Electron emission from metal surfaces exposed to ultrashort laser pulses

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The emission current and the charge emitted from a metal surface exposed to a picosecond laser pulse are determined. The competition between photoelectric and thermionic emissions is considered. It is shown that for intensities in excess of a critical value the emission current is entirely due to thermionic emission. At the same time, the emission pulse is practically undelayed relative to the laser pulse. This is due to the small specific heat of the degenerate electron gas which is practically thermally insulated from the lattice during the duration of the ultrashort laser pulse. It is noted that the electron-lattice relaxation kinetics can be investigated by measuring thermionic emission produced by ultrashort laser pulses.

1. INTRODUCTION

The emission of electrons from metal surfaces exposed to laser pulses is due to two phenomena, namely, the photoelectric effect (usually multiphoton) and the heating of the metal which results in thermionic emission. The relative importance of these processes depends on the intensity and length of the laser pulses. Both emission mechanisms have been extensively investigated in the millisecond and nanosecond pulse length ranges.[1-6] In particular, it is shown in[2,3] that, unless special measures are introduced to prevent the heating of the surface, the observed electron current is largely due to thermionic emission. At the same time, by using oblique incidence of the laser beam on the cathode,[4,5] and by reducing the laser pulse length,[7-9] it is possible to observe the multiphoton photoelectric effect. There is particular interest in experiments with picosecond laser pulses which can be used, for example, to observe[8,9] the theoretically predicted[10] reduction in the effective number of photons necessary for ionization as the field strength is increased.

The competition between the photoelectric effect and thermionic emission under laser illumination was investigated theoretically in[7] where it was shown that the optimum conditions for observing the nonlinear photoelectric effect against the background of thermionic emission corresponded to sufficiently short laser pulses. The results reported in[7] cannot, however, be directly applied to the important case of picosecond laser pulses because the pulse length is then less than the characteristic time for the transfer of energy between the electrons and the lattice, and this leads to an appreciable increase in electron temperature and thermionic emission current in comparison with the equilibrium case discussed in,[7] and to certain other features which complicate the separation of the photoelectric effect from the thermionic effect.

We shall briefly consider below the question of the competition between these two mechanisms under illumination by ultrashort pulses. It is important to note, however, that, independently of this question of competition, the study of thermionic emission under picosecond pulse illumination is of major interest in itself because it enables us to investigate directly the kinetics of relaxation between electrons and the lattice in metals. The experimental situation, in this case, is apparently simpler than in the case of evolution of nonequilibrium phonon distributions, which has also been investigated under picosecond illumination (see[11] and the reference therein).

2. BASIC EQUATIONS

The establishment of equilibrium between the electrons and the lattice in metals has been considered in a number of publications,[12-14] where it was shown that, during the relaxation process, the metal must be looked upon as a two-temperature system, and the heat transfer coefficient between the electron and phonon subsystems was calculated. According to[12], the energy transferred by electrons to the lattice per unit volume of the metal per unit time is

$$\Delta E=\alpha(T_e-T_i), \quad \alpha=\frac{\pi^2}{6}\frac{mns^2}{\tau_e T_e}, \tag{1}$$

where s is the velocity of sound and τ_e is the electron relaxation time (for example, in the formula for the electrical conductivity of a metal[15]) which is formally regarded here as a function of the electron temperature T_e. The expression given by (1) is valid when the temperatures T_e and T_i are much greater than the Debye temperature. Since, in this case, $\tau_e \sim T_e^{-1}$, the heat transfer coefficient α between the electrons and the lattice is independent of temperature. Estimates based on the electrical conductivity of typical metals give values of α of the order of 10^{17} erg/cm^3.sec.deg. The relaxation time for the phonon temperature, which is of the order of c_i/α (c_i is the specific heat of the lattice), is then $\sim 10^{-10}$, and for laser pulses of shorter duration the violation of equilibrium between the electrons and the lattice becomes important.

As usual,[1,12-14] we shall write down the energy balance equations for the metal absorbing the laser pulse in the form

$$\begin{aligned} c_e(T_e)\frac{\partial T}{\partial t}&=\chi\Delta T_e-\alpha(T_e-T_i)+f(\mathbf{r},t),\\ c_i\frac{\partial T_i}{\partial t}&=\alpha(T_e-T_i). \end{aligned} \tag{2}$$

The validity of the macroscopic description given by (2) can be justified by simple estimates. To calculate the thermionic emission current, we must solve (2) and calculate the electron temperature on the surface of the metal. Since the laser pulse length is small, it is clear that this can be done in the one-dimensional case because, in all the cases of interest in practice, the temperature distribution on the surface is determined by the intensity distribution within the laser beam.

