reliable estimate. It is noteworthy, however, that for those orientations in which a periodic train of oscillations is experimentally observed, the values of Δk calculated from the period of the oscillations and from the position of the high field maximum agree, to within a few percent.

The Δk values can readily be derived from the frequency and velocity of the sound waves, and the magnetic fields at which the prominent maxima occur. The velocities were determined in a separate experiment by measuring the time interval between successive reflections. The results, estimated to be accurate to better than 3% are given in Table I.

Table I

Specimen Number	Velocity (cms /sec x 10^{-5})
1 and 3	2.37
2	2.40
4	2.48
7 and 9	2.20
8	2.37
10	2.50
11	2.56
12	2.44

The velocity measured was that of the principal acoustic mode generated when an X-cut crystal was attached to the metal. Only when the direction of propagation is along a symmetry direction is this a true longitudinal mode.

The oscillations can conveniently be considered in four groups, according to their period, the fields at which they occur, and the range of magnetic field directions over which they can be observed. We shall call these Groups I, II, III, IV, and shall now discuss them in detail.

Group I consists of prominent and well-defined oscillations which appear only when the magnetic field is near the $[100]$ or $[111]$ directions. These oscillations appear very suddenly as the field is rotated, growing from zero to their full amplitude over a range of two or three degrees, and they rapidly dominate any others which are present. The regions over which Group I oscillations are observed are shown in figure 2b and examples are shown in figures 3 and 4, where the attenuation is plotted against magnetic field on a reciprocal scale.

When the magnetic field is near $[100]$, two well defined peaks are observed, with small subsidiary maxima. When $\underline{H}$ is along a tetrad axis, the Δk values for these two peaks vary from $2.4 \times 10^8 \text{ cm}^{-1}$ and $1.5 \times 10^8 \text{ cm}^{-1}$ to $2.1 \times 10^8 \text{ cm}^{-1}$ and $1.1 \times 10^8 \text{ cm}^{-1}$ respectively as the direction of propagation goes from $[100]$ to $[110]$. As the magnetic field is tilted away from (100) in the (100) plane, Δk increases slightly and the oscillations disappear

at an angle of about 20° . For the magnetic field in the (110) plane, however, Δk remains almost unchanged and the high field peak disappears at an angle of about 8° .

When the magnetic field is near [111], one observes an extremely prominent peak which occurs at about 5 kilogauss and corresponds to Δk values of between $2.6 \times 10^8 \text{ cm}^{-1}$ and $2.9 \times 10^8 \text{ cm}^{-1}$ when the field is along a triad axis. It is not possible to deduce any systematic correlation between the magnitude of Δk and the propagation direction from the results, since the variations are small and apparently random. As the magnetic field is tilted away from [111] in any direction, Δk increases and it reaches values up to $3.3 \times 10^8 \text{ cm}^{-1}$ before the oscillations disappear. This occurs at an angle of about 15° when the field is tilted towards [100] and about 18° towards [110]. There is also a smaller peak over this angular range which occurs at Δk values of roughly $1.2 \times 10^8 \text{ cm}^{-1}$.

Group II comprises a series of oscillations with a prominent peak at about 3 kilogauss. Although these are not in general periodic in reciprocal field, there are orientations in which they are, and figure 5 shows an example where the oscillations occur at almost exactly the relative positions predicted by the free electron theory. In figure 2c, the values of Δk corresponding to the high field maximum are plotted stereographically as a function of the normal to both the propagation and magnetic field directions. The angular range over which these oscillations can be observed is shown in figure 2b.

There is probably no significance in the fact that Group I and II oscillations cannot be observed simultaneously, since the former rapidly dominate the latter when they are present together, but there is a region between [100] and [111] in which no oscillations of either group are observed, at high magnetic fields.

Group III oscillations occur at fields of less than about 1500 Gauss and are associated with small sections of Fermi surface and subsidiary maxima from larger sections. A detailed analysis of these oscillations is hardly feasible or profitable, since they have an extremely complex form and show no obvious systematic variation with angle.

