

Temperature dependence of work function, thermionic emission constants in metals and modification of Richardson-Dushman Equation.

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Abstract

For proper modelling of the thermionic converters, and evaluation of its efficiency and power output (from a given input energy flux) it is necessary to estimate accurately the thermionic currents from the hot emitter surface. In this paper we derive the expression for the work function of a metal as a function of temperature considering thermal expansion and constant number of free electrons. We then modify the Richardson-Dushman equation for thermionic emission and explain the observed variation of the Thermionic emission constant from metal to metal. The theory also explains the observed rate of change of work function with temperature for tungsten and predicts dependence of thermionic emission constant and current density (at a given temperature) of a metal on crystallographic direction of the emitter surface. Application of the derived equation is discussed. The theory seems to be in more reliable agreement with experimental results than that of Seely.

1.0 Introduction

The emission of electrons from a hot metal surface (semiconductors are not considered in this article) was termed initially the "Edison Effect." [1] by the British scientist William Preece in 1884 and later the British physicist Owens Willans Richardson [2] studied it in details and called it "thermionic emission". Thermionic emission finds application in many practical devices such as , electron guns [3], vacuum tubes [4], cathode ray oscilloscope [5], thermionic converters [6] etc. Modelling the performances of these devices is based on Richardson-Dushman equation [2] of the emitted current density which is given by:

$$J = A_0 T^2 \exp(-W_0/k_B T) \tag{1}$$

where $A_0 = (4em\pi k_B^2/h^3)$ a constant assumed originally to be independent of metals.

W_0 = work function of metal.

In reality A_0 depends on type of metal (Table 2). It is seen from Table 2 that the thermionic emission constant A_0 is quite less than the theoretical value for most of the metals except cesium. This dependence has been often empirically given by:

$$A' = \lambda_B (1 - r_{av}) A_0 \tag{2}$$

so that the equation is often written as

$$J = A' T^2 \exp(-W_0/k_B T) \tag{3}$$

In presence of the electric field, E (That can exist between the electron emitting cathode (-ve) and the electron collecting anode (+ve)) there is Schottky [7] effect as a result of which W_0 is lowered to

$$W' = W_0 - e(eE/4\pi\epsilon_0)^{0.5} \tag{4}$$

and consequently the thermionic emission current density increases according to

$$J = A' T^2 \exp(-W'/k_B T) \tag{5}$$

This effect (Eq. 4) is known as Schottky effect in thermionic emission. This however, does not explain the quantitative dependence of A' on metal characteristics [8]. Also since the work function is the difference between the vacuum energy and the Fermi energy, E_F and the E_F depends on temperature T at which (which is usually > 1000 K) the thermionic emission

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takes place, the work function is also dependent on temperature. In modelling electron guns and other devices which depend on $J(T)$, it is very important to have a precise knowledge of A' and W' .

Seely (1941)[8] investigated the change of potential energy barrier at the surface of a metal with the thermal expansion of the metal due to heating. From the change of the normal maximum energy of an electron calculated as a function of the temperature of the metal he showed that the thermionic emission constant depends on metal characteristics. In this work we give a completely new derivation of the modification of the eqn. (5) due to thermal expansion of the metal from a different concept. We assume the total number of free electrons in a metal to remain constant as temperature rises and derive an expression for the variation of $W(T)$ with temperature. We substitute this in Eq. (5) in place of W' to arrive at the modified version of equation (5). We see that automatically A'' becomes metal-specific. We also find that the variation of $W(T)$ with temperature depends on metal characteristics. The theory is found to explain fairly well the observed A'' in several metals with exception of alkali metals. It also explains the observed temperature rate of change of work function in tungsten. Our derived equations can properly be used in modelling electronic devices based on thermionic emissions.

