COLD EMISSION OF ELECTRONS FROM THE SURFACE OF A METAL IN A STRONG RADIATION FIELD

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The cold-emission electron current from the surface of a metal due to the action of an electromagnetic wave is calculated. In the limiting case of low frequencies and high field strengths the well known expression for field emission of electrons is obtained, whereas for high frequencies an expression is obtained for the photocurrent due to simultaneous absorption of several protons. Numerical estimates are presented.

1. As the result of the development of methods of generating powerful coherent electromagnetic radiation, theoretical interest has been renewed in the classical problems of the ionization of atoms and solid materials in the field of a strong electromagnetic wave.[1-4] A specific feature of the problem formulation in this new stage is the assumption that the photon energy $\hbar\omega$ of the field is less (in some cases considerably less) than the ionization potential I. With this approach the difference between the concepts of tunneling field emission and the photoelectric effect, which arose in the earlier classical approach to the problem, essentially disappears. Of course in this case it is always necessary to consider the multiphoton photoelectric effect and to calculate the ionization probability by use of perturbation theory. However, it appears that successful use of perturbation theory can be expected only for the two-photon photoelectric effect [3], when

$$1 < I/\hbar\omega < 2.$$

For larger values of $I/\hbar\omega$ the difficulties of analytic calculation with perturbation theory are well known to increase so greatly that it becomes practically impossible.

The problems mentioned above naturally are close to the classical problem of the cold emission of electrons from a metal surface under the action of an electromagnetic field. In addition, a new formulation of the problem arises in the simultaneous consideration of the phenomena of tunneling field emission and surface photoelectric effect for $\hbar\omega < w$ (w is the work function). For the case $w/\hbar\omega < 2$ (the two-photon photoelectric effect) the problem has recently been discussed by Smith [5] (see also the work of Makinson and Buckingham [6]).

In the present article the cold-emission problem is solved by the method used by Keldysh.[2] We have obtained an expression for the photocurrent which in the limiting case of low frequencies ω and high field intensities F transfers to the well known expression for the field-emission current, and for high frequencies corresponds to the photocurrent due to simultaneous absorption of n photons and which is proportional to $(F^2)^n$ ($n \sim w/\hbar\omega$). The solution obtained is more accurate for larger values of the ratio $w/\hbar\omega$. Numerical estimates show, however, that even for n = 1 and n = 2 (respectively the one-photon and two-photon photoelectric effect) accurate calculations using perturbation theory give the same results in order of magnitude as the theory developed by us.

2. We assume the following simplified model of a metal, similar to that used by Tamm and Shubin [7] and Mitchell:[8] the electrons move in a constant potential with a discontinuity U_0 at the metal-vacuum surface. Thus, if the plane x = 0 coincides with the metal surface:

$$U(x) = \begin{cases} 0, & x > 0, \\ U_0, & x < 0. \end{cases}$$

The wave functions of an electron in such a field have the form

$$\psi_p(x) = a_p \begin{cases} e^{ipx/\hbar}, & x > 0 \\ b_p^+ \exp\{i(p^2 + 2mU_0)^{1/2} x/\hbar\} + b_p^- \times \exp\{-i(p^2+2mU_0)^{1/2} x/\hbar\}, & x < 0; \end{cases}$$

$$\psi_k(x) = d_k \begin{cases} e^{-kx/\hbar}, & x > 0 \\ f_k^+ \exp\{i(2mU_0 - k^2)^{1/2} x/\hbar\} + f_k^- \times \exp\{-i(2mU_0 - k^2)^{1/2} x/\hbar\}, & x < 0. \end{cases} \quad (1)$$

The coefficients b_p^+, b_p^-, f_k^+, f_k^- are determined from the conditions of continuity of the functions and their derivatives at the boundary $x = 0$; a_p and d_k are determined from the normalization conditions:

$$\int_{-\infty}^{+\infty} dx\psi_p{}^*\psi_{p'} = \delta(p-p') + \lambda_p\delta(p+p'),$$

$$\int_{-\infty}^{+\infty} dx\psi_k{}^*\psi_{k'} = \delta(k-k');$$

$$\lambda_p = mU_0 / [p^2 + |p|(p^2 + 2mU_0)^{1/2} + mU_0]. \tag{2}$$

The numbers p determine the states with positive energy ($\epsilon_p = p^2/2m$), and the numbers k—the states with negative energy ($\epsilon_k = -k^2/2m$). Here

$$-\infty < p < +\infty, \qquad 0 < k < \sqrt{2mU_0}.$$

In formula (1) and subsequently for brevity we have not written out the factors corresponding to the transverse motion (of the type $\exp[i(p_y y + p_z z)/\hbar]$). This is because we will later be able to see directly the results of a correct calculation of these factors.

