[29]This is in accordance with the results given in Sec. IV regarding the role of the spin Zeeman effect.

[30]This expression is exactly the same as expression (25) when replacing $\tilde{\theta}$ by $(V_c^{(1)}\epsilon_{xz}/\Delta)\cos 2\theta_m$.

[31]A more complete but semiclassical study of this ion has been given in a previous paper (Ref. 13).

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Theoretical and Experimental Investigation of Photoemission in the Region of Periodic Schottky Deviations

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The existence of deviation from the photoelectric Schottky line has been investigated for metals theoretically, and the experimental evidence for this deviation has been obtained for tungsten and molybdenum. A theoretical equation is derived based on a modified model which contains a classical image force and exchange and correlation forces. The theoretical solution indicates that the amplitude is inversely proportional to the frequency of the light source, whereas the phase or the period is found to be independent of the frequency of the light source. The experiments involved measuring the photoelectric emission current of tungsten and molybdenum as a function of electric field. Results were obtained for two different frequencies of light source for each sample. The periodic deviation from the Schottky line was observed clearly from a number of runs. A comparison of the experimental results with the theoretical prediction has been made. The agreement in the amplitude and the phase between them is very good.

I. INTRODUCTION

The periodic deviations from the Schottky line in thermionic emission were explained[1-6] as due to a periodic dependency of the transmission coefficient upon the intensity of the applied field. This periodic behavior of the transmission coefficient was interpreted as the result of interference of the electron waves reflected from the potential barrier at the surface of the metal. Since the same potential barrier is used in the theory of the surface photoelectric effect, and since the transmission coefficient enters the expression for the photocurrent in the same way, a periodic dependence of the photocurrent on the applied field, similar to the dependence of the thermionic current, is to be expected. Guth and Mullin[7] and Juenker[4] derived the periodic terms for photocurrent based on the potential barriers used for their thermionic-emission studies. A comparison between their results and the experimental data has not been available. However, since both the expressions for the thermionic periodic deviation from the Schottky line derived by Guth and Mullin[1,2] and by Juenker[4] fail to agree with the thermionic experimental results[8-14] the accuracy of their results on photoelectric emission presented a great suspicion which initiated the present work.

A modified potential barrier, which has been used to derive the periodic deviation from the Schottky line for thermionic emission,[15] and, which was found to be in excellent agreement with the experimental data, is used in this paper to derive the periodic deviation from the Schottky line for photoelectric emission. In view of the result of the theoretical derivation, it is found that the effect should be most easily observable for frequencies very near the threshold, for then the fraction of the current due to the periodic term has its largest value.

An experimental setup for photoelectric emission was built, since experimental data on photoelectric periodic deviations are not yet available in the quantity[16,17] which makes possible analysis of the counter part thermionic results. The agreement between the present theory and the present experimental results is very good.

II. PHOTOELECTRIC EMISSION FROM METALS

A periodic dependence of photoelectric emission on an applied field is expected to be from two parts: (i) for electrons with energies W and absorbing a photon $h\nu$ such that (for notation, see Ref. 15)

$$W + h\nu \geq V^{0\prime} ,$$

where

$$V^{0\prime} = V^0 - \frac{1}{x_0 + 1/2W_a} = \text{maximum}$$

potential barrier height at field E;

(ii) for electrons with energies W, tunneling through the top of the barrier after absorbing a quantum $h\nu$, i.e.,

$$V^{0\prime} - \delta \leq W + h\nu < V^{0\prime} ,$$

where δ is an infinitesimal energy value. For part (i) $[W + h\nu \geq V^{0\prime}]$, the photocurrent is

$$i \sim \int_0^\infty N(\epsilon) P(\epsilon, \nu) D(E, \epsilon)\, d\epsilon \quad , \tag{1}$$

where

$$\epsilon = W - V^{0\prime} + h\nu = W - W_a + \frac{1}{x_0 + 1/2W_a} + h\nu + 4b W_a^2 \quad ,$$

$N(\epsilon)$ = Fermi distribution which has been integrated over the two components parallel to the surface

$$= \frac{4\pi mkT}{h^3} \ln_{10} (1 + \exp\{-[\epsilon + h(\nu_0' - \nu)]/kT\}), \tag{2}$$

ν_0 = threshold frequency $= \phi/h$,

$$\nu_0' = \left(\phi - \frac{1}{x_0 + 1/2W_a}\right)/h \; ;$$

ϕ is the work function, $D(E, \epsilon)$ is the transmission coefficient [obtained in Ref. (15)]. The only unknown quantity in the expression Eq. (1) for this current is the transition probability $P(\epsilon, \nu)$. Mitchell[18] has shown that near the threshold the transition probability is independent of the electron energy. Hence

$$i \propto (4\pi mkT/h^3) \int_0^\infty D(E, \epsilon) \ln 1 + \exp\{-[\epsilon + h(\nu_0' - \nu)]/kT\}\, d\epsilon \quad . \tag{3}$$

