# Calculation of Band-Structure Effects in Field-Emission Tunneling from Tungsten*

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A calculation was made to determine whether band-structure effects in field emission, as described in the Stratton theory of tunneling from a metal, could explain the anomalous total energy distribution (TED) for the ⟨100⟩ directions of tungsten obtained by Swanson and Crouser. In applying the Stratton theory, a simple model for the tungsten energy bands based on Loucks's calculated relativistic augmented-plane-wave band structure was chosen. The results are in fair qualitative agreement with the experimental data of Swanson and Crouser with two important differences: (1) Swanson and Crouser observed merely a shoulder in the TED, but our results show a small peak; (2) Swanson and Crouser observed an anomaly centered around $\epsilon\cong-2.0d$, but our peak is located at $\epsilon\cong-2.5d$, where $d$ is determined by the applied field. These discrepancies between theory and experiment may be due to the crudeness of the band-structure model used.

## I. INTRODUCTION

FIELD emission was observed as early as 1897,[1] and after the advent of Sommerfeld's free-electron theory of metals, a quantum theory of the effect was developed; this was begun by Fowler and Nordheim[2] in 1928. In their original work, the surface potential was a step function at zero applied field and a triangular barrier at finite values of the applied field, but this was later modified by Nordheim[3] to a one-dimensional image potential. The reduction of the problem to one-dimensional motion prompted the use of the normal energy distribution, and it was several years before experimental results seemed to agree with Nordheim theory.[4] However, this agreement was discovered to be fortuitous by Young and Müller[5,6] when measurements performed with their increased-resolution retarding-potential analyzer were found to be strikingly different from the predictions of normal energy theory; this led to the realization that the use of this retarding-potential analyzer measured not the *normal* energy distribution but rather the *total* energy distribution of the emitted electrons. The total energy distribution derived by Young[5] was found by Young and Müller[6] to agree quite well with the new measurements. The detailed experimental study of the Fowler-Nordheim (FN) theory by Van Oostrom corroborates their work.[7]

The recent field emission experiments with tungsten and molybdenum by Swanson and Crouser[8–10] indicate that the FN theory as modified by Young[4] does not explain certain aspects of field emission from metals. They measured the total energy distribution of electrons field-emitted from tungsten and molybdenum crystals in various directions and found agreement with the FN theory for all but the [110] and [100] directions; for these directions, the total energy distribution curves were significantly different from theoretical predictions. The largest deviation from theory was found for the [100] direction. Figure 1 shows the total energy distribution according to Young for various values of $p=kT/d$ with $d=0.174$ eV; this is to be compared with Fig. 2 which presents the experimental results of Swanson and Crouser for the [100] direction of tungsten. While the shape of the experimental curves agrees with FN theory, both very near to the Fermi energy and far below it, there is an anomalous hump centered at $\epsilon\cong-2d\cong-0.35$ eV. This hump was found to disappear when a layer of low work function ZrO was adsorbed on the tungsten emitter. It is also interesting that the emission from the [116] direction (only 13° from the [100] direction) was found to be in good agreement with theory. By various experimental means, Swanson and Crouser were able to eliminate electron

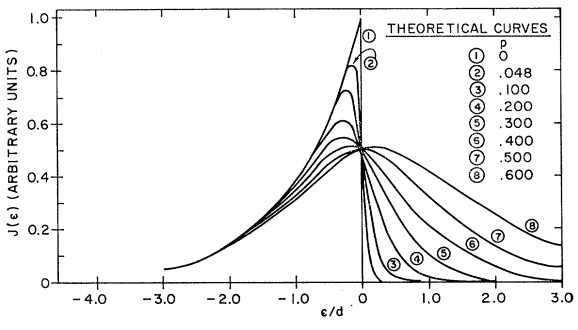


FIG. 1. Theoretical total energy distribution according to Young with $d=0.174$. $\epsilon=0$ corresponds to the Fermi energy.

