

## Anomaly of the thermal-field emission and total-energy distribution of the (012), (013), and (023) tungsten faces

J. K. Wysocki

*The Institute of Experimental Physics, University of Wrocław, Cybulskiego 36,  
50-205 Wrocław, Poland*

(Received 16 September 1981; revised manuscript received 25 October 1982)

The thermal-field-emission (TFE) current from the (012) tungsten plane was measured within a field-electron microscope with the compensated measuring setup. The current decreases with a temperature increase starting at 78 K, which is caused by the work-function increase. At still higher temperatures the current reaches its minimum in the (300–600)-K range, and further increases as “expected” because its thermal tail dominates. This result was confirmed by the measurement of total-energy distributions (TED). The work-function temperature coefficient for the (012) plane was calculated using the experimental data and the Murphy-Good equation for TFE and the Young equation for TED. A value of  $1.9(+0.6, -0.8) \times 10^{-4}$  eV/K was found. The properties of the (013) and (023) crystal planes are very similar to those of the (012) plane.

### I. INTRODUCTION

In 1969 we reported on an appearance of a minimum in the thermal-field-emission (TFE) current versus temperature dependence for some tungsten crystal planes.<sup>1,2</sup> This anomaly is most significant for the (012) plane. This observation is in partial agreement with the later measurements of field-electron energy distribution (FEED) for the W(012) plane performed by Czyżewski.<sup>3,4</sup> Our measurements were performed in a probe-hole field-emission microscope (FEM). Instead of the magnetic deflection used in previous works,<sup>1,2</sup> in the present study the rotation of the tip was employed to direct emitted electrons in the collector. It was shown<sup>5</sup> that the TFE characteristics measured from the W(111) plane fitted well with the Murphy-Good (MG) theory,<sup>6</sup> based on the free-electron model. Contrary to the tradition, the W(012) plane was not used as a standard for the testing of an electron energy analyzer, because it exhibits, unlike to the W(111) plane, the above-mentioned anomaly phenomena. Therefore the method for testing of an analyzer, based on the measurements of TFE and FEED from the W(111) plane for various temperatures was elaborated.<sup>7</sup> The aim of the present work is to find out the reason of TFE and FEED anomaly occurring on the W(012).

### II. THEORY

#### A. Basic equations

Our further considerations are based on following three equations describing the current densities (in

units of A/cm<sup>2</sup>): (i)  $j_{\text{FN}}$  for field emission according to the Fowler-Nordheim (FN) theory,<sup>8,9</sup> (ii)  $j(T)$  for the TFE according to the MG theory,<sup>6</sup> and (iii)  $j'(\epsilon)$ , for the FEED according to Young.<sup>10</sup> The currents are given by

$$j_{\text{FN}} = (4\pi me/h^3)d^2 e^{-c} \quad , \quad (1)$$

$$j(T) = j_{\text{FN}} [\pi p / \sin(\pi p)] \quad , \quad (2)$$

$$j'(\epsilon) = (j_{\text{FN}} e^{\epsilon/d}/d)(1 + e^{\epsilon/kT})^{-1} \quad , \quad (3)$$

where

$$p = kT/d \quad , \quad (4)$$

$$d = \hbar e F [2\sqrt{2m} \varphi^{1/2} t(y)]^{-1} \\ = 9.758 \times 10^{-9} F [\varphi^{1/2} t(y)]^{-1} \quad , \quad (5)$$

$$c = 4\sqrt{2m} \varphi^{3/2} v(y) / 3\hbar e F \\ = 6.832 \times 10^7 \varphi^{3/2} v(y) F^{-1} \quad , \quad (6)$$

$$\epsilon = E - E_F \quad .$$

The  $t(y)$  and  $v(y)$  functions are tabulated<sup>11</sup> where

$$y = e^{3/2} F^{1/2} \varphi^{-1} = 3.795 F^{1/2} \varphi^{-1} \quad . \quad (7)$$

Here  $h$  and  $k$  are Planck's and Boltzmann's constants, respectively,  $F$  (V/cm), the electric field strength,  $T$  (K), the temperature,  $\varphi$  (eV), the work function,  $E$ , the electron energy corresponding to its momentum perpendicular to the emitting surface,  $E_F$ , the Fermi energy, and  $m$  and  $e$ , the mass and charge of an electron.