Group IV consists of a few small peaks, occurring at high fields of 6 kilogauss and above and corresponding to Δk values of $4.0 \cdot 10^8 \text{ cm}^{-1}$ and greater. These oscillations are observed principally near [110] and are very sensitive to the magnetic field direction, but very small peaks at high fields can also be observed occasionally between [100] and [111], in the region where the Group I oscillations are not seen. An example of a Group IV oscillation for H near [110] is shown in figure 4. Because of their small amplitude and elusive nature it was not possible to make a detailed study of these oscillations.

3. (b) Absorption at High Fields

The limiting behaviour of the ultrasonic absorption in high transverse magnetic fields has been discussed by Pippard (1960). He finds that the

attenuation of longitudinal waves tends to a limit when the electron orbit diameter becomes much less than the acoustic wavelength and $\omega_c \tau$ is much greater than 1 for all electrons, provided that only closed orbits are present. ω_c is the electronic cyclotron frequency and τ the mean relaxation time around the orbit. In the presence of open orbits, however, the attenuation does not saturate, but continues to rise as H^2 , provided that the open orbit direction does not coincide with the direction of propagation. Kaner (1960) on the other hand, states that the attenuation saturates at high fields, whether or not open orbits are present, although it can show considerable anisotropy.

The principal difficulty in determining the directions of non-saturation from the experimental results in this case is that, although $\omega_c \tau$ is about 30 in a field of about 14 kilogauss, the ratio of the acoustic wavelength to the largest orbit diameter is only about 2 in this field. Consequently the attenuation has not necessarily reached its saturation value in the highest field and it is difficult, in some orientations, to decide whether the absorption is increasing towards a constant value, or indefinitely. At 78 Mc/s, the attenuation is still rising with field, at 14 kilogauss, if the field is directed near [100] or [110]. Further experiments at 45 Mc/s,* using fields up to 21 kilogauss, showed, however that the attenuation for H near [100] does saturate, although it is still rising with field when H is near [110], approximately as $H^{1.5}$ in the highest fields. The magnetic field directions for apparent non-saturation of the absorption are plotted stereographically in figure 2d.

* These experiments were carried out in collaboration with Mr. R. J. Kearney at Iowa State University.

3. (c) Anisotropy of Attenuation at High Magnetic Fields

The ultrasonic absorption was measured as a function of the magnetic field direction for all the specimens, in fields of about 13 kilogauss. The resulting rotation diagrams for propagation along $[100]$ and $[110]$ are shown in figure 6.

The results have two general features:

(1) When the magnetic field is in such a direction that the Group I oscillations can be observed, the attenuation at high fields is correspondingly large.

Furthermore the attenuation changes rapidly with orientation as the oscillations appear. This can be seen particularly clearly for propagation along $[100]$, for the region of high attenuation is just that over which the Group I oscillations are present.

(2) The attenuation is higher in the directions, near $[110]$, where it does not appear to saturate in high fields. This can be seen clearly in the rotation diagram for propagation along $[110]$, which also illustrates the general feature that the change in attenuation with magnetic field direction between regions of saturation and non-saturation is rather gradual.

3. (d) Dependence of the Attenuation on Propagation Direction

Since lead is superconducting at liquid helium temperatures, it is not possible to measure the dependence of the attenuation on propagation direction below the critical field directly, but an attempt can be made to extrapolate the results to zero field. This is a somewhat rough procedure, however, and

the significance of the results, which are given in the second column of Table II may be further reduced by possible differences in purity among the specimens, although resistance ratio measurements on three of them showed only small variations.

Another measure of the effect of propagation direction on the electronic absorption is given by the magnitude of the largest maximum in the magnetoacoustic oscillations observed for a particular specimen. This can be measured with much greater accuracy, and the results are displayed in the third column of Table II.

Table II

Specimen	Attenuation ($H = 0$)	Attenuation (Maximum)
	(nepers/cm)	(nepers/cm)
2	0.6	2.0
3	0.6	1.7
4	0.5	1.9
8	1.0	2.4
9 [100]	1.2	4.4
10 [111]	1.0	1.7
11 [110]	0.7	1.8
12	0.6	1.7

For comparison, the free electron value for the attenuation in zero field, taking the number of electrons to be 4 per atom, and ql as much greater than one, is approximately 8 nepers/cm. In a high magnetic field, on the other hand, the free electron theory predicts a value of 2.5 times the zero field value, taking ql as 20.