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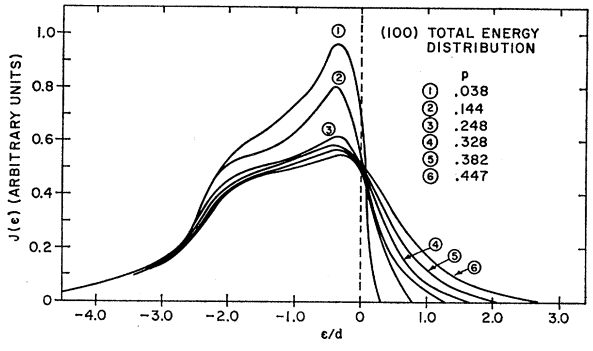


FIG. 2. Experimental total energy distribution for the [100] direction of tungsten according to Swanson and Crouser with $d=0.174$.

optical effects, magnetic field interactions, and collector patch-field effects as possible explanations of the anomaly. On the other hand, the angular localization of the anomaly suggests that it might be due to the details of the band structure of tungsten. The object of this work was to determine whether band-structure effects as included by Stratton[11] in his theory of tunneling from solids and applied to metals can be used to explain the anomaly in the tungsten [100] field emission.

In Sec. II the Stratton theory is discussed and appropriate formulas derived; in Sec. III the band structure of tungsten is discussed; in Sec. IV the Stratton theory is applied to tungsten using a simple model for the Fermi surface and energy bands of tungsten; and finally, in Sec. V, the results of these calculations are discussed.

## II. STRATTON THEORY

Stratton[11] has proposed a theory for the total energy distribution of field-emitted electrons that is expected to apply emitters of arbitrary band structure. It is assumed that: (i) the transmission coefficient depends on the state of the electron only through the energy outside the barrier associated with the $x$ direction and (ii) the tangential components ($P_y$ and $P_z$) of the electron quasimomentum are conserved when it tunnels through the barrier. Assumption (ii) implies that the $x$ directed energy outside the metal is $E_x=E(\mathbf{P})-E_\perp$, where $E(\mathbf{P})$ is the dispersion relation for the total energy and $E_\perp\equiv(P_y^2+P_z^2)/2m$. In general, $E_x\neq P_x^2/2m$ and this is the point of divergence from the derivation of the total energy distribution by Young in that it introduces the band structure of the solid through $E(\mathbf{P})$ and thus permits the use of other models besides the free-electron model.

In the following, the vacuum level is defined as the zero of potential energy. If $f(E)$ is the average number of electrons that occupy a quantum state of energy $E$ (the electron distribution function) and if $v_x=\partial E/\partial P_x$ is the $x$ component of velocity of an electron with energy $E$ and if $D(E-E_\perp)$ is the probability that an electron with $x$-directed energy $E-E_\perp$ will penetrate the potential barrier (the transmission coefficient), then the total energy distribution obtained by Stratton is

$$P_T(E)=-\frac{K}{2\pi}f(E)\int_0^{2\pi}d\phi\int_E^{E-E_m(E,\phi)}D(E_x)dE_x$$
$$=Kf(E)\left[\int_0^E D(E_x)dE_x-\frac{1}{2\pi}\times\int_0^{2\pi}d\phi\int_0^{E-E_m(E,\phi)}D(E_x)dE_x\right], \quad (1)$$

where $\phi$ is the polar angle in the plane perpendicular to the emission direction; $E_m(E,\phi)$ is the maximum value of $E_\perp$ for a given polar angle $\phi$, and $K=4\pi m/h^3$. The energy distribution has thus been divided into two terms, one which is independent of the detailed band structure of the metal and another which depends on the band structure through $E_m(E,\phi)$.

