SOME FEATURES OF THE INTERACTION BETWEEN SHORT LASER RADIATION PULSES AND MATTER

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Problems related to the competition between one- and many-photon interactions between radiation and matter are considered for short interaction times. The ionization of gases and electron emission from the surface of a metal under the action of short light pulses are considered as particular cases. It is shown that multiphoton ionization in dense gases (without cascade ionization) and the multiquantum surface photoeffect (without thermal emission of electrons) can be observed in "pure" form by shortening the pulse duration.

1. INTRODUCTION

IN the observation of different effects occurring when a pulse of electromagnetic interaction interacts with matter, a very important role may be played by the duration of the radiation pulse itself. In this article we wish to discuss the influence of the duration of the pulse on the observed effect, due to two competing physical processes of interaction between radiation and matter, occurring with different rates as functions of the radiation intensity S. Such competing processes can be, for example, single-photon and n-photon ($n > 1$) interaction processes, which are different in their physical nature but lead to the same experimentally-observable effect, namely that the probability of the first process (per unit time) is $w_i^{(1)} \sim S$, whereas that of the second is $w_2^{(2)} \sim S^n$. With this, if for some pulse duration τ the observed effect is due only to the first (single-photon) process (the n-photon process makes a negligibly small contribution), then by shortening τ and increasing the intensity S we can always attain the opposite situation, whereby the contribution of the first process is suppressed and the entire effect is governed by the n-photon interaction.

This circumstance is connected with the fact that the contributions from both processes are determined essentially by the total probabilities $p^{(1)} = w_1^{(1)}\tau$ and $p^{(2)} = w_n^{(2)}\tau$; shortening of τ and reduction of S make it possible in principle to decrease the probability $p^{(1)}$ practically without limit and at the same time increase the probability $p^{(2)}$.[1)] In the case when the observed effect has a threshold character, we can introduce the concept of the critical radiation-pulse duration τ_{cr}, at which the roles of the two different competing processes in the observed effect are reversed.

We shall consider further, from the foregoing point of view, the following two effects: ionization of gases, and emission of electrons from metal surfaces under the influence of laser-emission pulses.

2. IONIZATION OF GASES BY SHORT LIGHT PULSES

In the ionization of gases, two competing processes participate in the focus of the laser beam – cascade ionization and the n-quantum photoeffect (see the review [1]). For laser-radiation fluxes S smaller than $S_0 = c\hbar^2\omega^4/8\pi e^2\overline{v}^2$ (ω – radiation frequency, $\overline{v}$ – average electron velocity)[2)], the cascade ionization is a single-photon process [2] consisting of cascade multiplication of the free electrons of the gas as a result of acceleration by a (single-photon) antibremsstrahlung effect, with subsequent impact ionization of the atoms. The n-photon ionization process (the number n is equal to the integer part of $I/\hbar\omega + 1$, where I is the ionization potential of the atom and $\hbar\omega$ is the

[1)]Of course, we assume now that when the radiation density S increases, the first process will not cease to be single-photon.

[2)]For ruby-laser emission ($\hbar\omega = 1.78$ eV) and $v = 10^8$ cm/sec the intensity is $S_0 = 5 \times 10^{12}$ W/cm².

energy of the emission quantum) has been observed so far only in strong rarefied gases [3-7], when the mean free path of the electron is $l > a$ (a - linear dimension of the focusing region) (and the cascade ionization process is impossible.

The ideas raised in the introduction point to a method of experimentally separating the two discussed gas-ionization mechanisms; the method involves not variation of the gas pressure (as was done in [3-7]), but variation of the laser-emission pulse duration. With such a method of separation, it is possible to observe multiphoton ionization in a dense gas (i.e., when $l \ll a$) with the same visual effect (spark production) as in experiments on cascade ionization. The effect should have here a threshold character and there should exist a critical pulse duration τ_{cr} which depends on n and on the gas density N_a, namely, at pulse duration $\tau < \tau_{cr}$ the main contribution to the ionization is made by the n-quantum photoeffect, and at $\tau > \tau_{cr}$ it is made by the electron cascade.

We present quantitative estimates for the duration τ_{cr} and for the threshold radiation intensity S_{cr}^{thr} corresponding to this duration.

