THE DE HAAS–VAN ALPHEN EFFECT IN ZINC IN PULSED MAGNETIC FIELDS

V. A. VENTTSEL'

Submitted May 15, 1968

Zh. Eksp. Teor. Fiz. 55, 1191–1196 (October, 1968)

The experimentally measured dependence of the amplitudes of oscillations, connected with "butterfly" and "cigar" cross sections, on the direction and magnitude of a magnetic field and on the temperature is compared with the theoretical predictions. Current theoretical concepts of magnetic breakdown do not explain the angular dependence of the oscillation dependence in the (11$\bar{2}$0) plane. A qualitative explanation of the mechanism of magnetic breakdown is given for trajectories passing near the edge of the Brillouin zone.

INTRODUCTION

THE Fermi surface of hexagonal metals of the second group is sensitive to weak spin-orbit interaction. The presence of two atoms in the elementary cell leads to the result that the hexagonal boundary between zones I and II, and also between zones III and IV is not a Bragg reflection plane for electrons, and consequently the Fermi surface is located inside the doubled I-II and III-IV Brillouin zones.[1] Spin-orbit interaction removes the degeneracy on the hexagonal boundaries and the Fermi surface is seen to be included in four identical zones.[2] In a strong magnetic field a transition is possible from a single zone model of the Fermi surface and a double zone model, as the result of magnetic breakdown of the spin-orbital gap. In zinc, the value of the spin-orbital gap is small[2] ($\lesssim$ 0.03 eV), and the magnetic breakdown between the parts of the electron surfaces in zones II and IV was observed earlier.[3,4]

These electron surfaces in the model of nearly free electrons are located around the middle of the "horizontal" edge of the six-sided prism—the Brillouin zone (the hexagonal axis is directed along the "vertical"). The "butterfly," which consists of two intersecting discs of elliptical shape, is found in zone III. The axes of rotation of these discs are perpendicular to the "horizontal" edge of the Brillouin zone and form an angle of 65° with the hexagonal axis. The "cigar," which is extended along the edge and placed inside the "butterfly" in a combination of the zones, is located in zone IV. In the absence of spin-orbit interaction, halves of the "butterfly" and the "cigar" form a folding "cavity", which is not symmetric relative to the (0001) plane which coincides with the six-angle boundary of the Brillouin zone. The lateral boundaries of a prism are formed by planes of the (10$\bar{1}$0) type (six planes obtained by permutation of the first three indices). Planes of the (11$\bar{2}$0) type are perpendicular to the "horizontal" edges of the prism.

It is of interest to compare the amplitude oscillations observed experimentally with the theoretical predictions.

EXPERIMENTAL PROCEDURE

The method of measurements of the de Haas–van Alphen effect in pulsed magnetic fields goes back to the researches of Shoenberg[5] and is described in[4]. Resonance "bursts," which correspond to different extremal cross sections of the Fermi surface, are observed for different values of H and dH/dt, and this leads to errors in the determination of their amplitude. Actually, the emf developed in the measuring circuit consists of the sum of a high frequency signal associated with the oscillating dependence dM/dH, and of a low-frequency signal (~15 Hz), which is proportional to dH/dt, and which arises from the incomplete compensation of the turns of the measuring coil. The low-frequency signal has a large amplitude (up to 10 V) and can shift the operating points of the input circuit, which would lead to an unequal amplification factor for different values of dH/dt. To avoid this effect, the active resistance in the series circuit of the preamplifier was replaced by a choke. Since a resonance method was used in the experiment for the measurement of the de Haas–van Alphen effect, a capacitor was connected in parallel with the choke. The preamplifier had selective properties at the same frequency as the recording coil (33 kHz). In supplying the input of the preamplifier with a mixture of signals of high frequency ($u_1 \sim 10^{-3}$ V) and low frequency (50 Hz), the amplitude of the high-frequency signal did not depend on the value of the low-frequency up to $u_2 \approx 25$ V. The independence of the amplification factor of the signal, which is proportional to dM/dH, of the value of dH/dt is important in the comparison of the oscillation amplitudes associated with the different extremal cross sections, and also in the measurement of the Dingle factor from the dependence on the value of the magnetic field.