The external magnetic field is taken in the form of a plane wave incident on the metal surface. Up to optical frequencies the wavelength of the radiation is considerably larger than the principal characteristic length entering into the present problem, $\hbar/k_F$, where $k_F = [2m(U_0 - \epsilon_F)]^{1/2}$ and ϵ_F is the Fermi energy.[8] This fact permits us to neglect the spatial dependence of the radiation field, or, which is the same thing, to limit ourselves to the dipole approximation. It is easy to see that in this case the field components along the metal surface do not contribute to the ionization probability, since with respect to their transverse motion the electrons act as free electrons. Consequently we can limit ourselves to a perturbing potential of the form:

$$V = \frac{e}{c}\dot{A}(t)x, \quad A(t) = -F\frac{\sin\theta}{\omega}\sin\omega t,$$

where F is the electric field intensity of the wave and θ is the angle of incidence of the wave on the metal surface. Here the effect occurs only in the case where the electric field is polarized in the plane of incidence.

The wave function of an electron at an initial time $t = 0$ in a state ψ_k will be sought in the form

$$\Psi(x,t) = \exp\left\{\frac{i}{\hbar}\frac{k^2}{2m}t\right\}\psi_k + \int dpC_p(t)\Psi_p(x,t);$$

$$C_p(0) = 0. \tag{3}$$

As $\Psi_p(x,t)$ we choose, following Keldysh,[2] the orthonormal set of functions

$$\Psi_p(x,t) = \psi_{p-eA(t)/c}\exp\left[\frac{i}{\hbar}\int_0^t \left(p - \frac{e}{c}A(\tau)\right)^2\frac{d\tau}{2m}\right]. \tag{4}$$

These functions accurately account for the action of a strong radiation field on a free electron and, as Keldysh has shown,[2] lead mainly to a correct description of the tunneling and multiphoton processes. The correction to the expression for the photocurrent, due to the nonfree nature of the electron motion, will be introduced into the final result by the same means used by Keldysh.[2]

To determine the photocurrent we find the probability for transition of an electron from a state ψ_k to a state Ψ_p per unit time

$$\lim_{t\to\infty}\frac{|C_p(t)|^2}{t}.$$

The result obtained is integrated over the momenta of the emitted electrons and over all electron states inside the metal. It is necessary, of course, to take into account the factors $\exp[i(p_y y + p_z z)/\hbar]$ in the wave functions ψ_k and Ψ_p. It is evident beforehand to what result this leads. The quantity $C_p(t)$ will contain the factor $\delta(p_y - p'_y)\delta(p_z - p'_z)$. This means that the transition probability will be proportional to $\delta^2(p_y - p'_y)\delta^2(p_z - p'_z)$. After integration over the momenta of the emitted electrons we obtain $\delta^2(0) = L^2/(2\pi\hbar)^2$, where L tends to infinity. Here the total probability for emission of an electron increases infinitely, but the current density, which is proportional to the probability of removing an electron from a unit area of the surface, remains finite. In other respects the integration over momenta, and also over time, is carried out in exactly the same way as by Keldysh.[2]

Integration over the internal electron states in the metal is carried out on the assumption that the electrons are distributed according to a Fermi distribution. Since the transition probability depends only on the component of **k** along the x axis, $k_x \equiv k$, we have

$$\int d\mathbf{k} = 2\pi\int_{k_F}^{(2mU_0)^{1/2}} dk \int_0^{(k^2-k_F^2)^{1/2}} k_\perp dk_\perp = \pi\int_{k_F}^{(2mU_0)^{1/2}} dk\,(k^2 - k_F^2). \tag{5}$$