The quantities τ_{cr} and S_{cr}^{thr} should obviously be the solutions of the systems of two equations:

$$\tau = \theta_c(S), \quad w_n(S)\tau N_a = N_e^{thr}, \tag{1}$$

where $\theta_c(S)$ is the lifetime of one generation of electrons in the cascade-ionization process, $W_n(S)$ is the probability of n-photon ionization of the atom, N_e^{thr} is the threshold density of the electrons produced as a result of the ionization. For dense gases the value of N_e^{thr} is essentially determined by the condition $\alpha a \sim 1$ (α - coefficient of absorption of light in the produced plasma) and amounts to approximately 10^{17}—10^{18} cm^{-3} [1]; as will be shown later, the values of τ_{cr} and S_{cr}^{thr} depend little on N_e^{thr} so that it is not necessary to make the values of N_e^{thr} more precise.

For simplicity in the further analysis we shall assume that the gas density N_a is such that the electron loss due to their escape from the focusing region can be neglected ($l \ll a$, $D\tau \ll a^2$, D - coefficient of spatial diffusion of the electrons). We neglect also the energy loss of the electrons to elastic collisions (such losses are significant only for a light gas such as helium [1]). Then the lifetime $\theta_c(S)$ is determined from the condition (see [1])

$$\frac{e^2E_0^2}{m\omega^2}\nu_{eff}\,\theta_c = I/\alpha, \tag{2}$$

where $E_0^2 = 8\pi S/c$ is the square of the amplitude of the electric field of the light wave, ω is the frequency of the wave, e and m are the charge and mass of the electron, ν_{eff} is the effective frequency of the elastic collisions of the electrons with the atoms, and α is the probability that the electron will "jump through" the atom-excitation band [1]. Substituting in (2) the relation $\nu_{eff} = \sigma_{tr}\bar{v}N_a$ (σ_{tr} is the transport collision cross section and $\bar{v}$ the average velocity of the electrons) and introducing the symbol $\omega_t = eE_0/\sqrt{mI}$, we obtain $\theta_c(S)$:

$$\omega\theta_c = \frac{A}{N_a}\left(\frac{\omega_t}{\omega}\right)^{-2}, \tag{3}$$

where

$$A = \omega/\alpha\sigma_{tr}\bar{v}. \tag{4}$$

To determine the $w_n(S)$ dependence we assume that there is no resonance in the photoeffect on the atoms of the investigated gas, and that the number n is sufficiently large to be able to use the Keldysh formula [8,1]:

$$w_n = 2^{-3n}B\omega n^{3/2}(\omega_t/\omega)^{2n}. \tag{5}$$

Here B is a dimensionless quantity, the values of which for different atoms are close to unity. The values of $B^{1/n}$ will furthermore be assumed equal to unity. Substitution of (3) and (5) into the system (1) and solution of the latter yields

$$\omega\tau_{cr} = \left[\frac{(A/8)^n}{N_e^{thr}}\right]^{1/(n-1)}\frac{n^{3/2(n-1)}}{N_a}, \tag{6}$$

$$S_{cr}^{thr} = \frac{mc\omega^2 I}{8\pi e^2}\frac{8^{n/(n-1)}}{n^{3/2(n-1)}}\left(\frac{N_e^{thr}}{A}\right)^{1/(n-1)}.$$

It is seen from these formulas, in particular, that the critical duration τ_{cr} is approximately inversely proportional to the initial gas pressure p, whereas the threshold intensity S_{cr}^{thr} does not depend at all on the pressure[3)].

We present a numerical estimate for τ_{cr} and S_{cr}^{thr} at $n = 3$; this estimate will pertain, in particular, to the case of ionization of alkali-metal vapor (Na, K, Rb, Cs) by ruby-laser radiation ($\hbar\omega = 1.78$ eV). At a gas density $N_a = 10^{19}$ cm^{-3}, the probability α can be assumed to be of the order of 0.1 (see [1]), and therefore $A \approx 10^{23}$ cm^{-3}; With this [4)]

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3)The exact relation $\tau_{cr} \sim 1/p$ is distorted by the relatively weak dependence of the probability α on p [1]; the statement that S_{cr}^{thr} on the pressure is accurate if one neglects the exceedingly weak dependence of the quantity $\alpha^{1/(n-1)}$ on p.