The energy average transmission coefficient is defined as

$$\bar{D}(E) = \frac{1}{kT} \int_0^\infty D(E, \epsilon) \times \ln(1 + \exp\{-[\epsilon + h(\nu_0' - \nu)]/kT\})\, d\epsilon \quad . \tag{4}$$

By using $D(E, \epsilon)$ of Ref. (15), the integration can be carried through, leading to a complicated expression.

Since we are interested in the low-temperature range, i.e., $1/kT \gg 0$, if we use $\nu = \nu_0$ (where v_0 is the threshold frequency at zero field), the condition

$$\exp[-(h\nu - h\nu_0')/kT] \ll 1$$

is fulfilled for all values of the field above some relatively small values (since ν_0' decreases as E is increased). For large values of E, the condition may be fulfilled with ν chosen even less than ν_0. So with the field in which we are interested, i.e.,

$$(x_0 + 1/2W_a)^{-1} \ll 1 \; ,$$

to a very good approximation

$$\bar{D}(E) = \frac{[h(\nu - \nu_0')]^2}{2(kT)^2} \left\{ 1 - \frac{W_a^{1/2}}{2} \frac{(1 - 8bW_a)}{(1 - 4bW_a)^{3/2}} \frac{2}{h(\nu - \nu_0')} \times \frac{\cos\{A - \tan^{-1}[(\gamma + 2\ln 2)/\pi]\}}{[\pi^2 + (\gamma + 2\ln 2)^2]^{1/2} [\frac{1}{2}(x_0 + 1/2W_a)^3]^{1/2}} \right\}$$

$$= \frac{[h(\nu - \nu_0')]^2}{2(kT)^2} (1 + \Phi_2') \quad . \tag{5}$$

So the periodic deviation from the Schottky line is

$$\Phi_2' = \ln\bar{D}(E) - \ln\bar{D}(0) = -\frac{W_a^{1/2}}{h(\nu - \nu_0')} \frac{(1 - 8bW_a)}{(1 - 4bW_a)^{3/2}} \times \frac{\cos(A - 0.56)}{[\pi^2 + (\gamma + 2\ln 2)^2]^{1/2} [\frac{1}{2}(x_0 + 1/2W_a)^3]^{1/2}} \; , \tag{6}$$

where 0.56 in $\cos(A - 0.56)$ is due to

$$\tan^{-1}[(\gamma + 2\ln 2)/\pi] = 0.56 \quad .$$

For part (ii) $(V^{0\prime} - \delta < W + h\nu < V^{0\prime})$, the transmission coefficient can be obtained by replacing the excess of emission ϵ in $D(E, \epsilon)$ by

$$\epsilon' = -\epsilon = V^{0\prime} - (W + h\nu) \quad ,$$

which gives

$$D'(E, \epsilon') = \frac{\exp\{-2\pi[\frac{1}{2}(x_0 + 1/2W_a)^3]^{1/2}\epsilon'\}}{1 + \exp\{-2\pi[\frac{1}{2}(x_0 + 1/2W_a)^3]^{1/2}\epsilon'\}} - \frac{W_a^{1/2}}{2} \frac{(1 - 8bW_a)}{(1 - 4bW_a)^{3/2}} \times \frac{\exp(-2\pi\{\frac{1}{2}[x_0 + 1/2W_a]^3\}^{1/2}\epsilon')}{[1 + \exp(-2\pi\{\frac{1}{2}[x_0 + 1/2W_a]^3\}^{1/2}\epsilon')]^{3/2}} \times \cos[A + (\gamma + 2\ln 2)\{\tfrac{1}{2}[x_0 + 1/2W_a]^3\}^{1/2}\epsilon'] \quad . \tag{7}$$

