Mechanism determining the electron angular distribution during ionization of a low-density gas by an intense electromagnetic field

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The ionization of a gas by a radially varying high-amplitude electromagnetic field is analyzed. The electrons emitted from the beam focus during ionization of a low-density gas have an asymmetric angular distribution. The reason for the asymmetry is the drift velocity of the electrons produced in the field of the intense wave. An analogy is drawn between the electrons having this velocity and the appearance of electrons above the threshold in the multiphoton limit of nonlinear ionization. The theoretical results are compared with the experimental data available.

INTRODUCTION

There is both theoretical and experimental interest in resolving some questions concerning the ionization of a gas by intense electromagnetic fields, in particular, laser beams. In the intense fields which are produced by the focusing of microwave and IR beams, the photon energy is below the ionization potential of the gas atoms, and nonlinear ionization of the atoms can accompany the onset of an avalanche. This process, whose probability is a nonlinear function of the number of photons, usually acts primarily as a nucleating process in dense media, initiating the subsequent avalanche growth in the electron density through impact ionization. In a low-density gas, this would be the only mechanism which would result in ionization, so research on this mechanism, which was begun in Refs. 1 and 2, is developing quite rapidly.

Along with the progress in this research, however, there have been difficulties in explaining some experiments. In particular, the experiment carried out by Agostini *et al.*[3] indicated the formation of electrons "above the threshold," whose existence contradicted the basic positions of perturbation theory, which would lead one to expect an infinitely low probability for the formation of such electrons. Since then, many experiments have qualitatively confirmed the existence of above-threshold ionization (but only in the so-called multiphoton limit; see the bibliography in Ref. 4). In general, however, despite the large number of theoretical papers on these questions, there is still no rigorous quantitative theory for this process. This comment applies in particular to the tunneling limit of nonlinear ionization, for which there is no theoretical treatment. The results of the experiment carried out in Ref. 5, in a study of the angular and energy characteristics of the electrons emitted from the ionization region, speak in favor of an above-threshold mechanism.

In the present paper we develop an approach for reaching an understanding of the processes of which above-threshold ionization develops in the case of electrons produced by an intense electromagnetic field, with parameter values of the radiation and the medium corresponding to the tunneling limit of nonlinear ionization. On the basis of the model proposed here, we calculate the angular and energy distributions of the electrons emitted from the region acted on by the field. On the whole, these results agree with the experimental results of Ref. 5.

ABOVE-THRESHOLD IONIZATION

In a low-density gas in an intense electromagnetic field, such that the condition $n \ll (4.9\sigma_i T^{1/2})^{-1/2}$ holds [T is the gas temperature in kelvins, and σ_i (in square centimeters) is the cross section for electron-impact ionization of the atoms], the atoms are ionized only as a result of their direct interaction with the field. If $\hbar\omega < I_0$ holds, where I_0 is the ionization potential of the atoms and ω is the field frequency, so-called nonlinear ionization of atoms can occur, in which the absorption of more than one field photon is required for ionization (resonance levels are ignored). In intense fields, this process has a fairly high probability. Depending on the frequency ω and the amplitude E_0 of the field, the nonlinear ionization will be manifested in different ways. For example, at low frequencies and intense fields, such that the condition $\gamma = (I_0/2\varepsilon_0)^{1/2} \lesssim 1$ holds [γ is the adiabatic index,[1] and $\varepsilon_0 = e^2E_0^2/(4m\omega^2)$ is the average oscillation energy of an electron in the field of the wave], the ionization is a tunneling effect. A very large number of photons, $s = I_0/\hbar\omega + 1 \gg 1$, must be absorbed if an atom is to be ionized (in the limiting case of a zero frequency, the number of photons becomes infinite). In the opposite case, the atom is ionized if more than one photon is absorbed simultaneously.