Such modified eqn. is expected to be important for thermionic emission from metals with low work function of the order of 1 eV at temperatures of 1500 K or higher. Thermionic emission[9] is recently combined with energy selective vacuum tunnelling of high speed electrons (in the Fermi distribution) in special nano scale (geometry) design of materials with low work function of 1.0 eV for refrigeration (thermionic cooling). It is found that for nano materials with 5-15 nm size gaps only a small external voltage (1 – 3 V) is required to create large currents and a cooling power of 100 W/cm². Carbon nanotubes[10] (CNTs) intercalated with alkali metals (to reduce W), such as potassium are promising candidates for new thermionic-photoemission materials. In such materials also which are good candidates for combined thermionic and photo-thermal power generators, where the work function is found to be temperature dependent, the modified thermionic emission equation will be very useful in predicting accurately the thermionic current density at a given temperature and hence power derivable from a given solar flux. Below we first discuss the simple “derivation of

of Richardson-Dushman thermionic emission equation from a metal” which is available in any text book. Next we discuss the modification of the equation and explanation for the temperature and orientation dependence of work function of a metal surface. Then we discuss the variation and comparison of the thermionic emission constants with the present theory.

2.0 Derivation of Richardson-Dushman thermionic emission equation from a metal.

The derivation of Eq.(1) is given in a undergraduate or graduate solid state physics book. Only certain relations relevant[11,12] to the subsequent derivations are quoted here.

According to the derivation, $A_0 = 4em_e\pi K_B^2/h^3$ is independent of metal, unless one uses the effective mass of electron. However, we are not really supposed to use the effective mass since m refers to the mass of the electron as it is out of the metal surface and moreover, we are talking about the free electron in the metal (which are emitted) whose effective mass ($m_e = \hbar^2/(\partial^2 E/\partial k^2)$) is close to the free electron mass because of the relation $E = \hbar^2 k^2/2m_e$. We want to explore the reason for this variation from metal to metal.

The density of states defined as the number of quantum states per unit volume of the material per unit energy interval is for free electrons in metal given by[11]

$$g(E) = CE^{1/2} \tag{6a}$$

$$\text{Where } C = (m/\hbar^2)^{3/2} 2^{1/2}/\pi^2 \tag{6b}$$

This density of states is independent of temperature. The Fermi energy at 0 K is given by $E_{F_0} = (\hbar^2/2m)(3n/8\pi)^{2/3}$

Using the concept of $g(E)$, the total number, N of free electrons in the metal are given by:

$$\text{At temperature 0 K, } N(T = 0K) = V_0 \int_0^{E_{F_0}} g(E)dE \tag{7a}$$

$$\text{At temperature T, } N(T) = V \int_0^\infty g(E)f(E)dE \tag{7b}$$

Where $f(E) = 1/[1 + \exp((E - E_F)/k_B T)]$ is the well known Fermi-function.

Considering thermal expansion of the metal, $V = V_0(1 + 3\alpha T)$.

Where α = Linear thermal expansion coefficient. In general we shall use $V = V_0(1 + r\alpha T)$ where $r = 3$ for a three dimensional (sphere or cube) emitter, $r = 2$ for a two dimensional (plate or thin film emitter); $r = 1$ for one dimensional (thin wire emitter).

Thermionic emission takes place at high temperature (>1000 K). Since the work function is defined as the difference between the vacuum level and the Fermi-level, E_F and since E_F is dependent on temperature, even though E_F/k_B varies from about 15000 K to 110,000 K in metals, at temperatures above 1000 K (above which usually thermionic emission takes place), change of E_F with T will change the work function with T and this in turn will affect the thermionic emission. Our primary objective now is to obtain E_F as a function of T from the two equations (7a) & (7b). The 2nd objective is to see how this affects the Richardson-Dushman thermionic equation. The third objective is to see overall how the equation is modified when Schottky effect is considered, i.e., the modification of work function in presence of electric field.

Variation of work function of a metal with temperature and the consequent modification of Richardson-Dushman thermionic equation.

