Effect of pressure on the Fermi surface of zinc

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The effect of hydrostatic pressure up to 100 bar on cross sections of the Fermi surface of zinc is measured for a broad range of orientations. The matrix elements of the pseudopotential and their dependence on pressure are calculated. The experimental technique is described. The nature of the pressure dependence of several extremal cross sections is discussed.

1. INTRODUCTION

The shape and dimensions of the Fermi surface of a metal are determined by the type of crystalline structure and the lattice parameters, i.e., by the shape and size of the Brillouin zone and also by the effective lattice potential acting upon electrons. Under hydrostatic compression that does not lead to a polymorphic transformation the lattice parameters of a metal can be varied gradually, thus permitting an investigation of how the parameters of the electron energy spectrum depend on the lattice parameters.

In the present work, as in[1], measurements of the de Haas—van Alphen (DHVA) effect were employed to study the effect of pressure on the dimensions of particular parts of the Fermi surface of zinc. Then the pressure dependence of the matrix elements of the pseudopotential was calculated within the framework of the local pseudopotential theory. It is interesting to investigate zinc under pressure because of its highly anisotropic compressibility. The Fermi surface of zinc under normal pressure has been thoroughly investigated experimentally[2-5] by measuring the DHVA effect, and has been calculated theoretically by Harrison.[6,7] Figure 1 shows the form of the Fermi surface of zinc in the 1-OPW approximation.

Stark and Falicov[8] calculated the electron segments in the nonlocal pseudopotential approximation and concluded that the Fermi surface has no electron segments around the point L in the third and fourth Brillouin zones ("butterflies" and "cigars"). Rudin and Stark[9] have given a new interpretation of some extremal cross sections. The pressure dependence of the oscillation frequencies enables one, within the framework of the pseudopotential theory, to more definitely relate the observed oscillations to particular segments of the Fermi surface.

The possibility of determining experimentally the pressure dependence of parameters of the electron energy spectrum has been investigated by ourselves[1] and by other authors.[10,11] Gaĭdukov and Itskevich[10] investigated only the smallest cross section of the Fermi surface, a "needle" in the third Brillouin zone. O'Sullivan and Shirber investigated, in addition, the cross sections β and γ of the "monster" and performed calculations within the framework of the local-model pseudopotential. The respective results obtained for the needle are very different. To calculate the pressure dependence of all the pseudopotential matrix elements it is not sufficient to know the pressure dependence of only one or two cross sections of the Fermi surface. In[1] and in the present work we have determined the pressure dependence of all the cross sections of the Fermi surface, including the largest, and are consequently able to compare data obtainedfrom the different cross sections.

2. EXPERIMENTAL TECHNIQUE

To determine the behavior under pressure of large cross sections of the Fermi surface, the experimental work must be performed with a magnetic field that is homogeneous throughout the volume of the sample and the lattice must be rigorously periodic. These conditions necessitate the construction of a pressure vessel that will not distort the external magnetic field and the selection of a pressure-transmitting medium that will provide a purely hydrostatic pressure. Since the experiments were performed by a modulation technique to measure the DHVA effect,[12] with a large amplitude of modulation,[13] we had to renounce the use of massive fixed-pressure bombs. The application to solids at helium temperatures considerably reduced the oscillation amplitude; we therefore decided to confine ourselves to the region of small purely hydrostatic pressures transmitted by liquid helium. For these low pressures we used a thin-walled stainless steel tube (thereby considerably simplifying the construction), instead of a massive beryllium bronze bomb. On the other hand, under low pressures it became necessary to measure very precisely the small pressure-dependent variations of the DHVA oscillation frequencies. The initial pressure-dependence coefficient thus obtained for the areas of the extremal cross sections would necessarily be constant up to pressures commensurable with the elastic moduli of the crystal, at least for large cross sections.

