

INVESTIGATION OF THE FERMI SURFACE OF TIN BY THE METHOD OF CYCLOTRON RESONANCE

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Some problems relating to the experimental investigation of cyclotron resonance are considered. The anisotropy of the effective masses of the conduction electrons of tin in the (010) plane is studied in detail and in the (001) plane in less detail. Part of the masses found are ascribed to certain parts of the Fermi surface of tin constructed on the free electron approximation.^[14] The dimensions of the surface of the third hole band are determined.

CYCLOTRON RESONANCE INVESTIGATIONS

AN investigation of cyclotron resonance is one of the most promising means of studying the Fermi surface of a metal.^[1] An experimental study of cyclotron resonance spectra is a direct and most accurate method for measuring effective masses of conduction electrons, and also enables some properties of the Fermi surface to be determined.^[2,3]

In the first experiments^[4,5] aimed at observing the phenomenon of cyclotron resonance, the values of effective electron masses were calculated roughly from the single broad and rather indefinite minima in the dependence of the surface impedance of tin on magnetic field. Investigations by a more reliable method showed the existence of periodic minima in the variation of the surface impedance of tin^[6] and copper^[7] as a function of the inverse field, which should be peculiar to cyclotron resonance. A number of values of effective masses of electrons and holes have been obtained from these and also from more detailed studies of cyclotron resonance carried out recently on different metals.^[8,9] However, in none of the published papers have such well resolved spectra of sharp cyclotron resonances (consisting of groups of resonances, periodic in the inverse field, which reached several tens of orders) been obtained as was achieved^[2,3,11,12] by the method of frequency modulation.^[10]

Leaving aside trivial experimental drawbacks peculiar to some of the work (the bad quality of specimens, low sensitivity and imperfections in the methods of measurement), we shall cite here some important considerations either not taken

into account at all or only partially considered in all investigations except the work carried out by the method of frequency modulation.^[2,3,10-12] These omissions are evidently the main reason for the lack of clarity and poor resolution of the cyclotron resonance spectra shown in published work, making their interpretation very difficult and often ambiguous. For the same reason the physical meaning of the numerical results is partly lost. The trouble is that, in all these experiments, either a plane polarized wave was not used and the high frequency currents flowing in the surface of the specimen were not rectilinear^[8] (at least over a significant part of its surface), or the specimen did not have a plane (cylindrical) surface.^[9] This must have led to excitation of cyclotron resonances in different electron groups in different parts of the surface of the specimen. In fact, as is known from general considerations^[1] and can also be seen from the experiments described in the present work, the excitation of resonances for different singularities of the Fermi surface depends on the polarization direction of the wave relative to the crystallographic directions in the specimen. As a result, the spectra should have contained precisely twice as many groups of resonances (and furthermore of smaller amplitude) than would be observed with a plane polarized (transverse) electromagnetic wave and a plane specimen, other conditions being the same. Even if well-defined effective masses can be determined from such spectra, it remains impossible to determine the connection of these masses with definite singularities of the Fermi surface.

The use of a cylindrical specimen introduces another experimental complication because it is

hardly possible to prepare a cylindrical single crystal of sufficiently correct shape. The inaccuracy of specimen shape makes impossible the alignment of the magnetic field parallel to the whole surface of the specimen with the necessary accuracy of $\sim 10''$. As experiments carried out with the magnetic field parallel to the high-frequency current show, an inclination $20-30''$ of the field considerably changes the appearance of the spectrum of cyclotron resonances. Consequently, a difference in the inclination of the field relative to the different parts of the surface of the specimen of such an order of magnitude must lead to a sharp worsening in the resolution of the spectra. Besides, it is not possible to study anisotropy of effective masses in a cylindrical specimen by rotating the magnetic field relative to the specimen.

The considerations discussed lead to the conclusion that an investigation of cyclotron resonance has to be carried out on a specimen with a plane surface, and the high frequency currents flowing in it must be rectilinear. A strip resonator in which the only type of oscillations is a transverse electromagnetic wave (TEM) satisfies these conditions, and the specimen can be a plane rectangular strip of the resonance length or the plane base of the resonator placed near it (Fig. 1). Strip resonators described by the author^[10] were already used earlier^[2,3,11,12] and are used in the present work.

