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To cite this article: E A Mikhin et al 2017 J. Phys.: Conf. Ser. 917 092009

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Nonlinear electron emission in ultrashort laser pulses and rescattering suppression

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Abstract. The process of nonlinear electron emission from a metal surface under the action of femtosecond laser pulse with moderate intensity \(-10^{11}\) W/cm\(^2\) is considered. One-dimensional model is formulated, taking into account the advantage of the \(p\)-polarized light in the nonlinear emission. The time dependent Schrödinger equation with fixed equilibrium boundary conditions is solved in the half-space using the Laplace transform technique. The energy spectrum of emitted electrons is presented, including analysis of dependence on the laser pulse parameters. In the framework of classical mechanics, the impact of additional dc electric field on the rescattering of emitted electron on the metal surface is investigated. The model reproduces the fiches of electron energy distribution and can be used at arbitrary Keldysh parameters.

1. Introduction

The nonlinear cold emission of electrons from a metallic nano-needle under the action of a short laser pulse has a number of unique features that are very attractive for possible practical applications. For its experimental realization, it is sufficient to have moderate intensities of the laser field, since in the nano-needle, surface plasmons are generated at the metal-vacuum interface, which leads to a significant enhancement of the laser field near the tip [1]. Arising surface plasmon resonance is a collective oscillation of conduction band electrons that typically occurs at optical frequencies in noble metals. The characteristic size of the amplification region is several tens of nanometers, which makes it possible to obtain a practical point source of electrons.

A strong nonlinear dependence of the photoemission on the laser field strength leads to the formation of free electrons mainly at small intervals of time near the amplitude values of the field. As a result of nonlinear photoemission, electron bunches are formed with duration less than the period of oscillations of the laser pulse. Nonlinearity is also manifested in multiphoton above-threshold photon absorption, which leads to the ejection of electrons with energies much greater than the energy of the light quanta.

The emission of electrons from the nano-needle caused by the field of an ultrashort laser pulse can be used in a number of laser applications related to monitoring, control and diagnostics of various nano-sized objects [2, 3]. Generated in this way ultrashort coherent electron bunches are crucial to the free electron lasers, laser acceleration of relativistic electrons, picosecond cathodoluminescence, and femtosecond electron diffraction. They would enable exciting technological developments such as four-dimensional time-resolved electron microscopy, spectroscopy, and holography.
2. Theoretical model

The experimental data [4] show a dependence on the laser polarization, indicating that the emission yield is about one order of magnitude larger for the $p$-polarized light electric field normal to the surface than for the $s$-polarization. This result could apparently suggest, according to the model proposed by Broudy [5], that the extraction mechanisms are directly related to the component of the electric field normal to the surface. Appropriate analytical one-dimensional model of electron emission from a metal surface, under the influence of both a dc electric field and laser field illumination with exact solution was derived to electron emission for arbitrary values of laser field, laser frequency, dc electric field, and metal work function [6]. However, the considered electron emission from a metal-vacuum interface driven by a combination of a dc electric field and a light field was limited to a single carrier frequency.

We improve the model, describing the motion of electrons in a half-space under the influence of arbitrary pulsed laser field. The boundary conditions will ensure the asymptotic behavior of the metal electronic states outside its surface. We write down the one-dimensional Schrödinger equation for an electron in the laser pulse field, described by the vector potential $A(t)$ for the half-space outside the metal

$$i \frac{\partial \psi(x,t)}{\partial t} = \frac{1}{2} \left( \hat{p} + \frac{1}{c} A(t) \right)^2 \psi(x,t), \quad x > 0. \tag{1}$$

Here $\hat{p}$ is the momentum operator, $\psi(x,t)$ is a wave function. We use the atomic system of units in which $m = \hbar = e = 1$.

Consider laser field with a strength $F(t) \ll 1$, and assume that external electromagnetic field weakly distorts electron states in the metal. The initial states belonging to the solid target band structure are taken as exponentially decreasing into the vacuum. Under our assumptions, the wave function $\psi(x,t)$ satisfies the boundary conditions

$$\psi(0,t) = B \exp(-i E_0 t),$$
$$\psi'(0,t) = -\kappa B \exp(-i E_0 t).$$

Here $B$ is the normalization constant, $\kappa = \sqrt{2|E_0|}$, $E_0$ is the energy of an electron inside the metal, the prime in the second line of Eq. (2) denotes the derivative with respect to the space variable.