The 1×1–5-mm zinc sample was positioned within one of two compensated coils that were inserted into the pressure vessel. The cylindrical part of the vessel was a thin-walled stainless steel tube with a tight plug at one end. The other end was connected by an obturator to a stainless steel capillary tube that led to a helium tank. Pressure inside the vessel was measured with a standard pointer-type gauge. The same capillary carried leads from the pickup coil to the hot zone. The diameter

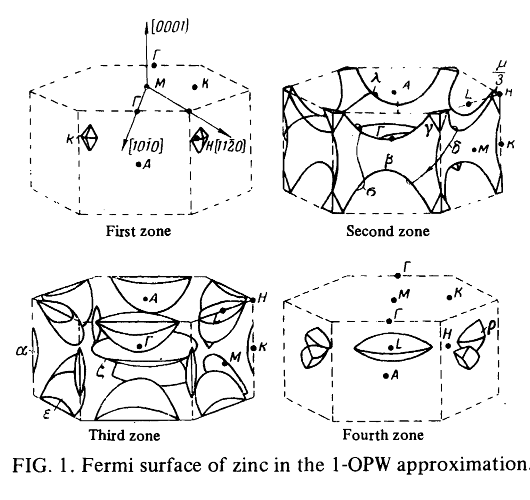

FIG. 1. Fermi surface of zinc in the 1-OPW approximation.

of the working vessel was 4 mm, with a 0.15-mm thick wall. The angular dependence of S and d ln S/dp was measured with a device providing for rotation, which was located inside a pressure vessel that in this case had an outside diameter of 11 mm. The pickup coil was placed inside a plastic drum; the rotation axes of the coil and drum were mutually perpendicular. The drum was rotated through an angle φ by means of the longitudinal displacement of a rod having a hinged connection to the rim of the drum. The hot zone contained both the obturator for passage of the measurement leads from the high-pressure zone and the stuffing-box packing around the rod. The angle of rotation was computed from the displacement of the rod and from the magnitude of the signal induced in a special goniometric coil having its axis perpendicular to the axes of both the drum and the pickup coil. The error in measuring the rotation angle φ was at most 1°.

The high-pressure vessel containing the sample was placed within a superconducting solenoid that produced fields up to 55 kOe, and within a modulation coil that generated fields up to 500 Oe at 467 Hz.

The modification, proposed by Windmiller et al.,[13] of the modulation method for measurement of the DHVA effect consists in using a large amplitude of modulation (comprising several periods of oscillation) and registering the signal in high harmonics, such as the twelfth, of the fundamental frequency. In this case the amplitude of the measured signal is determined not only by spectral parameters,[14] but also by the quantity $J_{12}(2\pi F_i h_0/H^2)$, where J_{12} is the twelfth-order Bessel function, F_i is the frequency of oscillations related to the i-th cross section of the Fermi surface, h_0 is the amplitude of the modulation field, $h = h_0\cos(2\pi ft)$, and H is the field of the superconducting magnet. The modulation amplitude h_0 can be selected to make any frequency F_i dominant in amplitude over the other frequencies. This is especially important when measuring metals with complicated Fermi surfaces having many different extremal cross sections for any magnetic field direction, and also when it is necessary to measure high-frequency oscillations, whose amplitudes are usually much smaller than those of the low-frequency oscillations.

In order to obtain the modulating field amplitude $h_0 \propto H^2$, which was required for the purpose of maintaining a constant argument of the Bessel function when varying the field H, we utilized a Hall pickup (DKhG-05M) that was placed in a magnetic field proportional to the main field H. The measuring current was also proportional to H; therefore an emf proportional to H^2 was induced between the Hall contacts. This emf was used to control the output of the acoustic generator. The signal induced in the test coil was carefully filtered to remove the fundamental modulation frequency, both by coils wound in series opposition and by means of a variometer. All harmonics except the twelfth were eliminated by a preamplifier where the grid circuit of a 6S2P vacuum tube included a high-Q loop ($Q \approx 150$) tuned to the frequency 12f = 5.6 kHz.

