THE EFFECT OF TEMPERATURE ON THE FIELD EMISSION FROM LEAD

By J. Żebrowski

Institute of Experimental Physics, University of Wrocław*

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Single micro-crystals of lead were grown on the tungsten emitter of a field emission microscope (FEM). The current-potential characteristics for the field emission from these crystals were measured at various temperatures. It was found that the slope of Fowler-Nordheim plots (FN) increases with increasing temperature. We suggest a correlation between phenomenon observed and the work function dependence on temperature. The temperature coefficient of the work function for lead, which is approximately $8 \times 10^{-4} \, \text{eV/K}$ was estimated.

1. Introduction

Investigations on the electron emission from metals with a low melting temperature are rarely performed because of difficulties in obtaining a clean surface. Melmed and Gomer [1] found that whiskers grown by the evaporation of a metal onto the FEM emitter can be used in field emission studies because they have a high volume and surface purity, a high mechanical strength and their radius of curvature is small. Using this method, whiskers of numerous metals [1] including lead [2] have been obtained.

In this study we used lead whiskers to measure the dependence of the field emission on temperature. At low temperatures the dependence of the field emission current on the work function and the electric field strength (E) is given by the Fowler-Nordheim equation [3]:

$$I_{\rm FN} = 1.54 \times 10^{-6} \, \frac{\beta^2 U^2}{\varphi t(y)} \, A \, \exp\left[-6.83 \times 10^7 \, \frac{\varphi^{3/2}}{\beta U} \, v(y) \right], \tag{1}$$

where the current (I), potential (U) and work function (φ) are expressed in ampers, volts and electronovolts, respectively. A represents the emitting area in square centimeters

^{*} Address: Instytut Fizyki Doświadczalnej, Uniwersytet Wrocławski, Cybulskiego 36, 50-205 Wrocław, Poland.

t(y) and v(y) are Nordheim function [4] and β is a geometrical factor $(E = \beta U)$. In the semilogarithmic coordinates relation (1) is a straight line (FN plot) with the slope of:

$$S = -6.83 \times 10^7 \frac{\varphi^{3/2}}{\beta} s(y), \tag{2}$$

where s(y) is another Nordheim function. The variation of the field emission current with temperature is described by the Murphy-Good (MG) equation [5]:

$$I_{\rm MG} = I_{\rm FN} \frac{\pi p}{\sin \pi p},\tag{3}$$

where $p = 8.82 \times 10^3 \ \varphi^{1/2} \frac{T}{E} t(y)$. In the MG theory, E and φ are assumed to be independent of temperature. In fact, the work function is temperature dependent [6]. This also was proved experimentally by field emission measurements [7, 8].

As a consequence of thermal expansion, also the quantities β and A in equation (1) change. The slope of the FN plot (2) will thus vary with temperature variation. The temperature change of the β factor can be estimated easily from the simple relation: $\beta = \beta_0(1 - \gamma \Delta T)$, γ denotes the coefficient of thermal expansion. Hence, equation (2) can be used for the determination of the dependence of the work function on temperature.

2. Experimental

Field emission microscopes and lead sources used in these investigations were described elsewhere [9]. Single micro-crystals of lead used as field emitters were obtained using the method proposed by Melmed [2], i.e., by the deposition of lead onto the tungsten emitter at 300-400 K with a deposition rate of 2-4 monolayers per minute. The presure of residual gases with the lead source operating was less than 5×10^{-10} Torr. The emitter temperature was evaluated by measuring the resistivity of the central portion of the loop to which the emitter was spot-welded. The subsequent stages of the growth of a single lead crystal are shown in Fig. 1.

Fig. 2. shows lead crystals with various symmetries and different orientations with respect to a substrate, which were grown within the FEM. It is commonly assumed that single crystals of this type are whiskers [1, 2, 10].

Photo b in Fig. 2 shows the same crystal as in photo a after heating in the presence of an electric field. The change in the emission pattern is caused by a change in the emitter shape. Sharp edges are formed because of the surface diffusion in an electric field and the electron emission is more pronounced. This process is called thermo-field build-up and it also occurs with field-emitters made from refractory metals [11–13]. This enhances the geometrical factor, β . Contrary to refractory metals, however, the build-up of the lead emitter occurs at a much lower strength of the electric field. To avoid the uncontrollable variation of the geometrical factor, β , all measurements were performed using lead emitters which were previously built-up by heating in an electrical field higher than that used for the meausrements. It was stated that for refractory metal emitters [14–16] the build-up