The dependence of W on T can be understood when we understand the change of E_F with T. To work out this change we rely on the fact that the total number of electrons in a given piece of metal at $T = 0 K$ is the same at $T = T K$. (The case where thermionic emission takes place without the electrons being replaced at the emitter (which is usually the cathode) will not be treated here). Now at $T = 0 K$, $E \leq E_{F_0}$ and the Fermi function $f(E) = 1$

Thus from eqs 7a and 7b we have

$$V_0 \int_0^{E_{F_0}} g(E) dE = V_0 (1 + r\alpha T) \int_0^\infty g(E) f(E) dE \tag{8}$$

Using the expressions for $g(E)$ and $f(E)$ Eq. (8) becomes

$$\int_0^{E_{F_0}} C E^{1/2} dE = (1 + r\alpha T) \int_0^\infty [1/(1 + \exp(E - E_F)/k_B T)] C E^{1/2} dE \tag{9}$$

The LHS of eq.(9) is independent of T while the RHS is dependent on T. Solution of this eq.(9) for E_F as a function of T is a non-trivial problem. We shall follow the Sommerfield expansion method (in the original expansion the factor $(1 + r\alpha T)$ was absent).

Our target here is to express E_F as a function of E_{F_0} and T. Once that is done we shall be able to express the work function W as a function of T and understand why the Richardson-Dushman thermionic constant A, varies from metal to metal instead of being a constant.

We write the RHS of Eq.(9) as

$$I = (1 + r\alpha T) \int_0^\infty f(E) C E^{1/2} dE = (1 + r\alpha T) [f(E) \int_0^\infty C E^{1/2} dE - \int_0^\infty \{df(E)/dE\} \int_0^\infty C E^{1/2} dE] dE \tag{9a}$$

$$= (1 + r\alpha T) \left[\int_0^\infty f(E) 2 C E^{3/2} / 3 \Big|_0^\infty - \int_0^\infty \{df(E)/dE\} 2 C E^{3/2} / 3 dE \right] \tag{10}$$

The first term in the square bracket is zero because $f(E)$ is zero at $E = \infty$ and unity at $E = 0$.

For evaluation of the second integral we need to express $E^{3/2}$ as a function of $E - E_F$ since this term $(E - E_F)$ is in $f(E)$.

$$E^{3/2} = E_F^{3/2} + (E - E_F)(E^{3/2})' + (E - E_F)^2 (E^{3/2})'' / 2! + (E - E_F)^3 (E^{3/2})''' / 3! + \dots \tag{11}$$

$$df(E)/dE = -[1/(1 + \exp(E - E_F)/k_B T)]^2 \exp((E - E_F)/k_B T) / k_B T \tag{12}$$

In Eq.(11) the successive ' indicates the order of derivatives evaluated at E_F .

The derivative in Eq.12 is an even function of $E - E_F$ and is significant only around $E = E_F \pm$ a few $k_B T$. Therefore, the integral in Eq.(10) is insignificant when E is less or greater than E_F by more than a few $k_B T$. The successive first three integrals are:

$$L_0 = - \int_0^\infty 2 C E_F^{3/2} \{ [1/(1 + \exp(E - E_F)/k_B T)]^2 \exp((E - E_F)/k_B T) / k_B T \} = 2 C E_F^{3/2} / 3$$

$$L_1 = - (E^{3/2})' \int_{-\infty}^\infty 2(E - E_F) [1/(1 + \exp(E - E_F)/k_B T)]^2 \exp((E - E_F)/k_B T) / 3 k_B T \tag{13}$$

Because of the term $E - E_F$ in eq.(13) the integral is an odd function of $E - E_F$. Thus the integral is zero. The third integral is

$$L_2 = (E^{3/2})'' \int_{-\infty}^\infty (E - E_F)^2 \{ [1/(1 + \exp(E - E_F)/k_B T)]^2 \exp((E - E_F)/k_B T) / 3 k_B T \} dE \tag{14}$$

$$L_2 = (E^{3/2})'' (k_B T)^2 \int_{-\infty}^\infty x^2 e^x dx / (1 + e^x)^{2/3}$$

Where $x = (E - E_F)/k_B T$

Now the standard integral $\int_{-\infty}^\infty x^2 e^x dx / (1 + e^x)^2 = \pi^2 / 3$