The FN equation in coordinates  $\log_{10}(i_{\text{FN}}/U^2)$  vs  $U^{-1}$  yields a straight line with a slope

$$m_{\text{FN}} = 2.967 \times 10^7 \varphi^{3/2} s(y) \beta^{-1}, \quad (8)$$

where  $s(y)$  is a function also tabulated,<sup>11</sup> and  $i_{\text{FN}}$  is a current, while  $\beta = F/U \text{ cm}^{-1}$  is a geometrical factor, and  $U$  denotes a tip-anode voltage. The MG equation for low temperatures may be approximated by<sup>5</sup>

$$j_1(T) \cong j_{\text{FN}} \exp[(\pi p)^2/6] \quad \text{for } 0 < p < 0.43 \quad (9)$$

for higher temperatures, when  $0.43 < p < \sim 1.5$ , the TFE current is given approximately by<sup>12</sup>

$$j_2(T) \cong 1.161 j_{\text{FN}} \exp[7.967 \times 10^{-2} (\pi p)^3]. \quad (10)$$

When the TFE current or FEED current as a function of temperature does not behave in accordance with the theory, one may conclude from Eqs. (1)–(7) that the only possible explanation of this effect is the temperature dependence of  $F$  and  $\varphi$ . The temperature dependence of the field strength may be caused either by (i) the change in the tip size, or (ii) by the surface-structure transition (SST) (both above effects are reversible), or, finally (iii) by the irreversible migration of surface atoms. (i) We have presented earlier<sup>5</sup> a method of the compensation of the temperature dilatibility of the tip  $[\Delta\beta(T)]$ . Compensation occurs by the appropriate change of the anode-voltage  $\Delta U$  to remain the field-strength constant. This method was used also in the present work. (ii) The SST was observed on clean (001) planes of tungsten and molybdenum,<sup>13</sup> and also by the adsorption, for example, hydrogen on the W(001).<sup>14</sup> The observations of the SST on W(001) carried out by Debe and King<sup>15</sup> by means of several methods, indicate that this phenomenon changes the work function too. Melmed *et al.*<sup>16</sup> have shown that the periodic displacements of the SST on the W(001) plane have vertical components. Their experiment was performed in the similar-to-ours temperature range of 15–460 K, and the field-evaporation method in FIM was used. The periodic preferential of surface atoms for the field is due to the vertical displacement and/or periodic variation of the binding energy. The SST phenomena for the W(012) plane were not investigated until now. If, with temperature, the surface atoms of this plane should indeed reveal the vertical reconstruction component, the presently investigated TFE anomaly would be caused by a field decreasing. Further, it must be due to the decreasing of roughness of the plane, or, in another words, due to a hiding of protruding atoms. But this interpretation is not consistent with that given by Melmed *et al.*<sup>16</sup> In addition, examples with some thermocouples, mentioned in the last chapter of this work, confirm the view that the SST

on the W(012) plane does not occur.

A temperature change of the band structure of a crystallographic direction could be measured effectively (measuring integral currents) as the corresponding work-function change. The theoretical considerations of Christensen and Feuerbacher<sup>17,18</sup> do not result in the conclusion that the band structure of the tungsten depends significantly on temperature. Cutler also shares the same opinion.<sup>19</sup>

The characteristics  $j(T)$  and  $j'(\epsilon, T)$  measured for increasing and decreasing temperature were the same in the temperature and field-strength limits used in the present experiment. This indicates that in the conditions of our experiment the appearance of an irreversible migration of surface atoms can be excluded.

#### B. Evaluation of the temperature dependence of the work function using TFE and FEED

Van Oostrom<sup>20</sup> and Swanson and Crouser<sup>21</sup> calculated the temperature dependence of the work function utilizing measured changes of FN slopes with temperature. The authors assumed that the TFE characteristics in coordinates  $\log_{10}[i(T)/U^2]$  and  $1/U$  are straight lines with slopes varying with temperature. The deviation from the theoretically predicted slope changes were supported to result from the temperature dependence of the work function. We would like to emphasize at this point that the TFE current in the above-mentioned coordinate system might be approximated with the straight line only for high fields and low temperatures. This could be easily proved using Eqs. (9) and (10). The slopes of “straight lines” can be expressed as follows: For low temperatures [Eq. (9)],

$$m_{\text{FN1}} = m_{\text{FN}} + [1.283 \times 10^8 T^2 \varphi^2 t^2(y) \beta^{-2}] U^{-1}, \quad (11)$$

and for high temperatures [Eq. (10)],

$$m_{\text{FN2}} = m_{\text{FN}} + [1.7 \times 10^{12} T^3 \varphi^{3/2} t^3(y) \beta^{-3}] U^{-2}. \quad (12)$$

It can be seen from these equations that slopes of “FN straight lines” for TFE depend on the abscissa  $1/U$ . They are not generally straight lines. The approximation is even worse for high temperatures, because the slope depends there on  $(1/U)^2$ .

