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Fermi Surface of Thallium from Magnetoacoustic Measurements

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Magnetoacoustic measurements, at frequencies up to 270 Mc/sec, have been made on single-crystal specimens of thallium. From the observed oscillatory behavior, the extremal dimensions of the Fermi surface have been obtained. These dimensions agree quite closely with the predictions of the free-electron model, assuming a single-zone scheme to take into account the effects of spin-orbit splitting. No unequivocal effects due to magnetic breakdown have been observed.

I. INTRODUCTION

A CONSIDERABLE amount of theoretical and experimental work has been devoted to elucidating the band structure of divalent metals with a hcp structure.[1] Such studies have revealed that the free-electron model provides a very good description of the band structure of these metals provided that the possible effects of spin-orbit splitting are taken into account.[2] This splitting removes the degeneracy associated with {0001} faces of the Brillouin zone for a hcp structure and necessitates the use of a single-zone, rather than the double-zone, scheme in constructing the appropriate Fermi surface. For the divalent metals, however, the effects on the band structure are relatively minor and in the case of zinc, for example, principally involve the connectivity of the hole surface in the second zone and the existence of small hole pockets in the first zone.[3]

For a trivalent hcp metal the effects of the change from a double zone to a single zone are much more marked. Hitherto, no studies of these effects has been undertaken. In the case of thallium, which is trivalent and has a high atomic number, the spin-orbit splitting should be quite large and for this reason it was chosen as the object of the present investigation. The results do, in fact, indicate that spin-orbit effects are considerable and that the extremal dimensions of the Fermi surface are in quite good agreement with the free-electron surface derived from a single-zone scheme. No effects clearly due to magnetic breakdown[4] are observable.

II. EXPERIMENTAL TECHNIQUE

Measurements were made by an automatic recording technique,[5] at frequencies up to 270 Mc/sec, using longitudinal waves. Because of the low Debye temperature of thallium, all data were taken at approximately 1.3°K to minimize the effects of phonon scattering. Glycerin was found to give satisfactory echoes in all cases.

Thallium is hcp at room temperature but at 234°C undergoes a transformation to a bcc structure which is stable to the melting point. To produce single crystals of the low-temperature hexagonal phase, it was found necessary to adopt a procedure due to Meyerhoff and Smith.[6] High-purity thallium (99.999+%), obtained from the American Smelting and Refining Company, was melted down into an ingot approximately 1 in. in diameter and 3 in. in length. This ingot was then vacuum annealed at 215°C, just below the transformation temperature, for a period of 3 days. The resulting grain growth produced quite large crystals of the hexagonal phase which, by judicious selection, provided adequate specimens for magnetoacoustic measurements. Samples approximately 1-cm sq by 3 mm thick were cut from the ingot by a Servomet[7] spark erosion apparatus. The specimen axes were, respectively, along $[0001]$, $[10\bar{1}0]$, and $[\bar{1}2\bar{1}0]$.

Data were taken at 5-deg intervals in field orientation for $\mathbf{q}\|[10\bar{1}0]$ and $\mathbf{q}\|[\bar{1}2\bar{1}0]$, while for $\mathbf{q}\|[0001]$ 3-deg intervals were used. The sound wavelength, appropriate to each propagation direction, was measured directly at 10 Mc/sec using conventional pulse techniques. Reduction of the transit-time data was accomplished using the expansion measurements of Erfling.[8]

III. THEORY

A. Oscillatory Attenuation

From the theory of the magnetoacoustic effect[9] it is known that, for sound propagating in a direction $\mathbf{q}$ with a transverse magnetic field $\mathbf{H}$, the attenuation is periodic in $1/H$. This period $\Delta(1/H)$ is related to some dominant orbit extremum in the direction $\mathbf{q}\times\mathbf{H}$ by the relation

$$k_{\text{ext}}=\frac{e}{\hbar c}\frac{\lambda}{\Delta(1/H)}, \qquad (1)$$

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[1] See, for example, Walter A. Harrison, Phys. Rev. **118**, 1190 (1960).