The reference voltage for the synchronous detector was developed by a generator of the n-th harmonic, where the sinusoidal voltage of frequency f was converted into pulses that excited the loop tuned to 5.6 kHz. Monochromatization of the produced signal was achieved with a type V6-4 resonant amplifier.

3. EXPERIMENTAL RESULTS

Pressure produced such a small change in the oscillation frequency F, that this change could be observed reliably only through the small phase shift of large numbers of quantum oscillations in the fixed magnetic field H. In a field H = const a pressure-induced change of the oscillation phase by the amount α radian corresponds to a change $\Delta F = \alpha H/2\pi$ in the oscillation frequency. The accuracy of $\Delta F/F$ measurements was determined by the errors in measuring the phase shift α (due to drift of the photoamplifier), the field H, and the frequency F. The accuracy achieved for $\Delta F/F$ was not poorer than 10%.

Identification of the experimentally observed oscillation frequencies with particular parts of the zinc Fermi surface cannot be accomplished unambiguously for all frequencies. Therefore we shall first present results pertaining to reliably interpretable oscillations.

The hole surface in the combined 1st and 2nd Brillouin zones—the so-called monster—has an extremely complicated shape and a large variety of extremal cross sections. The cross section β of the monster is minimal for **H** ∥ [11$\bar{2}$0] and is responsible for oscillations at $F = 4.46 \times 10^5$ Oe. Under pressure this cross section is enlarged with the coefficient $d\ln S/dp = +(42.5 \pm 0.5) \times 10^{-3}$ kbar^{-1}. We have previously[1] made a comparison with data of other authors[10,11] for the principal crystallographic directions. When the field is rotated in the basal plane oscillations corresponding to the cross section σ of the monster are observed, having the frequencies 3.4×10^7 Oe for **H** ∥ [10$\bar{1}$0] and 2.7×10^7 Oe for **H** ∥ [11$\bar{2}$0]. Under pressure this cross section is reduced with the coefficient $-(4.4 \pm 1.4) \times 10^{-3}$ kbar^{-1} for **H** ∥ [10$\bar{1}$0] and $-(5.1 \pm 0.4) \times 10^{-3}$ kbar^{-1} for **H** ∥ [11$\bar{2}$0]. In the (11$\bar{2}$0) plane oscillations with the minimal frequency 2.14×10^7 Oe were observed, corresponding to the cross section δ of the monster. These oscillations were observed at angles from 30° to 70° between the direction of **H** and the [0001] axis, but the pressure dependence of this cross section was reliably measurable only in the narrower interval $40° < \varphi < 60°$. Within the accuracy limits, $d\ln S/dp$ is constant at $\sim(-6 \pm 1) \times 10^{-3}$ kbar^{-1} over the entire angular range.

The electron segments of the zinc Fermi surface that are amenable to unambiguous interpretation are a needle located at the lateral edge of the third Brillouin zone and a lens at the center of this zone. We measured the minimal cross section of the needle for **H** ∥ [0001] and its pressure dependence, for which we obtained $d\ln S/dp = (282 \pm 7) \times 10^{-3}$ kbar^{-1}. According to O'Sullivan and Schirber,[11] $d\ln S/dp = 320 \times 10^{-3}$ kbar^{-1} for $\varphi = 0$, and when **H** deviates from the [0001] axis it decreases more rapidly than would be expected for an ellipsoid (for $\varphi = 20°$ we have $d\ln S/dp = 300 \times 10^{-3}$ kbar^{-1}). The lens is reduced only slightly under pressure; for **H** ⊥ [0001] the decrease is given by $d\ln S/dp = -(3.1 \pm 0.5) \times 10^{-3}$ kbar^{-1}. When **H** deviates 10° from the [0001] direction $d\ln S/dp$ varies within the limits of experimental errors [$d\ln S/dp = -(2.6 \pm 0.5) \times 10^{-3}$ kbar^{-1} for $\varphi = 10°$].