EXPERIMENT

The studies of cyclotron resonance were carried out by the method of frequency modulation^[10] on single crystal disks, 18 mm in diameter and 1–2 mm thick, made from extremely pure tin with the resistance ratio $\rho(20^\circ\text{C})/\rho(3.75^\circ\text{K}) = 1.4 \times 10^5$ (specimen No. 6 of^[13]). Single crystals were grown from the melt in polished quartz demountable formers; their natural surfaces did not receive any further treatment. The plane surfaces of the disks had the (010) and (001) orientation, checked by x-ray examination with an accuracy of $\sim 20'$. By rotating the disk contained in the strip resonator (Fig. 1) relative to the cavity, any direction of the high frequency currents \mathbf{J} relative to the crystallographic axes could be achieved. The magnetic field \mathbf{H} could also be directed in any way relative to the specimen by rotating (φ) and inclining (ψ) the electromagnet producing it.

Examples of cyclotron-resonance spectra for four different directions of the magnetic field are shown in Fig. 2. The direction of the field parallel to a crystallographic axis was determined with an

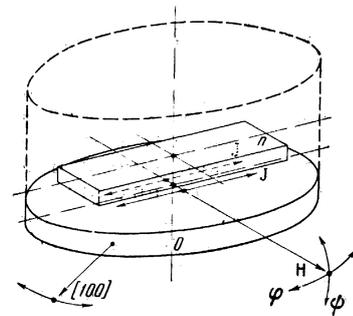


FIG. 1. Diagram of strip resonator. The disk specimen O serves as the base of the resonator, the volume of which is shown by the dashed line. The copper strip of the resonance length ($\lambda/2$) is placed over the specimen at a height of 1 mm. The high frequency currents \mathbf{J} , flowing in the specimen, are rectilinear.

accuracy of $\sim 1^\circ$ from the symmetry of such spectra made for several directions of the field on both sides of the axis. The magnet was set to make the field parallel to the specimen surface also by checking the symmetry of the cyclotron resonance spectra and by tilting the magnet for the greatest sharpness and best resolution.

An inclination of the field not only decreases the depth and broadens the resonances (to a different degree for different electron groups^[3]), but in many cases also shifts them relative to the field, leading to a change of effective mass m^* calculated from the formula

$$\mu = m^*/m_e = (e/m_e c \omega) / \Delta H^{-1} \quad (1)$$

(here $\omega = 5.967 \times 10^{10}$ cps is the frequency of the hf measuring field and ΔH^{-1} is the resonance period as a function of inverse magnetic field). Figure 3 shows this phenomenon for four electron groups with $\mathbf{H} \perp \mathbf{J}$. It can be seen that an inclination of even $3'$ produces a significant effect; when $\mathbf{H} \parallel \mathbf{J}$ the sensitivity to the inclination is even several times greater. The change in μ with field inclination is much more rapid than for rotation of the field in the corresponding crystallographic plane of the specimen when this plane coincides with its surface. The reason for this behavior is probably that an inclination of the field can lead to the appearance of a velocity component normal to the surface of the specimen among the electrons of the resonance section. This can change the conditions of cyclotron resonance in such a way that they appear optimal for a section of the Fermi surface parallel to the extremal direction (determined by the direction of \mathbf{H}), but slightly shifted relative to it in such a way as to compensate partially for the deterioration in the resonance conditions resulting from the appearance of a normal