[2] M. H. Cohen and L. M. Falicov, Phys. Rev. Letters **5**, 544 (1960).

[3] A. S. Joseph and W. L. Gordon, Phys. Rev. **126**, 489 (1962).

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

[5] J. A. Rayne, Phys. Rev. **129**, 652 (1963).

[6] R. W. Meyerhoff and J. F. Smith, J. Appl. Phys. **33**, 219 (1962).

[7] Metals Research Inc., Cambridge, England.

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

[9] A. B. Pippard, Proc. Roy. Soc. (London) **A257**, 165 (1960).

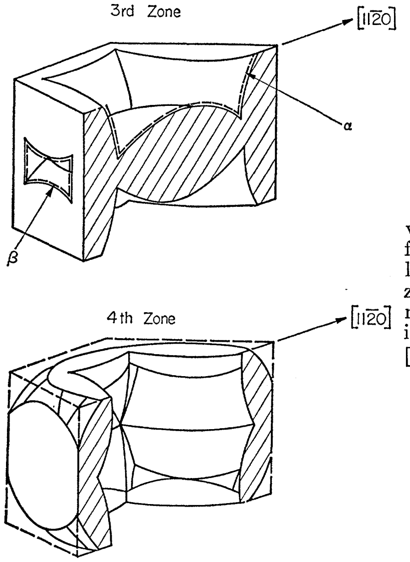

FIG. 1. Perspective view of third and fourth zones of thallium using single-zone free-electron model. The section is perpendicular to [11$\bar{2}$0].

where λ is the appropriate sound wavelength. As is usual in the interpretation of such experiments, we shall *assume* that the extremum measured is, in fact, the extremal dimension of the Fermi surface in the direction $\mathbf{q}\times\mathbf{H}$. The main justification for this assumption is that it seems to give results which seem to be eminently reasonable.

As will shortly become apparent, open orbits in thallium are possible when the magnetic field is in the basal plane. Under these conditions, when $\mathbf{q}$ is also in this plane, it is possible to obtain so-called resonant type oscillations.[10] Such resonant oscillations occur when the period of the open orbit in real space is equal to a multiple of the sound wavelength. This condition is fulfilled when the magnetic field satisfies the condition

$$H = c\hbar k_0/e\lambda n, \tag{2}$$

in which k_0 is the period of the open orbit in reciprocal space and n is an integer.

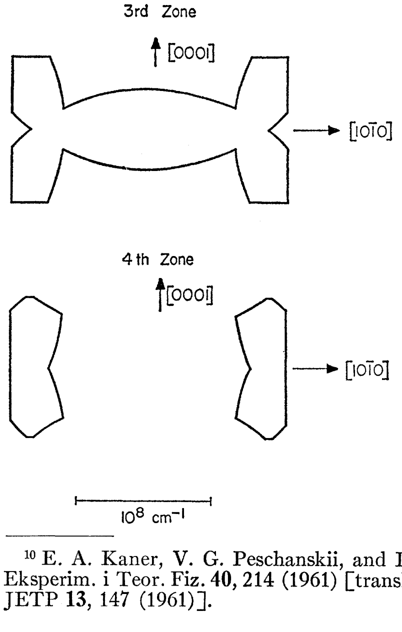

FIG. 2. Cross sections of third- and fourth-zone surfaces of thallium taken perpendicular to [10$\bar{1}$0].

B. Fermi Surface from Free-Electron Model

Harrison has constructed the Fermi surface for a trivalent hcp metal using a double-zone scheme.[1] In this case the first zone is full, the main part of the Fermi surface coming from a bamboo-like structure in the second zone. Small electron pockets exist in the third zone.

When a single-zone scheme is used, the nature of the surface is profoundly changed as may be seen from Fig. 1. Zones one and two are full as before. There are now, however, two-hole surfaces occupying zones three and four, respectively. The hole surface in zone three is a multiply connected corrugated cylinder with transverse membranes spaced roughly half as close as those in the double-zone surface. The fourth-zone surface is also corrugated cylinder, which can support open orbits. Cross sections of these surfaces, taken normal to [1$\bar{2}$10], are shown in Fig. 2. Electron surfaces also exist in the fifth and sixth zones, but the presence of these is somewhat conjectural in the presence of a finite crystal potential.