4)The value obtained for S_{cr}^{thr} satisfied the condition $S_{cr}^{thr} \ll S_0 = 5 \times 10^{12}$ W/cm² (see footnote 2)), and consequently the cascade-ionization process remains single-quantum. Further, S_{cr}^{thr} and τ_{cr} correpsond to a parameter value $(\omega_t/\omega)^2 = (A/N_a)/\omega\tau_{cr} \approx 10^{-2}$, i.e., much smaller than unity, and therefore the process of ionization by radiation should have, as before, the same character as the photoeffect, and not the character of tunnel ionization [8].

$$\tau_{cr} \approx 0.4\cdot 10^{-9}\,\text{sec},\quad S_{cr}^{thr} \approx 2.7\cdot 10^{11}\ \text{W/cm}^2. \qquad (8)$$

We have put in these estimates $N_e^{thr} = 10^{17}\,\text{cm}^{-3}$ and $I = 5$ eV.

3. ELECTRON EMISSION FROM A SURFACE OF A METAL UNDER THE INFLUENCE OF SHORT LIGHT PULSES

When a pulse of laser radiation is incident on the surface of a metal, the following phenomena can be observed: thermionic emission, sublimation of the metal, and surface photoeffect. In connection with the development of laser physics, it has become possible in principle to observe the multiquantum surface photoeffect [9,10]. However, an experimental observation of the effect is greatly hindered by thermionic emission [11-13].

Farkas et al.[14] proposed and developed experimentally a method for suppressing the thermionic emission of the electrons, by using oblique incidence of the laser beam on the surface of the metal. We propose a different method for suppressing thermal effects, based on the concepts developed above. Thermionic emission of electrons and sublimation of the metal, which are connected with absorption of radiation by free electrons of the metal, are essentially due to a single-photon mechanism and can therefore be excluded by choosing the proper duration τ of the radiation pulse. Our problem now consists of finding the critical pulse duration τ_{cr} satisfying the following conditions: When $\tau < \tau_{cr}$, the sublimation of the metal is completely eliminated and the electron emission during the duration of the pulse is due essentially only to the multiquantum photoeffect. Such a duration τ_{cr} exists when $n \geq 3$.

The sought critical duration τ_{cr} and the corresponding threshold intensity of the radiation S_{cr}^{thr} should be determined by solving a system of equations analogous to (1):

$$\tau = \theta_T(S),\quad j_n(S)\tau = q_{thr} \qquad (9)$$

Here q_{thr} is the threshold value of the charge emitted from a unit surface; in our problem it is determined by the sensitivity threshold of the charge indicator, and we shall assume in our numerical estimates $q_{thr} = 10^{-15}/F$ Coulomb/cm^2 (F - area (in cm^2) illuminated by the laser beam); $j_n(S)$ is the density of the electric current from the surface of the metal, due to the n-quantum photoeffect; $\theta_T(S)$ is the duration of a pulse of radiation with intensity S, during which thermionic emission of a charge βq_{thr} takes place, where $\beta < 1$ (the exact value of this quantity should be determined by the required accuracy of suppression of the thermionic emission).

To estimate the intensity of the thermionic emission of the electrons during the radiation pulse, we shall use Richardson's formula and substitute in it that value of the temperature, which is produced on the surface of the metal when radiation with constant intensity S is normally incident on it:

$$T = \frac{2(1-R)S}{K}\left(\frac{\varkappa t}{\pi}\right)^{1/2}, \qquad (9')$$

where K and κ are respectively the thermal-conductivity and the temperature-conductivity coefficients of the metal (we do not take into account here the dependence of K and κ on the temperature), R is the coefficient of reflection of the radiation from the surface, and t is the time elapsed from the start of the irradiation.

