Multiple Sondheimer oscillations in thin (110-face) tungsten plates

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The known Sondheimer size effect [E. H. Sondheimer, Phys. Rev. **80**, 401 (1950)] is used to investigate the scattering of conduction electrons from various sections of the Fermi surface by the surface of a thin tungsten plate. The magnetic field **H** was directed along the [110] axis of the crystal. The Fuchs specularity parameters are calculated from the experimental data for different surface states of the crystal, and an explanation is proposed for the differences between them. The mean free paths are calculated for the carriers of these groups. It is demonstrated that this procedure has a high "resolving power" for groups with close values of $|\partial S/\partial p_z|$.

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1. INTRODUCTION

Conduction-electron scattering by metal surfaces has been the subject of a large number of studies. The interest in this scattering is due, in particular, to its substantial influence on the kinetic properties of thin metallic samples. Many methods were developed for the study of this influence, such as that of the static skin effect, of transverse focusing of the conduction electrons, and others.[2-4] These methods were used to measure the coefficients of specular reflection of conduction electrons from various faces of tungsten and some other single crystals, and to estimate the probability of interband transitions in such scattering. The static skin effect method, being integral, was found to be most sensitive to scattering with transition to other bands, if the sign of the carrier is reversed thereby. In contrast, the transverse-focusing method is sensitive to any scattering with loss of the tangential component of the quasimomentum. We have previously investigated[5] the scattering of conduction electrons by a surface, using a method based on the known size effect—the Sondheimer oscillations. Being a differential method, just as that of transverse focusing, it makes it possible to determine in certain cases the scattering parameters from substantially different sections of the Fermi surface (FS),[6] estimate the carrier mean free paths, and determine the curvatures of the FS sections responsible for the excitation of the oscillations. The Sondheimer method expands considerably thereby the possibilities of experimentally investigating scattering of conduction electrons from surfaces.

In Ref. 5 we succeeded in investigating the scattering of only group; this was possibly due to the small number of experimental points used in the successive Fourier analysis, and to the insufficient accuracy with which their coordinates were determined. In the present paper, with more accurate parameters, we carried out a similar investigation on the (110) face of tungsten, which has furthermore a higher specularity than the previously investigated (100) face.

2. EXPERIMENT

We used in the experiment W1 samples of thickness $d = 160$ μm and W2 samples 250 μm thick, with residual resistivity ratio $\rho_{300\,\mathrm{K}}/\rho_{4.2\,\mathrm{K}} \geqslant 10^5$. The samples were cut along the [110] direction. Since it was necessary to vary in the course of the experiment the state of the surface, one of the samples, W1, was placed in a glass bulb and mounted vertically accurate to 3°, and the bulb was evacuated to 10^{-11} Torr. Sample W2 was placed directly in liquid helium. All the measurements were performed in a superconducting magnet in the form of Helmholtz coils. This magnet form made it possible to monitor, by rotating the sample around the axis, the direction of the magnetic field in the (110) plane relative to the surface.

The standard modulation technique was used in the experiment to measure the first derivative dR_{xx}/dH of the diagonal component of the magnetoresistance R_{xx}, which was next differentiated by an analog device. The signal obtained, as well as a signal proportional to the intensity of the magnetic field H, was punched on a tape, and simultaneously recorded with an LKD4 automatic x-y recorder. The time rate of change of the magnetic-field intensity was maintained accurate to within 2%; this rate changed by less than 1% from measurement to measurement. The useful signal was therefore equal, with sufficient accuracy, to the second derivative d^2R_{xx}/dH^2 multiplied by a certain constant. In the course of the experiment we recorded up to 1500 points with four significant figures, and the measurement results were computer-reduced by a program that determined the Fourier-expansion coefficients of the useful signal. To check on the reliability of the Fourier analysis, the inverse Fourier transforms of the results were taken; the agreement with the original curve was good even in the small details.