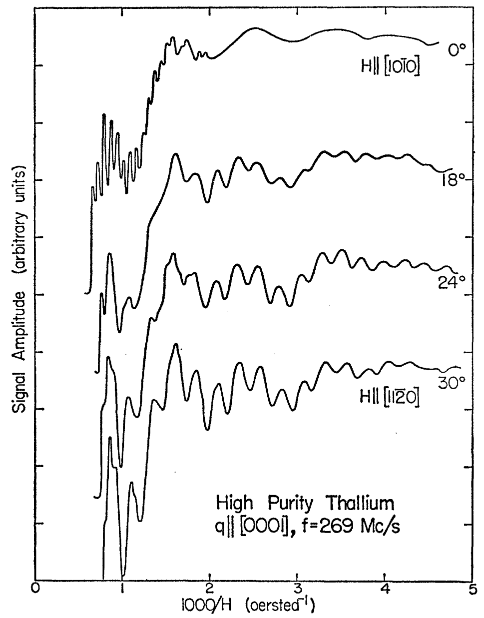

FIG. 3. Oscillatory attenuation of longitudinal 269 Mc/sec sound in thallium as a function of $1/H$. The propagation direction is [0001]. *Note added in proof.* In a preliminary communication, viz., J. A. Rayne [(Physics Letters 2, 128 (1962)] the directions [10$\bar{1}$0] and [1$\bar{2}$10] were inadvertently interchanged for $\mathbf{q}\|$[0001].

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

IV. RESULTS AND DISCUSSION

Representative data for each propagation direction are given in Figs. 3 through 5. It will be noted that in all cases pronounced oscillatory behavior is found and that usually more than one period is observed. From the number of oscillations it is possible to make an estimate of the electronic mean free path in the thallium. Thus, we have

$$l=\pi(n+\tfrac{1}{2})\lambda, \tag{3}$$

where n is the order of the last oscillation clearly observable. For $q\|[1\bar{2}10]$ one typically has $n\sim 30$, so that $l\sim 1.5$ mm, corresponding to a resistance ratio of about 100 000 to 1. The dominant oscillations are clearly periodic in $1/H$ and from the observed value of $\Delta(1/H)$, the extremal dimensions were calculated using Eq. (1). The resulting wave-number vectors (i.e., extremal *radii*) are shown in Fig. 6 and Table I.

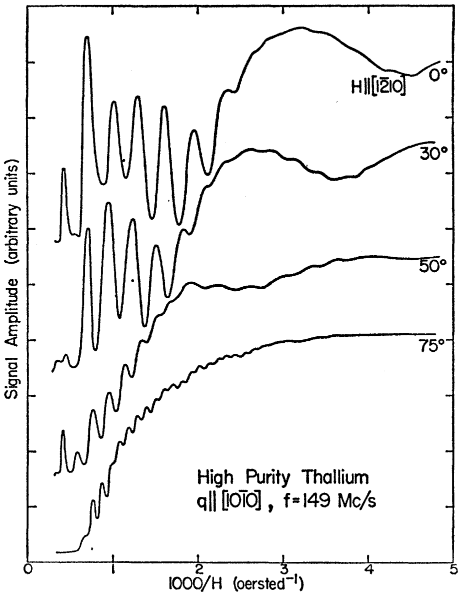


FIG. 4. Oscillatory attenuation of longitudinal 149 Mc/sec sound in thallium as a function of $1/H$. The propagation direction is $[10\bar{1}0]$.