Near the direction **H** ∥ [0001] oscillations were observed at the frequencies $F_L = 0.51 \times 10^7$ Oe, $F_J = 1.1 \times 10^7$ Oe, and $F_K = 1.7 \times 10^7$ Oe, which are usually identified, respectively, with the cross sections of the monster (γ), of the cigar in the fourth Brillouin zone, and of the butterfly in the third zone.[2-4]

Rudin and Stark[9] have suggested another interpretation of these frequencies, because calculations reported in[8] indicate the absence of elements of the zinc Fermi surface that are called a butterfly and a cigar. They suggest[9] that these frequencies result from magnetic breakdown between the monster in the second Brillouin zone and "pockets" in the first zone; this cross section is far from the (0001) plane and only the magnetic-breakdown cross section is extremal, not the initial cross sections of the monster and pockets that comprise it. Figure 2 shows the angular dependence of $d \ln S/dp$ for oscillations designated by L, J, and K as in[4].

The L-series oscillations are of the single-period type for H ∥ [0001] and for **H** forming angles greater than 40° with the [0001] axis; at smaller angles these oscillations have the clearly pronounced character of a curve with beats, in agreement with the data of Joseph and Gordon.[2] For $\varphi = 0$ under pressure the L-series frequency is reduced $[d \ln S/dp = -(18.9 \pm 0.5) \times 10^{-3}$ kbar$^{-1}]$, while for $\varphi > 40°$ it is increased; $d \ln S/dp = +(22.3 \pm 0.5) \times 10^{-3}$ kbar^{-1} is reached for $\varphi = 70°$. In the region where oscillations with beats were observed, the data obtained for the pressure dependence of the frequency F corresponds to the average frequency of the oscillations; in Fig. 2 this region is denoted by a dashed curve.

Oscillations of the J and K series, which also exhibit beats when **H** deviates from [0001], are observed within a range of at most 20°. Under pressure the frequencies of these oscillations also decrease but the angular dependence of $d \ln S/dp$ in this case is similar in character to that for the L oscillations.

4. DISCUSSION OF RESULTS

It is difficult, within the framework of the existing ideas about the shape of the zinc Fermi surface, to account for the observed steep angular dependence of $d \ln S/dp$ for L-series oscillations and for the similar dependence in the cases of J- and K-series oscillations. If the L oscillations are related to the γ cross section of the monster, the sign reversal of S(p) that accompanies $\varphi > 40°$ rotation from the [0001] direction would indicate that when the monster is approximated by a hyperboloid[3] the compressibility of the hyperboloid axes is marked by strong anisotropy ($\sim 10^2$, which is about 20 times greater than the anisotropy of compressibility along the crystallographic axes).

For the surfaces of the butterfly and the cigar in the 1-OPW approximation the area of an extremal cross section should increase with pressure. To account for the observed areal decrease one would have to assume considerable enhancement of the psuedopotential matrix element $W_{10\bar{1}1}$, in contradiction with data obtained for $W_{10\bar{1}1}$ from the cross section β of the monster. Moreover, the steep angular dependence of $d \ln S/dp$ for the cross sections of the butterfly and cigar, which are usually approximated by ellipsoids, is also possible only with anomalously high anisotropy of the axial compressibility of the ellipsoids.

On the basis of the interpretation of the L, K, and J oscillations that is given in[9] the angular dependence of $d \ln S/dp$ for the corresponding cross sections could not be predicted without detailed computer calculations, although internal consistency can be required for these oscillations. According to[9] the cross sections associated with the J, K, and L oscillations are related by $S_L \approx 2S_K - S_J$, from which it follows that

$$\frac{dS_L}{S_L} = \frac{2S_K}{S_L}\frac{dS_K}{S_K} - \frac{S_J}{S_L}\frac{dS_J}{S_J}.$$

For **H** ∥ [0001] we have $dS_K/S_K = -14 \times 10^{-3}$ kbar^{-1} and $dS_J/S_J = -11 \times 10^{-3}$ kbar^{-1}, while for dS_L/S_L we would expect -24×10^{-3} kbar^{-1}, which is close to the experimental value -18.9×10^{-3} kbar^{-1}. Figure 2 shows that the angular dependence of $d \ln S/dp$ for the J and K oscillations resembles that for the L oscillations, thus also indicating their internal consistency. Earlier data[15] on the effective mass m/m_0 and the Dingle factor x for these oscillations do not conflict with the magnetic breakdown model.