If the WKB approximation is used this becomes

$$P_T(E)=Kd(E)f(E)e^{-c(E)}\times\left[1-\frac{1}{2\pi}\int_0^{2\pi}e^{-d^{-1}(E)E_m(E,\phi)}d\phi\right], \quad (2)$$

where $c$ and $d^{-1}$ are the usual coefficients in the Taylor series expansion of the logarithm of the WKB transmission coefficient. It is now evident that the band-structure effects, which enter by means of the integral in Eq. (2) are negligible unless $d^{-1}(E)E_m\lesssim 1$. If this criterion is not met, the expression for $P_T(E)$ reduces to the result derived by Young, which we shall denote by $P_1(E)$.

## III. BAND STRUCTURE OF TUNGSTEN

The band structure of tungsten has been studied both theoretically and experimentally by a large number of

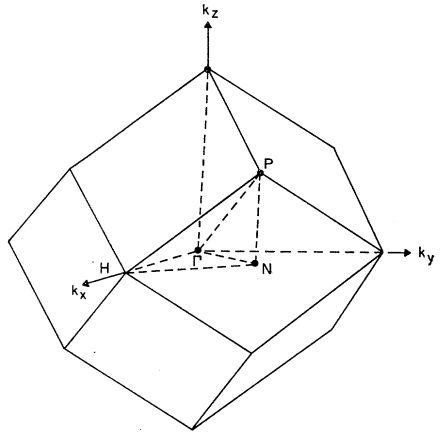


FIG. 3. Symmetry points in the Brillouin zone for a bcc lattice.

[11] R. Stratton, Phys. Rev. **135**, 794 (1964).

investigators and there is general agreement between theory and experiment regarding the shape and dimensions of its Fermi surface. However, much less is known about lower-energy bands than about the Fermi surface. In order to facilitate the description of the Fermi surface and references to it, the notation of Wigner, Boukaert, and Smoluchowski[12] for the symmetry points and directions in the Brillouin zone is used; this notation is given for a bcc lattice in Fig. 3.

The first tungsten band-structure calculations were done by Manning and Chodorow[13] who used the cellular method. More recently, Lomer[14] proposed a model for all of the chromium-group metals, based mainly on Wood's[15] augmented plane wave (APW) calculations for iron. This model consisted of a central body which tapered off smoothly in the $\Gamma H$ ([100]) directions but Lomer[16] later revised this to an electron octahedron centered on $\Gamma$ joined to hole octahedra at the symmetry points $H$ by spherical protrusions from the central octahedron in the $\Gamma H$ directions. In the necks, joining the central octahedron to the spherical protrusions, are electron lenses which have been invoked to account for the anomalies observed by Swanson and Crouser. There are also small hole pockets at $N$. Although Lomer limited the validity of this revised model to molybdenum, nonrelativistic APW calculations by Mattheiss[17]

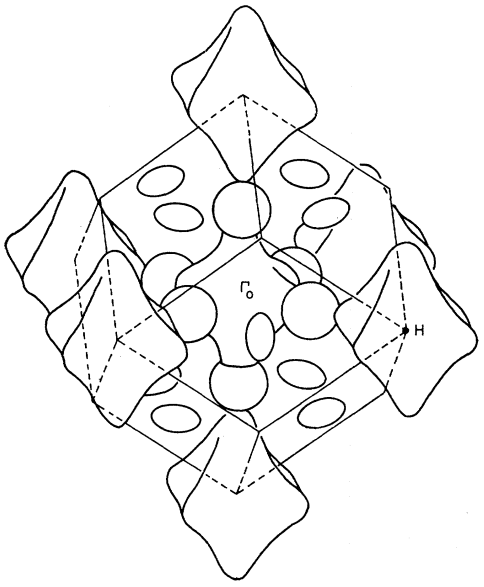


FIG. 4. Sketch of the revised Lomer model for the Fermi surface of tungsten.