We now follow the same procedure as in part (i): Since T is very low, i.e.,

$$h(\nu - \nu_0')/kT \gg 0 \; ,$$

$$\exp[-h(\nu - \nu_0')/kT] \ll 1 \; ,$$

and since $x_0 + 1/2W_a \gg 1$, $\bar{D}'(E)$ becomes approximately

$$\bar{D}'(E) = -\frac{h(\nu - \nu_0')}{(kT)^2} \frac{W_a^{1/2}}{2} \frac{(1 - 8bW_a)}{(1 - 4bW_a)^{3/2}} \times \frac{\cos\{A + \tan^{-1}[(\gamma + 2\ln 2)/2\pi]\}}{[4\pi^2 + (\gamma + 2\ln 2)^2]^{1/2} [\frac{1}{2}(N_0 + 1/2W_a)^3]^{1/2}} \; , \tag{8}$$

so the periodic deviation from the Schottky line due to this tunneling contribution is

$$\Phi_2'' = -[W_a^{1/2}/h(\nu-\nu_0')][(1-8bW_a)/(1-4bW_a)^{3/2}] \times \frac{\cos(A+0.302)}{[4\pi^2+(\gamma+2\ln 2)^2]^{1/2}[\frac{1}{2}(x_0+1/2W_a)^3]^{1/2}}, \quad (9)$$

where 0.302 in $\cos(A+0.302)$ is due to

$$\tan^{-1}[(\gamma+2\ln 2)/2\pi] = 0.302.$$

Combining Eqs. (6) and (9), the total periodic deviation from the Schottky line is

$$\Phi_2 = \Phi_2' + \Phi_2'' = -\frac{W_a^{1/2}}{h(\nu-\nu_0')}\frac{(1-8bW_a)}{(1-4bW_a)^{3/2}}[\pi^2+(\gamma+2\ln 2)^2]^{-1/2} \times \{\tfrac{1}{2}[x_0+1/2W_a]^3\}^{-1/2}[\cos(A-0.56) + 0.5623\cos(A+0.302)]. \quad (10)$$

Substituting

$$[x_0+1/2W_a]^{1/2} \approx (3.587\times 10^4/E^{1/2})^{1/2}$$

into Eq. (10) and with some numerical calculation, we have

$$\Phi_2 = -[0.056\times 10^{-6}W_a^{1/2}(1-8bW_a)/h(\nu-\nu_0') \times(1-4bW_a)^{3/2}]E^{3/4}[\cos(A-0.56) + 0.5623\cos(A+0.302)]. \quad (11)$$

Equation (11) shows that the periodic deviation from the Schottky line Φ_2 is a function of applied electric field E, potential barrier height W_a, the coefficients of the exchange and correlation energies b and $h(\nu-\nu_0')$.

It is clear that A in Eq. (20) of Ref. 15 does not involve $h(\nu-\nu_0')$, therefore, the phase of periodic deviation from the Schottky line is independent of the frequency of the light source. However, A is still a function of E, W_a, and b.

III. EXPERIMENTAL PROCEDURE

There is no significant experimental result available for the photoelectric periodic Schottky deviation at present.[16,17] Therefore, an experimental setup has been built to measure the photoelectric emission from the single crystals of tungsten and molybdenum.

Samples of $\frac{1}{4}$ in. diameter, $\frac{1}{2}$ in. in length were edged in a ⟨111⟩ direction and then electropolished by the chemical solution. The samples were mounted on a copper block which was silver soldered to the Ultek 80335 dual liquid-nitrogen feedthrough so that the temperature of the samples could be maintained at the low temperature. In order to insulate the sample from ground (copper