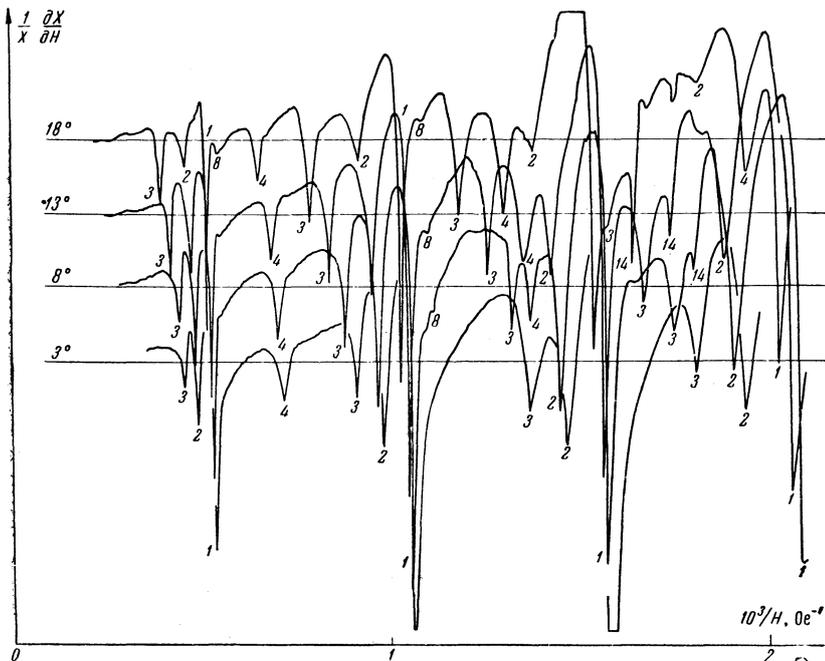


FIG. 2. Traces of cyclotron resonances in a single crystal of tin, the plane surface of which has the (010) orientation. The angle of rotation of the magnetic field in the plane of the specimen relative to the (001) axis is shown to the left of the curves; H is perpendicular to J . Groups of resonances periodic in the inverse field are numbered (the corresponding effective masses in Fig. 4 are indicated by the same numbers); X is the reactive part of the surface impedance of the specimen; the scale of the ordinate axis is linear.

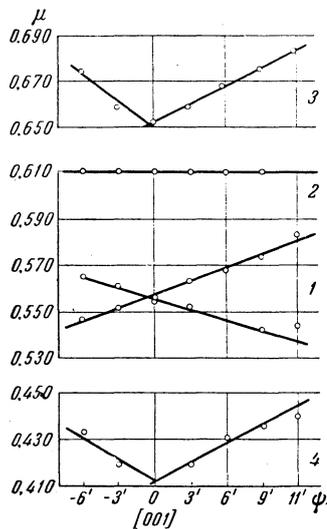


FIG. 3. The dependence of the effective masses, determined by Eq. (1), for several groups of cyclotron resonances, on the inclination of the magnetic field to the plane of the specimen. The numbers of the masses are the same as in Figs. 2 and 4.

component of the electron velocity. We should point out the broadening of some resonances observed on inclining the magnetic field, as the example of mass 1 in Fig. 3 shows. According to the point of view introduced, it can be suggested that optimal conditions for resonance can be achieved in this case by shifting the section from the extremal position in two directions.