The calculations of temperature-dependent changes of the work function  $\varphi(T)$ , based on the data obtained in the present work, were performed in the following way: We treat  $\varphi$  as a  $T$ -dependent function. Then we differentiate Eqs. (2) and (3) and arrive at the final formulas:

$$[\Delta\varphi(T)]_{\text{MG}} = -\delta_{\text{MG}}2\varphi/[3c + 1 + \pi p \cot(\pi p)] , \quad (13)$$

$$[\Delta\varphi(T)]_Y = -\delta_Y2\varphi/(3c + 1) , \quad (14)$$

$$\delta_{\text{MG}} = \frac{T_0 \sin(\pi p_0)}{T \sin(\pi p)} \left[ 1 + \frac{\Delta i(T)}{-i_0} \right] - 1 , \quad (15)$$

$$\delta_Y = \{ [i'(\epsilon_0, T)]_{\text{expt}} - i'(\epsilon_0, T_0) \} / i'(\epsilon_0, T_0) , \quad (16)$$

where  $\delta$  is a relative difference of the theoretical and experimental values of TFE or FEED currents, respectively. The role of indices MG and Y is to indicate that the respective parameters were calculated from MG and Young equations, respectively. Equation (15) was derived from the definition

$$\delta_{\text{MG}} = \{ [i(T)]_{\text{expt}} - i(T) \} / i(T) ,$$

where one had taken into account that

$$[i(T)]_{\text{expt}} = i_0 + \Delta i(T) ,$$

$$i_{\text{FN}} = i_0 \sin(\pi p_0) / \pi p_0 \text{ for } T_0 .$$

$[i'(\epsilon_0, T)]_{\text{expt}}$  denotes distribution current measured at the Fermi energy ( $\epsilon=0$ ) in any temperature, but lower than the inversion temperature<sup>22</sup>

$$T^* = d/kT .$$

The current  $i'(\epsilon_0, T_0)$  corresponds to the lowest temperature  $T_0$  in one series of measurements. We assume that theoretical and experimental values of the currents are equal,

$$i'(\epsilon_0, T_0) = [i'(\epsilon_0, T_0)]_{\text{expt}} .$$

When the emitter is a free-electron metal and its atomic surface structure, work function, and electron structure do not depend on temperature, the

FEED characteristics in the temperature range  $T < T^*$ , cross each other in one point at the Fermi level, and  $i'(\epsilon_0, T) = \text{const.}$  The variation of this value with temperature, we use for calculation of  $\varphi(T)$ .

### III. RESULTS

The investigations were carried out in a device constructed to measure the field-emission currents<sup>5</sup> and the energy distributions as well.<sup>7</sup> The tube was immersed in liquid nitrogen and was supplied with the titanium pump with a hot cathode proper for the pumping of hydrogen. The total pressure of residual gases after immersion of the tube into liquid nitrogen was lower than  $1.5 \times 10^{-10}$  Torr. The emission current from the clean tip was very stable. The TFE characteristics as a function of temperature were reproducible. We consider these observations as a proof of good vacuum conditions and lack of tip contaminations. The tip was cleaned before measurement of each set of characteristics with a 2-sec flash at 2200 K.

The characteristic  $i(T)$  for the W(111) plane behaves according to Eq. (2) (cf. Fig. 8 of Ref. 5). A minimum of the TFE current for the W(012) plane occurs, approximately at room temperature (Fig. 1). The TFE current was measured in the compensated circuit<sup>23</sup> to increase the accuracy. Figure 1 shows, for example, that the initial current  $i_0 = 5.3 \times 10^{-9}$  A corresponding to the lowest temperature  $T_0$ , was compensated down to the value  $i_{0c} = 2.2 \times 10^{-10}$  A, while the sensitivity of the electrometer increased 1 order of magnitude. The difference between any current value and the value  $i_{0c}$  is just the current variation  $\Delta i(T)$  in Eq. (15). Figure 2 shows the TFE characteristic taped in conditions similar to