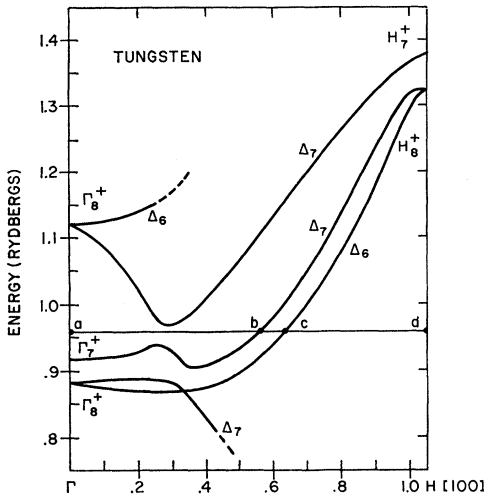


FIG. 5. Relativistic energy bands for tungsten (Ref. 18).

as well as by Loucks[18] show that its major features are also those of the Fermi surface of $W$. Figure 4 is a sketch of the model. The size-effect measurements of Walsh and Grimes[19] and the de Haas–van Alphen measurements of Sparlin and Marcus[20] indicate, however, two important differences between the revised Lomer model and the measured Fermi surface of tungsten: (a) Whereas in the Lomer model, the electron jack (consisting of the central octahedron and the spherical protrusions from it) contact the hole octahedra at $H$, Walsh and Grimes have found that there is a separation between them which is equal to about 5% of the $\Gamma H$ distance. (b) While Sparlin and Marcus[20] were able to observe orbits corresponding to the lenses in the Fermi surface of molybdenum, they were not able to observe the corresponding orbits in their experiments with tungsten. Relativistic APW calculations by Loucks[21] agree with these results. From Fig. 5, one sees that, because of spin-orbit coupling, the lower $\Delta_7$ band is split into two parts, the higher one having a small hump at about 0.03 Ry or 0.4 eV below the Fermi surface. The lens in the Fermi surface of molybdenum is a manifestation of the dipping of the upper $\Delta_7$ band below the Fermi level, which does not happen in tungsten, but the existence of this hump shows that there are lenses in the energy surfaces of tungsten at about 0.4 eV below the Fermi energy. That this is almost the same as the energy at which the anomalous shoulder observed by Swanson and Crouser occurs adds to the plausibility of their hypothesis that it is a result of this hump in the $\Delta_7$ band.

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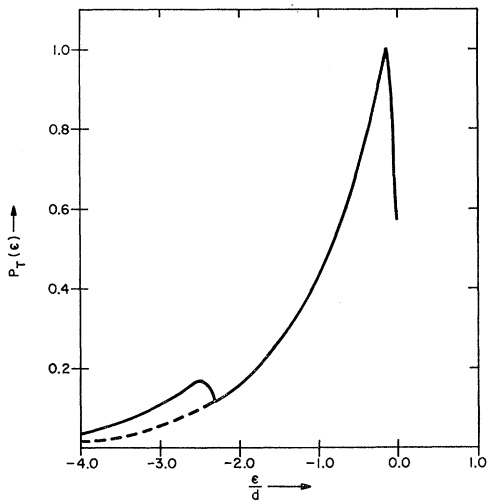


FIG. 6. Comparison of the total energy distribution according to the model of Sec. IV (solid curve) with the total energy distribution according to Young (broken curve) for $p=0.038$.

## IV. APPLICATION OF STRATTON THEORY TO TUNGSTEN

In applying the Stratton theory to a particular case, it is necessary to make two model approximations, one for the surface potential $V(x)$ and another for the energy surfaces. The potential model used in the following work is the one-dimensional classical-image potential. The expansion coefficients $c$ and $d$ are therefore,

$$c=[6.83\times10^{7}\phi^{3/2}/F]v(y) \quad \text{eV},$$
$$d=\frac{9.76\times10^{-9}F}{\phi^{1/2}t(y)} \quad \text{eV}, \tag{3}$$

where $F$ is the electric field in V/cm, $\phi$ is the work function in eV, and $v$ and $t$ are slowly varying functions of $y=3.79\times10^{-4}F^{1/2}/\phi$ and tabulated in a review article by Good and Müller.[22] A model for the energy surfaces is suggested by the general features of the computed Fermi surface of tungsten and other details of its band structure.