The oscillations with the J, K, and L frequencies thus appear to be of identical origin, most likely as a result of magnetic breakdown. However, the same data do not provide a basis for asserting that near the point L of the Brillouin zone the Fermi surface has no electron parts which in the H ∥ [0001] direction could be responsible for lower-amplitude oscillations, the latter being unobservable against the background of higher-amplitude oscillations with the J and K frequencies. These oscillations could be a natural continuation of the C branch, which is interpreted in[4] as the branch associated with the butterfly.

The pressure experiments enable us to determine the dependence of the pseudopotential on the lattice parameters. We calculated the pseudopotential matrix elements in the local approximation for p = 0 and 1 kbar, and we determined the pressure dependence of the pseudopotential form factors, $\Delta w_q/\Delta p$. The standard procedure yielded the secular equation

$$\begin{vmatrix} T_k - \varepsilon & & W_{q_i} \\ & T_{k-q_1} - \varepsilon & \\ & \cdots & \\ W_{q_i}^* & & T_{k-q_i} - \varepsilon \end{vmatrix} = 0, \qquad (1)$$

where $T_{k-q_i} = (\mathbf{k} - \mathbf{q}_i)^2/2$ is the kinetic energy of free electrons in atomic units, ϵ is the energy of electrons in the lattice, and W_{q_i} is the matrix element of the pseudopotential on the q_i-th Bragg reflection plane for electrons. Utilizing the experimental values of the areas of the extremal cross sections, we are able to determine the set of numbers w_q that best satisfy the equations (1) for different cross sections of the Fermi

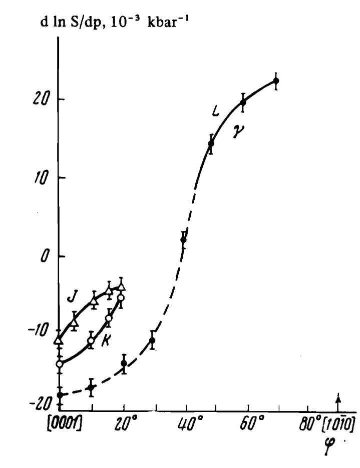

FIG. 2. Angular dependence of d ln S/dp for J, K, and L oscillations.

surface. The interpretation of the obtained numbers w_q depends on the assumed theoretical model and is, generally speaking, ambiguous.

The pressure-induced variation of the lattice parameters has two consequences: a) variation of the geometric parameters—the dimensions of the Brillouin zone and of the free-electron sphere radius, and b) variation of the pseudopotential. For the purpose of separating these effects easily we employed a simplified scheme of calculation. Although we could not thus achieve the best agreement of the numbers w_q obtained at different points of the Brillouin zone, we had a clearer procedure for calculating the pressure dependence of w_q. The simplifications were: 1) The Fermi energy was taken to be $k_F^2/2$ (in atomic units), while the radius of the Fermi sphere was determined only from the dimensions of the Brillouin zone and was not used as an adjustable parameter; 2) the number of mixed plane waves was limited to three.

In zinc the free-electron sphere intersects four Bragg planes of electron reflection: (0001), (0002), $(10\bar{1}0)$, and $(10\bar{1}1)$. However, the structure factor vanishes on the (0001) plane, so that experiments can determine only the three form factors w_{0002}, $w_{10\bar{1}0}$, and $w_{10\bar{1}1}$. This can be done most easily by considering three extremal cross sections of the Fermi surface—that of the lens (whose size is determined mainly by the matrix element W_{0002}), that of the needle in the direction $\mathbf{H} \parallel [0001]$ (determined by $W_{10\bar{1}0}$), and that of the cross section β of the monster in the direction $\mathbf{H} \parallel [11\bar{2}0]$ (determined by the matrix elements W_{0002} and $W_{10\bar{1}1}$). With the purpose of testing for internal consistency, in addition to these main cross sections we utilized cross sections of the lens and needle in other orientations together with the cross section σ of the monster and the maximal cross section of a disc of the butterfly. Values under pressure were calculated for cross sections obtained by extrapolating for p = 1 kbar. Table I gives the geometric parameters employed for the calculations.