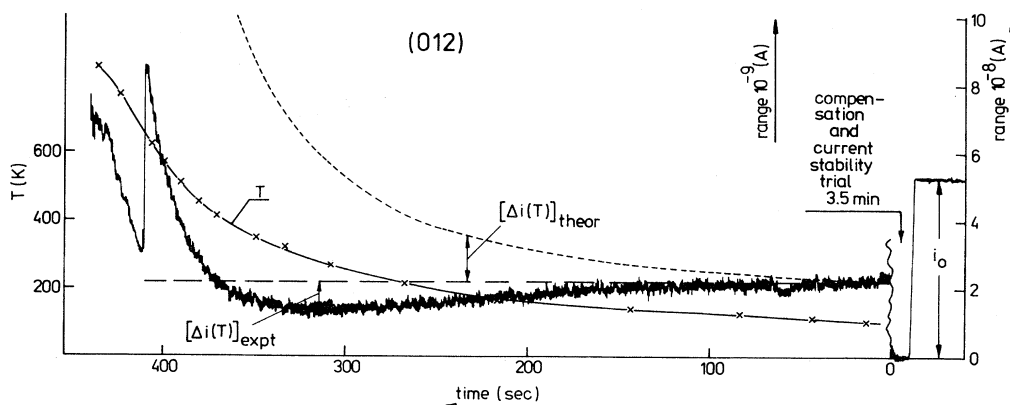


FIG. 1. Recorder tape of the TFE current increase from the W(012) plane as a function of temperature (the curve with crosses). The discontinuity of the tape at  $T = 650$  K corresponds to the change of the current range to that of  $3 \times 10^{-9}$  A. The short-dashed line is the theoretically predicted current increase according to Murphy and Good (Ref. 6). The horizontal dashed line indicates the starting compensated current.

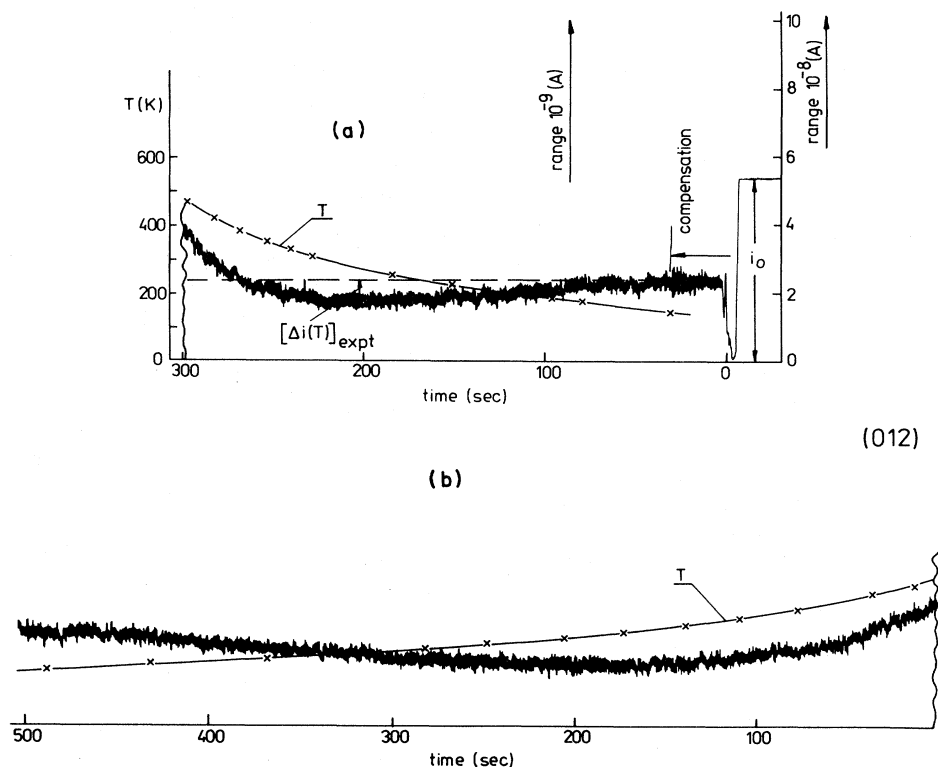


FIG. 2. Recorder tapes showing the changes of the TFE-compensated current from the W(012) plane caused by (a) a temperature increase, (b) a temperature decrease.

