Above-threshold photoelectric effect at a metal

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A multiphoton photoelectric effect at a metal in an intense laser field is analyzed. The light is propagating along the surface of the metal. An interpretation is offered for the above-threshold peaks in the spectrum of photoelectrons. The Coulomb interaction of these electrons with the image potential of the electron cloud in the metal is taken into account in this interpretation. The observed electron spectrum can be explained on the basis of a multiple scattering of an electron by the image potential, with a field photon being captured in each scattering event. It is also shown that the above-threshold peaks arise in fields far weaker than the corresponding fields in the case of atoms (in above-threshold ionization).

1. Introduction. Recent experiments[1–3] have revealed that the spectrum of electrons resulting from the multiphoton photoelectric effect at a metal has the same features as in the case of above-threshold ionization of atoms (ATI).[4–6] This spectrum consists of some fairly narrow, equidistant lines separated by intervals equal to the photon energy $\hbar\omega$. The maximum of the envelope of these lines shifts up the energy scale with increasing power of the laser light. The number of observed lines also increases with increasing laser power.

In contrast with ATI, however, the intensity of the laser light at which the above-threshold photoelectric effect at a metal is observed is $\approx 10^9$ W/cm^2. The threshold intensity required for ATI is higher by at least three orders of magnitude ($\approx 10^{12}$ W/cm^2 for neodymium-glass lasers). We do not yet have a reliable explanation for this difference.

Experiments[1–3] have shown that the photoelectric effect gives rise to an electron cloud near the cathode, and this cloud is confined near the surface by image forces. The potential of the corresponding Coulomb field which the photoelectron experiences is proportional to the number of electrons in the cloud. Because of the Coulomb interaction with the image, the photoelectron is not a free particle, and it is capable of absorbing (and emitting) field quanta.

In this letter we analyze the above-threshold photoelectric effect on the basis of a model of multiple Coulomb scattering of an electron by the image potential, accompanied by the pickup of a field photon in each scattering event. A similar approach was taken in our earlier paper[7] in a description of ATI in the case of atoms.

2. Statement of the problem. A photoelectron localized near the cathode surface is in the field of intense laser light and simultaneously in the Coulomb field of the image of the electron cloud. The interaction of a photoelectron with a wave is taken into account by the Keldysh approach.[8]

The potential energy of a photoelectron in the image field is described by the operator ($\hbar = c = 1$)

$$\hat{V} = -Z_{\text{eff}} e^2/[\rho^2 + (z+a)^2]^{1/2}, \tag{1}$$

where $Z_{\text{eff}}e$ is the effective image charge, and z and $\rho = (x^2+y^2)^{1/2}$ are respectively the transverse and longitudinal (with respect to the metal surface) coordinates of the electron (the origin of coordinates is at the surface of the metal, so the z coordinate of the center of the image is $-a$).

The transverse dimension of the cloud, a, is found from the equilibrium condition[9]

$$Z_{\text{eff}} e^2/a^2 = eE_{\text{ext}},$$

where E_{ext} is the strength of the extraction field which accounts for the saturation photocurrent (this field is calculated below).

Interaction (1) in the photoelectron-metal system is taken into account in the procedure for iterating the amplitude for the transition of an electron from an near-threshold initial state to a highly excited final state in the continuum.

The state of the photoelectron in the continuum in the presence of a laser wave is specified by the wave function

$$\Psi_p(\mathbf{r},t) = \exp[i(\mathbf{p}_{\parallel} \vec{\rho} + p_{\perp} z - \epsilon_p t)] \cdot \exp\left\{ i\left[\frac{e\mathbf{E}_0 \cdot \mathbf{p}}{m_e\omega^2} \cos \omega t + \frac{(eE_0)^2}{8m_e\omega^3} \sin 2\omega t\right]\right\}, \tag{2}$$

where $\mathbf{p}_{\parallel}$ and $\mathbf{p}_{\perp}$ are respectively the tangential and transverse components of the electron momentum with respect to the metal surface, $\epsilon_p = p^2/2m_e$ is the kinetic energy of the electron as the wave field is turned off adiabatically, $\mathbf{E}_0$ is the electric field of the wave, which is directed along the z axis, and ω is the wave frequency. The wave function in (2) is a nonrelativistic analog of the Volkov solution, and the basis of eigenfunctions of the problem to be solved is constructed on the basis of this wave function (this is the Keldysh method).