EXPERIMENTAL RESULTS

The data given earlier[4] on the angular dependence of the oscillation amplitudes associated with the "butterfly" and "cigar" cross sections in the (11$\bar{2}$0) plane were made more accurate, and in the range of angles $45° < \varphi < 70°$ it was possible to measure the amplitude of much weaker oscillations against the background of strong oscillations associated with the cross section of large γ. These weak oscillations are associated with the "cigar" cross section. Thus the total picture of amplitude oscillations associated with the "butterfly" and "cigar" cross sections is shown in Fig. 1 (solid line). The scatter of amplitudes for different samples is of the order of 10 relative units, the noise level is also of about 10 units. Both the

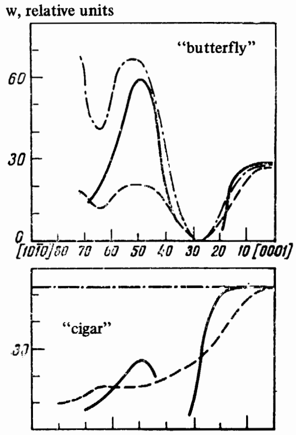

FIG. 1. Oscillation amplitude w in relative units for frequencies corresponding to the "butterfly" and "cigar" cross sections in the (11$\bar{2}$0) plane. The solid curve represents the experimental results; the dash-dot, the calculation from the formulas of I. Lifshitz and Kosevich; [6] the dotted curve, the amplitude decreased by the factor $(1-W)^2$, where W is the probability of breakdown of the spin-orbital gap in the (0001) plane.

"butterfly" and the "cigar" were observed only in the two angle ranges $0 < \varphi < 20°$ and $35° < \varphi < 65°$.

It is of interest to compare the obtained angular dependence of the amplitude w with that calculated from the formula of I. Lifshitz and Kosevich.[6] For this purpose, it was necessary to determine the value of the effective mass for these trajectories from the w(T) dependence. For the "butterfly" in the angular range $50° < \varphi < 70°$, the values of m/m_0 were given in[4]. These are given in the table, together with new data.

Results of measurements of the effective mass m/m_0 and the Dingle factor x in the (11$\bar{2}$0) plane for the "butterfly" and the "cigar." (Accuracy of measurement of $m/m_0 \sim 10\%$, of x, ~20%).

φ°	"butterfly"		"cigar"	
	m/m_0	x	m/m_0	x
7	0.46	2,2	0,37	2.3
20	0,49	1,2	0.37	2,1
40	0,57	—		
50	0.70	0.7		
57,5	0,79	1.2		
65	0.90	1.9		
70	0,77	1,5		

The dependence of the oscillation amplitude on the value of the magnetic field gives the Dingle factor x, which describes the decrease in the amplitude of oscillations due to the scattering of the electrons by impurities and lattice imperfections. It was discovered in[4] that there is an increase in the Dingle factor in regions of possible magnetic breakdown of the spin-orbit gap, and it was suggested that the increase in x can serve as a qualitative characteristic of the presence of magnetic breakdown. The value of the Dingle factor for the "butterfly" and "cigar" is given in the table, together with the already published values of x for the "butterfly" in the range of angles $50° < \varphi < 70°$.

COMPARISON OF THEORY AND EXPERIMENT

The dash-dot line in Fig. 1 shows the amplitude of the oscillations, computed from the formulas of[6] with account of resonance method applied for the measurement of the de Haas–van Alphen effect in pulsed magnetic fields. It was tentatively assumed that $w_{comp} = w_{exp}$ for the "cigar" at $\varphi = 0$. Comparison of the experimental curve with the computed one shows that it is necessary, in the case of the "cigar," to call upon the mechanism of magnetic breakdown for explanation of the two gaps in the curve $w_{exp}(\varphi)$. For the "butterfly," the vanishing of the oscillations for $\varphi > 70°$ is evidently connected only with the magnetic breakdown, while the gap in the region of 30°, in addition to the magnetic breakdown mechanism, has another explanation: close to 25°, the ratio $m/m_0 = 0.5$ and the amplitude of the oscillations should vanish, because of the factor $\cos(\pi m/m_0)$.[6] The large value of the Dingle factor for the "butterfly" at $\varphi \approx 20°$ gives a basis for supposing that both these mechanisms operate simultaneously.

In the case of a single plane of Bragg reflection, the probability of breakdown has the form $W \sim \exp(-\Delta^2/\hbar\omega\epsilon_0 \sin\varphi)$, where φ is the angle between the direction of H and the normal to this plane.[7] The amplitude of the oscillations, associated with the "butterfly" and "cigar" cross sections, would decrease here by a factor of $(1 - W)^2$;[8] if we were dealing with the (0001) plane, the amplitude would have the form pictured in Fig. 1 as the dotted line. The experimentally observed dependence $w(\varphi)$ is also not explained by this model.

As the result of magnetic breakdown between the "butterfly" and the "cigar," a "cavity" cross section should appear between the "butterfly" and the "cigar." This cross section was never observed, however, in the (11$\bar{2}$0) plane.

Thus the experimental data do not agree with the predictions of existing theory of magnetic breakdown in two points: 1) the absence of oscillations in the (11$\bar{2}$0) plane, which could be connected with the "cavity" cross section; 2) the presence of two angular intervals where the oscillations connected with the "butterfly" and "cigar" are observed, and two intervals where these oscillations are not observed.