4. Further Information on the Electronic Structure of Lead

In order to interpret the experimental results in detail, a model of the electronic structure of lead is required. Such a model has been provided by the results of previous investigations and a brief review of some of the existing evidence concerning the Fermi surface in lead will be given before the magnetoacoustic measurements are discussed in detail.

An experimental study of the anisotropy of the De Haas-Van Alphen effect, together with a consideration of the nearly free electron model led Gold (1958) to propose, as a model for the Fermi surface, a modification of the latter consistent with his results. Lead has 4 conduction electrons per atom and the Brillouin zone, which is shown with the nearly free electron Fermi surface in figure 7, contains 2 electrons per atom. It is therefore probable that there are regions of Fermi surface in more than one zone. Gold suggested that the first zone is full and, just as in the free electron model, electrons overlap all the zone faces, leaving a single hole surface in the second zone. No very detailed study of this surface was made, but, since the periods of the De Haas-Van Alphen oscillations which were assigned to it were almost

constant, it was proposed that it is roughly spherical with a radius of $0.57 \frac{2\pi}{a} \text{ cm}^{-1}$ in $\underline{k}$ space, where $a \approx 4.94 \text{ \AA}$, the lattice constant of lead. It was recognized, however, that there was considerable uncertainty in the shape of this surface, and it is noteworthy that the free electron surface departs considerably from a sphere.

A surface formed by the overlapping of electrons across the zone edges was proposed for the third Brillouin zone. This surface, shown in figure 8, is multiply connected and may be thought of simply as the thickened up zone edges of the Brillouin zone. The evidence for this surface consisted of low frequency oscillations with an angular dependence consistent with the variation of area of the arms of the surface, and oscillations with the period and angular variation characteristic of the orbit ξ in figure 8.

Gold also observed beat patterns in the oscillations attributed to the arms of the third zone surface, and suggested that these were due to the occurrence of oscillations arising from the small electron surfaces in the fourth zone. Harrison (1960) showed, however, that these could be accounted for in detail by orbits around the arms of the multiply connected surface in the third zone and he and Gold (1960) suggested that the fourth zone was probably empty. Aubrey (1960) discovered that this hypothesis also improves the agreement with the estimate of the total Fermi surface area from the anomalous skin resistance in a polycrystalline sample. Beat

patterns in the low frequency De Haas-Van Alphen oscillations and the effective mass data obtained by Aubrey (1960) from cyclotron resonance experiments, suggest that the arms of the multiply connected surface are not cylindrical but taper in such a way that there is a minimum area near the zone corners.

More recently Alekseevski and Gaidukov (1961) have determined the magnetic field directions for which open orbits exist, by making a detailed study of the magnetoresistance of lead single crystals. They proposed that the Fermi surface consists of a network of $[111]$ cylinders, although this is incompatible with the work of Gold. It was shown, however, (Mackintosh (1960b)) that the complex orbits ρ and τ of figure 8 can exist on the multiply connected surface and Young (1962) has shown in detail how these can account for the magnetoresistance data. Young (1962) and Khaikin and Mina (1962) have also studied cyclotron resonance in more detail and conclude that their results are consistent with Gold's model.

5. Discussion

5. (a) The Fermi Surface of Lead

The Group I oscillations can be accounted for very satisfactorily by orbits on the multiply connected surface in the third zone. The orbits η and ξ in figure 8 can only exist when the magnetic field is near $[100]$, and θ and σ only near $[111]$. If we take the arms to be cylindrical, with a radius given by Gold's value of $0.19 \frac{2\pi}{a} \text{ cm}^{-1}$, we can calculate approximately

the angular ranges over which these orbits should exist. For the θ and σ orbits the result is 15° and 17° , as the field is tilted in the $[100]$ and $[110]$ directions respectively, which compares well with the experimentally determined values of 15° and 18° .