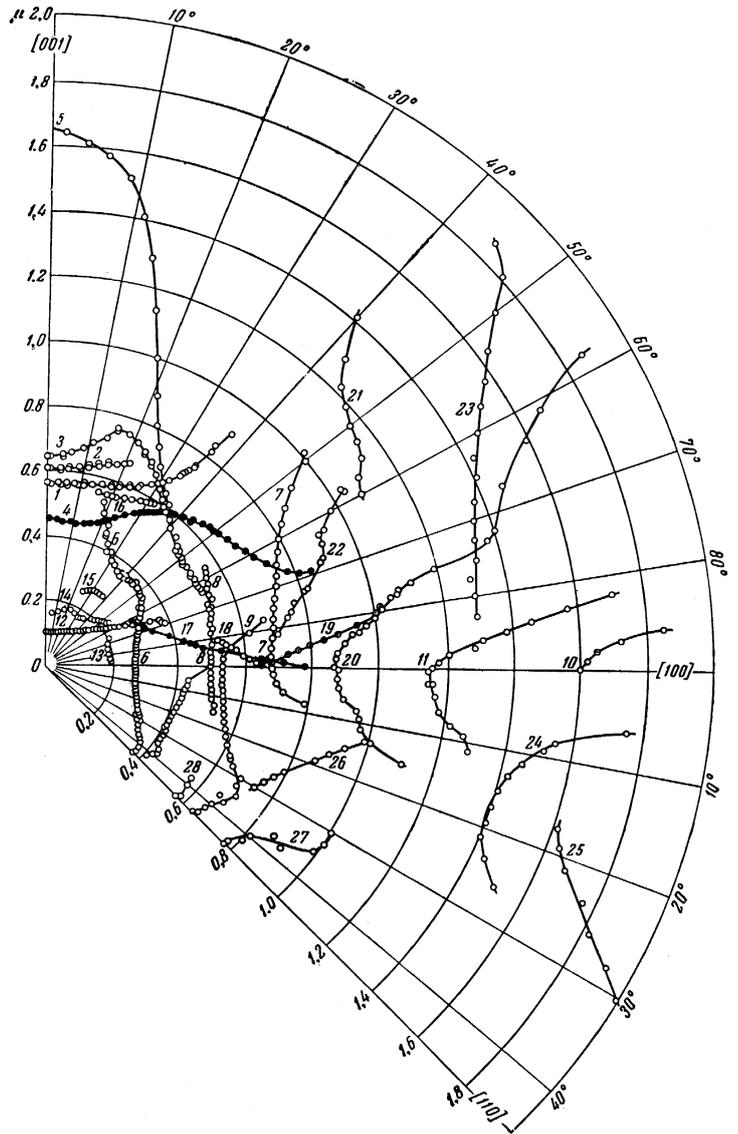
The investigations of cyclotron resonance were carried out on single crystals of tin with plane surfaces coinciding with (010) and (001). Spectra, similar to those shown in Fig. 2 were recorded for different directions of the high-frequency

currents and of the magnetic field in these planes for a specimen temperature of $\sim 2^\circ \text{K}$. The field was rotated through $1-3^\circ$ from spectrum to spectrum. The angles of rotation of the field and of the specimen from the original position were measured with an accuracy of $\sim 20'$. The magnetic field strength was measured with a Hall e.m.f. probe, calibrated by nuclear resonance. The values of effective mass μ of the electrons were calculated by Eq. (1). The accuracy of measuring the absolute values of μ for about ten groups of deep and sharp resonances was $\sim 1\%$; it decreased to $3-5\%$ for less clear resonance. The relative changes in μ could be measured with an accuracy several times greater.

The measurements of the effective masses of conduction electrons in tin (including holes, which are not differentiated from electrons in the present experiments) are shown in a polar diagram (Fig. 4). On the diagram are given masses whose existence was determined in some interval of directions on a number of spectra. The masses corresponding to the deepest resonances are shown by the numbers up to 10. A few shallow cyclotron resonances ($\sim 1\%$ of the overall number), which were not established as belonging to the measured groups of resonances with strongly marked angular dependence, were not used in the calculations. This could lead to the loss of small masses ($\mu < 0.1$), the detection of which is made difficult by the presence of high order resonances from heavier masses.*

*This experimental difficulty can be removed by using thinner specimens in which the large electronic orbits are cut off.^[12]

FIG. 4. Results of the investigation of the anisotropy of effective masses of electrons in tin in the (010) plane – the upper part of the diagram, and in the (001) plane – lower part of the diagram (this plane was studied in less detail). The masses numbered from 1 to 10 correspond to the deepest resonances.