3. DISCUSSION OF RESULTS

Figure 1 shows plots of d^2R_{xx}/dH^2 for sample W1 with atomically clean surface (curve 2) and with a surface coated with a submonolayer film of the residual gases (curve 1). The transition from curve 1 to curve 2 occurred after the sample was heated to 2500 K. The monotonic part of the magnetoresistance R_{xx}, which was also measured in the experiment, increased by 1.4 times at the same time. The initial state of the sample was restored after maintaining the apparatus at room

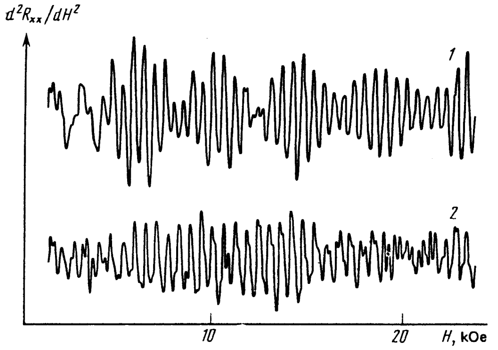

FIG. 1. Plots of the second derivative $\partial^2 R_{xx}/\partial H^2$ of the magnetoresistance vs the magnetic field H. Curve corresponds to a sample whose surface is coated with a submonolayer impurity film, and curve 2 to a sample with a cleaned surface.

temperature. This was evidence that the change of the shape of curve 1 was due to the change in the surface state and not in the volume of the sample. It is seen from Fig. 1 that removal of the submonolayer film of the residual gases from the surface ("cleaning" the surface) altered curve 1 in two ways: first, the amplitude of the oscillations decreased somewhat, and second, the "modulation" clearly noted on the curve disappeared. Figure 2 shows the results of the Fourier analysis of the respective curves. Curve 1 shows clearly four harmonics corresponding to four carrier groups; only two of them are noticeably expressed in curve 2. In addition, there appeared the heretofore unobserved harmonics 2β, 2γ, and 2δ, which are multiples of the harmonics β, γ, and δ, respectively.

For quantitative estimates, we solved Eq. (2.6) in Ref. 5; the expression obtained for the oscillating part of the conductivity is

$$\sigma_{\rm osc}^{\pm}=\left(\frac{c}{H}\right)^2\frac{2}{dh^3}\sum\int dp_z S\frac{|m||v_z|}{(\gamma\mp i)^2}\sum_{s=0}^{\infty}\{\rho^s(1-\rho)^2\exp[-(2s+1)\xi(\gamma\mp i)]$$
$$+\rho^{s+1}(1-\rho)^2\exp[-(2s+2)\xi(\gamma\mp i)]\}. \qquad (1)$$

here $\sigma_{\rm osc}^{\pm}$ is the oscillating part of the conductivity ($\sigma^{\pm}=\sigma_{xx}\pm i\sigma_{xy}$), ρ is the Fuchs specularity parameter, v_z is the z-component of the carrier velocity ($z \parallel \mathbf{H}$), S is the area of the intersection of the FS with the plane $p_z=$ const, $\gamma=\nu/\Omega$, where Ω is the cyclotron frequency, ν is the collision frequency, l is the carrier mean free path, m is the effective mass, and

$$\xi=2\pi|\partial S/\partial p_z|^{-1}deH/c.$$

The summation is over all the sheets of the FS. We have assumed that both surfaces reflect the conduction electrons in like manner.

It was observed in Ref. 5 that the main contribution to the conductivity oscillations is made by electrons from section A of the hole octahedron; The integral (1) was correspondingly evaluated for the FS section with the maximum value of $|\partial S/\partial p_z|$. In the present paper, however, the shapes of the curves of Fig. 1 and, in particular, the absence of a pronounced dependence of the oscillation amplitude on the magnetic-field intensity make it necessary, when evaluating the integral (1), to take into account also the contribution of the limiting points of the Fermi surface. In this case we obtain for the oscillating part of the conductivity $\sigma_{\rm osc}^{\pm}$

$$\sigma_{\rm osc}^{\pm}=\left(\frac{c}{H}\right)^4\frac{1}{2\pi^2 d^3 e^2 h^2}\sum_i F^{(i)}\sum_{s=0}^{\infty}\left\{\frac{\rho^{2s}(1-\rho)^2}{(2s+1)^2}\exp[-(2s+1)\xi^{(i)}(\gamma\mp i)]\right.$$
$$\left.+\frac{\rho^{2s+1}(1-\rho)^2}{(2s+2)^2}\exp[-(2s+2)\xi^{(i)}(\gamma\mp i)]\right\}, \qquad (2)$$
$$F^{(i)}=(\partial S^{(i)}/\partial p_z)^6/(\partial^2 S^{(i)}/\partial p_z^2)^4. \qquad (3)$$