The angular dependence of the Group I oscillations near $[100]$ is more complex. The three largest orbits from the multiply connected surface which can contribute to the oscillations in this region are η , ξ , and γ of figure 8. There are also, of course, possible orbits from the hole surface. The angular region in the (100) plane over which the Group I oscillations are observed agrees very well with the value of just over 20° for the range of the β De Haas-Van Alphen oscillations observed by Gold. This value also agrees well with the calculated value of 21° for the angular extension of the η and ξ orbits. Young (1962) has suggested however that the β oscillations might be associated with the γ orbit, in which case all three orbits should cease to exist at about the same magnetic field direction, which would account for the rapid decrease of attenuation at this point (see figure 6). As the field is tilted towards $[110]$ however, the prominent high field peak, which we associate with the orbit η , decreases rapidly in amplitude and vanishes at an angle of about 10° . This behaviour is at first sight, somewhat puzzling, since the model predicts an angular range of about 25° for this orbit. Perhaps the most likely explanation is that the shape of the orbit becomes less favourable for contributing strong oscillations

as the magnetic field moves away from $[100]$ towards $[111]$. This would happen for instance, if the arms of the multiply connected surface had the shape of the free electron model of figure 7, in which case no well defined extremal length exists when the magnetic field is tilted away from $[100]$. The increase in the Δk value for the η orbit when the magnetic field is along $[100]$, as the sound propagation direction goes from $[110]$ to $[100]$ indicates that the orbit is somewhat square, rather than circular, so that, as in the free electron model, the Fermi surface follows the zone edges.

The Δk values and angular ranges of the Group I oscillations are compared with the predictions of the connected cylinder model in Table III, in the cases where a fairly unambiguous identification can be made.

The η and σ orbits, which are the best identified, give Δk values larger than the theoretical, which indicates that the phase of these oscillations is probably shifted to higher fields. This is not surprising, however, since the large amplitude of these oscillations, as compared with those of Group II, indicates that the deformation of the multiply connected surface under strain is large, and this may be taken as evidence that the free electron theory is breaking down. Furthermore the η and ξ orbits are probably not circular, and this again leads to a phase shift.

Table III

Orbit	$\Delta k \times 10^{-8} \text{ cm}^{-1}$		Angular Range	
	Theory	Experimental	Theory	Experimental
σ [111]	2.2	2.9-2.6	{ [100] 15° [110] 17°	15°
θ [111]	1.2	~1.2		18°
η [100]	1.8	2.4-2.1	[111] 25°	10°
			[110] 21°	20°
ξ [100]	0.8	{ 1.5-1.1	[111] 25°	23° (?)
			[110] 21°	20°
γ [100]	0.4-1.3		[110] 30°	
			[111] 28°	

Notes

(a) The crystallographic directions in the Angular Range column indicate the direction of tilting of the magnetic field.

(b) The theoretical values are based on a model of interconnected cylinders, following the zone lines.

(c) The γ orbit has the approximate shape of a four cornered star, on this model.

(d) The angular range of 23° for the ξ orbit as the field is tilted towards [111] comes from the range over which Group II oscillations are not observed.

The Group II oscillations probably arise from the hole surface in the second zone. The dimensions shown in figure 2c are generally unreliable since they are based only on one high field oscillation. In three cases, however, a short train of three or four oscillations was observed, periodic in $\frac{1}{H}$, and the Fermi surface extensions calculated from the periods are shown encircled in figure 2c. The values 1.6 and $1.7 \times 10^8 \text{ cm}^{-1}$ for the extension in the $[110]$ directions can be compared with the free electron hole surface value of $1.74 \times 10^8 \text{ cm}^{-1}$. In the $[100]$ direction the value of $1.8 \times 10^8 \text{ cm}^{-1}$ is somewhat smaller than the free electron value of $1.94 \times 10^8 \text{ cm}^{-1}$.

The Δk values determined from the final peaks appear to indicate a bulging of the Fermi surface towards the $[111]$ face of the Brillouin zone, whose extension in this direction is $2.2 \times 10^8 \text{ cm}^{-1}$. The free electron surface is concave towards $[111]$ and has an extension in a central plane of only $1.27 \times 10^8 \text{ cm}^{-1}$. Although the zone boundary would tend to cause the hole surface to bulge towards it, it seems likely either that non-central orbits are contributing to the oscillations or that there is a marked phase shift to higher fields for orbits which pass near $[111]$ on the hole surface.

