CYCLOTRON RESONANCE IN TUNGSTEN

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Cyclotron resonance is studied in a tungsten single crystal at a frequency of 9.38 GHz and a magnetic field lying in the (110) and (001) planes. A number of resonances with effective masses of (2.2–0.24) m_0 are detected. Some of the orbits can be ascribed to orbits originating on the electron "jack" of the Fermi surface model for tungsten[1-3]. The mean velocity of electrons moving along an "octahedron" of the electron "jack" is determined on basis of the mass on the central section of the surface and is found to be $\overline{v} = (0.71 \pm 0.04) \times 10^8$ cm/sec.

THE experimental investigation of the electronic properties of transition metals such as tungsten is of undisputed interest, stimulated by the progress attained in the theoretical calculation of their energy spectra[1-3]. According to the calculation, the Fermi surface of tungsten consists of an electronic "jack," which has a shape close to that of a regular octahedron whose vertices are topped by spherelike formations, and a set of equivalent hole octahedra. In addition to these main surfaces, there can also exist a number of additional electronic and hole surfaces of small volume. However, the theory is still unable to express any definite opinions concerning the latter.

The real Fermi surface of tungsten is apparently close to the described model. According to the results of a study of the radio-frequency size effect[4], the extremal dimensions of the main parts of the surfaces are close to theoretical, and their anisotropy agrees with the model. Comparison of the anisotropy and of the value of certain extremal sections of the electron and hole surfaces, obtained on the basis of a study of the de Haas–van Alphen effect (the experiments, unfortunately, were not published) with the predictions of the theory, carried out by Louks[3], also offer evidence in favor of the latter. Brandt and Rayne[5] investigated only small sections, which give large periods of the de Haas–van Alphen effect, one of which can be connected with the "neck" of the electronic "jack."

Cyclotron resonance of the Azbel'-Kaner type was investigated in tungsten by Fawcett and Walsh[8] in the 8 mm range. They found a set of effective masses, but the poor resolution of the resonances and the notions concerning the spectrum, which were still unclear at that time, greatly hindered the interpretation of the results and their comparison with the Fermi-surface model. Walsh[8] determined with the aid of a very elegant experiment—cyclotron resonance on "jumping" orbits in a normal field the sign of the carriers, which in the case of a field parallel to the [111] axis had an effective mass $0.84m_0$ and $0.063m_0$. Both turned out to be electrons.

In this paper we present an investigation of cyclotron resonance on tungsten in the 3 cm band. Owing to the apparently better quality of the samples, it became possible to observe a number of new hitherto unobserved masses and to compare certain masses with different orbits on the Fermi surface. Unfortunately, the difficulty connected with insufficient resolution of the resonances were not completely eliminated.

EXPERIMENT

The measurements were made by the frequency-modulation method[9] at a frequency 9.38 GHz. We investigated single-crystal samples of tungsten in the form of strips measuring 11.4 × 4 × 1 mm; the orientation of the crystallographic axes in the samples is indicated in Fig. 1. The samples were made of tungsten purified by zone melting, with a resistance ratio $\rho(293°K)/\rho(4.2°K) \approx 55\,000$.[1)] The initial crystal was in the shape of a cylindrical rod of 5 mm diameter with the [1$\bar{1}$0] axis along the cylinder axis; samples of the required shape were obtained by electrolytic polishing on a flat-polishing machine, without using any mechanical working. (Preliminary experiments have shown that no cyclotron resonance is observed in samples prepared by mechanical working.) Since the prepared samples had rounded corners, the sample was placed in a screen of annealed copper foil ~50 μ thick, as shown in Fig. 1c, to improve the resolution of the cyclotron resonance. Only the central section on one side of the sample ~3 mm wide and equal to the entire length of the sample was left open. The change in the

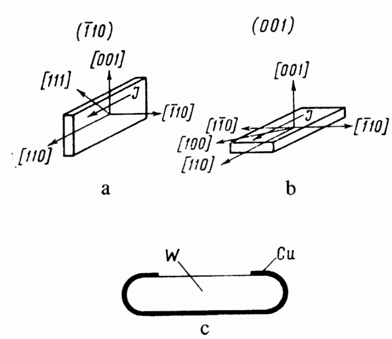

FIG. 1. Orientation of the crystal axis (a, b) and position of sample in the screen (c).