DISCUSSION OF RESULTS

1. The asymmetry of the "cavity" relative to the (0001) plane leads to the result that for all directions of H in the (11$\bar{2}$0) plane, with the exception of $H \perp [0001]$, the extremal cross section of the "cavity" lies in the plane k_z = const, which does not pass through the "horizontal" edge of the Brillouin zone. The extremal crosssections of the "butterfly" and the "cigar" always lie in the plane $k_z = k_0$, which passes through the edge of the Brillouin zone. The distance between these planes Δk_z can be significant; thus, in the nearly free electron model, for H parallel to the axis of rotation of one of the "butterfly" discs ($\varphi \approx 65°$), Δk_z was estimated as $10^{-2}A^{-1}$ for a thickness of the "cigar" of $\sim 10^{-1}A^{-1}$. The magnetic breakdown decreases the amplitude of the oscillations associated with the "butterfly" and "cigar" but does not lead to the appearance of oscillations corresponding to the "cavity" cross section, since the energy gap $\Delta_{3,4}(k) = |\epsilon_3(k) - \epsilon_4(k)|$ is minimal at $k_z = k_0$ the

"cavity" cross section is not extremal and does not make a contribution to the de Haas—van Alphen effect. Breakdown of the gap $\Delta_{3,4}(\mathbf{k})$ for k_z corresponding to the extremal cross section of the "cavity" begins in zinc at much higher magnetic fields. For magnesium, which has the same model of a Fermi surface and a spin-orbit gap that is an order of magnitude smaller, oscillations were observed corresponding to the "cavity" cross section, at fields of ~60 kOe.[9]

2. The complicated angular dependence of $w(\varphi)$ for the "butterfly" and "cigar" can be connected with the passage of the plane containing the electronic orbit through the edge of the Brillouin zone. The Bragg reflection planes (0001) and (10$\bar{1}$0) intersect at this edge, which is responsible for the division of the surfaces of zones III and IV.

If the plane in which the electronic orbit lies passes through the edge of the Brillouin zone, then it is not possible, without a rigorous theoretical analysis, to predict the dependence $W(\varphi)$. However, the contribution from electrons moving along such orbits is small, since the thickness of the layer of trajectories giving a contribution to the oscillations of the magnetic moment $\delta k_z \sim b\sqrt{\hbar\omega/\epsilon_0}$ (b is the value of the order of the dimensions of the Brillouin zone, ϵ_0 the order of the boundary energy). Therefore, in addition to electrons that intersect the edge of the Brillouin zone, it is necessary to consider also electrons whose orbits lie in planes removed from the edge by a value $\lesssim \delta k_z \sim 10^{-2} A^{-1}$ ($b \sim A^{-1}$, $\hbar\omega \sim 10^{-3}$ eV, $\epsilon_0 \sim 10$ eV). Such orbits intersect the planes (0001) and (10$\bar{1}$0) at some distance from the edge and if the distance between the points of intersection is large, it can be considered independently the probability of magnetic breakdown on each of the planes.

A similar situation arises for a small departure of the magnetic field from the normal to the edge of the Brillouin zone at an angle θ of the order of several degrees, which is always experimentally possible. This departure is not important in the determination of the values of the extremal cross sections, inasmuch as, close to the direction of symmetry,

$$S = S_0 + \frac{1}{2}\frac{d^2S}{d\theta^2}\theta^2,$$

but it also leads to the intersection of the orbits with the planes (0001) and (10$\bar{1}$0) at some distance from the edge of the Brillouin zone.

Figure 2a shows the cross sections of the "butterfly" and "cigar" with the plane $k_0 + \delta k_z$ = const. The "butterfly" and "cigar" are realized both in the removal of the degeneracy at the points 1, 3, 5, 7 and for magnetic breakdown at all these points. We denote the probability of breakdown at the (10$\bar{1}$0) plane by W_1, and at the (0001) plane by W_2. Then the total probability of motion along the "butterfly" and "cigar" orbits is equal to $[W_1W_2 + (1 - W_1)(1 - W_2)]^2$. The trajectories of the "cavity" exist in the breakdown gap only on one of the planes, and the probability of such motion is equal to $[W_1(1 - W_2) + W_2(1 - W_1)]^2$. The course of $W_1(\varphi)$ and $W_2(\varphi)$ can be predicted if the behavior of $\Delta_{3,4}(\mathbf{k}) = |\epsilon_3(\mathbf{k}) - \epsilon_4(\mathbf{k})|$ is known on the (0001) and (10$\bar{1}$0) planes near the edge of the Brillouin zone.