The absence of oscillations from the hole surface over a region between $[111]$ and $[100]$, as shown in figure 2b, is, at first sight rather puzzling. A possible reason is that there is destructive interference between the

Group I and Group II oscillations in this region, so that the Group II oscillations can only appear when the η , ξ , and γ orbits no longer exist. The tentative value of 23° for the angular range of the ξ orbit in Table III is assigned on this basis.

The Group III oscillations are presumably subsidiary maxima of the high field peaks and oscillations arising from orbits such as ξ , ω , and γ around the arms of the multiply connected surface. No more detailed identification appears to be possible at the moment.

The Group IV oscillations for $\underline{H}$ near $[110]$ are probably associated with the orbit τ , which can exist over a range of angles near $[110]$. The Δk values of around $4.0 \times 10^8 \text{ cm}^{-1}$ which are observed for these oscillations agree roughly with the expected extension of the τ orbit. The other Group IV oscillations may be associated with extended orbits on the multiply connected surface, but they have not been studied in sufficient detail to allow positive identification.

The regions of apparent non-saturation of the attenuation at high fields agrees fairly well with the two dimensional regions of open orbits deduced by Alekseevski and Gaidukov (1961) from their magnetoresistance measurements. The one dimensional regions, due to the orbits ρ , μ , and λ have not been observed in this work, but this is not particularly surprising since these regions have a very small angular range and measurements were taken

only at intervals of five or ten degrees. It seems likely, therefore, that the attenuation does not saturate, as predicted by Pippard (1960), if there are open orbits present which do not run along the direction of propagation. This conclusion is supported by measurements on tin (Mackintosh (1960b) and to be published). It is desirable to carry out further experiments on very pure lead at higher fields to check this conclusion, and to seek the directions of non-saturation in the (111) plane.

The absolute magnitude of the attenuation is somewhat smaller than the free electron theory predicts. This result emphasizes the fact that the free electron value gives only a rough approximation to the ultrasonic absorption, since both the Fermi surface and its deformation under strain are strongly affected by the presence of the lattice. When the attenuation saturates at high fields, it does so at values of typically 2 to 5 times the extrapolated zero field value, which can be compared with the corresponding ratio of 2.5 for the free electron model, for a ql of 20.

The only feature of the dependence of attenuation on propagation direction which seems to be sufficiently clear to be worthy of comment is the relatively high absorption for propagation along [100], particularly in a magnetic field. In zero field, at high ql values, the electrons which cause most of the attenuation are those which can stay in phase with the sound wave, and are therefore moving almost normal to the direction of propagation. For this orientation, such electrons are located on the

η , ξ , and γ orbits and on the hole surface. The electrons on the multiply connected surface are associated with regions of high deformability under strain, and these regions have low curvature normal to $[100]$, so that many electrons contribute significantly to the attenuation. Analogous considerations apply in a magnetic field. No other direction of propagation has an equal number of potent orbits.

The magnetoacoustic results and all the other experimental data are therefore consistent with a Fermi surface of lead composed of two sheets. The hole surface in the second zone has dimensions in the $[100]$ and $[110]$ directions which appear to be somewhat smaller than the free electron model. Such a shrinking of the hole surface would be consistent with, and indeed required by, the non-existence of electrons in the fourth zone and the reduced volume of the electron surface in the third zone, since Luttinger (1960) has shown that, even taking account of electron-electron interactions, the Fermi surface must contain an integral number of electrons. There is some indication, which may be spurious, of a bulging of the hole surface towards $[111]$.

There is strong evidence for the multiply connected surface in the third zone, especially from the range of existence of the Group I oscillations associated with the η , σ , and θ orbits, which have not previously been observed. There is evidence that the η and ξ orbits are not circular and an indication, from the behaviour of the oscillations

from the η orbits, that the arms may have approximately the cross sectional shape of the free electron Fermi surface. In addition oscillations have been observed which can be associated with the orbit τ , and the apparent directions of non-saturation of attenuation are consistent with the model. Further experiments at higher frequencies and magnetic fields should provide more detailed information and help resolve the remaining discrepancies.