Now $(E^{3/2})'' = 3/2 (E^{1/2})' = (3/2)(1/2)(E^{-1/2})$ at $E = E_F$

$$= (3/4) E_F^{-1/2}$$

Thus $L_2 = (\pi^2/8) E_F^{-1/2} (k_B T)^2$

Retaining terms only up to 2nd order, thus the integral of Eqs.(9a) & (10) becomes

$$I = (1 + r\alpha T) [2 C E_F^{3/2} / 3 + (\pi^2/12) E_F^{-1/2} (k_B T)^2] \tag{15}$$

Thus eq.(9) becomes

$$(2/3) C E_{F_0}^{3/2} = (1 + r\alpha T) [(2/3) C E_F^{3/2} + C (\pi^2/12) E_F^{-1/2} (k_B T)^2] \tag{16}$$

$$E_{F_0}^{3/2} / E_F^{1/2} = (1 + r\alpha T) [E_F + (\pi^2/12) (k_B T)^2 / E_F] \tag{17}$$

On the LHS of Eq.(17) we can take $E_F = E_{F_0}$. Then Eq.(17) becomes

$$E_{F_0} = (1 + r\alpha T) [E_F + (\pi^2/12) (k_B T)^2 / E_F]$$

$$E_F - E_{F_0} = -r\alpha T [E_F] - (1 + r\alpha T) (\pi^2/12) (k_B T)^2 / E_F \tag{18}$$

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Replacing E_F by E_{F_0} on the RHS will not affect the RHS much, because even at T between 1000 and 2000 K (where most of the thermionic emissions take place), $\alpha T \sim 0.03$ for and $(k_B T/E_F)^2$ is between 0.01 and 0.0001 for most metals and we get

$$E_F - E_{F_0} = -\alpha T [E_F] - (1 + \alpha T) (\pi^2/12) (k_B T)^2/E_{F_0} \quad (19)$$

Now the work function W_0 of a metal (at 0 K) is the minimum energy required for an electron at energy E_{F_0} to escape from the surface of the metal.

$$W_0 + E_{F_0} = E_v (\text{Vacuum energy of electron}) \quad (20a)$$

$$\text{Or } W_0 = E_v - E_{F_0} \quad (20b)$$

Now with rise in temperature, E_v does not change but E_F . The new work function W at temperature T should be defined (in analogy with eq.(20b)) by

$$W(T) = E_v - E_F(T) \quad (21)$$

Using Eqs. (19) - (21) we see that

$$W(T) = W_0 + \alpha T E_{F_0} + (1 + \alpha T) \pi^2 (k_B T)^2 / 12 E_{F_0} \quad (22)$$

The rate of change of work function with temperature is given by

$$dW(T)/dT = \alpha E_{F_0} + 2(1 + \alpha T) \pi^2 (k_B T) / 12 E_{F_0} + \alpha \pi^2 (k_B T)^2 / 12 E_{F_0} \quad (23)$$

Eq. 22 gives us the temperature dependence of the work function of a metal. W_0 corresponds to T = 0 K. Because of the quantities, α and E_{F_0} , this dependence on T is metal specific. The eq. says that $W(T)$ increases with T. Though the 2nd term in Eq. 22 makes the major contribution to the dependence of $W(T)$ on temperature, the 3rd term is also important when E_{F_0} is small. $W(T)$ has been observed to increase [10,15] in metals with T and this is consistent with Eq.(22). The quantitative comparison of eq.(22) with observed variation of $W(T)$ from metal to metal has been reserved for our next paper.

Inserting the expression for $W(T)$ in Eq.1 we get the modified Richardson-Dushman thermionic equation as:

$$J = (4em\pi k_B^2/h^3) T^2 \exp(-W/k_B T) = A_0 T^2 \exp(-[W_0 + \alpha T E_{F_0} + \{1 + \alpha T\} (\pi^2/12) (k_B T)^2/E_{F_0}]/k_B T)$$

Finally,

$$J = A_0 \exp(-\alpha E_{F_0}/k_B) T^2 \exp(-[W_0 + \{1 + \alpha T\} (\pi^2/12) (k_B T)^2/E_{F_0}]/k_B T) \quad (23a)$$