If we confine our attention to the one-dimensional problem and transform to dimensionless variables

$$\Theta_{e,i}(\xi,\ \eta)=(\alpha\chi)^{1/4}[I(1-R)]^{-1}T_{e,i}(x,\ t),$$
$$\xi=x(\alpha/\chi)^{1/4},\quad \eta=t/\tau$$

(τ is the pulse length, I is the maximum intensity in the laser pulse, and R is the reflecting power of the surface), we obtain the following set of equations:

$$\delta\Theta_e\frac{\partial\Theta_e}{\partial\eta}=\frac{\partial^2\Theta_e}{\partial\xi^2}-\Theta_e+\Theta_i-\varepsilon g(\eta)e^{-\varepsilon\xi},$$
$$\frac{\partial\Theta_i}{\partial\eta}=\mu(\Theta_e-\Theta_i). \tag{3}$$

In (3) we have taken into account the fact that at temperatures much smaller than the Fermi energy ϵ_F, the electron specific heat is a linear function of temperature. Moreover, we have neglected the temperature dependence of the light absorption coefficient κ. The dimensionless function $g(\eta)$ describes the temporal form of the laser pulse and is normalized to unity at the maximum.

The solutions of (3) depend on the following three dimensionless parameters:

$$\delta=\frac{I(1-R)}{\alpha\tau\sqrt{\alpha\chi}}\frac{\pi^2nk^2}{\varepsilon_F},\quad \mu=\frac{\alpha\tau}{c_i},\quad \varepsilon=\varkappa\sqrt{\frac{\chi}{\alpha}}.$$

Estimates show that, for experiments with picosecond pulses, we usually have $\delta \ll 1$, $\mu \ll 1$. By increasing the pulse length, we can transform to the case $\mu \sim 1$, and by increasing the intensity to the case $\delta \sim 1$ (it must be remembered that, for $\mu\delta \sim 1$, the approximate formula for c_e must be replaced by the exact one).

3. LIMITING CASES

The problem defined by (3) is nonlinear and, in general, requires numerical solution. This solution will be obtained in the following sections. Here, we confine our attention to a detailed analysis of some simple limiting cases.

We begin with the physically most interesting case $\delta \ll 1$, $\mu \ll 1$. It is readily shown that, in this case, the electron temperature on the surface of the metal is given by

$$\Theta_e(0,\eta)\approx\frac{\varepsilon}{\varepsilon+1}g(\eta). \tag{4}$$

It is clear from this formula that the dependence of the electron temperature on time repeats the form of the laser pulse without delay. This is so because the specific heat of the electron gas insulated from the lattice is very small ($c_e \sim \mu\delta c_i$, $\mu\delta \ll 1$) and, therefore, the electron temperature cannot follow the energy flux density in the laser pulse. In actual fact, there is a very small delay between the electron temperature and the laser pulse (of the order of $\delta\tau$). However, this delay is so small that the usual method of separating thermionic emission from the photoelectric current, based on the delay of the former, cannot be used in the present case.

When the parameter μ is not too small, it is a simple matter to calculate the corrections to (4) due to the heating of the lattice by energy transfer from electrons. The necessary formulas are given in[16].

We now consider one further limiting case, namely, that of an infinitely large absorption coefficient $\epsilon \to \infty$ (surface absorption). For $\mu \sim 1$ and $\delta < 1$, this was considered in detail in[1]. We note that an increase in μ leads to an increase in the electron temperature, since the energy lost through phonon generation, which is equal to the absorbed power for $\delta \ll 1$, is proportional to the temperature difference between the electrons and the lattice. Therefore, the simple formula given by (4) tends to underestimate the electron temperature.

In the limiting case corresponding to (4), we do, in fact, neglect the specific heat of the degenerate electron gas. This is definitely inaccurate for short times, but it is not difficult to establish a simple particular solution of (3) describing this stage when the intensity is a linear function of time. We shall not consider this in detail because the initial stage does not contribute appreciably to the emitted charge.

We note that the requirement $\delta \ll 1$ imposes a restriction on the maximum laser intensity. The inequality for I_{max} can be written in the form

$$I_{max}(1-R)\ll 5\cdot 10^{20}\tau \tag{5}$$

where I_{max} is in W/cm^2. When this condition is not satisfied, the complete set of equations given by (3) must be solved.

4. NUMERICAL SOLUTION

The set of equations given by (3) was integrated numerically for a broad range of values of the parameters δ, μ, and ϵ. The resulting solutions were then used to calculate the emission current and the total emitted charge. Most of the calculations were carried out for pulses $g(\eta)$ of triangular shape with unit half-width. The temperature of the surface as a function of time is shown in Fig. 1. When the electron temperature T_e is much less than the Fermi energy [in dimensionless variables, this means that $\Theta_e \ll (3\mu\delta)^{-1}$], the shape of the temperature pulse repeats the shape of the laser pulse with good accuracy. For high intensities, or long pulses, the temperature pulse spreads out and becomes delayed relative to the laser pulse; at the same time, the temperature pulse becomes asymmetric.

Figure 2 shows the dimensionless temperature Θ_e at the maximum as a function of δ for two values of the parameter μ and $\epsilon = 1$. For other values of ϵ, the values of Θ are obtained by multiplying by $2\epsilon/(\epsilon+1)$.