No. of mass in Fig. 4	Conditions for observing resonance*	μ along [001] or [100] axis			Orbit in the nomenclature of Fig. 5, from [14]**
		present work	measurement***	calculation[15]	
1	H ⊥ J	0.56	0.70	0.38	4a ζ
2	H ⊥ J				3β
3	H ⊥ J; H ∥ J, φ ≥ 20°				
4	H ⊥ J; H ∥ J, φ ≥ 25°				
5	H ∥ J				
6	H ⊥ J; H ∥ J, 80° ≥ φ ≥ 20°	0.27	0.45	0.32	4b ξ (?)
7	H ⊥ J	0.67	1.27	0.42	4a η
8	H ⊥ J				3β
9	H ⊥ J	0.53		~0.2	3ε
10.11	H ∥ J				
12	H ⊥ J	0.10	0.09	0.061	3δ
13	H ∥ J				
14	H ⊥ J	0.16	0.15	0.089	5μ
15.16	H ⊥ J				
17-19	H ∥ J				
20-28	H ⊥ J				

*φ is the angle between H and the [001] direction.
 **The figure denotes the zone number; 4a is the hole zone of Fig. 5, 4b the electron zone.[14]
 ***Measured by the de Haas – van Alphen effect.

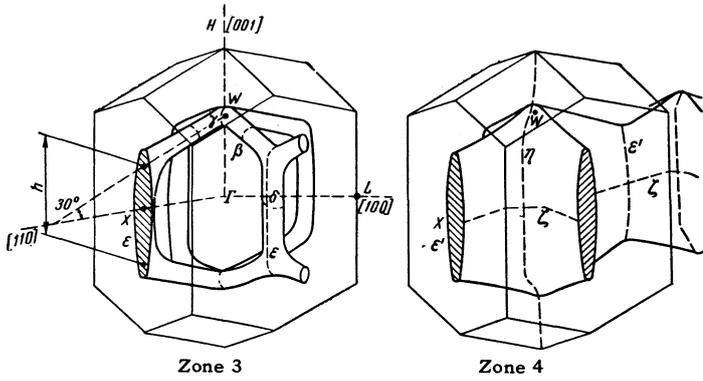


FIG. 5. The hole surfaces of the third and fourth zones of the Fermi surface of tin.^[14] Dimensions in units of $2\pi/a$: segment $\Gamma L = 1$; $\Gamma X = 1/\sqrt{2}$; $\Gamma W = 0.65$; $\Gamma H = 1.19$.

Some characteristics of the behavior of the cyclotron resonances, and of the corresponding effective masses shown in Fig. 4, are listed in the table, together with the conditions of their observation. The resonances observed for $\mathbf{H} \perp \mathbf{J}$ occur at the extremal sections of the Fermi surface, and for $\mathbf{H} \parallel \mathbf{J}$ at elliptical points of support.^[1] There are cases of the observation of resonances for both these polarizations (and also for intermediate cases); these resonances must belong to a non-central Fermi surface.

DISCUSSION

The Fermi surface of tin is evidently very complicated; some of its topological features were explained by Alekseevskii and Gaïdukov,^[13] while a fuller investigation was undertaken by Gold and Priestley^[14] using the de Haas-van Alphen effect. Separate details of the structure of the Fermi surface, proposed by Gold and Priestley on the basis of the free electron model are confirmed by a number of experiments.^[12-14] Judging from their angular dependences and magnitudes (Fig. 4), the results of the present work make it possible to relate some of the effective masses found to certain features of the Fermi surface,^[14] the corresponding comparisons are shown in the table.

The open hole surface of the fourth zone and the multi-connected hole surface of the third zone, with which the comparison of the data obtained is the fullest, are shown in Fig. 5. We shall calculate some dimensions of the third zone. From the range of directions of the magnetic field ($\pm 70^\circ$ from the [001] axis) within which resonance 12 on orbit δ is observed, we can calculate the ratio of the diameter d of tube δ to its height h :*

$$d/h = \operatorname{tg} 20^\circ = 0.36.$$

The height h can be calculated from the data of Fig. 5 and is $h = 0.48(2\pi/a)$ where $a = 5.796 \text{ \AA}$ at 4° K ; from this $d = 0.17(2\pi/a)$. From the range

* $\operatorname{tg} = \tan$.