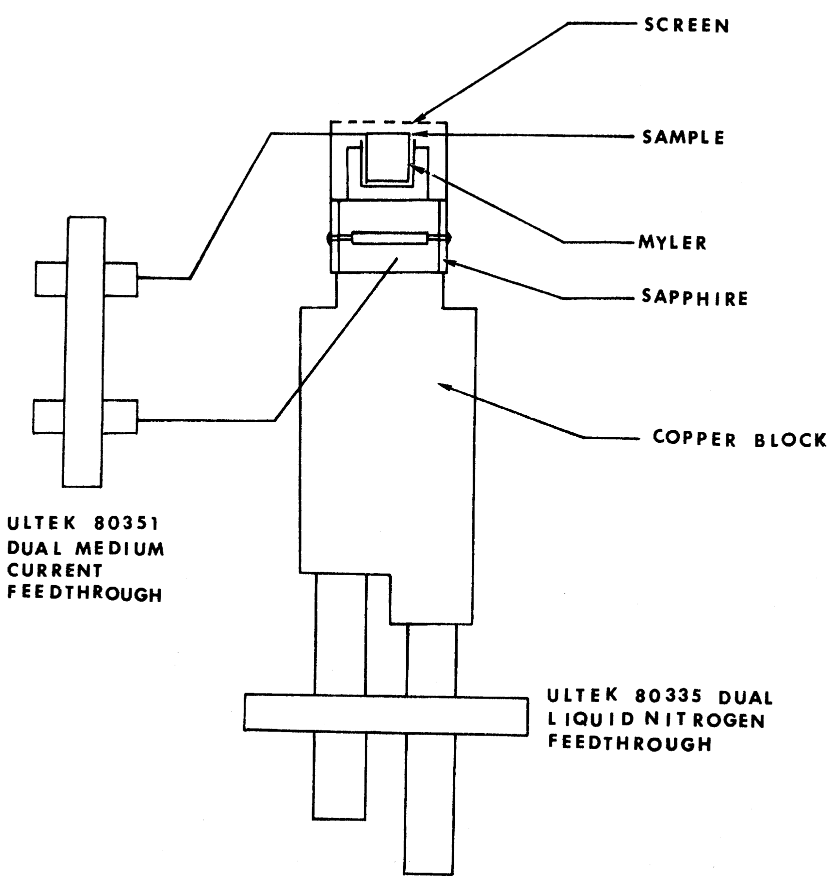

FIG. 1. Schematic diagram of the experimental setup inside the vacuum system.

block) and to maintain the smallest temperature difference between the samples and the copper block, a mylar piece 5/1000 in. thick was wrapped outside the sample before it was mounted to the copper block.

A screen used to collect the emission electrons from the samples was mounted to the same copper block but was insulated from it by four pieces of sapphire. The sample was connected to one end of the Ultek 80351 dual medium current feedthrough by a copper wire for emitted current; the other end of this current feedthrough was connected to the screen for applying the high voltage across the sample. The whole system was put into the General Electric vacuum system with absorption pump and 25-L/S triode ion pump, and the pressure inside the system was maintained at ultrahigh vacuum of about 10^{-9} to 10^{-10} Torr (Fig. 1).

A light source of Oriel Deuterium lamp with Suprasil window (50 W), lined up with a Bausch and Lomb monochromator, was focused on the sample surface. The photocurrent measurements were made by varying the applied electric field. A Keithley 640 electrometer was used to measure the photocurrent, while a Fluke 415 A power supply was used for the high electric field. The schematic diagram of the experimental setup is shown in Fig. 2.

The Ultek 80351 dual medium current feedthrough can stand the high voltage up to 12 kV; since the distance between the sample surface and the screen was set to be 5/1000 in., the electric field on the sample was able to be applied to as high as 945 kV/cm.

In order to eliminate the possible intensity fluctuation of the light source, the time constant was required to be small, in other words, the shunt resistance should be decreased; then the current for a stable reading should be increased; this was accomplished by removing our entrance slit of the monochromator. The emission current so obtained was in the order of 10^{-12} A. The corresponding shunt resistance used was 10^{10} Ω. With this higher current and lower resistance, the time constant required for each run was about 30 to 40 min.

Therefore, with all equipment well calibrated, the maximum error in emission current determinations was estimated at 0.1%.

However, the emission current was still very small and any electrical or mechanical noise could cause an instability of the reading of current. In order to reduce the possible noise to the smallest amount, a large box covered with heavy duty aluminum foil was built to contain the vacuum system, light source, and the shunt resistance of the 640 Keithley electrometer.

IV. RESULTS AND DISCUSSION

A direct comparison between Eq. (11) and Juenker's results[4] is made as follows:

Both results are similar in form and show that the amplitude of the periodicity is inversely proportional to $h(\nu - \nu_0')$; they also indicate that the phase and the period of thermionic and photoelectric deviations are identical. However, because of the different potential model employed in the derivation, the two predicted results are different in amplitude and in phase. Our equation will be substantiated by the experimental results.

It has been mentioned in Sec. III that in order to have stable current, the entrance slit is removed and the exit slit chosen to be 3 mm, so the bandpass of the monochromator is large. On the other hand, in order to have a high percentage of the periodic deviation from the Schottky line, $\lambda = 2850$ and 2900 Å are chosen on the monochromator for tungsten, whereas $\lambda = 2950$ and 3000 Å are used for molybdenum.