those of Fig. 1. The current minimum occurs both for an increase and a decrease of temperature [Figs. 2(a) and 2(b)]. Identical results were obtained for the (021) plane and qualitatively similar results for (013) and (023) planes. This phenomenon was observed on three different tungsten tips. Figure 3 presents the temperature variations of the current for the (013) plane caused by the sudden temperature changes. Just before this measurement, the tip was cleaned with flash and then cooled down to 78 K. The test of current stability was not carried out in this experiment. The loop-heating current was turned on, exactly after compensation. Within 14 sec the tip temperature reached 402 K and the emission dropped suddenly. After further 34 sec (at  $T=412$  K) the heating of the loop was turned off and the emission current went up to the initial value with an accuracy of 0.7%. It is possible to repeat this part of the experiment many times with the same result. However, for temperature 830 K the current did not reach its initial value after cooling of the tip. A reason for this is the freezing of changes in atomic order of the tip surface. In the case under consideration, the 13% decrease of the current was found. In some other cases (not quoted here), an in-

crease of the current was observed. The reason for that is the random migration of the surface atoms at higher temperatures. For temperatures lower than about 400 K, the surface (in the atomic sense) is quite stable. The support of this hypothesis is given also by other experiments. (i) Swanson<sup>24</sup> found for the W(013) plane that the rms percent fluctuations of the current reveal a maximum in the (800–900)-K temperature range. 300 K is the noise threshold. (ii) Nishigaki and Nakamura<sup>25</sup> studied surface self-diffusion on the tungsten tip using FIM. In the (78–300)-K range no surface-atom displacements were observed on the tip region lying between the  $\langle 011 \rangle$  and  $\langle 001 \rangle$  crystallographic directions.

Measurements presented in Figs. 1–3 were carried out without compensation of the temperature-induced changes in the tip size. This correction was performed for the measurements depicted in the next figures. The FEED measurements for  $\langle 012 \rangle$  and  $\langle 021 \rangle$  crystallographic directions are plotted in Fig. 4. These directions form an angle of  $36^\circ 52'$ . The similarity of both families of curves evidence that rotation of the tip does not distort the electron optics of the spectrometer. FEED's of the W(111) plane inserted in the work,<sup>7</sup> as well as FEED for the

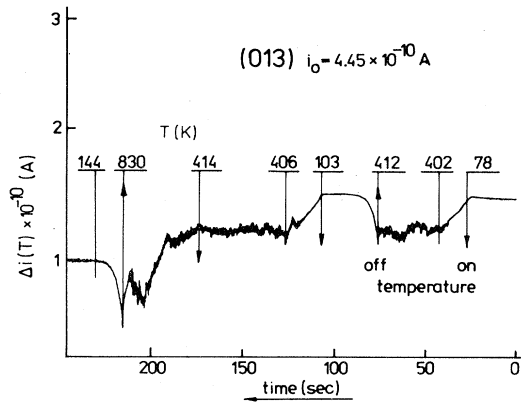


FIG. 3. Recorder tape of the TFE-compensated current variation with time from the W(013) plane, at sudden temperature changes. Arrows pointed down, or up, correspond to turnings on, or off, of the loop-heating current, respectively.

W(001) plane shown in Fig. 5 evidences that the spectrometer was tested in a proper way. Figure 5 shows the dependence of enhancement factor  $R$  on the energy. The maximum value of this factor equals 2.3 for  $\epsilon = -0.35$  eV coincides well with the other data.<sup>26,27</sup> The arguments quoted above and the way of analyzer testing presented in Ref. 7 indicate that untypical energy distributions obtained here for the (012) plane cannot be considered as resulting from apparatus.

The results concerning the dependence of the TFE current on temperature and the FEED current corresponding to the Fermi energy, are presented in Fig 6. In those measurements first the TFE current was measured for a given temperature, then the FEED characteristic was taken, and, afterwards the TFE current was measured again. With such a sequence of measurements the stability of the measuring system and the consistency of TFE and FEED could be controlled. The dependence of the work function on