The solution of Eq. (1) with the boundary conditions (2) in the positive half-space ($x > 0$) can be found using the Laplace transform $f(s,t) = \int_0^\infty \exp(-sx)\psi(x,t) \, dx$, where $f(s,t)$ is transform of the function $\psi(x,t)$, $s$ is a complex variable. In the transform space Eq. (1) takes the form

$$i \frac{\partial f(s,t)}{\partial t} = \frac{1}{2} \left( -is + v(t) \right)^2 f(s,t) + \left\{ s \frac{\alpha(t)}{2} + \frac{\beta(t)}{2} + iv(t) \alpha(t) \right\}. \tag{3}$$

For more evident written we introduce notations: $\alpha(t) = \psi(0,t)$, $\beta(t) = \psi'(0,t)$ and $v(t) = A(t)/c$. When enter additional parameters for a classical electron displacement under the action of the laser field and the integral from the kinetic energy:

$$a(t_1,t_2) = \int_{t_1}^{t_2} v(t) \, dt_2 \quad \text{and} \quad S(t_1,t_2) = \int_{t_1}^{t_2} \frac{v^2(t_2)}{2} \, dt_2. \tag{4}$$
respectively, the solution of Eq. (3) can be written as

$$f(s,t) = -i\int_0^T \left( s\frac{\alpha(t_1)}{2} + \frac{\beta(t_1)}{2} + iv(t_1)\alpha(t_1) \right) \exp \left\{ i \left( \frac{s^2(t-t_1)}{2} + is\alpha(t_1) - S(t,t_1) \right) \right\} dt_1. \tag{5}$$

Substitution $s = ip$ into Eq. (5) gives the solution of the original Eq. (1) in the momentum representation, and the calculation of spectral distribution for the emitted electrons as a function of the pulse parameters is reduced to the integral

$$f(p,T) = -iB\int_0^T \left( \frac{ip}{2} + iv(t_1) \right) \exp(-iE_0t_1) \exp \left\{ i \left( -\frac{p^2}{2}T - p\alpha(T,t_1) - S(T,t_1) \right) \right\} dt_1. \tag{6}$$

Here $T$ is duration of the pulse. With the aid of the obtained function $f(p,T)$, the electron energy spectrum can be written as

$$\frac{dN}{dE} = \frac{f(\sqrt{2E},T)^2}{\sqrt{2E}}. \tag{7}$$

Eq. (7) describes a spectrum of emitted electrons having definite initial energy. However, we need take into account that electrons in the metal are distributed over the initial energies. According to the Sömerfeld theory, the number of electron states in a unit volume of a metal per unit energy interval is determined as

$$\frac{dn}{dE} = \frac{1}{\pi^2} \frac{\sqrt{2(E_0 - U_0)}}{1 + \exp\left( \frac{E_0 - E_f}{k\theta} \right)} \tag{8}$$

Here $\theta$ is the metal temperature, $E_f$ is the Fermi energy, $U_0$ is the energy corresponding to the bottom of the conduction band, $k$ is the Boltzmann constant.

After integration over the initial electronic states energy and taking into account Eq. (8), we obtain the final equation that gives the energy spectrum of the emitted electrons

$$\frac{dN}{dE} = \frac{1}{\pi^2} \int_{U_0}^{E_f} \frac{E_0 - U_0}{E} \left( 1 + \exp\left( \frac{E_0 - E_f}{k\theta} \right) \right)^{-1} f_{E_0}(\sqrt{2E},T)^2 dE. \tag{9}$$

The integration in Eq. (9) is carried out from the bottom of the metal conduction band $U_0$ to the energy $E_f$. At the room temperature, the population of electronic states above the Fermi level can be neglected.