The superscript (i) indicates that this expression pertains to the i-th limiting point. Just as before, we have assumed that $\nu \ll 1$ and that the FS has axial symmetry. Taking Eq. (2.28) of Ref. 5 into account

$$\frac{dR_{xx}}{dH}=\frac{2}{H}R_{\rm mon}-R_{\rm mon}^2\frac{d\sigma_{\rm mon}^{+}}{dH}, \qquad (4)$$

where $R_{\rm mon}$ is the monotonic part of the magnetoresistance

$$R_{xx}=R_{\rm mon}+R_{\rm osc}, \qquad (5)$$

we obtain

$$\frac{d^2R_{xx}}{dH^2}=\frac{2}{H^2}R_{\rm mon}-R_{\rm mon}^2\frac{d^2\sigma_{\rm osc}^{+}}{dH^2}; \qquad (6)$$

$$\frac{d^2\sigma_{\rm osc}^{+}}{dH^2}=\frac{c^2}{H^4}\frac{2}{dh^3}\sum_i F_1^{(i)}\sum_{s=0}^{\infty}\{\rho^{2s}(1-\rho)^2\exp[-(2s+1)\xi^{(i)}(\gamma-i)]$$
$$+\rho^{2s+1}(1-\rho)^2\exp[-(2s+2)\xi^{(i)}(\gamma-i)]\},\qquad F_1^{(i)}=\frac{(\partial S^{(i)}/\partial p_z)^4}{\partial^2 S^{(i)}/\partial p_z^2}. \qquad (7)$$

In (6) and (7) we differentiated only the rapidly oscillating functions. Just as in Ref. 5, Eqs. (2) and (7) describe conductivity changes periodic in $\mathbf{H}$, with a period

$$\Delta H=(c/de)|\partial S^{(i)}/\partial p_z|. \qquad (8)$$

For the ratio of the first and second harmonics we obtain the expression

$$G_2/G_1=\rho e^{-d/l}. \qquad (9)$$

Formulas (3.4), (3.5), and (3.6) of Ref. 5, which we need to calculate the specularity coefficients, remain unchanged:

$$\rho'=\rho''G_1''G_2'/G_1'G_2'', \qquad (10)$$

$$\frac{G_1''}{G_1'}=\left(\frac{R_{\rm mon}''}{R_{\rm mon}'}\frac{1-\rho''}{1-\rho'}\right)^2, \qquad (11)$$

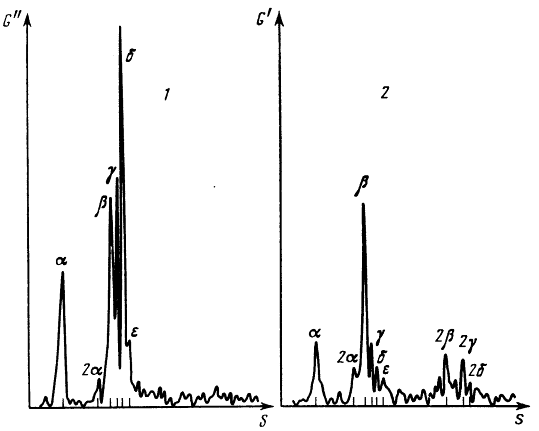

FIG. 2. Results of Fourier analysis of curves 1 and 2 of Fig. 1. The different Greek letters denote the contributions of carriers from different sections of the FS, and the numbers preceding them correspond to multiple harmonics.

TABLE I.

Group	$\frac{\hbar}{2\pi}\left\lvert\frac{\partial S}{\partial p_z}\right\rvert$, Å^{-1}	G_1''/G_2''	G_1'/G_2'	G_1'/G_1''	ρ''	ρ'	l, mm
α	0.596	4.79	1.63	0.48	0.2±0.05	0.6±0.05	0.15±0.01
β	0.250	15.40	4.19	0.97	0.1±0.05	0.35±0.05	0.35±0.05
γ	0.228	20.42	1.31	0.26	0.05±0.05	0.7±0.05	⩾1.5
δ	0.219	17.40	1.44	0.10	0.05±0.05	0.75±0.05	⩾1.5
ε	0.205	3.93	1.16	0.43	0.2±0.05	0.6±0.05	0.23±0.01

$$\rho''=\frac{1-(R'_{\mathrm{mon}}/R''_{\mathrm{mon}})\,(G_1''/G_1')^{1/2}}{1-(R'_{\mathrm{mon}}/R''_{\mathrm{mon}})(G_1''/G_1')^{3/2}\,(G_2'/G_2'')}\,. \tag{12}$$