Proceeding as indicated above, we obtain the final formula for the current density in the following form:

$$j = \frac{me\omega^{3/2}U_0^{1/2}}{32\pi^6\hbar^{3/2}}\gamma^{3/2}\lambda^{5/4}\int_{\gamma_F}^{\gamma} dq\left[\frac{q}{(1+q^2)^{1/2}}\right]^{5/4}\frac{(1-q/\gamma)^{1/2}}{q^2}$$

$$\times(q^2 - \gamma_F^2)\left|J\left((\gamma - q)\frac{(q\lambda)^{1/2}}{(1+q^2)^{1/4}}\right)\right|^2$$

$$\times\sum_{n=n_0}^{\infty}\theta\left[\left(\frac{n-\lambda/2}{\lambda}\right)^{1/2} - q\right]\left(n - \frac{\lambda}{2} - \lambda q^2\right)^{-1/2}$$

$$\times \exp\left\{-2\left[n\left(\sinh^{-1}-\frac{q}{(1+q^2)^{1/2}}\right)+\frac{\lambda q^3}{2(1+q^2)^{1/2}}\right]\right\}. \tag{6}$$

Here

$$\gamma=\frac{(2mU_0)^{1/2}\omega}{eF_x},\quad \gamma_F=\frac{(2mw)^{1/2}\omega}{eF_x},\quad w=U_0-\varepsilon_F,$$

$$\lambda=\frac{e^2F_x^2}{2m\hbar\omega^3}=\frac{w}{\hbar\omega\gamma_F^2},\qquad \theta(x)=\begin{cases}1, & x>0,\\ 0, & x<0,\end{cases}$$

$$h_0=\mathrm{E}\left[\lambda\left(\frac{1}{2}+\gamma_F^2\right)+1\right],\quad F_x=F\sin\theta,$$

and E is the symbol for the integral part.

The function $J(x)$ is determined by the following integral:

$$J(x)=\lim_{\varepsilon\to 0}\int_{-\infty}^{+\infty}\frac{dz\,e^{-z^2}}{(z-i\varepsilon)^2(z-ix)^{1/4}}. \tag{7}$$

The appearance of this integral is due to the fact that the matrix element has two singular points, the position of one of them depending on the variable of integration k. For $k = (2mU_0)^{1/2}$ these singular points coincide. Ultimately only the asymptote of $J(x)$ for large x will be actually needed. It is easy to see that for $x \gg 1$

$$J(x)\simeq -2i^{1/4}\sqrt{\pi}/x^{1/4}.$$

For small x the function $J(x)$ is represented by the series:

$$J(x)\approx -{}^8/_5(1+\sqrt{-i})A+{}^{16}/_9\, i(1+\sqrt{i})Bx+\ldots;$$

$$A=\int_0^\infty\frac{dz\,e^{-z^2}}{z^{1/4}},\qquad B=\int_0^\infty dz\,e^{-z^2}z^{3/4}. \tag{8}$$

The derivation of Eq. (6) rests essentially on the use of the method of steepest descents in integration over time. This limits the region of applicability of the results. The corresponding condition has the form

$$\frac{\lambda\gamma_F}{(1+\gamma_F)^{1/2}}=\frac{w}{\hbar\omega\gamma_F(1+\gamma_F^2)^{1/2}}\gg 1. \tag{9}$$

Formula (6), and also the subsequent formulas, are not completely true for angles θ near $\pi/2$, i.e., for almost grazing incidence of the light. This is due to the fact that we do not take into account reflection and refraction of the light at the metal-vacuum interface. In the approximation of geometrical optics this can be done by the same method used by Mitchell.[8]

The expression for the current is considerably simplified in the limiting cases $\gamma_F \gg 1$ and $\gamma_F \ll 1$. For $\gamma_F \ll 1$ a very large number of terms are important in the sum over n. This permits us to transfer from a summation to an integration. As a result we have

$$j\approx\frac{\sqrt{6\pi}}{16\pi^5}\frac{(2m)^{3/4}U_0w^{1/4}}{\hbar^{5/2}}e^{3/2}F_x^{1/2}\times\exp\left(-\frac{4}{3}\lambda\gamma_F^3\right)\Phi\left(\frac{\gamma}{\gamma_F},\lambda\gamma_F^3\right),$$

$$\Phi\left(\frac{\gamma}{\gamma_F},\lambda\gamma_F^3\right)=\int_1^{\gamma/\gamma_F}d\xi\,\frac{\xi^2-1}{\xi^{5/2}}\exp\left[-\frac{4}{3}\lambda\gamma_F^3(\xi^3-1)\right]. \tag{10}$$