The amplitude for the process which corresponds to a multiphoton photoelectric effect followed by scattering of the photoelectron by the image Coulomb potential is given by

$$A_{\mathbf{p}'}(t) = -i \int^t dt' \int \frac{dp}{(2\pi)^3} \langle \Psi_{\mathbf{p}'}(\mathbf{r},t') | \ \hat{V} | \ \Psi_{\mathbf{p}}(\mathbf{r},t') \rangle A_{\mathbf{p}}(t')$$

$$-i \int^t dt' \int \frac{d\mathbf{p}}{(2\pi)^3} \, 2\pi \, \frac{Z_{\text{eff}} e^2}{| \mathbf{p}'_{\parallel} - \mathbf{p}_{\parallel} |} \exp[-|\mathbf{p}'_{\parallel} - \mathbf{p}_{\parallel}| a][|\mathbf{p}'_{\parallel} - \mathbf{p}_{\parallel}|$$

$$+ i(p'_{\perp} - p_{\perp})]^{-1} . \exp[i(\epsilon_{p'} - \epsilon_p)t'] \exp(-i\Delta z \cos\omega t') A\mathbf{p}(t').$$

Here $A_{\mathbf{p}}(t)$ is the initial amplitude for the photoelectric effect from the metal into a state of an electron with a kinetic energy $\epsilon_p = n_0\omega - A$ (n_0 is the minimum number of photons required for the multiphoton photoelectric effect, and A is the work function of the metal), $\Delta z = (eE_0\lambda/\omega)(\Delta p_{\perp}/m_e)$ is a dimensionless parameter governed by the intensity and frequency of the wave and also by the value of the increment $\Delta p_\perp = p'_\perp - p_\perp$ in the z component of the electron momentum due to the scattering, and $\lambda = 1/\omega$.

In principle, the scattering of an electron by the image potential during the laser pulse can be accompanied by an absorption (or emission) of some arbitrary number n of field photons. Under the experimental conditions of Ref. 2, however, with $eE_0\lambda/\omega \gtrsim 1$, we have $\Delta z \sim (n\omega/m_e)^{1/2} \ll 1$, so the most probable process is one involving the pickup of one photon in each scattering event.

With this circumstance in mind, we can easily formulate a recurrence relation for the probability amplitude for n-fold scattering accompanied by the pickup of a field photon in each scattering event:

$$A_{\mathbf{p}_n}(t) = i\pi\delta(\epsilon_{p_n} - n\omega - \epsilon_p)\exp[i(\epsilon_{p_n} - n\omega - \epsilon_p - i\tilde{\alpha})t]$$
$$\times \int \frac{dp_{n-1}}{(2\pi)^3}\left(\frac{eE_0\lambda}{2m_e\omega}\right)2\pi \frac{Z_{\text{eff}}e^2}{|\mathbf{p}'_{n\|} - \mathbf{p}_{n-1\|}|}$$
$$\times \exp(-|\mathbf{p}_{n\|} - \mathbf{p}_{n-1\|}|a)i\pi\delta[\epsilon_{p_{n-1}} - (n-1)\omega - \epsilon_p]A_{\mathbf{p}_{n-1}}, \tag{4}$$

where $A_{\mathbf{p}_{n-1}}$ is the probability amplitude for the absorption of the $(n-1)$st photon on the energy surface, and $\tilde{\alpha} \approx +0$ corresponds to the adiabatic imposition of the wave field at $t \to -\infty$. In deriving (4) we used the pole approximation in a composite matrix element.[7]

Let us describe the procedure for evaluating the integral in (4). The integration over the z component of the intermediate momentum $\mathbf{p}_{n-1}$ is carried out with the help of a Dirac δ-function which expresses energy conservation and which is written in the form

$$\delta[\epsilon_{p_{n-1}} - (n-1)\omega - \epsilon_p] = m_e/\{2m_e[(n-1)\omega + \epsilon_p] - p^2_{n-1\|}\}^{1/2}$$
$$\times\{\delta[p_{n-1\perp} - \sqrt{2m_e[(n-1)\omega + \epsilon_p] - p^2_{n-1\|}}] + \delta[p_{n-1\perp}$$
$$+ \sqrt{2m_e[(n-1)\omega + \epsilon_p] - p^2_{n-1\|}}]\}. \tag{5}$$