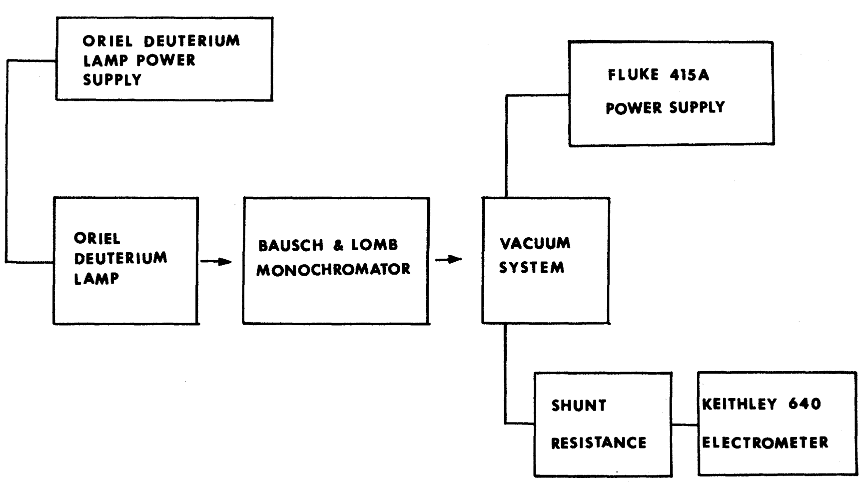

FIG. 2. Schematic diagram of the electric circuit for the experiment.

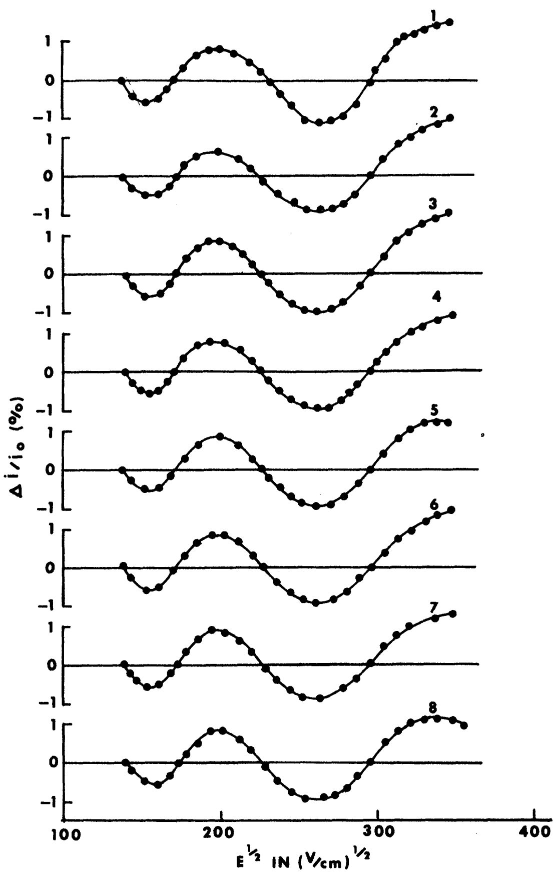

FIG. 3. Experimental results of the periodic Schottky deviations for tungsten. Data were obtained by using the mean wavelength $\lambda = 2650$ Å.

A. Tungsten

(i) At $\lambda = 2850$ Å, the emission current is of the order of 10^{-12} A. It is clearly found that the experimental data deviate in a regular manner from a straight line. In order to show this fluctuation more clearly than is possible on a curve of i against $E^{1/2}$, the deviation of the experimental data from a reference line was plotted against $E^{1/2}$. The reference line was chosen arbitrarily since the deviation is regular and periodic.

Eight sets of the experimental results for this frequency are reduced in Fig. 3 for the range of 140 to 350 $(V/cm)^{1/2}$. Average values of these eight curves are shown in Fig. 4.

The band-structure calculation has been reported by Mattheiss.[19] Among the various bands which intersect the Fermi surface, the minimum occurs at 0.73 Ry while the Fermi level is at 0.85 Ry. This difference (0.12 Ry) when added to the work function of tungsten, which 4.5 eV = 0.33 Ry, gives us 0.45 Ry for W_a. The value of b was chosen to be $3/64W_a$ which was obtained by matching the theoretical expression and the experimental data for thermionic periodic Schottky deviation.[15] With these values of W_a and b, the theoretical periodic Schottky deviation is calculated from Eq. (11) for a chosen frequency.

Since the threshold frequency of tungsten is 2760 Å, it is impossible to obtain the emission current with wavelength $\lambda = 2850$ Å as shown on the monochromator. However, because of the removal of the entrance slit and the use of a wide exit slit, the bandpass of the monochromator is about 500 Å, for instance, with $\lambda = 2850$ Å on the monochromator reading, the light with the wavelength from 2500 to 3000 Å would more or less pass through the monochromator (Fig. 5). Since the threshold wavelength is 2760 Å, the actual bandpass which effects the emission of the tungsten surface is from 2500 to 2760 Å, i.e., 260 Å. Investigating the spectrum of this bandpass, one finds that $\lambda = 2650$ Å is about the mean value. Therefore this wavelength will be chosen for our theoretical calculations.

Since

$$\Phi_2 = \ln\bar{D}(E) - \ln\bar{D}(0) = \ln i - \ln i_0,$$

$\Delta i/i_0 \approx \Phi_2$, the theoretical calculations for $\Delta i/i_0$ can be obtained from Eq. (11).