5. (b) Evaluation of the Method

Experimentally, there are few severe difficulties in making extremely accurate measurements of the change in the ultrasonic absorption when a magnetic field is applied to a metal, although measuring the absolute electronic attenuation is generally rather difficult, unless the metal is superconducting. The change in attenuation can readily be measured to better than one percent, and the positions of the maxima and minima located with great accuracy.

The interpretation of the results is a more difficult task however, particularly for polyvalent metals with several sheets of Fermi surface, since the interaction between orbits from different surfaces can be quite complicated. The total attenuation is not simply the sum of the contributions expected from each surface separately, but complicated interference effects occur, which tend to shift the maxima and blur out the oscillations, particularly at high fields. This was shown explicitly by a calculation of the

ultrasonic attenuation in a metal with two spherical sections of Fermi surface of different sizes (Mackintosh 1960b). In both of these respects the magnetoacoustic effect contrasts unfavourably with the De Haas-Van Alphen effect. It should be possible, however, to sort out the various contributions fairly well by using extremely pure materials and high frequencies and observing the magnetoacoustic oscillations down to very low fields. A major difficulty in this work was the high superconducting critical field of lead, which effectively removed the low field oscillations, but this can be overcome to a considerable extent by the use of higher frequencies.

Although Fermi surface parameters cannot be deduced from the magnetoacoustic effect as readily as they can from the De Haas-Van Alphen effect, the method does have some advantages. From the change in the oscillations with the direction of propagation keeping the field direction fixed, it is possible to obtain detailed information about the shapes of orbits which cannot readily be determined by other means. Furthermore, it is probably most useful for examining large orbits which show up clearly at high fields, and these are generally the most significant in determining the shape of the Fermi surface. This is in marked contrast to the De Haas-Van Alphen effect, in which the smaller orbits with long periods are the most prominent, and this fact makes the two methods conveniently complementary, as exemplified by the study of the multiply-connected surface.

In addition, open orbit directions can apparently be determined by observing the magnetic field directions for which the attenuation does not saturate, although it may be more convenient to do this by measuring the transverse magnetoresistance. Non-saturation of ultrasonic attenuation appears to be a sure indication of open orbits, however, (Pippard (1960)), whereas non-saturation of magnetoresistance can be due to equality of electrons and holes, and careful experiments must be carried out to distinguish the two possibilities.

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Figure Captions.

- Figure 1. Schematic diagram of the apparatus.
- Figure 2. (a) Stereographic projection of the specimen axis.
(b) Angular directions of the magnetic field for which the Group I and Group II oscillations are observed.
(c) Δk values for the hole surface deduced from the Group II oscillations. The circled values are derived from the period of a series of oscillations.
(d) Magnetic field directions for which the absorption does not saturate at 45 Mc/s. and 21 kilogauss.
- Figure 3. Magnetoacoustic effects in lead at 78 Mc/s. and 1.2°K.
- Figure 4. Group I magnetoacoustic oscillations observed for propagation along [110].
- Figure 5. The Group II oscillations when the propagation direction is along [110] and the magnetic field is along [110].
- Figure 6. Rotation diagrams for propagation along [100] and [110] at 78 Mc/s. 13 kilogauss, and 1.2°K.
- Figure 7. The nearly free electron Fermi surface for lead (after Harrison (1960)).
- Figure 8. Electron and hole orbits on the multiply connected surface in a magnetic field.

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Fig. 3 Magnetoacoustic effects in lead at 78 Mc/s. and 1.2°K.

Fig. 4 Group I magnetoacoustic oscillations observed for propagation along $[110]$.

Fig. 5 The Group II oscillations when the propagation direction is along $[110]$ and the magnetic field is along $[110]$.

Fig. 6 Rotation diagrams for propagation along [100] and [110] at 78 Mc/s. 13 kilogauss, and 1.2°K.

Fig. 7 The nearly free electron Fermi surface for lead (after Harrison (1960)).

Fig. 8 Electron and hole orbits on the multiply connected surface in a magnetic field.