Secular equations (1) for each concrete cross section were solved on a computer, with which we determined values of $\mathbf{k} = \mathbf{k}(\varphi, \theta, |W_q|)$ lying on the Fermi surface; here $\mathbf{k}$ is the wave vector, and φ and θ are polar coordinates. For each given direction we then integrated with respect to the angles and determined the cross-sectional area $S(|W_{q_i}|)$ as a function of the matrix elements W_{q_i}. A graph of the function was compared with experimental values of S. This procedure was employed for both p = 0 and p = 1 kbar; the pressure dependence of W_q was determined from the graph. An example of the results obtained by this calculation is shown in Fig. 3, where the extremal cross-sectional area of the needle for $\mathbf{H} \parallel [0001]$ is represented as a function of $W_{10\bar{1}0}$.

At p = 0 we have the area $S = 0.42 \times 10^{-4}$ at. units, which corresponds to $|W_{10\bar{1}0}| = 0.266 \times 10^{-2}$ Ry on the lower line of Fig. 3. Under pressure the area increases with the coefficient 0.282 S kbar^{-1} and at p = 1 it becomes 0.54×10^{-4} at. units, which corresponds to $|W_{10\bar{1}0}| = 0.269 \times 10^{-2}$ Ry on the upper line. The same figure shows values of S (1 kbar) obtained from the data of Itskevich[17] and of O'Sullivan and Schirber.[11] At p = 1 kbar these values should correspond to $W_{10\bar{1}0} = 0.278 \times 10^{-2}$ Ry and 0.263×10^{-2} Ry, respectively. Since from theoretical considerations $dw/d(q/k_F)$ should be greater than zero, while q/k_F decreases under pressure, from data of Gaĭdukov and Itskevich[10] and from the present work it follows that $w_{10\bar{1}0} < 0$ and increases in absolute value under pressure. However, from Schirber's data it follows that $w_{10\bar{1}0} > 0$ and decreases in absolute value under pressure; we would expect it to vanish at p ~ 100 kbar, thus producing a change in the topology of the Fermi surface. (By a simplified method of calculation[17] Itskevich obtained $d \ln w_{10\bar{1}0} = 2.8 \times 10^{-2}\ \text{kbar}^{-1}$).

TABLE I. Brillouin zone parameters used to calculate cross sections of the zinc Fermi surface, obtained from[2, 16]

p, kbar	Brillouin zone parameters (atomic units)		
	a	b	k_F
0	0.8350	0.6842	0.8393
1	0.8352	0.6850	0.8397

TABLE II. Calculated form factors of the pseudopotential and their pressure dependence

	Direction of q		
	(0002)	$(10\bar{1}0)$	$(10\bar{1}1)$
$q/2k_F$	0.815	0.863	0.954
$\|S\|$	1	1/2	$\sqrt{3}/2$
w_q, Ry	−0.0592	−0.00532	+0.0404
$\frac{d\|w_q\|}{\|w_q\|dp} \cdot 10^2$, kbar^{-1}	−0.51±0.34	+1.1±0.4	−0.11±0.05

TABLE III. Areas of Fermi surface cross sections calculated with the matrix elements given in Table II

Cross section	Direction of H	S_{calc}, at. units	Δ, %
Lens	0001	0.56	0
Lens	$10\bar{1}0$	0.190	2.5
Lens	$11\bar{2}0$	0.189	2.5
Monster (β)	$11\bar{2}0$	0.0012	0
Needle	0001	0.000042	0
Needle	$10\bar{1}0$	0.00063	10
Monster (σ)	$11\bar{2}0$	0.078	10
Butterfly	$10\bar{1}1$	0.091	25

Here $\Delta \equiv (S_{exp} - S_{calc})/S_{exp}$.