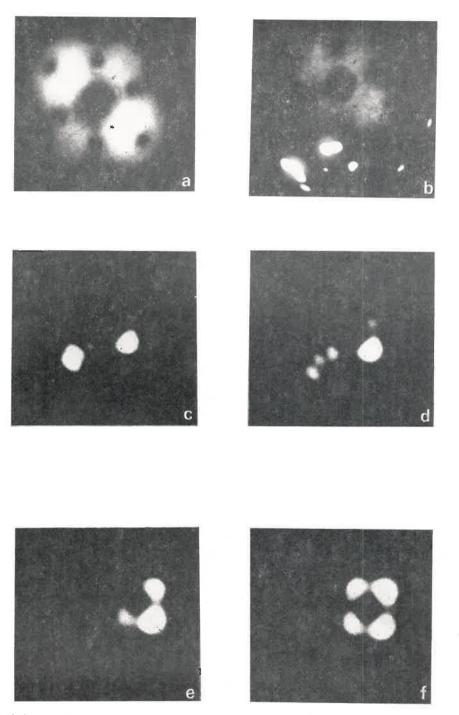
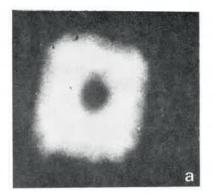
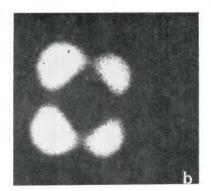
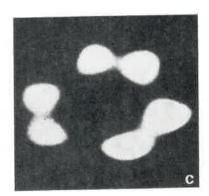


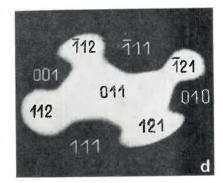
Fig. 1. Emission patterns of subsequent stages of lead crystal growth. Photograph a shows clean tungsten.

Photographs b-f correspond to 1, 3, 5 10 and 15 minutes of lead deposition time









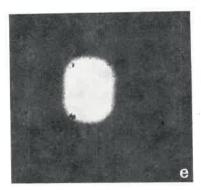


Fig. 2. Emission patterns of lead crystals with various orientations. a — (100) orientation before build-up, b — (100) orientation after build-up, c — (111) orientation, d — (110) orientation, e — another example with two-fold symmetry

form is preserved in the presence of an electric field even at high temperatures. One can expect also that for built-up lead emitters there exists a temperature range where they are stable.

3. Results and discussion

The FN plots shown in Fig. 3 correspond to a lead crystal having emission patterns shown in Fig. 2 a and b. Curves 1 and 2 were measured at liquid nitrogen temperature. One was first measured before build-up (Fig. 2a) and the second after thermo-field build-up (Fig. 2b). The FN plot corresponding to the build-up emitter is shifted to the region of lower voltages and its slope significantly changes compared to the plot derived before

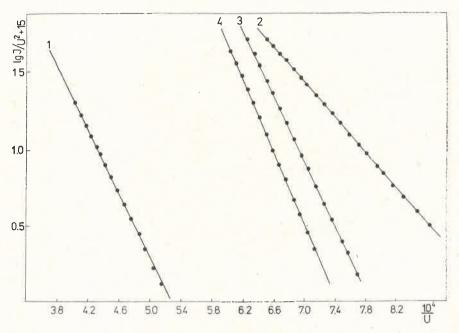


Fig. 3. Fowler-Nordheim plots of lead crystal at different temperatures: curves I and 2 at 80 K before and after build-up, curves 3 and 4 at 300 and 400 K after build-up

build-up. This indicates that the geometrical factor, β , also changes distinctly. FN plots 2 and 3 correspond to the build-up emitter and were measured at 300 and 480 K, respectively.

We found that the heating of the build-up emitter in the presence of an electric field up to 480 K does not affect its shape. After cooling down to 300 K the unchanged FN plot was measured. An increase in temperature above 480 K most likely changes the emitter shape because after lowering the temperature to 300 K a different FN plot was obtained. Therefore, we assumed that in the temperature range between 300–480 K the shape of the build-up emitter remained unchanged. The slope change occurring in the FN plot observed in this temperature region seems to be connected with the temperature variation of the

work function and/or with changes in the geometrical factor, β , because of the thermal expansion.

Fig. 4 shows the dependence of the slope of FN plots on temperature, measured in the temperature range stated above. During measurements the emitter always was kept in the presence of an electric field.

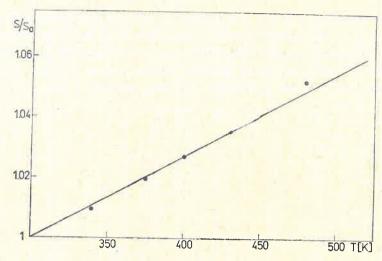


Fig. 4. The dependence of the slope of FN plots on temperature

After each measurement the temperature of the emitter was lowered to 300 K, and the FN plot was measured at this temperature. No changes in this plot were observed.