[1)]The initial crystals of the tungsten were graciously furnished by the Institut für Metallphysik und Reinstoffe der Deutschen Akademie der Wissenschaften, Dresden.

direction of the normal to the surface of the sample on its open part, measured with the aid of a double microscope, was ~10'. Because of this, the screen could not eliminate completely the possibility of appearance of effects connected with the inclination of the magnetic field to the surface of the sample through a small angle [10,11]. Nonetheless, the spectra obtained with the screen were much simpler than without it.

During the time of the experiments the sample served as the resonant element of a strip resonator [9]. The currents in the sample were linearly polarized and directed alongside of the crystal, that is, in both cases in the $[1\bar{1}0]$ direction.

An electromagnet was used to produce the magnetic field. The field intensity was measured with a Hall pickup calibrated during the time of the experiment against a nuclear magnotometer with running water [12]. The magnetic field could be rotated in the plane of the sample through any angle, and could also be inclined $\pm 2^\circ$ to this plane. The parallelism between the surface and the field was determined from the cyclotron resonance effect itself, and the accuracy of the setup was ~10', which agreed with the estimate of the non-planarity of the investigated section of the crystal.

The sample temperature was ~1.5°K during the time of the experiment.

RESULTS OF EXPERIMENTS

Figure 2 shows a sample recording of the cyclotron-resonance spectrum, on which one sees clearly three series of resonances, corresponding to the masses I, O, and P. The absolute value of the effective mass was determined from such spectra by means of the formula

$$\mu = \frac{m^*}{m_0} = \frac{e}{m_0 c \omega \Delta H^{-1}},$$

where ΔH^{-1} are the periods of the cyclotron resonances. The accuracy with which μ is determined was limited essentially to the line width of the cyclotron resonance and amounted to ~3%. At those field directions where there were two masses of close value (or masses that were practically integral multiples of each other), the error was larger, owing to the insufficiently good resolution.

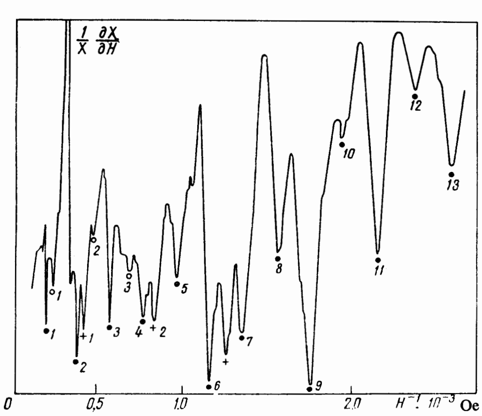

FIG. 2. Plot of the cyclotron-resonance spectrum at H ∥ $[1\bar{1}0]$ for a sample whose surface coincides with the (001) plane: ●—resonance I, ○—resonance O, +—resonance P.

The anisotropy of the investigated masses in the planes (110) and (001) is shown in the diagrams of Fig. 3 and 4. The position of the crystallographic axes was determined from the symmetry of the picture with accuracy ~1°. The experimental points in both planes were plotted in the angle range ~180° and then, in view of the symmetry of the crystal, referred to a single quadrant; as expected, the results of the measurements in two symmetrical quadrants coincided within the accuracy of the experiment.

1. The strongest resonance, which is seen in the (110) plane, is the resonance C with effective mass $\mu_C = 0.83$. According to Walsh [8], this resonance is connected with electrons moving along a trajectory on which the velocity component along the field changes direction six times, and the average drift velocity is equal to zero. Such properties can be possessed by the central orbit passing through the "octahedron" of the electronic "jack."

According to Louks [3], the orbit C exists in the region of magnetic-field directions, determined by the inequalities $\sphericalangle$ H, $[1\bar{1}0] \gtrsim 15^\circ$ and $\sphericalangle$ H, $[001] \lesssim 20^\circ$; practically the same values of the boundaries of this region are obtained by experiment, namely $15.5^\circ \pm 1^\circ$ and $17^\circ \pm 1^\circ$, respectively.