3. Electron spectrum

To consider the impact of the laser field on the spectrum of emitted electrons, let us determine the shape of the pulse. In the few-cycle regime, the electric field $F(t)$ can be written as $F(t) = f(t)\cos(\omega t + \varphi)$, where $f(t)$ is the pulse envelope, $\omega$ is the laser carrier frequency. In this parameterization the electric field depends on the carrier absolute phase $\varphi$, although the envelope is the same for all the pulses. Let us assume for further examination the Gaussian envelope, so

$$F(t) = F_0 \exp\left( -\frac{t^2}{\tau^2} \right) \cos(\omega t + \varphi). \tag{10}$$
Here $F_0$ is the amplitude of the laser field strength, $\tau$ determines the laser pulse duration.

**Figure 1.** Theoretical and experimental [8] photoelectron spectrums for tungsten target averaged over the carrier-envelope phase.

**Figure 2.** The maximum electron energy as a function of the ratio the ac field strength to the dc electric field.

The magnitude of the laser pulse field influences efficiently on the electron emission dynamics. Increasing the field strength leads to the greater yield of electrons according to higher photoemission probability and stretching the electron energy spectrum as the manifestation more effective electron acceleration by the laser field. Emitted electron spectrum is shown in figure 1 for different strengths laser pulse. Solid curves represent the results of calculations within the framework of our model. Two amplitude values of the electric strength of the laser field of 6.9 V/nm and 8.7 V/nm correspond to these curves in the order from below to the top. Other parameters taken to calculate the electron spectrum were $\lambda = 800 \text{ nm}$, $\varphi = 0$, $\tau = 6.5 \text{ fs}$ and $E_f = -4 \text{ eV}$. The Keldysh parameter was 1.8 and 2.3, and the nonlinear emission was intermediate between the multiphoton emission and the tunneling, more similar to the first one. This is confirmed also by existence characteristic peaks in the electron spectrum, which of them demonstrates the result of fixed number absorbed photons of the laser field. The minimum number of photons required for above-threshold electron emission is $K = \left(\left\lfloor \frac{E_f}{\omega} \right\rfloor \right)$, where $\left\lfloor \ldots \right\rfloor$ is the integer part of a number. In the present case $K = 3$, and the position of the first peak in figure 1 is located according to this number. Subsequent peaks show a lower yield of electrons and are separated by almost equidistant gaps $\sim \omega$. The photon spectrum of incident laser light is broad enough when one has a very short pulse, and in addition some contribution to the photoemission is produced by electrons having initial energy below the Fermi level. As a result, the energy spectrum of the emitted electrons is broadened, and peaks in the spectrum are not strictly equidistant.

Experimental realization of electron nonlinear cold emission from a metal surface implies usage relatively large laser fields. However, increasing laser intensity leads to heating of target and growth of thermoemission instead of the nonlinear field process. To overcome this difficulty, cozily to operate with ultrashort laser pulses of moderate intensity and magnify the field, using effect of plasmon superfocusing on the tip of a nanoneedle. The gain coefficient for the electric field strength by this mechanism can reach $\sim 10$ [7], and its magnitude strongly depends of a tip radius, thus specially prepared targets with nanostructured surfaces are exploited. Nevertheless, even with special treatment of the target surface and excitation of plasmons in it, the electron yield per laser pulse is small, and for a set of representative statistics, a large number series of laser pulses are used. Along with this, an additional dc electric field is applied, which sometimes is comparable in magnitude to the laser field strength. The field gain effect for electromagnetic wave we incorporate in the model as additional factor in Eq. (10), at the same time, the impact of ad substantial electric field remains out of frame present consideration.
To demonstrate the typical shape of the experimental electron spectrum, we plotted two of them in the figure 1, following the data [8] for cold nonlinear electron emission from the tungsten nanoneedle with radius of tip ~50 nm stimulated by laser radiation. Under these conditions, plasmons generation gives a considerable magnification of the field near the needle apex. The experimental ac electric field strengths of 4.32 V/nm and 4.98 V/nm correspond to these curves in the order from below to the top. The dc electric field strength in experiment was 0.8 V/nm. Other laser field and metal parameters are exactly the same as in the numerical simulations. Experimental electron spectrums have the local peaks separated with the gaps nearly equal the laser frequency as it is seen also in theoretical dependencies that indicates domination the laser field over the dc field and shows multiphoton character of emission. Because experimental conditions essentially differ from the theoretical model parameters, it would be very odd to expect exact coincidence of the results, and we plotted these curves together only to show evident correlation of the dependencies, but not to explain directly the experimental data.