Fig. 5 shows the dependence of the field emission current on temperature at a constant voltage of 1580 V. As can be seen from Fig. 5, exactly the same curve is obtained by both the increase and decrease of the temperature. From Fig. 6 it follows that the relationship between the field emission current at a constant voltage and the temperature has an exponential character.

It can be estimated from Fig. 5 that an increase in emitter temperature from 300 to 480 K causes a decrease in the emission current of $\Delta I = -55 \times 10^{-9}$ A. The ratio of this current change to the emission current at 300 K (I_0) is $\frac{\Delta I}{I_0} = -52.38 \times 10^{-2}$. Using the

MG equation (3), we calculated the relative change of the emission current $\left(\frac{\Delta I}{I_0}\right)_{MG} = 6.68 \times 10^{-2}$ for the temperature range 300-480 K. Taking into account the influence of thermal expansion on the emitting area. A, and on the geometrical factor, β , [7] we found that

$$\left(\frac{\Delta I}{I_0}\right)_A = 1.01 \times 10^{-2}$$
 and $\left(\frac{\Delta I}{I_0}\right)_{\beta} = -4.14 \times 10^{-2}$ for the temperature range considered.

The calculated values for the emission current change are one order of magnitude less than the measured value. Consequently, the observed current decrease is caused predominantly by an increase of the work function.

Let us assume that in the temperature range considered the work function increases linearly with increasing temperature: $\varphi = \varphi_0 + \alpha \Delta T$. Taking also into account the change of β caused by the thermal expansion we derive from (2):

$$\frac{S}{S_0} = 1 + \left(\frac{3}{2} \frac{\alpha}{\varphi_0} + \gamma\right) \Delta T,\tag{4}$$

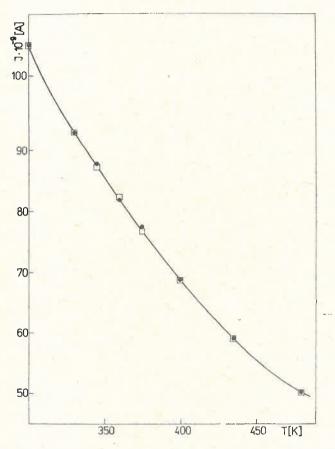


Fig. 5. The dependence of the emission current at a constant voltage of 1580 volts on the emitter temperature, measured at increasing (circles) and decreasing (squares) temperatures

where S_0 , φ_0 , β_0 are the corresponding values at 300 K. Neglecting the influence of temperature on the pre-exponential term in the FN equation we obtain from (1):

$$\log \frac{I}{I_0} = 9.96 \times 10^7 \, \frac{\varphi_0^{3/2}}{\beta_0} \, v(y) \left(\frac{3}{2} \, \frac{\alpha}{\varphi_0} + \gamma \right) \frac{\Delta T}{U} \,. \tag{5}$$

The nature of relationships (4) and (5) is in good agreement with the experimental results shown in Figs. 4 and 6. Both these results can be used to evaluate the temperature coefficient of the work function for lead.

Taking $\varphi_0 = 4.0 \text{ eV}$ [17] and $\gamma = 28.15 \times 10^{-6} \text{K}^{-1}$ we found that $\alpha = 8 \times 10^{-4} \text{ eV/K}$. For comparison, the temperature coefficients of the work function for several metals having the fcc structure are presented in Table I. It can be seen from this table that the value of the temperature coefficient determined in this study for lead is of the same order of magnitude as the values found for other metals. In most cases however, this coefficient is negative. Only silver, similarly to lead, has a positive work function temperature coefficient. It was found for tungsten [7, 8, 22] that both the value and the sign of the coefficient are different

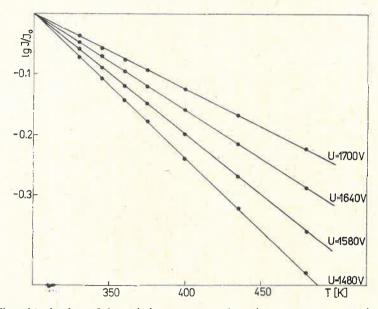


Fig. 6. Semilogarithmic plots of the emission current vs. the emitter temperature at various fixed voltages given in the figure. I_0 — denotes the emission current at 300 K

for various single crystal planes. For most planes this coefficient is negative, except for the (111) and (116) planes. According to Herring and Nichols [6], the temperature dependence of the work function is a consequence of many processes such as: (i) thermal expansion of the lattice, (ii) influence of atomic vibrations on the internal electrostatic potential and on the chemical potential, (iii) variation of the heat capacity of the electron gas, (iv) temperature change of the dipol moment of the surface layer. There is not any theory which takes into account all the above factors. Thus, neither the value nor the sign of the temperature coefficient of the work function can be determined theoretically.