Favoring the conclusion that the resonance C is realized on the central part of the electronic surface in the form of a regular octahedron is also the relatively large amplitude of the resonance. The reason lies in the fact that, as can be readily seen, when **H** ∥ $[1\bar{1}1]$ the effective mass for the octahedron does not depend on p_H (under the very likely assumption that the velocity on the boundary of the octahedron is independent of the momentum). It is therefore necessary to expect that the extremum of the mass with respect to p_H will be very broad and that a large number of electrons will take part in the resonance C.

Since the form of the orbit corresponding to the resonance C is known, we can calculate the average velocity of the electrons v_C on the central part of the "jack." From geometrical considerations, the velocity directed along the normal to the Fermi surface makes an angle 19° 28' with the plane of the orbit in momentum space. Therefore

$$\bar{v}_C = \frac{\bar{v}_\perp}{\cos 19°28'}, \qquad \bar{v}_\perp = \frac{1}{2\pi}\frac{P}{\mu m_0},$$

where P is the perimeter of the orbit in momentum space; for the octahedron it can be readily calculated from the value $k_{111} = 0.415$ Å^{-1} [4]. As a result of the calculation we obtain $\bar{v}_C = (0.71 \pm 0.04) \times 10^8$ cm/sec.

2. At directions $\sphericalangle$ **H**, $[110] \approx 0-19^\circ$ there exists in the (110) plane a strong resonance I, comparable in amplitude to the resonance C (in that region where they are observed simultaneously). When **H** ∥ $[1\bar{1}0]$ we have $\mu_I = 1.50 \pm 0.05$. In the plane (001), the resonance I is observed at $\sphericalangle$ **H**, $[110] \approx 0-5^\circ$. A characteristic feature of the resonance I is that it is observed when **H** ∥ **J** in the (110) plane and for both polarizations in the (001) plane. This means that when the electron moves in the skin layer it has a sufficiently large velocity component along the magnetic field. The resonance I has a moderate sensitivity to the angle between the field and the sample surface; its amplitude decreases noticeably at an inclination ~10'.

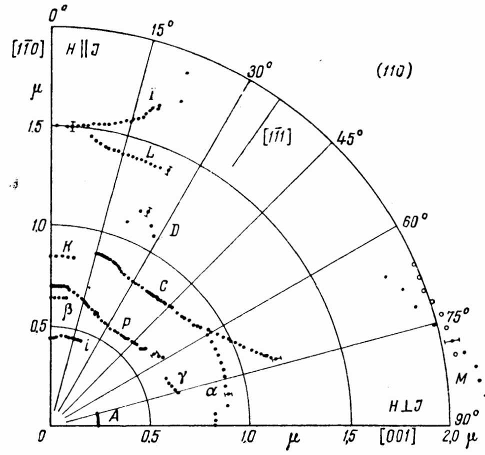

FIG. 3. Anisotropy of the effective masses of the carriers in tungsten in the (110) plane. The bars at certain points indicate the absolute measurement error.

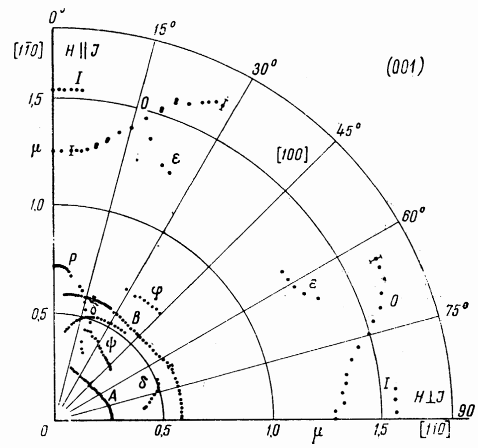

FIG. 4. Anisotropy of the effective carrier mass in tungsten in the (001) plane. The bars at certain points indicate the absolute measurement error.

According to the Fermi-surface model (Fig. 5), there should exist at H ∥ {110} three orbits with extremum values of the mass G, K, and I, each of which lies in an interval of variation of the momentum p_H bounded by the self-intersecting orbit; the mass should tend to infinity on approaching these orbits. Of all the three orbits, only the electrons moving on the orbit I can produce a resonance having the indicated properties, for the following reasons:

First, the orbit I is non-central; the velocity of the electrons on it is directed at an angle to the plane of the orbit. Since v_H is of alternating sign, the average velocity $\bar{v}_H$ along the field is apparently small, in accord with the relatively weak dependence on the inclination of the field.