For $\lambda\gamma_F^3 \gg 1$ the function $\Phi \approx 2/(4\lambda\gamma_F^3)^2$, so that

$$j\approx\frac{2\sqrt{6\pi}}{4^4\pi^5}\frac{U_0}{w}\frac{e^{7/2}F_x^{5/2}}{(2m)^{1/4}w^{7/4}\hbar^{1/2}}\exp\left(-\frac{4}{3}\frac{\sqrt{2m}\,w^{3/2}}{\hbar eF_x}\right). \tag{11}$$

The condition $\lambda\gamma_F^3 \gg 1$ coincides with the condition for applicability of the quasiclassical approximation, which is usually used in obtaining the expression for the field-emission current.

Formula (11) differs from the usual tunneling formula by the factor (see, for example, Bethe and Sommerfeld[9])

$$\frac{\sqrt{6\pi}}{8\pi^3}\frac{U_0}{w}[\lambda\gamma_F^3(1+\gamma_F^2)^{1/2}]^{-1/2}. \tag{12}$$

This discrepancy in the preexponential factor is due to the approximate nature of the $\Psi_p(x, t)$ functions chosen. Having determined the form of the factor (12) in the limiting case $\gamma_F \ll 1$, we will use it as a correction to the expression for the current for arbitrary values of γ_F. If we take into account this factor, formula (11) gives

$$j=\frac{e^3F_x^2}{16\pi^2\hbar w}\exp\left(-\frac{4}{3}\frac{\sqrt{2m}\,w^{3/2}}{\hbar eF_x}\right). \tag{13}$$

Of course, the method used does not permit taking into account the action of the image force on the removed electron, a deficiency which is reflected in formula (13).

In the opposite limiting case, $\gamma_F \gg 1$, we can limit ourselves to only the first term in the sum over n. This gives

$$j=\frac{\delta e}{4\sqrt{6}}\frac{m\omega^2}{\hbar}\left(\frac{w}{\hbar\omega}\right)^{1/2}\exp(2n_0-\lambda\gamma_F^2)\left(\frac{e^2F_x^2}{8mw\omega^2}\right)^{n_0}, \tag{14}$$

where

$$\delta=\mathrm{E}[\lambda(1/2+\gamma_F^2)+1]-\lambda(1/2+\gamma_F^2).$$

This formula describes the photoelectric effect with absorption of n_0 photons: $n_0 \sim \lambda\gamma_F^2 = w/\hbar\omega$. For $\delta \ll 1$ the contribution from the first term in the summation over n can become smaller than the contribution from the second term. As a result we obtain a formula similar to (14) but with the substitutions $n_0 \to n_0 + 1$, $\delta \to 1 + \delta$. Thus, we accomplish a continuous transition from processes with absorption of n_0 photons to processes with absorption of $n_0 + 1$ photons.

As we have already shown above, formula (14) turns out to be more accurate for large values of n_0. Its applicability for small n_0 is due to the fact that condition (9) is not fulfilled simultaneously with the condition $\gamma_F \gg 1$. However, comparison with the corresponding expression for the photocurrent obtained by Smith[5] in second-order perturbation theory shows that even in this case ($n_0 = 2$) the errors are minor. Thus, for sodium ($w = 2.28$ eV) for $\omega = 3 \times 10^{15}$ cps, formula (14) gives $j \sim 10^{-29} F_X^4$ A/cm^2 (F_X is expressed in volts per meter). Smith's formula[5] for this case leads to the value $j \sim 10^{-30} F_X^4$ A/cm^2. For platinum ($w = 6.2$ eV, $n_0 = 4$) for the same frequency ω we obtain from (14) $j \sim 10^{-72} F_X^8$ A/cm^2. For $F_X \sim 10^9$ V/m (which is quite attainable in a focused laser beam) an emission-current density of the order of 1 A/cm^2 is obtained. The difficulties of experimental observation of the photocurrent in the field of the laser are discussed to some extent by Smith.[5]

It should be noted that for microwave frequencies the tunneling formula (13) becomes applicable starting with comparatively low fields (~ 300 V/m).

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