Inasmuch as the band structure term in the Stratton formula depends only on cross sections of the energy surfaces in the direction of emission and not on their detailed topology, it is possible to adopt a simple model in applying it to tungsten. The central octahedron and the spherical protrusions were approximated by a spherical energy surface whose effective mass is large enough to cause the band-structure term to vanish. Recent cyclotron resonance measurements of the effective mass by Hermann[23] for this surface range from about 1.2 to 2.7. Similar values can also be deduced from an analysis of the computed $\Delta_6$ and $\Delta_7$ bands for tungsten. The lenses were approximated by spherical energy surfaces with effective mass $r=0.30$, measured in units of the free-electron mass. This value is that obtained by Sparlin and Marcus[20] for a neck orbit but it is assumed to be the same as the value that would be obtained for an orbit around the rim of the lens, as is the case with molybdenum, even though in tungsten the lens does not appear until about 0.4 eV below the Fermi energy (which is the reason for the lens orbit's not being experimentally observable for tungsten). The experimental effective masses are cyclotron effective masses but for circular orbits they are identical with the dynamic (or band structure) effective masses.

One could employ a more realistic model by using ellipsoidal energy surfaces of the form

$$E(\mathbf{p})=\frac{P_x^2}{2m_3}+\frac{P_y^2}{2m_2}+\frac{P_z^2}{2m_1},$$

in which case

$$E_m(E,\phi)=\frac{r_2E}{\sin^2\phi+(r_2/r_1)\cos^2\phi}$$

must be used in Eq. (2). $r_1$ and $r_2$ are effective masses for orbits in the $x$-$y$ plane and $x$-$z$ plane, respectively, and they are measured in units of the free-electron mass. In this case, the integral would have to be evaluated numerically and the improvement of the model for energy surfaces of tungsten would provide no greater information than the simpler model to be used as far as the [100] emission is concerned.

It was assumed that the total energy distribution $P_T(E)$ is the sum of $P_1(E)$ and $P_2(E)$, contributions from the main energy surface and the lenses, respectively. The lenses correspond to the energy surfaces of electrons in a filled band while the central sphere corresponds to the energy surfaces of electrons in a partially filled band. $P_1(E)$ is the usual total energy distribution for electrons field-emitted from a metal while $P_2(E)$ is obtained by noticing that the main contribution to the electron emission from the filled band will be from the top of that band so $B(E_x)$, the logarithm of the transmission coefficient, may be expanded about the top of the filled band instead of about the Fermi energy as for $P_1(E)$. Thus, if $c'$ and $d'$ denote the values of $c$ and $d$ as previously defined, evaluated at the top of the filled band ($E=E_H$), and one uses the fact that for the lens $E_m(E,\phi)=r(E_H-E)$ for $E<E_H$ and $E_m(E,\phi)=0$ for $E>E_H$, the following is obtained for $P_2(E)$:

$$P_2(E)=6Kd'e^{-(c'+E_H/d')}f(E)e^{E/d'}(1-e^{-r(E_H-E)/d'}), \quad E<E_H \tag{4}$$
$$=0, \quad E>E_H.$$

This differs from Eq. (32) of Stratton's paper which was previously derived by Fischer[24] only in the substitution

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of $E_H-E$ for $E$ and multiplication by a factor of 6, the differences being due: (a) to the use of the bottom of the conduction band as the reference level for potential energy by Stratton; (b) to the fact that the lens does not appear in the energy surface until the energy $E=E_H$ is reached; (c) to the existence of six lenses in the equivalent $\bar{\Gamma}\bar{H}$ directions. The total energy distribution is therefore,

$$P_T(\epsilon)=P_1(\epsilon)\left[1+\frac{6d'}{d}(1-e^{-r(E_H-\epsilon)/d'})\times\exp\left(\frac{d-d'}{dd'}\epsilon+\frac{\zeta-\epsilon_H}{d'}+c-c'\right)\right],\quad \epsilon<\epsilon_H$$
$$=P_1(\epsilon),\quad \epsilon>\epsilon_H \qquad (5)$$