Here G_n is the amplitude of the n-th harmonic in the Fourier expansion of the useful signal. All the single-primed parameters correspond to the state of the crystal with atomically clean surface, and the doubly primed to those with a surface coated by a submonolayer impurity film. The mean free path of the carriers is now given by

$$l=d/\ln\,(\rho G_1/G_2). \tag{13}$$

It must be noted that in the limit $d/l\to 0$ the foregoing results are in full agreement with Goland's results.[7]

Now, using the results of the Fourier analysis of the oscillations and these formulas we can estimate the values of $|\partial S/\partial p_z|$, the specularity parameters, and the mean free paths for all carrier groups of curves 1 and 2 of Fig. 2. The results of these estimates are summarized in Table I. The experimental errors (except those in the mean free paths, which will be discussed below) were determined by the signal/noise ratio and did not exceed 10%.

The close values of $|\partial S/\partial p_z|$, as well as of the specularity parameters of groups γ and δ, have led us to the assumption that these groups are connected with two FS sections that are symmetric about the [100] axis, and that the appearance of the difference between the periods is due to the slight tilt of the (100) plane relative to the [110] axis. Since the tilt could be the result of insufficient accuracy of setting the sample in the tube, and we were unable to calculate it, the experiment on sample W2 was performed directly in liquid helium. In this case the sample was so mounted that the deviation of H from the normal to the sample surface did not exceed 0.5°. The measurement results are shown in Figs. 3 and 4. Comparison with Figs. 1 and 2 has shown that when the sample was rotated in the magnetic field the harmonics γ and δ merged into one, the harmonic γ, thus confirming our assumption. The strong decrease of the amplitude of the γ peak is due to the high degree

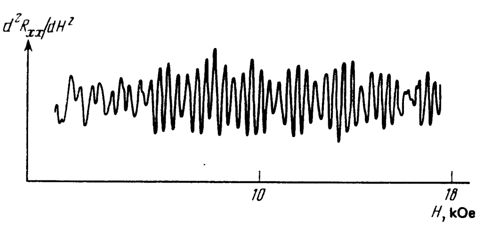

FIG. 3. Plot of $\partial^2 R_{xx}/\partial H^2$ vs H for crystal W2. The deviation of the magnetic field H from the normal to the crystal surface does not exceed 0.5°.

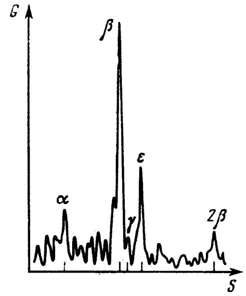

FIG. 4. Results of the Fourier analysis of the curve of Fig. 3.

of specularity of the (110) face after electric polishing, a fact already noted by Tsoĭ and Razgonov.[8] In addition, the results for this sample are in good agreement with results of an investigation of the Gantmakher-Kaner effect on the (110) face of tungsten.[9]

We have attributed the oscillations of groups α and ε, following Ref. 10, to sections A' of the hole octahedron and J of the electron Jack, respectively [see Fig. 5(a)]. The mean free paths of the carriers of these groups were calculated from the formula (see Ref. 5)

$$l=d/\ln\,(2\sqrt{2}\rho G_1/G_2). \tag{13′}$$

Since, as already mentioned, there are no expressions for the dependence of the oscillations on curves 1 and 2 on the magnetic field intensity, and the combined contribution of the oscillations of groups β and γ to the overall picture predominates, we have, using the results of Ref. 11, assigned these groups to the limiting points of the hole ellipsoids. The doubling of the peak γ on curve 1 of Fig. 2 means that the vicinity of the limiting point of group γ has no symmetry plane passing through this limiting point and parallel to the [100] direction. This condition is fully satisfied by the limiting point M. We have accordingly assigned the group β to the limiting point Q. Indeed, the experimentally obtained values of $|\partial S/\partial p_z|$ for this group agree with good accuracy with the value calculated in Ref. 11 (−0.223 Å^{-1}). Unfortunately, the agreement for group M is much worse (we recall that in Ref. 11 the value for group M is $(\hbar/2\pi)\,|\partial S/\partial p_z| = -0.128$ Å^{-1}).