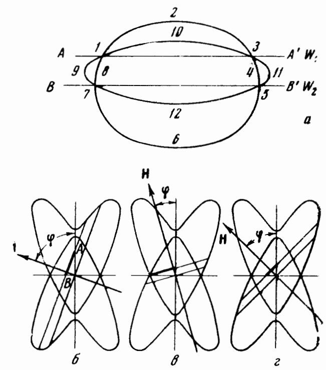

FIG. 2. a—cross section of the "butterfly" and "cigar" in the plane k_z = const, which does not pass through the edge of the Brillouin zone. b, c, d—cross section of "butterfly" and "cigar" in the (1$\bar{1}$20) plane, which passes through the center of the "horizontal" edge of the Brillouin zone. The boldface lines AB show the distance between the points of intersection of the orbit with the planes (10$\bar{1}$0) and (000$\bar{1}$)

An exact expression for $\Delta_{3,4}(\mathbf{k})$ is unknown, but it follows from the symmetry properties of a hexagonal lattice with two atoms on an elementary cell that, without account of the spin-orbit interaction, $\Delta_{3,4} = 0$ on all (0001) planes including the edge of the Brillouin zone. From the continuity of $\Delta_{3,4}(k)$, it can be assumed that on the (10$\bar{1}$0) plane the quantity $\Delta_{3,4} = 0$ on the "horizontal" edge of the Brillouin zone, and increases according to some law as one goes away from the edge, asymptotically approaching $\Delta_{3,4}(\mathbf{k}) = 2|V_{10\bar{1}0}|$ far from the edge. So far as the spin-orbit splitting is concerned, it is known that it is equal to zero in the (0001) plane along lines uniting the center of the hexagon with the centers of the edges, and is maximum at the corners of the hexagon.[2] In Fig. 2, this means that the gap at points 5 and 7 is large as one approaches the points 11 and 9, and tends to zero as the points come close together and to points 6 and 12 (i.e., as they approach the line of the points of degeneracy).

Thus, $W_1 \sim 1$ for $\varphi = 0$ ($\cos\varphi = 1$, while $\Delta_{3,4}$ is of the order of spin-orbital splitting) and decreases to zero as $\varphi \to \pi/2$ both because of the factor $\cos\varphi$ and because of the growth of the gap $\Delta_{3,4}$ (Fig. 2b,c). The probability W_2 should have an inverse dependence on angle: $W_2 \sim 1$ for $\varphi \approx \pi/2$ and decreases with decrease in φ. Such a dependence for $W_1(\varphi)$ and $W_2(\varphi)$ means that at angles close to zero and $\pi/2$, oscillations connected with the "butterfly" and "cigar" cross sections should not be observed. For $\varphi \sim 45°$ (Fig. 2d) the distance between the points of intersection is minimal and there evidently exists some unique gap $\Delta_{3,4} \lesssim 2|V_{10\bar{1}0}|$, the probability of the breakdown of which is small; in this range of angles, motion along the "butterfly" and "cigar" orbits is also more probable.

These qualitative considerations of the mechanism of the magnetic breakdown between "butterfly" and "cigar" in zinc explain the angular dependence of the oscillation amplitudes for these cross sections and make more convincing the assumption of the connection of the increase in the Dingle factor with the presence of magnetic breakdown.

The author is grateful to L. F. Vereshchagin for interest in the work, to A. A. Slutskin, A. I. Likhter and A. P. Kochkin for discussion of the results.

[1]W. A. Harrison, Phys. Rev. 118, 1190 (1960).

[2] M. H. Cohen and L. M. Falicov, Phys. Rev. Lett. **5**, 544 (1960).
[3] V. A. Venttsel', A. I. Likhter and A. V. Rudnev, ZhETF Pis. Red. **4**, 216 (1966) [JETP Lett. **4**, 148 (1966)].
[4] V. A. Venttsel', A. I. Likhter and A. V. Rudnev, Zh. Eksp. Teor. Fiz. **53**, 108 (1967) [Sov. Phys. JETP **26**, 73 (1968)].
[5] D. Shoenberg, Phil. Trans. Roy. Soc. (London) **A255**, 85 (1962).
[6] I. M. Lifshitz and A. M. Kosevich, Zh. Eksp. Teor. Fiz. **29**, 730 (1955) [Sov. Phys. JETP **2**, 636 (1950)].
[7] A. A. Slutskin, Zh. Eksp. Teor. Fiz. **53**, 767 (1967) [Sov. Phys. JETP **26**, 474 (1968)].
[8] A. P. Kochkin, Zh. Eksp. Teor. Fiz. **54**, 603 (1968) [Sov. Phys. JETP **27**, 324 (1968)].
[9] R. W. Stark, Phys. Rev. **162**, 589 (1967).

Translated by R. T. Beyer
133