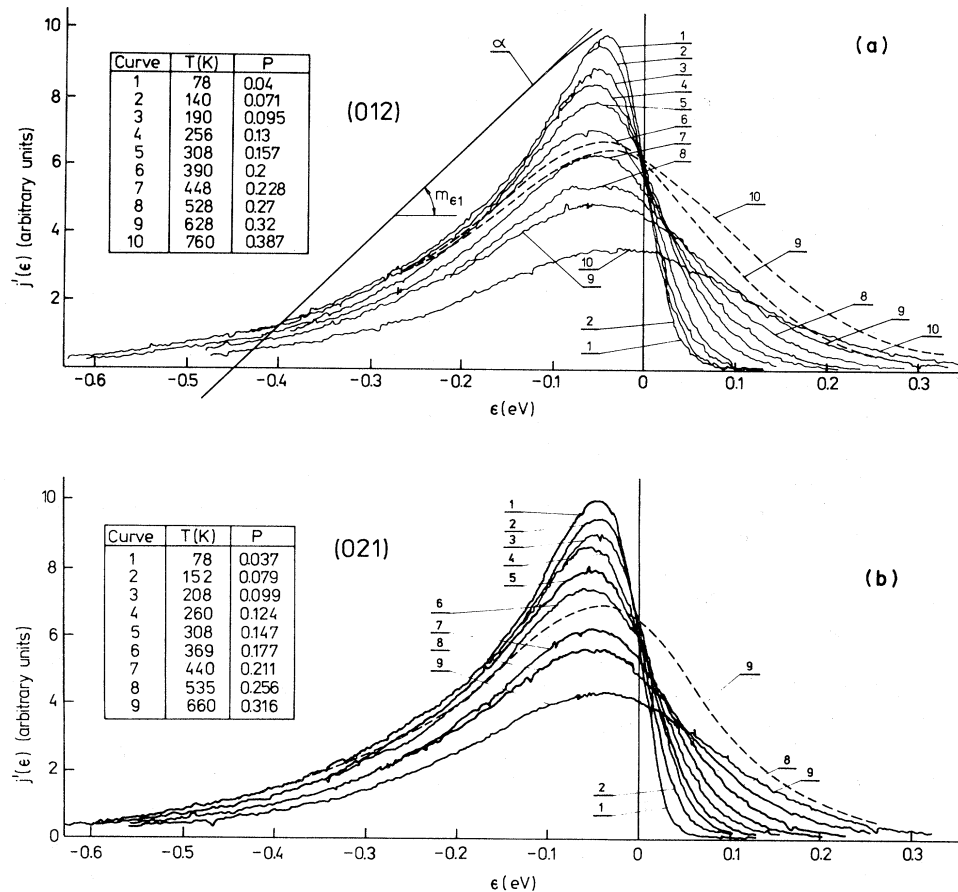


FIG. 4. Experimental and theoretical (dashed curves) total-energy distributions for (a) (012) plane, and (b) (021) plane. Straight line  $\alpha$  denotes the trailing edge of the FEED in semilogarithmic coordinates.

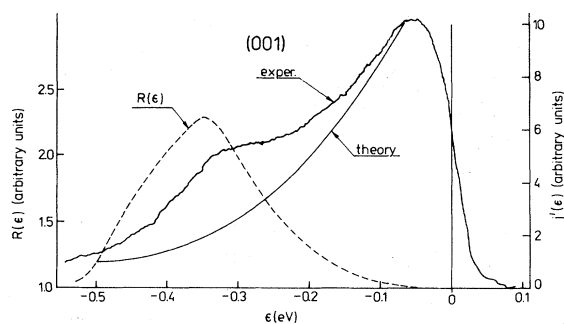


FIG. 5. Enhancement factor  $R(\epsilon)$ , experimental and theoretical total-energy distributions from W(001) at 78 K.

temperature calculated according to Eqs. (13)–(16) using the data presented in Fig. 6, is shown in Fig. 7. It can be seen in this figure that the points belonging to the one particular measurement set are not scattered. The curves corresponding to various measurement sets are visibly different. This indicates the occurrence of a systematic error. There are two possible reasons of such an error.

(i) The use of a constant value of the work function for the (012) plane equal 4.32 eV: This value of the work function was determined at the beginning of the measurement series and could change slightly during the experiment. Assuming for FEED measurements that the work function is 0.4% higher [Fig. 4(a)], and 0.3% lower [Fig. 4(b)], for these measurements shown in Fig. 7, we can obtain the  $\varphi(T)$  relation which agrees well with this relation obtained from TFE measurements.

(ii) An inaccuracy in estimation of  $i'(\epsilon_0, T_0)$  in

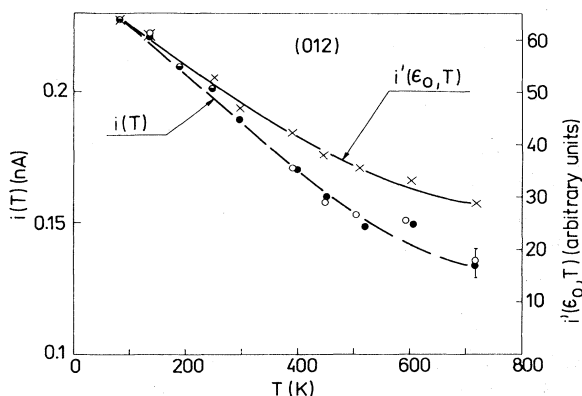


FIG. 6. Distribution current from the Fermi energy (crosses) and thermal-field emission (circles) as a function of temperature. Filled circles correspond to the TFE measurements before the FEED measurements, and open circles denote those made after the TFE measurements.