$\pm 12^\circ$ from the [100] axis within which resonance 9 is visible on orbit ϵ , the diameter Δ of tube β can be evaluated:

$$d/(h + 2\Delta) = \operatorname{tg} 12^\circ = 0.21,$$

whence

$$\Delta = 0.17(2\pi/a) = d.$$

We find the area of cross section of tube δ (assuming it to be a circular cylinder) to be $0.23(2\pi/a)^2$, which is somewhat larger than the value $0.014(2\pi/a)^2$ calculated from the period of the A-oscillations measured by Gold and Priestley.^[14] Oscillations corresponding to the β orbit were not observed by Gold and Priestley,^[14] but the cited calculation of their period by Harrison^[15] gives a value close to the area of the δ orbit, in agreement with the equality obtained above, $\Delta = d$.

The area of the ϵ orbit can be calculated from the dimensions of the zone:

$$(h + 2\Delta)d = 0.14(2\pi/a)^2,$$

while the O-oscillations^[14] give $0.10(2\pi/a)^2$. These differences, which somewhat exceed the possible error, can be explained by tubes δ and β not being cylindrical but narrowed in the middle.

The dimensions found for the third zone lead us to expect that cyclotron resonance will exist for sections of tubes β in the range of rotation of the field by $\sim 20^\circ$ from the [001] axis and by $\sim 35^\circ$ from the [100] axis in the (010) plane, and the effective masses should be close in magnitude. Curves 2 and 8 of Fig. 4 correspond to these conditions. On rotating the field in the (001) plane, resonance 8 in tubes β gives two branches: one disappears for an angle of $\sim 20^\circ$, while the other remains up to the [110] direction, the effective mass decreasing. This also agrees with the configuration of the Fermi surface of the third zone (Fig. 5).

A further comparison of the experimental data of Fig. 4 with the model of the Fermi surface proposed by Gold and Priestley^[14] is, of course, still

possible, but the difficulties increase and the reliability of the comparison is lost. The trouble is evidently by no means only in the incompleteness of the experimental data, but also in the imperfection of the model. Thus, we should point out one sharp disagreement: cyclotron resonances which should be observed continuously over the whole (010) plane are absent in Fig. 4, although such a resonance (with a depth that is not small) should occur at the central extremal section of the closed electron surface of the fourth zone, according to Gold and Priestley's model. It is possible that this surface should be increased in the $\langle 110 \rangle$ directions due to a small deformation, until it reaches the borders of the principal zone, as discussed in [14]. The surface would then become open in these directions, analogously to the hole surface of the fourth zone (Fig. 5) and resonance 6 of Fig. 4 can supposedly be ascribed to it.

We should point out that the heavy masses, observed in small ranges of directions of the field, probably correspond to complicated orbits encompassing several cells of the open surface of the fourth zone.

CONCLUSIONS

The present investigation shows that the study of cyclotron resonance in conduction electrons of a metal is becoming one of the most productive methods of studying the Fermi surface. Information about the structure of the Fermi surface, obtained by this method, is extremely broad and detailed, and although it does not enable the Fermi surface to be constructed directly, it gives ample material for control and correcting of models. Incidentally, use of the effect of cutting-off large electron orbits, [12] observed in cyclotron resonance, makes it possible to construct directly separate sections of the Fermi surface.

The measurement of effective masses of electrons by the method of cyclotron resonance has, evidently, no equal in sensitivity and accuracy. In this connection it is worth noting that the measurement of effective masses by any worse method giving some mean or even tentative value of the masses, is senseless because the orders of magnitude are known beforehand, while the whole value of the measurements, as the experiments of the present work show, lies just in the detailed information about the anisotropy of the effective masses. This consideration also refers fully to measurements of cyclotron resonance by various methods of low sensitivity or with poor resolution.

At present the best method for investigating cyclotron resonance is the method of frequency modulation used in the present work [10] in conjunction with the use of strip resonators and highly perfect specimens. The main reason for the difficulty in using experimental results of the present work for establishing the Fermi surface of tin is its great complexity. It is natural to expect that this problem will be solved considerably more easily in the case of metals with a simpler structure. This is borne out by the investigation of cyclotron resonance in lead.

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