The total probabilities for the ionization of atoms were calculated in Refs. 1 and 2 and in some subsequent studies of nonlinear ionization. In addition, energy and angular distributions of the electrons which were produced were found in Ref. 2. It follows from those results, in particular, that the vector **E** of the wave defines a predominant direction for the electron emission, while the distribution with respect to the number of absorbed photons is concentrated near the ionization threshold. This result corresponds to a kinetic energy of approximately zero for the electrons which are produced. It was shown in Ref. 3, however, that the distribution of photoelectrons observed during six-photon ionization of xenon atoms contains monoenergetic electrons not only with the energy corresponding to the ionization threshold, $s\hbar\omega \approx I_0$, but also with an energy exceeding the threshold by an amount which is a multiple of the photon energy (above-threshold ionization): $I_{\text{eff}} = \hbar\omega(I_0/\hbar\omega + 1 + s_0)$. The

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maximum observed energy depends on the intensity (photon density) of the radiation: With increasing intensity, the maximum energy of the photoelectrons detected increases.

The theoretical models which were subsequently developed have largely explained the observed effects for $s > 1$, i.e., in the multiphoton limit. However, there has been no theoretical work on above-threshold ionization for the case $s \gg 1, \gamma \ll 1$ (e.g., for radiation in the IR or microwave range), and there has been little experimental work (e.g., Refs. 5 and 6). Note, however, that since multiphoton ionization and tunneling ionization are cases of a common nonlinear-ionization process, above-threshold effects, e.g., the appearance of high-energy particles, should be observed in the tunneling limit.

This conclusion is implied, in particular, by the experimental results of Ref. 5, where a study was made of the angular and energy characteristics of the electrons emitted from a region in which a gas was being ionized by the linearly polarized beam from a neodymium laser ($\lambda = 1.06\ \mu$m) with a power density up to 10^{15} W/cm^2. That study was based on a model in which electrons produced with approximately zero energy were radially expelled from the region in which the field was acting (the "field region") by a nonrelativistic, nonlinear radiation-pressure force as a result of a transverse variation of the oscillating field $\mathbf{E}(r)$:

$$\mathbf{f}(r) = -\nabla U(r) = \frac{e^2}{4m\omega^2}\nabla |E^2(r)|, \qquad (1)$$

where $U(r)$ is the ponderomotive potential, and m is the mass of an electron. Since the force depends on the square of the field, the polarization of the field drops out of the equation of motion; as a result, there should be a uniform emission of electrons along all radii. Here the theoretical model runs into a contradiction with the experimental results of Ref. 5, according to which the ratio of the number of electrons emitted in the direction perpendicular to $\mathbf{E}$ to the number emitted along $\mathbf{E}$ lies in the interval 0.5–0.7. The relativistic correction to the force changes the theoretical model and the results calculated for the angular distribution only insignificantly. The assumption that the electrons are produced with a vanishing initial energy thus does not agree with the experimental results, and the reason for the observed asymmetry in the electron emission should be sought in the ionization process itself.

Below we propose a model of a drifting electron in an effort to show that the electrons produced in an intense field have a fairly high energy, comparable to ε_0, when the parameter values of the atoms and the radiation correspond to the case $\gamma \lesssim 1$. We then work from this model to calculate the angular distribution of electrons emitted from the field region; the results agree in general with the experimental results of Ref. 5.

MODEL OF A DRIFTING ELECTRON

In the model problem for tunneling ionization, there is a large probability $W(E)$ that in intense fields an atomic electron will be on the outer side of the potential barrier which forms. In this state, the Coulomb field of the nucleus can be ignored. The motion of the electron beyond the barrier and its interaction with the radiation field can be described by classical electrodynamics. This conclusion follows from two circumstances. First, the intense field is characterized from the quantum-mechanical point of view by large occupation numbers of photon states, so one can ignore the quantum structure of the field. Second, since the characteristic energies of the motion of an electron beyond the barrier are large ($\sim\varepsilon_0$), the de Broglie wavelength corresponding to this motion is much smaller than the wavelength of the radiation. It is even smaller in comparison with the length scales of the ionization region. It thus becomes possible to treat the motion of the electron classically. From the equation of motion we thus find the following result for the velocity of an electron outside the Coulomb well in the field of a linearly polarized wave:

$$v = \frac{eE_0}{m\omega}(\cos\omega t_0 - \cos\omega t), \qquad (2)$$

where ωt_0 is the phase of the field at which the electron is on the outer boundary of the potential barrier. Only in the case $\omega t_0 = \pi/2$ does an electron remain in the same place (while oscillating); at all other phases, the electron has an unclosed trajectory, and it has a positive velocity v_D, on the average over a period (there is a drift). The reason for this drift is the field, but if the field is turned off, or if the electron escapes from the field region, the discontinuous drift velocity converts into a constant velocity. For example, if the field is turned off in accordance with $E(t) = E_0 \sin\omega t \exp(-\alpha t)$, the electron is left at $t \to \infty$ with a velocity

$$v_D = \frac{eE_0}{m(\alpha^2+\omega^2)}(\omega\cos\omega t_0 - \alpha\sin\omega t_0)\exp(-\alpha t_0).$$

If the field is turned off adiabatically slowly ($\alpha \ll \omega$), the absolute value of the drift velocity will lie in the following interval, depending on the phase ωt_0:

$$\frac{eE_0}{m\omega} \geqslant |v_D| \geqslant \frac{\alpha}{\omega}\frac{eE_0}{m\omega}\exp\left(-\frac{\alpha\pi}{2}\right) \approx 0.$$

From the quantum-mechanical standpoint, the acquisition of energy by an electron in a field which is decaying in time means that the electron is absorbing photons of a different frequency from the radiation field. The decaying field gives rise to nonmonochromatic components in the Fourier transform of the field. Stimulated absorption and emission of photons from such a field may lead to changes in the energy and momentum of the electrons, so an electron would be left with a certain amount of kinetic energy when the radiation disappeared. Since the photon energies for radiation in the microwave or IR range satisfy the condition $\hbar\omega \ll I_0 \ll \varepsilon_0$, there will be nearly no monochromatic structure in the distribution of above-threshold electrons; their distribution will be continuous.

At a certain time, the field thus allows an atomic electron to tunnel through the potential barrier, but whether the electron acquires the energy required for ionization is determined entirely by the phase ωt_0.

CALCULATION OF THE ANGULAR DISTRIBUTION

We now use the model proposed above to calculate the angular distribution of the electrons which are emitted from the region in which a low-density gas is ionized by an intense electromagnetic field. We assume that the field is nonuniform over the beam cross section. Because of the drift velocities and also because of the nonrelativistic, nonlinear radiation-pressure force, which is uniform along any radius, the electron trajectories will have fairly complex shapes. Their angular distribution upon emission from the field region will be anisotropic. To calculate this distribution we need to trace the trajectory of each particle and then sum all electrons over all possible initial states. The trajectory of an electron can be found by integrating the equations of motion. However, under the assumption that the motion is planar in this case, a complete solution of the problem can be found most easily from energy and momentum conservation.[7]

We assume that the transverse distribution of the field is Gaussian: $E(r) = E_{0m}\exp(-r^2/a^2)$, where the constant a specifies the profile of the field, and r is the radial coordinate. The initial state of the electrons is characterized by the coordinates (r_0,ψ_0) (the point of creation) and $\dot\varphi$ (the drift velocity). The origin for the scale ψ_0 is the straight line along the field direction (Fig. 1).