Using $W_a = 0.45$ Ry, $b = 3/64W_a$, and $\lambda = 2650$Å in Eq. (11), the theoretical prediction for $\Delta i/i_0$ is obtained and plotted in Fig. 4 for comparison. The good agreement in amplitude and in phase between theoretical and experimental results is indicated clearly in this figure.

(ii) A similar result is obtained also for $\lambda = 2900$ Å. Figure 6 shows the set of six curves. Their average is shown in Fig. 7.

Theoretical calculations for $\Delta i/i_0$ based on Eq. (11) by using $W_a = 0.45$ Ry, $b = 3/64W_a$, and $\lambda = 2660$ Å is shown in Fig. 7. Again, the agreement is very good.

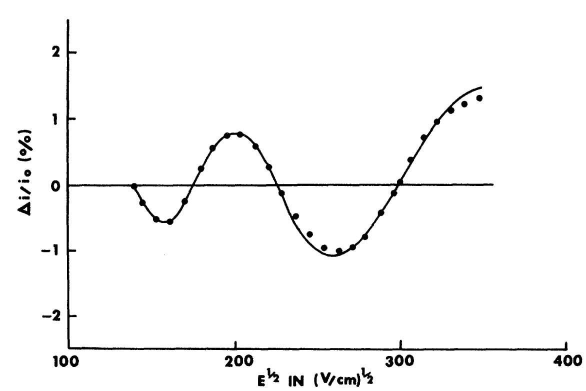

FIG. 4. Periodic Schottky deviations for tungsten. Solid curve is the theoretical prediction at $\lambda = 2650$ Å and the solid points are the average values of Fig. 3.

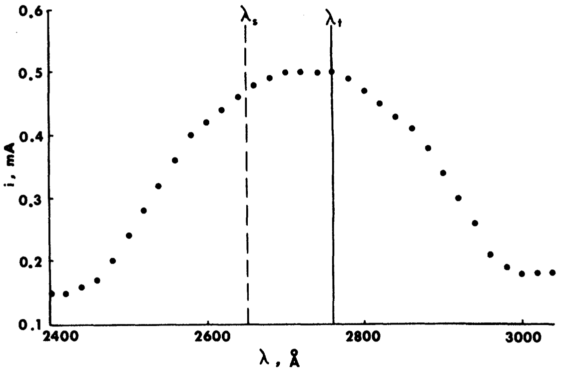

FIG. 5. Measured spectrum for the monochromator at λ = 2850 Å. Mean value of the wavelength is chosen to be 2650 Å.

The positions of maxima, minima, and zeros of the periodicities for both theoretical and experimental results are summarized in Table I.

B. Molybdenum

(i) As we set λ = 2950 Å on the monochromator, the emission current is of the order of 10^{-12} A. The periodicities occur clearly in the range of 140 to 350 $(\mathrm{V/cm})^{1/2}$. Figure 8 shows the deviation of the experimental data from the reference

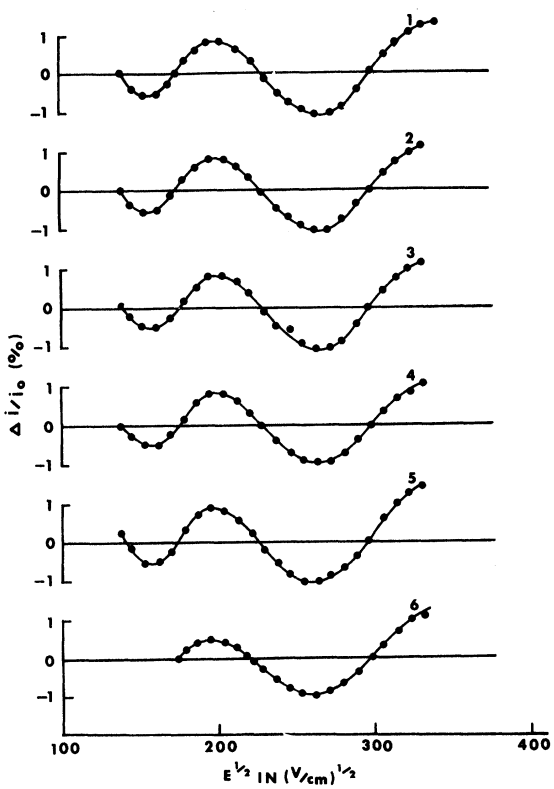

FIG. 6. Experimental results of the periodic Schottky deviations for tungsten. Data were obtained by using the mean wavelength λ = 2660 Å.