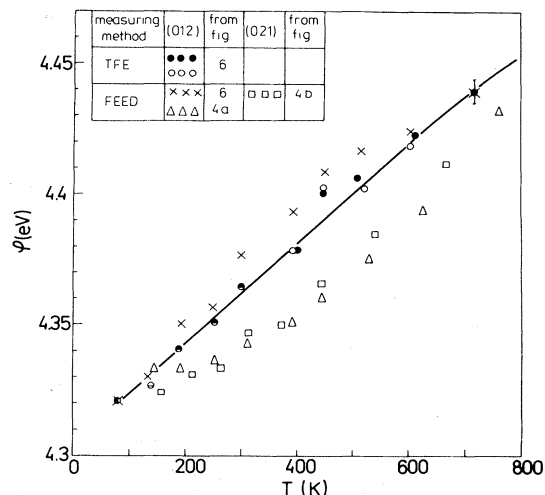


FIG. 7. Work function for the (012)-type planes as a function of temperature. Calculations are based on the experimental data (Figs. 4 and 6, according to the table inserted in this figure) and theory [Eqs. (13)–(16)], respectively.

Eq. (16) caused by considerable steepness of the distribution curve at  $\epsilon=0$ : Since in measurements of the work function  $\varphi(T)$  by means of TFE, both the initial current  $i_0$  and its temperature increment  $\Delta i(T)$  are measured with the satisfying accuracy,<sup>5</sup> this measurement was considered a most reliable one. From the plot in Fig. 7 one may calculate that  $d\varphi/dT = 1.9_{-0.8}^{+0.6} \times 10^{-4}$  eV/K.

#### IV. CONCLUSIONS

In the present work we showed that the thermal-field current and the current proportional to the energy distribution of electrons emitted for the W(012) plane are smaller than those predicted by the theory. The similar behavior is observed also for (013) and (023) planes.

A decrease of the TFE current was observed for the iridium emitter covered with hydrogen or carbon monoxide<sup>28</sup> and oxygen or ammonia.<sup>29</sup> The phenomenon was connected by the authors with the dependence of adsorbate dipole momentum on temperature. Yamamoto *et al.* observed a field-emission current decrease for a tungsten emitter caused by the adsorption of residual gases (mainly hydrogen).<sup>30</sup>

The stability of measured currents, lack of any influence of sudden temperature changes on reproducibility of measurements, lack of any difference between characteristics taken with both increasing and decreasing temperature, exclude in our opinion the possibility to relate observed phenomenon with ei-

TABLE I. Work-function temperature coefficients.

Plane	$d\varphi/dT$ (eV/K)	References
(111)	$+3.5 \times 10^{-5}$	21
(111)	$+(6 \pm 3) \times 10^{-6}$	20
(111)	0.0	5,7
(013)	$-3.2 \times 10^{-5}$	21
(012)	$+1.9^{+0.6}_{-0.8} \times 10^{-4}$	This work

ther apparatus effect or contamination. The comparison of characteristics based on our experimental data with those obtained from theoretical equations (2) and (3), leads unequivocally to the statement that the dependence of the work function on temperature is responsible for the observed anomaly. This hypothesis is also supported by the appearance of the minimum of electromotive force for specific temperatures and for some thermocouple types. For example for W-Mo (Ref. 31) and W-W at 26% Re (Ref. 32) thermocouples, the minimum of electromotive force occurs at 1000 and 200 K, respectively. Obviously surface-structure transition does not occur in thermocouples.

We have checked on the value of the computed coefficient  $d\varphi/dT$  and Eqs. (13)–(16) used to the calculation of  $d\varphi/dT$ , putting  $\varphi(T)$  back into Eqs. (2) and (3), respectively. Thus we have obtained a satisfactory agreement between calculation functions

$i[T, \varphi(T)]/i_0$  and  $i'[\epsilon_0, T, \varphi(T)]/i'_0(\epsilon_0, T_0)$ , with the measured ones, presented in Fig. 6, respectively. The similar checking of the characteristic from Fig. 1 has shown a difference between the calculated and measured values at the minimum of the characteristic of about 30%. The error increases with temperature due to the following effects. The temperature of the tip apex is lower in comparison to the measured temperature of the middle section of the loop, at the running temperature. Secondly, anode voltage correction (to compensate the electrical field-strength change at the tip, due to the emitter thermal expansion<sup>5</sup>) could not be performed for the continuously changing temperature. Finally, the elliptical functions  $v(y)$  and  $t(y)$ , depending on the work function, were treated as constant values for measured temperatures, this being obviously an approximation.