We dwell finally on the cause of the difference between the specularity coefficients of the "clean" sur-

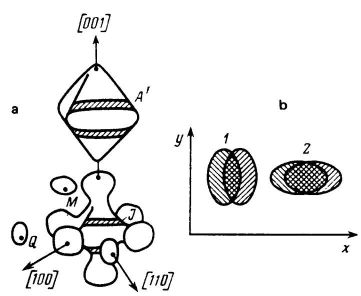

FIG. 5. a) Fermi surface of tungsten.[11] The marked sections are those whose carriers are responsible for excitation of oscillations. b) Illustration explaining the causes of the different values of the Fuchs specularity parameter ρ' for groups β and γ.

face for the very close groups β and γ. It must be noted first that in the case of the Sondheimer effect the additional condition for specular reflection is conservation of the total quasimomentum of the carrier from the FS, and not only of its tangential component. This means that when an electron from a certain section of the FS must land, after being scattered by the plate surface, on the same section, owing to the differential character of the effect. This condition can be satisfied only in a magnetic field whose direction coincides with the normal to the surface, while the normal coincides with a high-order symmetry axis, when the projections of the Fermi-surface sections responsible for the effect coincide. Deflection of the field away from the normal is equivalent to a shift of these projections relative to each other; the specular-reflection condition for the Sondheimer effect will then be satisfied only in a region common to both. Thus the surface, remaining specular in the sense of Ref. 12, becomes more diffuse for the carriers that contribute to the oscillations.

We introduce now viscinities of two reference points whose projections are ellipses and whose areas are equal; the major axis of one ellipse is perpendicular to the major axis of the other [e.g., directed along x for one and along y for the other; see Fig. 5(b)]. We assume further that deflection of the field shifted the projections along the x axis. For equal values of the shift, the total area of projection 1 is then smaller than the total area of the projection 2. Indeed, the specularity of the surface will be higher for the carriers from the region 2. A similar situation is realized in our case. The vicinity of the limiting point of the hole ellipsoid has on the (110) plane an "ellipse" whose major axis is directed along the [100] axis. The projection of the vicinity of the limiting point M in the same plane, however, is more elongated in the [110] direction. The shift of these projections in the [110] direction [which is equivalent to a tilt of **H** from the normal in the (100) plane] leads to larger specularity for the group M.

Estimates of the carrier mean free path in several measurement runs yielded substantially different values of the scatter of these values about the mean value; the scatter was quite substantial, in particular, for groups γ and δ. To find the reason for this, we investigated the sensitivity of Eqs. (13) and (13′) to measurement errors. After simple transformations we obtained the following expression for the linear increment of the mean free path in the case of a linear increment of the argument of the logarithm in the denominator of Eq. (13):

$$\Delta l \propto -\frac{d}{(k\rho G_1/G_2)\ln^2(k\rho G_1/G_2)}\Delta\left(\rho\frac{G_1}{G_2}\right), \tag{14}$$

where $k = 1$ or $2\sqrt{2}$, depending on whether (13) or (13′) is used. Noting from (9) that $k\rho G_1/G_2 = e^{d/l}$ and is always larger than unity, we ultimately obtain

$$\Delta l \propto -\frac{l^2}{d}\Delta\left(\rho\frac{G_1}{G_2}\right). \tag{15}$$

for groups α and β we have $l \propto 2d$ and Eqs. (13) and (13′) are relatively stable; for groups γ and δ we have $l \gg d$ and Eq. (13) is unstable.

We have thus investigated scattering of four carrier groups by the (110) face and determined the FS sections responsible for their excitation. We calculated the experimental values of $|\partial S/\partial p_z|$ at the limiting points Q and M of the hole ellipsoids of the Fermi surface of tungsten, and compared them with the theoretical results of Ref. 11. We estimates the mean free paths for these groups. We have demonstrated the high "resolving power" of the Sondheimer oscillation method, namely, we separated groups whose $|\partial S/\partial p_z|$ differed by about 3.5% (γ and δ in the text), a resolving power better than that of any other differential method. The result here agree well with results by others.

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