5. TOTAL EMITTED CHARGE. CONDITIONS FOR THE OBSERVATION OF THE NONLINEAR PHOTOELECTRIC EFFECT

We shall now calculate the thermionic emission current. In practice, one is interested in the case where the electron temperature is much less than the electron

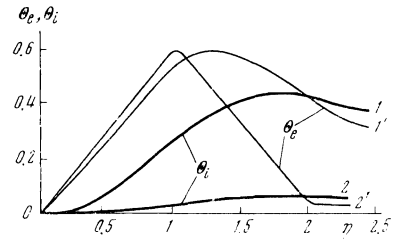

FIG. 1. The functions $\Theta_{e,i}(0, \eta)$ for $\delta = \mu = 1$ (curves 1 and 1') and $\delta = 0.03$ for $\mu = 0.1$ (curves 2 and 2').

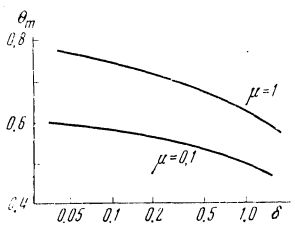

FIG. 2

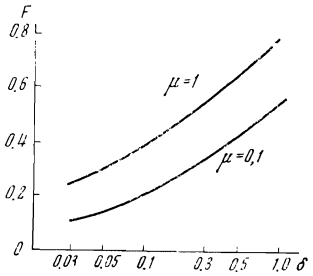

FIG. 3

FIG. 2. Values of $\Theta_e(0, \eta)$ at the maximum for different δ and μ for $\epsilon = 1$.

FIG. 3. Plots of $F(\delta, \mu)$.

work function. The emission current can be calculated from the Richardson formula by integrating the current density over the area of the illuminated spot on the metal surface. The emitted charge is obtained by successive integration with respect to time. It is important to remember that uncertainties in the constant A and fluctuations in the work function φ introduce substantial uncertainties into the results of these calculations. For the limiting case given by (4), the total charge is given by

$$q_t \approx 1.2 \cdot 10^{20} \tau S \varphi^2 \sigma^{-1/2} \left[\frac{\varepsilon}{p(\varepsilon+1)}\right]^{1/2} \exp\left[-\frac{p(\varepsilon+1)}{\varepsilon}\right], \qquad (6)$$

where we have assumed that the beam shape is Gaussian of radius r_0 and the integrals are evaluated by the Laplace method. In the above expression $S = \pi r_0^2$, $p = 1.2 \times 10^4 \varphi(\alpha\chi)^{1/2}/I(1 - R)$, φ is the work function in electron-volts, q_t is the charge in esu, I_m is the intensity at the maximum of the laser pulse, and σ is the second derivative of $g(\eta)$ at the maximum. For A we assume the value 3.6×10^{11} esu. Estimates based on (6) show that, for typical values (E = 0.01 J, $S = 0.01$ cm^2, $\tau = 10^{-11}$ sec), a charge of the order of 10^{-5} esu ($\sim 10^5$ electrons), which is readily recorded experimentally, can be obtained for a reflection coefficient $R \approx 0.8$.

The numerical calculations can be represented by the following formula which is analogous to (6):

$$q_t \approx 1.2 \cdot 10^{20} \tau S \varphi^2 F(\varepsilon, \delta, \mu) \left(\frac{\Theta_m}{p}\right)^{1/2} \exp\left(-\frac{p}{\Theta_m}\right).$$

In this expression, Θ_m is the maximum electron temperature. Calculations have shown that the function $F(\epsilon, \delta, \mu)$ changes by less than 10% when ϵ is varied between unity and ten. This can be neglected. The dependence of F on δ and μ is shown in Fig. 3.

We must now estimate the charge emitted by the metal surface as a result of the multiphoton photoelectric effect. The emission current density is $j_n = B_n I^n$, where n is the integral part of the number $1 + \varphi/\hbar\omega$. Different authors obtain different results for the constant B_n and, therefore, there are uncertainties in the values of j_n. For a triangular pulse of half-width τ and Gaussian intensity distribution over the beam cross section, the total emitted charge is

$$q_p = [(1-R) I_m]^n B_n \tau S [n(n+1)]^{-1}.$$

By setting $q_p = q_t$, we can determine the intensity I_m^* of the laser radiation above which thermionic emission predominates over photoelectric emission. Calculations performed for silver (n = 3 for $\hbar\omega = 1.78$ eV) with $B_3 = 3.5 \times 10^{-42}$ esu [17] yield $(1 - R) I_m^* \approx 3 \times 10^9$ W/cm^2. Similar results are obtained for other metals. We may, therefore, conclude that absorbed flux densities of the order of 10^9–10^{10} W/cm^2 can be regarded as the limiting values for which the photoelectric effect can still be observed against the thermionic emission background under illumination by ultrashort laser pulses.

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