The subsequent integration is simplified by the following circumstances. First, the amplitude $A_{\mathbf{p}_{n-1}}$ is a smooth function of the tangential component of the momentum $\mathbf{p}_{n-1}$ and can in principle be removed from the integral. Second, since the linear dimension of the cloud, a, is a macroscopic quantity, we have $p_\| a \gg 1$ in any state of the photoelectron. For this reason, the integral is dominated by that region of the variable $\mathbf{p}_{n-1\|}$ in which the condition $\mathbf{p}_{n-1\|} \approx \mathbf{p}_{n\|}$ holds. This statement means that the tangential component of the electron momentum is essentially unchanged by the absorption of a photon: $p_{n\|} \approx p_{n-1\|}$ and $\varphi_n \approx \varphi_{n-1}$ (here φ_{n-1} and φ_n are the azimuthal angles of the momenta $\mathbf{p}_{n-1\|}$ and $\mathbf{p}_{n\|}$, respectively, reckoned in the plane of the metal surface). The acceleration of the electron resulting from the absorption of field photons occurs along the z axis (along the wave polarization direction): $p_{n\perp} \approx (2m_e n\omega)^{1/2}$. As a result, the 3D problem on the behavior of the electron near the metal surface is essentially reduced to a 1D problem.

With these arguments in mind, we can derive an expression for the spectral and angular distribution of the intensity of the photoelectrons produced in the field of the laser wave along the polarization direction of this wave:

$$\frac{dN}{d\epsilon_p d\Omega_p} \sim \sum_{n=1} \left[\frac{eE_0\lambda\!\!\!/}{2\omega}\frac{Z_{\rm eff}e^2/a}{am_e}\left(\frac{\rm Rye}{n\omega}\right)^{1/2}\right]^{2n} n^{1/2}\delta(\epsilon_p - n\omega). \tag{6}$$

Here $\alpha = e^2/\hbar c$ is the fine-structure constant; $\mathrm{Ry} = m_e e^4/2\hbar^2$; and the coefficient of Ry in the expression in square brackets is e=2.718... . It follows from (6) that the photoelectron spectrum is an equidistant series of peaks separated by intervals $\hbar\omega$. In a weak field, such that the combination of parameters in the coefficient of $n^{-1/2}$ in square brackets in (6) is less than one, the absolute height of these peaks falls off monotonically with increasing n, starting at low values of the index. In an intense field, for which the opposite condition holds, the height of the peaks in the above-threshold photoelectric effect increases with n up to $n = n_{\rm max}$ and then falls off slowly with a further increase in n. The value of $n_{\rm max}$, which determines the position of the maximum of the envelope of the peaks, is found from the condition

$$n_{\rm max} = \left(\frac{eE_0\lambda\!\!\!/}{2\omega}\right)^2\left(\frac{Z_{\rm eff}\,e^2/a}{\alpha m_e}\right)^2 \mathrm{Rye}/\omega. \tag{7}$$

A numerical estimate of $n_{\rm max}$ for the parameter values of Ref. 2 is given below.

3. Conclusion. It follows from this analysis that the model of multiple Coulomb scattering of photoelectrons by the image potential of the electron cloud in the metal gives a description of the basic features of the above-threshold photoelectric effect.

1. It explains the origin and nature of the energy spectrum of the photoelectrons as a function of the intensity and frequency of the laser wave. For a fixed value of n the peak height is

$$\frac{dN_n}{d\epsilon_p d\Omega_p} \sim I^n Z_{\rm eff}^{2n}\omega^{-5n}. \tag{8}$$

2. The quantity $n_{\rm max}$, which determines the positions of the peaks of maximum height, depends on the intensity and frequency of the wave and also on the strength of the applied field:

$$n_{\rm max} \sim I Z_{\rm eff}\omega^{-5}E_{\rm ext}. \tag{9}$$

3. As an example we find a numerical estimate of $n_{\rm max}$ for the following parameter values:[2] $\lambda = 1064$ nm, $I = 0.3$ GW/cm^2, $Z_{\rm eff} \approx 10^7$ (this quantity is measured directly in corresponding experiments), and $E_{\rm ext} = 10$ kV/cm. It follows from (7) that we have $n_{\rm max} \approx 10$ in this case. This estimate agrees satisfactorily with the experimental observation $n_{\rm max\,exp} \approx 5$–10. It also follows from this result that the wave intensity required for observation of the above-threshold photoelectric effect is much lower than that required in the case of above-threshold ionization.

4. The width of the angular distribution of photoelectrons falls off with increasing n as $n^{-1/2}$ $(p_{\parallel}/p_{n\perp} \sim n^{-1/2})$. This result can be checked experimentally.

We note in conclusion that the acceleration of electrons which was observed in Ref. 10 was apparently also due directly to the role played by the Coulomb image potential in the metal.

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