As is evident from Figs. 6(a) and 6(b), there is marked anisotropy of the extremum for $\mathbf{q}\|[10\bar{1}0]$ and $[1\bar{2}10]$. In both cases, for H in the basal plane, an orbit such as α in the third zone is clearly involved, the extremal dimension being along [0001]. As can be seen from Table I, the magnitude is consistent only with the *single*-zone scheme and thus provides a convincing demonstration of the importance of spin-orbit splitting in thallium. Actually spin-orbit effects do not remove the degeneracy along $[10\bar{1}0]$,[4] so that for $H\|[1\bar{2}10]$ the *double*-zone dimension should be observed. Since the gap across the {0001} zone faces increases quite rapidly with angle, however, very precisely oriented crystals and fields would be required to observe double-zone orbits experimentally at low fields.

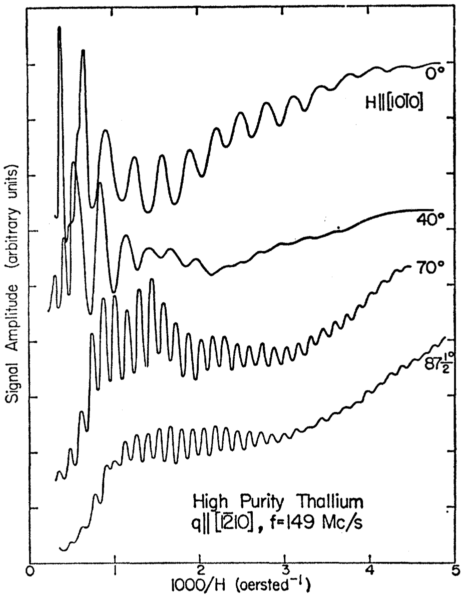
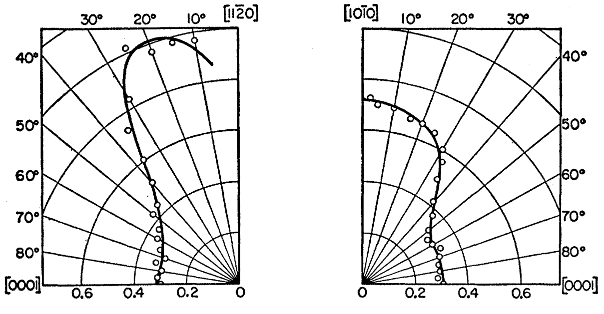


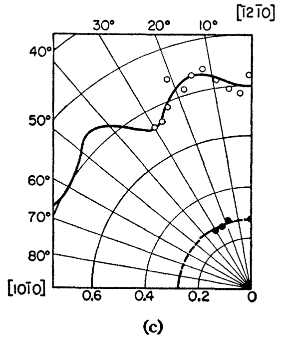
FIG. 5. Oscillatory attenuation of longitudinal 149 Mc/sec sound in thallium as a function of $1/H$. The propagation direction is $[1\bar{2}10]$.

At high fields for $\mathbf{q}\|[1\bar{2}10]$, $H\sim[10\bar{1}0]$, pronounced attenuation peaks occur. It is believed that these are resonance oscillations associated with open orbits in the



(a)

(b)



(c)

FIG. 6. Extremal values of wave-number vector for Fermi surface of thallium obtained from magnetoacoustic data.

Table I. Comparison of extremal wave numbers for the Fermi surface of Tl with single-zone free-electron model.

Propagation direction	Field direction	k_{ext} [a] experimental	Free-electron value[b]
$[0001]$	$[10\bar{1}0]$	0.80	0.88–1.05
		0.32	0.44
		0.07	?
	$[11\bar{2}0]$	0.73	0.69–0.76
		0.26	0.19–0.26
		0.07	?
$[1\bar{2}10]$	$[10\bar{1}0]$	0.31	0.26–0.43
	$[0001]$	0.73	0.69–0.76
$[10\bar{1}0]$	$[11\bar{2}0]$	0.31	0.26–0.43
	$[0001]$	0.80[c]	0.88–1.05

[a] All in units of 10^8 cm^{-1}.
[b] The range of values corresponds to the possible extrema of the free-electron model.
[c] Extrapolated value.