where

$$\epsilon=E-\zeta,\ P_1(\epsilon)=\frac{1}{1-p}\times\exp\left[p\ln\left(\frac{1-p}{p}\right)\right]\frac{e^{\epsilon/d}}{e^{\epsilon/pd}+1},$$

and

$$p=kT/d;$$

$P_1(\epsilon)$ has been normalized to unity. Using $r=0.30$, $\epsilon_H=-0.40$ eV, $F=4.1(10)^7$ V/cm and $-\zeta=4.80$ eV this becomes (for $\epsilon<\epsilon_H$)

$$P_T(\epsilon)=P_1(\epsilon)[1+5.82(1-e^{0.32(2.30+\epsilon/d)})e^{0.04\,\epsilon/d}]. \qquad (6)$$

$P_T(\epsilon)$ is plotted for comparison with $P_1(\epsilon)$ in Figs. 6 and 7 for $p=0.038$ ($T=77$°K) and $p=0.248$ ($T=500$°K), respectively, with a field of $4.1(10)^7$ V/cm. From these figures it is evident that the Stratton theory with our model has at least qualitative agreement with the anomalous measurements of Swanson and Crouser.

## V. DISCUSSION OF RESULTS

The Stratton theory with our model for the energy surfaces of tungsten yields total energy distribution curves whose deviation from those obtained for free-electron model is of the same order of magnitude as the anomaly reported by Swanson and Crouser. However, whereas the experimental anomaly has the form of a shoulder in the energy distribution curve at $\epsilon/d\cong-2.0$, the present work shows a peak at $\epsilon/d\cong-2.5$. This peak can be made to coincide with the observed anomaly simply by using a higher value of $E_H$ in the foregoing calculations; inasmuch as the quantity $E_H$ is not yet known with any certainty, it is possible that the value actually chosen is too low. While our results are very sensitive to the value of $E_H$, they are almost independent of $r$ as far as the position of the peak is concerned; the height of the peak, however, does depend significantly on $r$.

In considering the significance of this work relative to the validity of the Stratton theory, three model approximations for the $\Delta_7$ band from which the anomalous emission is presumed to originate should be remembered. (1) The portion of the band at $k\gtrsim0.4$ Å$^{-1}$ has been neglected, resulting in a discontinuous slope at $\epsilon/d=-2.3$; the effect of including the neglected portion of the $\Delta_7$ band in addition to eliminating the discontinuity in the slope would be to lower the relative height of the peak. (2) The disappearance of the lens below $\epsilon/d\cong-3.5$ has been neglected; this obviously does not affect the total energy distribution significantly in the range of the Swanson and Crouser results since it occurs far in the wing of the distribution. (3) It has been pointed out by Gadzuk[25] that the anomalous emission is supposedly originating from $d$-like bands, whereas we have not used any wave functions explicitly in our calculations; i.e., this form of the WKB approximation depends only on the nature of the surface barrier. Since the tunneling of $d$ electrons differs from that for $s$ electrons, taking account of the $d$-band characteristics would cause the Stratton theory to give a less pronounced hump in the total energy distribution due to smaller tunneling probability of $d$ electrons. Such a general approach has been suggested by Gadzuk which uses an additional angular-momentum-dependent potential term in the WKB transmission coefficient and a higher density of states for $d$ bands. Besides these band-structure approximations, we have made one other approximation in assuming that the total field emission current which comes from two overlapping bands can be calculated as the sum of contributions from each of these bands, considered independently of each other.