Second, the region of existence of the resonance I does not contradict the Fermi-surface model according to which the orbit I in the (110) plane exists when ∢ **H**, [1$\bar{1}$0] $\lesssim$ 35° and in the (001) plane when ∢ **H**, [1$\bar{1}$0] $\lesssim$ 10°.

The orbits G and K cannot give this resonance for the following reasons: The orbit G is central, and the resonance of the orbit K in the (001) plane cannot be seen if **H** ∥ **J** ∥ [1$\bar{1}$0], since the orbit enters in the skin layer "at angle" at which the velocity is perpendicular to the field. Furthermore, according to the estimate, the mass μ_K is of the same order as μ_C at **H** ∥ [1$\bar{1}$1]. The indicated features of the orbit K correspond to the mass denoted by the same letter in Fig. 3.

The orbit I has the form of a "dumbbell." Knowing the mass μ_I and the velocity v_C, and assuming that: a) the "octahedron" is not strongly distorted near the edge, b) the mass of the electron moving over the sphere does not depend on the momentum p_H, and c) when the electron moves over the "sphere" it describes practically a full circle, we can estimate μ_B, where the index B denotes the effective mass of the electron moving over the "sphere." From the relation

$$\mu_I \approx 2\mu_B + \frac{1}{2\pi m_0} \frac{P}{\bar{v}_C \cos 54°45'},$$

where P is the length of the path over the "octahedron," we obtain $\mu_B \approx 0.5$.

3. On the (110) plane, the electrons moving along the orbit I enter the skin layer twice during each cycle, which can lead to the appearance of a broad singularity (compared with the peaks of the cyclotron resonance) in the dependence of the impedance on the field, arising when the period of the microwave field is equal to the time of motion of the electron between the points of its entrance into the skin layer (the mechanism of this phenomenon is similar to the mechanism causing the oscillations of the impedance in weak fields[13].) An estimate similar to that given above yields for the field at which the singularity should be observed a value corresponding to the resonant field for electrons of mass ~$0.5m_0$. It is possible that the described phenomenon explains the existence of the resonance i, which is observed in practically the same field-direction interval as the resonance I. The line width of the resonance i is larger by 2—3 times than the width of the other resonances, and $\mu_i = 0.45$.

4. When the field turns in the (110) plane from the [1$\bar{1}$0] direction, there appears on the "jack" still one more orbit—L (see Fig. 5), lying between two self-intersecting orbits. Judging from the interval of the directions and the value of the mass μ_L, the resonance L pertains to this orbit (Fig. 3).

In the (110) plane at ∢ **H**, [001] ≈ 0–24°, there is observed the resonance M with mass $\mu_M = 2.2$ at **H** ∥ [001]. What is striking is the possibility of attributing the resonance M to the central orbit passing through the four "spheres" and the "octahedron" (the E orbit in[3]). Unfortunately, however, this explanation is refuted by the fact that there exists a region in which the resonances M and C are observed simultaneously, and this should not happen, as can be readily seen from the model (Fig. 5).

The origin of the resonance O, which is observed in the (001) plane, is likewise not clear. Taking into account the interval of the directions and the mass, it might be attributed to the orbit G passing through two "spheres" and the "octahedron." This orbit is central and the velocity along it lies always in the plane of the orbit. In contradistinction, the resonance O is observed both for **H** ⊥ **J** and **H** ⊥ **J**.

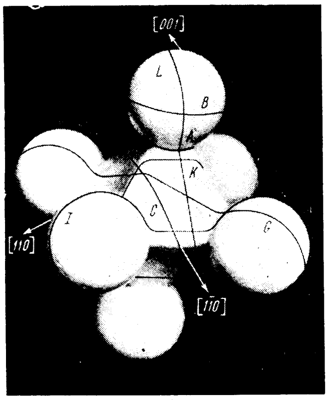

FIG. 5. Model of the electronic "jack." The indicated orbits correspond to the resonances with the same indices on the diagrams as in Fig. 3 and 4.