fourth zone. The period of the single-zone structure along $[0001]$ is $k_0=2\pi/c$. Taking $c=5.51$ Å, we have $k_0=1.14\times10^8$ cm^{-1} and, hence, from Eq. (2) $H=5000/n$, where n is an integer. The observed field at the peaks corresponds quite closely to the values of (4) for $n=2, 3$, viz., 2500, 1670. As the field is tilted out of the basal plane these peaks diminish in amplitude, as would be expected when the open orbits degenerate into extended orbits. Beyond about 10° tilt angle the peaks no longer appear. No peak corresponding to $n=1$ is observed, since the attenuation at such fields becomes too large. Similar, though less pronounced peaks, occur for $\mathbf{q}\|[10\bar{1}0]$, $H\sim[1\bar{2}10]$.

For both $\mathbf{q}\|[1\bar{2}10]$, $\mathbf{q}\|[10\bar{1}0]$ with the magnetic field nearly in the basal plane, a period roughly half that of the dominant oscillation is visible at high fields. The corresponding extremum is approximately that corresponding to a double-zone orbit dimension along $[0001]$. It is, thus, tempting to assign the appearance of these oscillations to a magnetic breakdown effect. For $H\|[10\bar{1}0]$, $\mathbf{q}\|[1\bar{2}10]$, however, the gap due to spin-orbit splitting is a maximum and it seems difficult to believe that a field of several kilogauss is sufficient to cause transitions across the gap.[11] At the moment no reasonable explanation of these oscillations can be advanced.

For both orientations with $H\sim[0001]$, the oscillatory behavior is undoubtedly due to orbits around the hexagonal cups formed in either or both the third and fourth zones. This assignment is confirmed by the extrema obtained for $\mathbf{q}\|[0001]$, which agree quite closely with the basal plane dimensions obtained from the other propagation directions. In both cases the agreement with the free-electron model is quite good, as may be seen from Table I. Another satisfactory feature of the observed oscillatory behavior is that for $H\sim[0001]$, the amplitude is much larger for $\mathbf{q}\|[1\bar{2}10]$ than for $\mathbf{q}\|[10\bar{1}0]$. Such behavior results from the hexagonal orbits involved, giving a much larger extremal path length in the former than in the latter case.

For $\mathbf{q}\|[0001]$, the oscillatory behavior involves three clearly defined periods, of which only two are shown in Fig. 6(c). As mentioned above, the short-period oscillation is certainly associated with hexagonal orbits in the third and fourth zones. The intermediate period, which is roughly isotropic, probably arises from orbits such as β in the third zone. From the observed isotropy, the shape of these indentations is considerably different from those predicted by the free-electron model. The long-period oscillations are probably associated with pockets of electrons in the fifth and sixth zones. A detailed comparison of these dimensions with the free-electron model seems hardly profitable, since the probable effects of the finite crystal potential on the theoretical dimensions are quite large.

It is noteworthy that the extremal dimensions of the Fermi surface in the basal plane [Fig. 6(c)] do not conform closely to the hexagonal cross section expected from the free-electron model. These deviations are also presumably explicable in terms of the effects of the finite crystal potential. As may be seen from Table I, however, the agreement between theory and experiment is generally satisfactory and demonstrates the essential correctness of the former quite effectively.

V. CONCLUSIONS

Magnetoacoustic measurements have been made in single-crystal thallium at frequencies up to 270 Mc/sec using longitudinal waves. The extremal dimensions of the Fermi surface agree very well with the free-electron model, assuming a single-zone scheme to take into account the effects of spin-orbit splitting. No well-defined effects due to magnetic breakdown were observed.

ACKNOWLEDGMENTS

The author would like to thank Paul Hodder who helped prepare the thallium single crystals used in these experiments. Thanks are due also to Dr. M. Priestley, Dr. A. Mackintosh, and Dr. L. Falicov for helpful discussions.

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[11] *Note added in proof.* Evidence of magnetic breakdown in thallium for magnetic fields ~20 kG has been reported; see A. R. Mackintosh, L. E. Spanel, and R. C. Young, Phys. Rev. Letters **10**, 434 (1963).