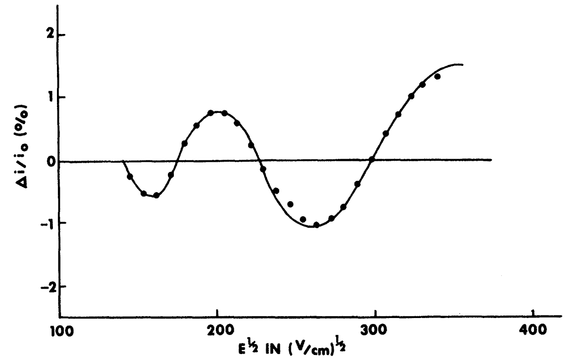

FIG. 7. Periodic Schottky deviations for tungsten. Solid curve is the theoretical prediction at λ = 2660 Å; solid points are the average values of Fig. 6.

line versus $E^{1/2}$. The set of eight curves in Fig. 8 is essentially the same in phase and in amplitude. Figure 9 is obtained by taking the average of these eight curves.

The band structure of molybdenum is not available at the present time. An alternate way to determine W_a is to match the theoretical and the experimental results of the thermionic periodic

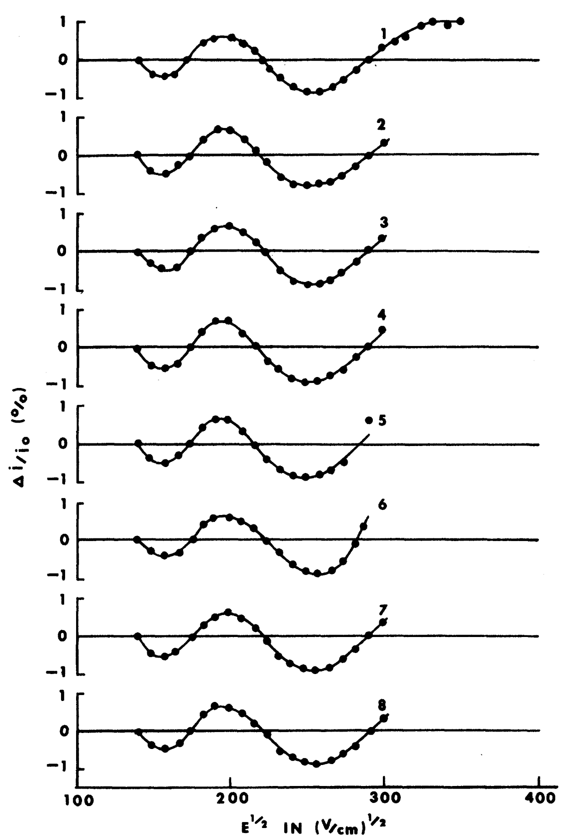

FIG. 8. Experimental results of the periodic Schottky deviations for molybdenum. Data were obtained by using the mean wavelength λ = 2780 Å.

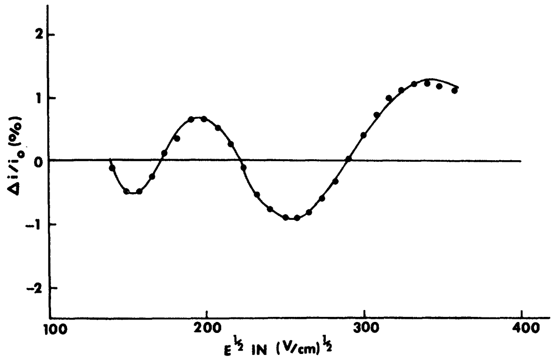

FIG. 9. Periodic Schottky deviations for molybdenum. Solid curve is the theoretical prediction at $\lambda = 2780$ Å; solid points are the average values of Fig. 8.

Schottky deviation which has been done by the authors[15]; the result is $W_a = 5.44$ eV. At the same time, b is obtained to be $1/16W_a$.[15]

The theoretical calculation for $\Delta i/i_0$ is obtained from Eq. (11) by employing $W_a = 5.44$ eV $= 0.4$ Ry, $b = 1/16W_a$, and $\lambda = 2780$ Å (the wavelength λ is chosen as the mean value from the spectrum). The calculated result is plotted in Fig. 9 for comparison. Good agreement is found from this comparison.

(ii) The deviation of the experimental data from the reference line versus $E^{1/2}$ for $\lambda = 3000$ Å is shown in Fig. 10. At this frequency, similar to the case of $\lambda = 2950$ Å, the phase and the amplitude of these nine curves are almost the same. The mean value of these curves is shown in Fig. 11. The theoretical prediction for $\Delta i/i_0$ from Eq. (11) by using $W_a = 0.4$ Ry, $b = 1/16W_a$, and $\lambda = 2800$ Å is included in Fig. 11. The degree of agreement is excellent.