Eq. (23) gives us the modified thermionic emission equation with the constant A_0 changed to a new constant A'

$$A' = A_0 \exp(-\alpha E_{F_0}/k_B) \quad (23b)$$

Now in Eq. (23b) the term E_{F_0}/k_B is a fundamental constant of metal and is metal specific though absolutely temperature independent. E_{F_0} is given by

$$E_{F_0} = (h^2/2m)(3n/8\pi)^{2/3} = 3.65 \times 10^{-19} n^{2/3} \quad (\text{ineV}) \quad (24b)$$

$n = \text{number density of free electrons} = n = z N_A \rho / M$. $z = \text{valency}$; $\rho = \text{density}$; $M = \text{atomic mass}$. $N_A = \text{Avogadro number}$. n can also be calculated from the following relation: $n = xz/a^3$. Where $x = \text{number of atoms per unit cell of the metal crystal}$. $z = \text{valency of the metal}$. $a = \text{lattice constant of the metal}$. n of some metals calculated from the second relation is given in Table 1. E_{F_0} is then computed from Eq.(24b). The nearly temperature independence of the thermal expansion coefficient α can be understood [11] from the following considerations:

$$\alpha = (\gamma C_\gamma^{ion} + C_\gamma^{el})/3B \quad (25)$$

Where γ is the overall Gruneisen parameter of the metal; this is primarily of the order of unity and temperature independent. B the bulk modulus. At temperatures above 300 K, C_γ^{ion} assumes the value of $3R$. C_γ^{el} is usually much smaller than C_γ^{ion} . The electronic contribution C_γ^{el} is very negligible at high temperature. Thus α can be assumed to be fairly independent of temperature in a metal.

This new thermionic constant A' is dependent on two quantities: α and E_{F_0} which are both metal specific. Eq. (23a) adds also temperature dependent modification of the thermionic equation that is again metal specific. It is to be noted that the Schottky effect (lowering of work function by the presence of electric field, E on to the metal surface) modification of the thermionic emission equation that is well-known is not added to eq. (23a). With Schottky- effect W_0 is lowered to [11].

$$W_0' = W_0 - e(e E/4\pi\epsilon_0)^{0.5} \quad (26)$$

the final modified equation for thermionic equation is given by:

$$J = A' T^2 \exp(-[W_0' + \{1 + \alpha T\} (\pi^2/12) (k_B T)^2/E_{F_0}]/k_B T) \quad (27)$$

Discussion

For materials with low work function W_0 [13,16] and low E_{F_0} the above modified thermionic equation (27) is going to be important in modelling devices made of such thermionic cathodes. The temperature dependent work function is given by Eq.22 at $E = 0$. This can be measured accurately from photoemission experiment. Whereas Eq. (27) will provide temperature dependent work function as measured from thermionic emission experiment. The two work functions thus will not be exactly the same. Let us consider the second term in Eq. (22) for tungsten. This gives the first order temperature effects on $W(T)$. Table 1 gives relevant data for some metals which are used to compute E_{F_0} given in Table 2.

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We have seen in Table 2 that somewhat closer agreement between the experimental and calculated thermionic constant is obtained for $r = 2$ in tungsten. Then the first term in Eq. (22) gives the value αE_{F_0} (see Tables 1 & 2) as $7.9 \times 10^{-5} eV/K$. This is not in bad agreement with the measured value [15] of $6.3 \times 10^{-5} eV/K$ for the rate of change of work function with temperature. Surprisingly, but without proper explanation, this measured value is very close to the theoretical value of $r\alpha E_{F_0}$ for $r = 1.5$, half way between $r = 1$ and $r = 2$. Interestingly, for this value of r (i.e., 1.5), the measured value (60) of A' also agrees with the value (60.48) calculated from Eq. (23b) for tungsten.