The above calculations do not take into account the reflection of electrons on the metal surface, whereas it is known that this effect substantially increases the maximum final energy in the analogous process of nonlinear ionization of atoms [9]. A complete model involving the rescattering process should be quantum mechanical, but the upper limit of the electron spectrum can be estimated using the classical equations of motion, taking into account the vibration motion of an electron in the laser field. After the emission from the metal, when the direction of the field is changed, the electron can return and be reflected from the surface of the metal. This process increases the final drift velocity of the emitted electrons and influences on the observed electron spectrum. Thus, rescattering leads to the appearance of a high-energy spectral plateau, beginning near the energy $2U_p$ and abruptly terminating at the energy ~ $10U_p$ [10].

An additional accelerating dc electric field in the nonlinear emission experiments impede return of electron back to the metal surface and at some critical ratio $\alpha$ of the laser field amplitude to the dc field this effect completely disappears. The result of the classical simulation of the maximum electron energy as a function of $\alpha$ is shown in figure 2. When $\alpha < 1.57$, the dc electric field completely suppress rescattering, and the maximum energy of the electron is $2U_p$. With a decrease in the relative strength of the dc electric field, part of the emitted electrons returns to the metal surface and is rescattered on it. However, the dc electric field, in this case, also inhibits the returning electrons, making the collision conditions with the metal surface far from optimal to obtaining the maximum possible energy. With the decrease of the electric field (at $\alpha > 1.57$), the maximum energy of the emitted electron increases to ~ $10U_p$.

In figure 2 the small circles indicate the estimation of the maximum electron energy for experimental field ratios [8]. These energies have evident correlation with experimental cutoff. Significantly to note that due to the nonlinearity of emission, a large part of the free electrons is formed near the maximum of the laser field strength. For this group of electrons, the rescattering disappears at a relatively weak dc electric field, since before changing the direction of the laser field they already gain some velocity in the direction from the metal surface. Thus, the ones shown in
figure 2 the estimates refer only to the limiting values of the electron energies and do not characterize the very form of the spectrum.

Strong nonlinear dependence the probability of the electron photoemission on the laser field strength leads to a sharp change in the electron currents during a period of field oscillation. When the laser pulse duration greatly exceeds the period of oscillation, series of sub-femtosecond electron pulses are formed, which follow one after the other with the laser field period. For the few-cycle laser pulses, the process looks different since a variation a carrier wave phase with respect to the envelope strongly modulates the peaks of the laser field strength impact the spectrum of the emitted electrons. This effect does not influence the main parameters of photoemission and the overall shape of the electron spectrum, but changes the positions and heights of the local energy maxima. Figure 3 shows the results of calculation the electron spectrum for different carrier phases, where the peaks modulation is clearly-visible on all three curves.

Increasing the electron energy the phase effects are manifested to a greater extent. To demonstrate this, the inset in figure 3 shows the normalized to the unity maximum magnitude of the second and sixth peak as a function of phase. These dependences are qualitatively consistent with experimental data [11].

4. Conclusions
We have formulated the one-dimensional analytical model for the nonlinear field electron emission from a metal surface under the ultrashort laser pulse action. In this framework we investigated the influence the laser pulse parameters on the energy spectrum of the emitted electrons. The model is predicted the general shape of the electron energy distribution and it gives a reasonable qualitative description the dependences on the laser pulse parameters. Our model was developed for ac electric field, but experimental measurements were produced sometimes in the presence of essential additional dc electric field. Accelerating electrons by additional dc electric field reduces the rescattering effect, and decreases their maximum energy. Thus, dc electric field can be used to control the form of the energy spectrum. For the case of complete suppression of the rescattering, our model allows us to make a qualitative comparison with the experimental electron spectrum and give its interpretation. In the next paper we shall present a more complete quantum-mechanical description of the nonlinear electron emission taking into account wave packet rescattering as the next step in the development of the theory.

Acknowledgments
This work was supported by RFBR (grant No 16-32-00255) and Ministry of Science and Education RF (project No. 3.6369.2017).

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