Table II summarizes some characteristics of

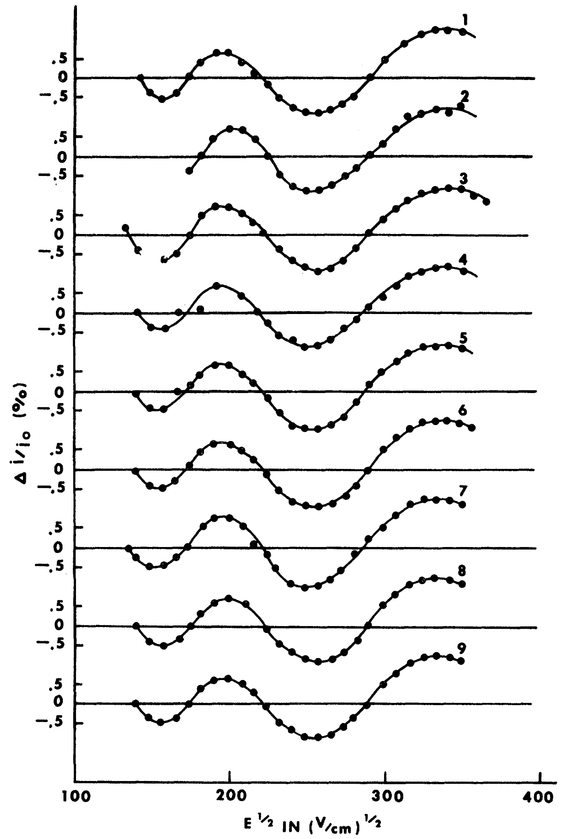

FIG. 10. Experimental results of the periodic Schottky deviations for molybdenum; data were obtained by using the mean wavelength $\lambda = 2800$ Å.

the periodicities for both theoretical and experimental results.

It is useful to show the effect of b on the amplitude and the phase of the periodicities. Equation (20) (of Ref. 15) shows that the phase and period

TABLE I. Calculated and observed positions of the maxima, minima, and zeros of the periodic Schottky deviations in terms of $E^{1/2}$ for tungsten.

	Zero 1	Min 1	Zero 2	Max 1	Zero 3	Min 2	Zero 4
Theoretical	141	160	175	200	225	260	300
Experimental I ($\bar{\lambda} = 2650$ Å)	140	158	175	200	226	264	298
Experimental II ($\bar{\lambda} = 2660$ Å)	140	158	174	200	227	264	298

TABLE II. Calculated and observed positions of the maxima, minima, and zeros of the periodic Schottky deviations in terms of $E^{1/2}$ for molybdenum.

	Zero 1	Min 1	Zero 2	Max 1	Zero 3	Min 2	Zero 4	Max 2
Theoretical	140	155	171	195	222	255	290	340
Experimental I ($\bar{\lambda} = 2780$ Å)	140	156	170	194	220	254	290	
Experimental II ($\bar{\lambda} = 2800$ Å)	140	154	170	194	221	256	290	336

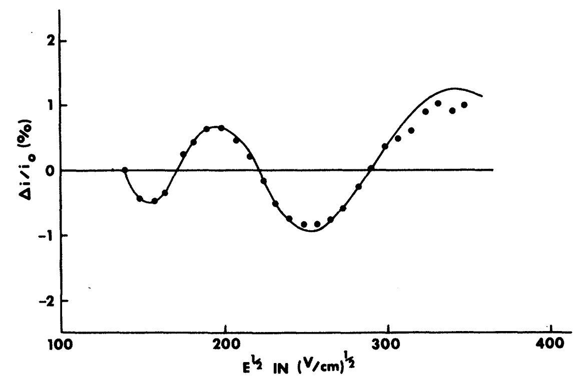

FIG. 11. Periodic Schottky deviations for molybdenum; solid curve is the theoretical prediction at $\lambda = 2800$ Å; solid points are the average values of Fig. 10.

do not depend too much on b. On the other hand, from Eq. (11), it is clear that the amplitude depends sensibly on b. Physically, the amplitude of the periodicity depends upon the shape of the potential barrier near the surface of the metal; in this region, the exchange and correlation forces play the important role. To illustrate this we may select $b=0$ in Eq. (11) for Φ_2; the result shows there is about 15% deviation from the experimental data in amplitude. This deviation can not be compensated for by adjusting the value of W_a because the phase of the periodicity is largely determined by the value of W_a.[15]

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