If the same procedure that we have used for tungsten were employed in calculating the total energy distribution for the ⟨100⟩ direction of Mo, the only difference

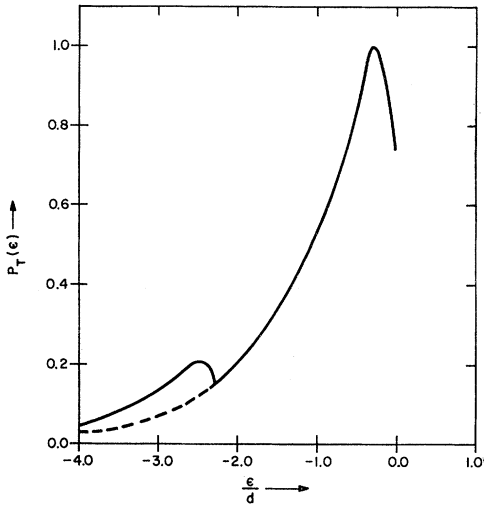


FIG. 7. Comparison of the total energy distribution according to the model of Sec. IV (solid curve) with the total energy distribution according to Young (broken curve) for $p=0.248$.

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would be in the value of $\epsilon_H$. If we chose $\epsilon_H \cong -0.15$ eV, then excellent agreement with experimental results would be obtained in that the theoretical total energy distribution would then have a peak at about the same energy as that observed by Swanson and Crouser.[10] Since we do not really know what $\epsilon_H$ is, however, and because of the approximations mentioned above, this result must be treated with reservation. Also, the accuracy of this procedure for estimating $\epsilon_H$ depends on the approximations mentioned above.

The modifications suggested by Gadzuk and improvements in our band-structure model might result in better agreement with the experimental results. However, in view of the doubtful accuracy of the WKB approximation as applied to this problem and the validity of separating the energy of an incident electron into transverse and normal part for any but a parabolic energy band, a different approach would seem to be more promising, e.g., the formulation of the problem in terms of scattering theory or more exact treatment of tunneling from periodic structures in a WKB approximation.[26]

## ACKNOWLEDGMENTS

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# Fourier-Series Representation of the Pt Fermi Surface*

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The Fourier-series representation is shown to give an accurate representation of the band structure of transition metals in a restricted energy interval, e.g., near the Fermi energy. A 19-term fit to the fifth and sixth bands in fcc Pt is used to compute the de Haas–van Alphen areas and effective masses of all orbits expected for the Fermi surfaces associated with these bands.

## INTRODUCTION

MANY methods may be used to calculate energy bands in metals through the entire Brillouin zone and over all energies of interest. The interpretation of various experimental quantities in terms of these energy bands is complicated. Most quantities that can be determined experimentally are averages over some region of momentum space and/or energy. If such averages are found from quantities derived directly from band calculations, much computer time is expended. Even with the comparatively fast interpolation techniques[1] the process requires considerable computational time.

For many properties, e.g., transport phenomena, all that is required is a knowledge of the band structure (momenta and velocities) in the immediate vicinity of the Fermi surface. Even in such a restricted energy range, the time required to derive momenta and velocities ($\nabla_k E$) from the band-structure calculations is substantial. Therefore, a parametrization scheme is required capable of giving a faithful representation of the constant-energy surfaces near the Fermi energy using relatively little computational time. Moreover, experimental momenta and velocities are modified by many-body effects. A properly constructed scheme should have sufficient generality to include such effects.

Such a representation is particularly useful in converting the extremal areas and effective masses measured in de Haas–van Alphen experiments into Fermi momenta (radii) and Fermi velocities. The inverse process of comparing the results of band-structure calculations with experiment is also of interest. For closed, single-valued surfaces possessing inversion symmetry, the problem of converting areas and masses into radii and velocities has been solved by the present authors.[2–4] The problem has also been treated by Foldy.[5] The techniques make use of a series expansion of a theorem due to Lifshitz and Pogorelov.[6] The terms in the series are appropriate combinations of spherical harmonics such that the point-group symmetry of the surface is maintained in each order of the expansion.

One of the surfaces to be discussed here is the open fifth-band hole surface of Pt. This surface, because it is open, does not satisfy the conditions for application of the Lifshitz-Pogorelov theorem. The proper representa-

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