The total energy and momentum are

$$\varepsilon=\frac{m\dot r^2}{2}+\frac{M^2}{2mr^2}+U(r)=\text{const}, \qquad (3)$$

$$M=mr^2\dot\varphi=\text{const}, \qquad (4)$$

where $U(r) = \varepsilon_{0m}\exp(-r^2/a^2)$ is the potential energy corresponding to the nonrelativistic, nonlinear radiation-pressure force. From Fig. 1 we find

$$\dot r_0=v_0\cos\psi_0\cos\varphi,\quad \dot r_0\dot\varphi=v_0\sin\psi_0\cos\varphi,$$

where $v_0 = 2m^{-1/2}[\varepsilon_{0m}\exp(-r_0^2/a^2)]^{1/2}$. We then find the following expressions for the initial energy and momentum:

$$\varepsilon_0=\varepsilon_{0m}(1+2\cos^2\varphi)\exp(-r_0^2/a^2),$$
$$M_0=2r_0\sin\psi\cos\varphi(m\varepsilon_{0m})^{1/2}\exp(-r_0^2/2a^2).$$

To calculate $\Delta\psi$, the angle through which the electrons revolve, we separate variables in Eq. (3), integrate it, and use Eq. (4). As a result we find

$$\Delta\psi=\int\frac{Mdr}{r^2[2m(\varepsilon_0-U(r))-M^2/r^2]^{1/2}}+\text{const}. \qquad (5)$$

Using the initial energy and momentum, we can rewrite the expression for $\Delta\psi$ in the form

$$\Delta\psi=\int_{r_0}^{\infty}\frac{2^{1/2}r^{-2}r_0\sin\psi_0\cos\varphi dr}{\{1+2\cos^2\varphi(1-r_0^2\sin^2\psi_0/r^2)-\exp[(r_0^2-r^2)/a^2]\}^{1/2}}. \qquad (6)$$

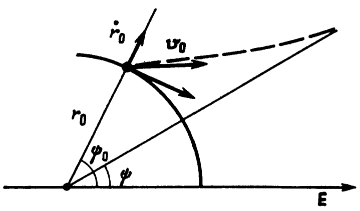

FIG. 1. An electron produced at the point (r_0, ψ_0) with a velocity v_0 is ejected by a radial force from the region acted upon by the field.

It follows from this result that the revolution angle does not depend on the maximum average oscillation energy of the electron, ε_{0m}, or thus on the amplitude of the ionizing field.

If the direction of the electron drift velocity is such that an electron is obliged to overcome force (1) at the beginning of its trajectory, the integral must be broken into two parts:

$$\Delta\psi=2\Delta\psi\int_{r_{min}}^{r_0}+\Delta\psi\int_{r_0}^{\infty}. \qquad (7)$$

Here $r_{\min}$ is the minimum distance from the trajectory of the electron to the axis of the radiation beam. This minimum distance is found from the equation

$$1+2\cos^2\varphi(1-r_0^2\sin^2\psi_0/r^2)-\exp[(r_0^2-r^2)/a^2]=0.$$

The total angle through which an electron is deflected as it is emitted from the ionization region is thus

$$\psi=\psi_0+\Delta\psi. \qquad (8)$$

RESULTS AND COMPARISON WITH EXPERIMENT

The calculations from expressions (6)–(8) were carried out numerically because of the large volume of initial data. The calculation procedure can be outlined as follows: The overall volume of the ionization region was broken up into small cells (2°, $0.05r_0$). One point selected in each cell was assigned in succession the entire set of initial energies ($\sim\cos\varphi$), where φ was varied at a step of 2°. The result was to generate a three-dimensional file of initial parameter values. For each point in this file, an integration was carried out (over r). With increasing r_0, the area of the cell increases, and the number of electrons produced in it increases accordingly. Consequently, each successive value of the area was multiplied by an appropriate value of r_0. In the calculations, the integration was limited by the point $r_0 = 4$, at which the average oscillation energy is lower than the maximum energy ε_{0m} by a factor ≈ 50. At the same distance, cells 2° in width were constructed along the angular variable. An electron striking any cell was detected and summed with the electrons which arrived there later. For example, during the summation over all the initial parameters, a distribution of electrons with respect to emission angle was constructed in the ψ cells. Figure 2 shows the result for the particular case of a field profile with $a = 1$.