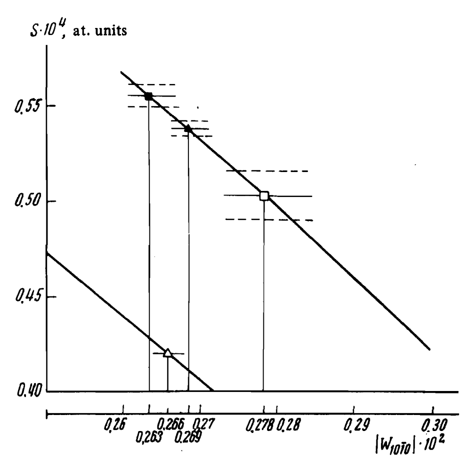

FIG. 3. S(W) of the needle for H‖[0001]; lower line at p = 0, upper line at p = 1 kbar. Data: ■—from[11], □—from[17], ▲ and Δ—present work.

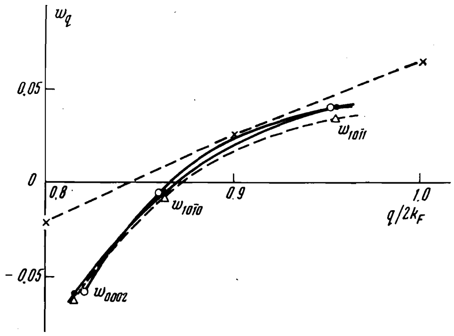

FIG. 4. Dependence of w_q on $q/2k_F$ at p = 0 (●) and p = 10 kbars (○); Δ–data from[7]; X–Heine-Abarenkov model pseudopotential.[18]

Similar calculations performed for other cross sections enabled us to obtain all the matrix elements of the pseudopotential. The dependence of $w_{\mathbf{q}}$ on $q/2k_F$ is shown in Fig. 4; because of the small pressure dependence of $w_{\mathbf{q}}$ it was plotted at 10 kbar for the sake of clarity. The same figure represents Harrison's data in[7], obtained by treating experimental data in[2]; the model pseudopotential of[18] is also plotted. Numerical values pertaining to the pseudopotential and its pressure dependence are given in Table II.

For the butterfly and cigar near **H** ∥ [0001] it was found to be impossible to select numbers $W_{\mathbf{q}}$ that would bring about agreement with the observed frequencies of the J and K oscillations. Even in the 1-OPW approximation and neglecting spin-orbit splitting for the butterfly-cigar system in the [0001] direction, the calculated cross section is somewhat smaller than the experimental value, which is 0.0293 at. units. The situation only becomes worse when the matrix elements are brought in. The maximal cross section of a butterfly disk for **H** ∥ [10$\bar{1}$1] is in better agreement with the chosen model, although the discrepancy is still large (see Table III).

Table III gives the areas of several extremal cross sections calculated in the 3-OPW approximation with the matrix elements given in Table II. Better agreement could be obtained by utilizing k_F as an adjustable parameter or by employing a nonlocal potential. However, the mathematical treatment would thereby become considerably more complicated and there would be a loss of simplicity in interesting results regarding the pressure dependence of $w_{\mathbf{q}}$.

The observed areas of extremal cross sections and their pressure dependence are therefore determined sufficiently well with the chosen model of a local pseudopotential, except for the cross sections in a magnetic field parallel to the [0001] direction, which have usually been regarded as connected with a butterfly and a cigar. For these cross sections the pressure data and the angular dependence of the pressure coefficient indicate that extremal magnetic-breakdown cross sections of the monster are here the more likely cause of magnetic-susceptibility oscillations. However, this does not prove the absence of a butterfly-cigar system like that in the band structure of zinc, because, as previously, evidence favoring this system is provided by the C branch,[4] which has still not been successfully investigated under pressure.

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