For comparison, in Table I the work-function temperature coefficients are listed. They have been calculated according to MG or Young theory.

#### ACKNOWLEDGMENTS

The author is very grateful to Professor Z. Sidorski for a critical reading of the manuscript, Professor T. Madey for helpful discussion, and the Polish Academy of Sciences for partial financial support.

<sup>1</sup>J. K. Wysocki, Acta Phys. Polon. **35**, 195 (1969).  
<sup>2</sup>J. K. Wysocki, Acta Phys. Polon. **A42**, 129 (1972).  
<sup>3</sup>J. J. Czyzewski, Surf. Sci. **33**, 589 (1972).  
<sup>4</sup>J. J. Czyzewski, Surf. Sci. **39**, 1 (1973).  
<sup>5</sup>J. K. Wysocki, Surf. Sci. **104**, 463 (1981).  
<sup>6</sup>E. L. Murphy and R. H. Good, Phys. Rev. **102**, 1464 (1956).  
<sup>7</sup>J. K. Wysocki, J. Phys. E **15**, 1376 (1982).  
<sup>8</sup>R. H. Fowler and L. W. Nordheim, Proc. R. Soc. London Ser. A **119**, 173 (1928).  
<sup>9</sup>L. W. Nordheim, Proc. R. Soc. London Ser. A **121**, 626 (1928).  
<sup>10</sup>R. D. Young, Phys. Rev. **113**, 110 (1959).  
<sup>11</sup>H. C. Miller, J. Franklin Inst. **282**, 382 (1966).  
<sup>12</sup>J. K. Wysocki, in Proceedings of the First Seminar on Surface Physics [Acta Univ. Wratislav. **29**, No. 380, 99 (1977)].  
<sup>13</sup>T. F. Felter, R. A. Barker, and P. J. Estrup, Phys. Rev. Lett. **38**, 1138 (1977).  
<sup>14</sup>R. A. Barker, P. J. Estrup, Phys. Rev. Lett. **41**, 1307 (1978).

<sup>15</sup>M. K. Debe and D. A. King, Surf. Sci. **81**, 193 (1979).  
<sup>16</sup>A. J. Melmed, R. T. Tung, W. R. Graham, and G. D. W. Smith, Phys. Rev. Lett. **43**, 1521 (1979).  
<sup>17</sup>N. Egede Christensen and B. Feuerbacher, Phys. Rev. B **10**, 2349 (1974).  
<sup>18</sup>B. Feuerbacher and N. Edge Christensen, Phys. Rev. B **10**, 2373 (1974).  
<sup>19</sup>P. H. Cutler, private communication.  
<sup>20</sup>A. van Oostrom, Phys. Lett. **4**, 34 (1963).  
<sup>21</sup>I. W. Swanson and L. C. Crouser, Phys. Rev. **163**, 622 (1967).  
<sup>22</sup>L. W. Swanson, L. C. Crouser, and F. M. Charbonnier, Phys. Rev. **151**, 327 (1966).  
<sup>23</sup>R. Klein and L. B. Leder, Phys. Rev. **124**, 1046 (1961).  
<sup>24</sup>L. W. Swanson, Surf. Sci. **70**, 165 (1978).  
<sup>25</sup>S. Nishigaki and S. Nakamura, Jpn. J. Appl. Phys. **15**, 1647 (1976).  
<sup>26</sup>L. W. Swanson and L. C. Crouser, Phys. Rev. Lett. **16**, 389 (1966).  
<sup>27</sup>E. W. Plummer and J. W. Gadzuk, Phys. Rev. Lett. **25**, 1493 (1970).

<sup>28</sup>H. F. Kempin, doctoral thesis, University of Munich, 1978 (in German) (unpublished).

<sup>29</sup>K. Klapper, doctoral thesis, University of Munich, 1978 (in German) (unpublished).

<sup>30</sup>S. Yamamoto, S. Fukuhara, H. Okano, and N. Saito,

Jpn. J. Appl. Phys. 15, 1643 (1976).

<sup>31</sup>S. Mróz and E. Chrzanowski, Prib. Tek. Eksp. 4, 176 (1968) (in Russian).

<sup>32</sup>T. E. Madey, private communication.