We have calculated the values of A' for 10 metals and compared with the experimental values as given in Table 2 for $r = 1$ and for some $r = 2$. For either $r = 1$ or $r = 2$ we find that the agreement is fairly good except for Cs, Na & Ca which have high α . It is interesting to note that for four metals Cr, W, Pt & Mo the agreement is quite good only with $r = 2$. It may be mentioned that at temperatures of thermionic emission, cesium usually evaporates unlike other metals. This evaporation of metal may cause problem in the experimental measurements and also it is not taken into account in the above derivation. Another factor is that the errors in the experimental values are not precisely known so as to include effect of reflection [16] from the anode. Also the dimensionality (whether a thin wire, thin-film surface, volume emitter, sphere or cube) of the experimental cathodes is not precisely known. The dimensionality can affect the value of $(A')_{exp}$ because the density of states for one dimensional and two dimensional free electron system are different from that given by Eq. (6a) and also because of the fact that the factor 3α strictly applies for a volume emitter instead of a thin wire emitter. Another contribution to the remaining discrepancy between the measured values of A and those calculated using the formulations above, is that the effective number of free electrons in a metal is not exactly what we calculated in Table 1 using the valency of the atom. Thermionic emission from such systems will be treated separately.

In our derivations above, $E_{F_0} = \left(\frac{h^2}{2m} \right) (3n/8\pi)^{2/3}$ is assumed to depend on free electron mass. This is based on spherical Fermi surface in k -space. Actually in metals Fermi surface is not spherical [11]. In the case of thermionic emitter E_{F_0} should depend on the effective mass m_{ef} of electron in the metal crystal rather than free electron mass. m_{ef} depends on crystallographic direction. Thus from our equation we see that A'' and hence J at a given T should depend on the metal face. In fact such observation has been made.

Thus in modelling thermionic (and/or photo-thermal) power converters consideration of the metal face will be important.

Despite some noticeable success in explaining the thermionic emission data with the above theory, we feel that some more works are necessary in this line to derive similar expressions considering the density of states for one and two dimensional thermionic emitter systems and see if the remaining discrepancy between the experimental and theoretical values can be addressed better.

In our next paper we shall present application of the above equations in modelling photothermal power converts using solar energy.

Table 1. Computation of number of free electrons, n per unit volume of the following metals using equation $n = xz/a^3$. The data of the first 5 columns are taken from ref. [10]

Metal	Type of crystals	No. of atoms x/unit cell	Lattice constant $a(\text{\AA})$	Valency of atom, z	$n/m^3 \times 10^{28}$
Ca	Fcc	4	5.58	2	4.61
Cr	Bcc	2	2.88	1	8.5
Cs	Bcc	2	6.05 (78 K)	1	0.91
Fe(Iron)	Bcc	2	2.87	2	17.0
Ta	Bcc	2	3.31	2	11
W	Bcc	2	3.16	2	12.67
Pt	Fcc	4	3.92	1	6.6
Na	Bcc	2	4.23	1	2.64
Mo	Bcc	2	3.15	1	6.4
Ni	Fcc	4	3.52	2	18.34

Table 2: Comparison of calculated and experimental values of thermionic emission constant A' for metals in Table 1.

Metal	α ($^{\circ}\text{C}$) $\times 10^{-6}$		$n \times 10^{28}/\text{m}^3$	E_{Fo} (eV)	$(A'')_{\text{th}} \times 10^4$ Amp/ m^2 Eq.(23b) r=1 r=2		$(A'')_{\text{exp}} \times 10^4$ Amp/ m^2 Ref	
		Ref.						
Ca	22.5	[11]	4.60	4.69	35.3		60	[12,8]
Cr	6.2	[14]	8.37	7.05	72.3	43.5	48	[12]
Cs	97	[11]	0.90	1.59	20		162	[12]
Iron	11.7	[11]	16.92	11.2	26.3		26	[12]
Ta	6.5	[14]	11.03	8.38	63.8		55	[12]
W	4.3	[14]	12.67	9.2	76	48	60	[12,8]
Pt	9	[14]	6.64	5.96	64.4	34.6	32	[12,8]
Na	71	[11]	2.64	3.24	8.3		41	[12]
Mo	5	[14]	6.4	5.84	85.5	61	55	[8]
Ni	13	[14]	18.34	11.8	20.3		27	[8]

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