The resonance D is possibly connected with the orbit passing through three "spheres" and the "octahedron," but information concerning this resonance are extremely skimpy, since the broad paramagnetic-resonance peak does not make it possible to trace its anisotropy.

5. An analysis of the resonances having masses smaller than μ_C is difficult. On the one hand, the experimental picture is very complicated, owing to the superposition of resonances from the carriers with nearly-equal masses and higher orders of resonances of large masses; on the other hand, there are not enough characteristic features which make it possible to use the anisotropy to identify the resonances with definite sections of the Fermi surface.

The resonance P observed in the (110) plane at $\mathbf{H} \parallel [1\bar{1}1]$ has the same mass as one of those observed by Walsh[8] in a normal field. It was established in[8] that this resonance is connected with electrons moving over a central orbit with threefold symmetry. In the same investigation, however, in a tangential field, several poorly resolved resonances with merely equal masses were observed, whereas we observed one broad peak. We cannot state therefore that the resonance P is an electron resonance.

The anisotropy of the masses of P and C is very similar, with the exception of the resonance P is observed up to the $[1\bar{1}0]$ axis. It is therefore possible that the orbit P belongs to the hole octahedron.

In the (001) plane there is observed a resonance B with almost isotropic mass $\mu \approx 0.52-0.55$. This resonance can be seen when $\mathbf{H} \perp \mathbf{J}$ but not when $\mathbf{H} \parallel \mathbf{J}$. The value of the mass μ_B agrees with the mass on the "sphere" of the electronic "jack," an estimate of which was obtained by using the value of the mass μ_I. The foregoing circumstances make it possible to ascribe the resonance B to that part of the surface, but other possibilities of its interpretation may also arise.

The resonance A has the smallest mass of all the observed resonances. It is connected, according to the conditions of observation, with an orbit for which the velocity lies in its plane. The mass $\mu_A \approx 0.23$ (at $\mathbf{H} \parallel [001]$) is close to that expected for a resonance on the "neck" of the electronic "jack," calculated under the assumption that the velocity on the "neck" equals the velocity on the "octahedron." However, two circumstances cast doubts on such an interpretation: the excessively slow growth of the mass when the field is rotated away from the [100] axis and even a slowing down of the rate of the growth when the field approaches the $[1\bar{1}0]$axis, and the absence of a resonance when $\mathbf{H} \parallel [1\bar{1}0]$. From an examination of the model we can conclude that when $\mathbf{H} \parallel \{110\}$ the orbit on the "neck" should acquire a velocity component parallel to $\mathbf{H}$.

In addition to the already described resonances, we observed a large number of resonances (α, β, γ, δ, ϵ, φ, ψ) which cannot be interpreted in any manner at the present. It is not excluded that some of the resonances are connected with extraneous effects, such as the inclination of the field to a part of the sample surface which is not fully plane, or to errors in the analysis of the experimental data, due to insufficient resolution. Both sources of errors can be eliminated by using better quality samples.

CONCLUSION

The investigation of the cyclotron resonance has shown that tungsten has a very complicated Fermi surface, on which a large number of extremal values of the effective mass exist for any direction of the magnetic field. On the electronic "jack" surface obtained as a result of calculations of the energy spectrum[1-3], there should exist a whole series of extremal orbits, some of which can be set in correspondence with the experimentally observed resonances (C, L, I, K, and possibly also B, A, and D). Unfortunately, there are no published effective-mass calculations with which to compare the experimental results. It is not excluded that on the main electronic surface of the "jack" there can exist additional mass extrema which cannot be observed on the basis of intuitive general considerations only. In any case, it seems to us that the masses M and O also pertain to the surface of the "jack," but it is impossible to indicate for them suitable trajectories for the time being. At the same time, some resonances, which should be observed on the surface, were not observed in the experiments.

A large number of resonances with effective mass $\mu \sim 0.7-0.3$ points to the existence, besides the main parts of the Fermi surface, also of additional parts with smaller volume. However, the insufficient resolution of the resonance lines does not make it possible to draw any definite conclusions.

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