Integrating the thermionic-current density over the pulse-duration time τ, we obtain the following expression for the charge q emitted during that time from a unit surface:

$$q = A_0\left(\frac{2(1-R)S}{K}\right)^3 \frac{2\pi k}{\varkappa\varphi}\left(\frac{\varkappa\tau}{\pi}\right)^{5/2} \exp\left[-\frac{\varphi K(\pi/\varkappa\tau)^{1/2}}{2k(1-R)S}\right], \qquad (10)$$

where $A_0 = 4\pi k^2 em/h^3 = 120$ A/cm^{2}deg^2, φ is the work function of the metal, and k is Boltzmann's constant. This formula is true if

$$\frac{\varphi K}{2(1-R)S}\left(\frac{\pi}{\varkappa\tau}\right)^{1/2} \gg 1, \qquad (11)$$

which essentially coincides with the condition $S \ll S_{thr}^s$, where S_{thr}^s is the threshold intensity of radiation for sublimation of the metal. Indeed, it is easy to show that

$$S_{thr}^c = \frac{\lambda\rho}{1-R}\left(\frac{\varkappa}{\pi\tau}\right)^{1/2}, \qquad (12)$$

where λ is the specific sublimation energy and ρ is the density of the metal. Substituting for S in the right side of (11) the intensity (12), we obtain a ratio

$$\frac{\pi\varphi K}{2k\lambda\rho\varkappa} = \frac{\pi}{2}\frac{\varphi/k}{\lambda/C}$$

(C - specific heat of the metal) which is of the order of unity for all metals.

Formula (10) enables us to determine $\theta_T(S)$, provided we substitute in place of q the quantity βq_{thr}, replace τ by θ_T, and solve the resultant equation with respect to θ_T. In this case condition (11) is satisfied for all real values of S, τ, and q_{thr}, and by the same token the critical duration τ_{cr} determined from (9) will automatically satisfy the condition that the sublimation be excluded.

For the current density $j_n(S)$ due to the n-quantum photoeffect we shall use a formula derived in [10]:

$$j_n(S) = 2^{-3n} B \frac{em\omega^2}{\hbar} n^{1/2} \left(\frac{\omega_t}{\omega}\right)^{2n}, \qquad (13)$$

where the dimensionless quantity B, just as in (5), is close to unity and is henceforth assumed equal to unity; $\omega_t^2 = e^2E_0^2/m\varphi = 8\pi e^2 S/mc\varphi$.

Substituting the functions $\theta_T(S)$ and $j_n(S)$ in (9) and eliminating S, we obtain the following equation for τ_{cr}:

$$\omega\tau_{cr} = n^{1/(n-2)} \left(\frac{em\omega/\hbar}{q_{thr}}\right)^{2/(n-2)} \times \left\{\frac{\pi e^2 K}{2mc\omega^2 k(1-R)} \left(\frac{\pi\omega}{\varkappa}\right)^{1/2} \Big/ \left[\ln Z + \frac{5n-6}{2n}\ln(\omega\tau_{cr})\right]\right\}^{2n/(n-2)}, \qquad (14)$$

with Z defined as

$$Z = \frac{16A_0 k\varphi^2}{\pi^2 n^{3/2n}\varkappa\beta q_{thr}} \left(\frac{\varkappa}{\pi\omega}\right)^{5/2} \left(\frac{mc\omega^2(1-R)}{e^2K}\right)^3 \left(\frac{q_{thr}}{em\omega/\hbar}\right)^{3/n}. \qquad (15)$$

The threshold intensity corresponding to the duration τ_{cr} is

$$S_{cr}^{thr} = \frac{mc\omega^2\varphi}{\pi e^2 n^{1/2n}} \left(\frac{q_{thr}}{em\omega/\hbar}\right)^{1/n} \frac{1}{(\omega\tau_{cr})^{1/n}}. \qquad (16)$$

We present a numerical estimate of τ_{cr} and S_{cr}^{thr} for silver (φ = 4.7 eV, K = 1 cal/cm-sec-deg, κ = 1.7 cm^2/sec, R = 0.8) and for ruby-laser emission ($\hbar\omega$ = 1.78 eV, n = 3); we put q_{thr} = 10^{-15} Coul/cm^2, β = 0.1.

Recognizing that ln Z $\approx 10^2$, the calculation of τ_{cr} by formula (14) can be carried out by an iteration method, discarding during the first stage the term with $\ln(\omega\tau_{cr})$. In the second stage we obtain here $\tau_{cr} \approx 0.9 \times 10^{-9}$ sec, $S_{cr}^{thr} \approx 1.2 \times 10^8$ W/cm^2. (We note that such values of S and τ lead, in accordance with (9′) to heating of the surface of the metal by approximately 250°C.)

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