In [23] the effect of the thermal expansion on the work function (φ_u) , and lattice correction, $(\delta \varphi_{hkl})$, in jellium approximation [24] has been considered. It was found that the temperature coefficient, $\alpha_u = \frac{d\varphi_u}{dT}$, is always negative. However, for metals having the fcc structure, $\alpha_{hkl} = \frac{d(\delta \varphi_{hkl})}{dT}$ can be either negative or positive, depending on the crystallographic direction.

TABLE I

Metal	$\alpha \times 10^4 \frac{\text{eV}}{\text{K}}$	References
(100) Cu	-13.8	
(110) Cu	- 5.2	
(111) Cu	- 3.4	[18]
(112) Cu	- 6.5	
(221) Cu	- 4.0	
(100) Ni	- 1.7	[19]
(111) Ni	- 1.7	
Hg	- 6.6	[20]
Ag ^a	+18.7	
Ag ^a Ag ^b	+ 3.6	[21]

^a Solid state:

Calculations performed for Pb (100) and Pb (110) give the following values of the temperature coefficient of the work function -5.92×10^{-5} eV/K and 6.62×10^{-5} eV/K, respectively. Since in the theory [23] only thermal expansion was considered, the agreement between experimental results obtained in this work and the theoretical calculations appears to be relatively good.

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REFERENCES

- [1] A. J. Melmed, R. Gomer, J. Chem. Phys. 34, 1802 (1961).
- [2] A. J. Melmed, J. Chem. Phys. 42, 3332 (1965).
- [3] A. G. J. van Oostrom, Philips Res. Rep. Suppl. Nr 1 (1966).
- [4] R. H. Good, E. W. Müller, in *Handbuch der Physik*, edited by S. Flüge, Springer-Verlag, Berlin 1956, Vol. 21, p. 176.
- [5] E. L. Murphy, R. H. Good Jr., Phys. Rev. 102, 1464 (1956).
- [6] C. Herring, M. Nichols, Mod. Phys. 21, 185 (1949).
- [7] A. van Oostrom, Phys. Lett. 4, 34 (1963).
- [8] L. W. Swanson, L. C. Crouser, Phys. Rev. 163, 622 (1967).
- [9] J. Żebrowski, Acta Phys. Pol. A50, 307 (1976).
- [10] M. M. P. Janssen, J. Modhuysen, Surface Sci. 42, 86 (1973).
- [11] W. P. Dyke, F. M. Charbonnier, R. W. Strayer, R. L. Floyd, J. P. Barbour, J. K. Trolan, J. Appl. Phys. 31, 790 (1960).
- [12] J. P. Barbour, F. M. Charbonnier, W. W. Dolan, E. E. Martin, J. K. Trolan, Phys. Rev. 117, 1454 (1960).
- [13] P. C. Better, M. Charbonnier, Phys. Rev. 119, 85 (1960).
- [14] D. M. Pautov, J. L. Sokolskaya, Zh. Tekh. Fiz. 41, 1999, (1971).

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- [15] L. W. Swanson, L. C. Crouser, J. Appl. Phys. 40, 4741 (1969).
- [16] A. W. Crewe, D. N. Eggenberger, J. Wall, L. M. Welter, Rev. Sci. Instrum. 39, 576 (1968).
- [17] W. S. Fomenko, I. A. Podtsherniaeva, The Emission and Adsorption Properties of Elements and Chemical Compositions, Moskva, Atomizdat 1975 (in Russian).
- [18] E. H. Blevis, A. C. Crowel, Phys. Rev. 133, A580 (1964).
- [19] K. Christmann, G. Ertl, D. Schober, Z. Naturforsch. A29, 1516 (1974).
- [20] S. Trassatti, Applic. Surface Sci. 1, 341 (1978).
- [21] V. G. Bolshov, Zh. Tekh. Fiz. 26, 1150 (1956).
- [22] A. R. Hutson, Phys. Rev. 98, 889 (1955).
- [23] A. Kiejna, K. F. Wojciechowski, I. Żebrowski, J. Phys. F. 9, 1361 (1979).
- [24] N. D. Lang, W. Kohn, Phys. Rev. B3, 1215 (1971).