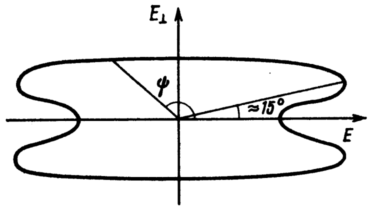

FIG. 2. Angular distribution of electrons emitted from a region in which a gas is ionized by an intense electromagnetic field with a Gaussian transverse cross section. The plane of the figure is the polarization plane of the field; the horizontal line is the polarization axis.

The energy spectrum can be found from Eqs. (3) and (4). We will not go through that calculation here. We can, however, assert on the basis of general energy considerations that the maximum energy of the emitted electrons will reach $3\varepsilon_{0m}$ in the direction along the polarization of the field, and ε_{0m} in the perpendicular direction.

According to (2), the maximum drift velocity is reached at $\omega t_0 = 0$. At $\omega t_0 = 0$, however, the probability for an atomic electron to tunnel is zero, since there is no external field, and the electron is blocked by the nuclear potential. A tunneling probability $W(E)$ must be introduced in the calculations to deal with this circumstance. As a result, the number of particles produced in the phase $\omega t_0 = 0$ is zero, and the maximum energy of an electron emitted in the $\psi = 0$ direction is less than $3\varepsilon_{0m}$. If the field amplitude is sufficiently high, however, then even at small values of ωt_0 the field value $E = E_0 \sin \omega t_0$ becomes sufficient to cause a significant probability for the tunneling of an atomic electron.

The photon energy in Ref. 5 was ≈ 1 eV. The ionization potential of helium is $I_0 = 24.49$ eV, and the average oscillation energy of an electron in a field of intensity 10^{15} W/cm^2 and frequency $\omega = 2 \cdot 10^{15}$ rad/s[1] is 40 eV. Since we have $\gamma \approx 0.5$, in accordance with the tunneling approximation, all the results found here can be used to describe the experiment of Ref. 5. For example, the ratio of the number of particles in the ψ interval from 45° to 135° to the number in the interval from $-45°$ to $+45°$ is ≈ 0.55. To within the error involved here, this result agrees with the value of 0.5–0.7 which was reported in Ref. 5. In Fig. 2 we see two peaks, near 15° and 165°. They could not be seen in Ref. 5, however, since the particles were collected in a solid angle of 1.5 sr. The maximum energy detected in Ref. 5 was above 100 eV. Recalling the earlier discussion of the maximum electron energy, we see that the value found here agrees fairly well with the energy found in that earlier study.

We note in conclusion that if the atoms are ionized by a circularly polarized field then the distribution of electron drift velocities will be isotropic, since there is no special field direction. As a result, the distribution with respect to ψ should be totally symmetric for electrons emitted from the gas ionization region.

[1] L. V. Keldysh, Zh. Eksp. Teor. Fiz. **47**, 1949 (1964) [Sov. Phys. JETP.

[2] A. I. Nikishov and V. I. Ritus, Zh. Eksp. Teor. Fiz. **50**, 255 (1966) [Sov. Phys. JETP **23**, 168 (1966)].

[3] P. Agostini, F. Fabre, G. Mainfray *et al.*, Phys. Rev. Lett. **42**, 1127 (1979).

[4] N. B. Delone and M. V. Fedorov, Usp. Fiz. Nauk **158**, 215 (1989) [Sov. Phys. Usp. **32**, 500 (1989)].

[5] B. V. Borkhem and R. Mavaddat, Izv. Akad. Nauk SSSR, Ser. Fiz. **43**, 313 (1979).

[6] S. Chin, F. Yerqeau, and P. Lavingne, J. Phys. B **18**, L213 (1985).

[7] L. D. Landau and E. M. Lifshitz, *Mechanics*, *3rd ed.*, Pergamon, New York, 1976.

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