Energy and angular distributions of electrons resulting from barrier-suppression ionization of atoms by strong low-frequency radiation

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Using the Keldysh–Faisal–Reiss approximation, analytical expressions are found for the probability of barrier-suppression ionization of atoms and for the energy and angular distributions of the outgoing photoelectrons in the field of strong low-frequency laser radiation. The case in which the radiation is circularly polarized is considered. The results agree with the previously obtained distributions for tunneling of atoms by the field of low-frequency radiation for small values of the intensity. © *1995 American Institute of Physics.*

1. INTRODUCTION

Experiments that describe the interaction of atoms with intense laser radiation are customarily treated in two regimes, multiphoton and tunneling. A new mechanism, barrier-suppression ionization, was first observed by Augst *et al.*[1] It is found to be most effective for ionization of noble gas atoms by the radiation field of neodymium and titanium–sapphire lasers with intensities from 10^{13} to 10^{16} W/cm^2.

We refer to fields with such parameters as "above-barrier fields," since the perturbed energy of the initial atomic state exceeds the maximum of the effective potential barrier

$$V_{\rm eff}(x) = -Z/x - Fx \tag{1}$$

in the direction x in which the electron is removed.[2] Thus,

$$E_n(F_{\rm BSI}) = V_{\rm eff}^{\max}. \tag{2}$$

Here F is the strength of the laser radiation field, Z is the charge of the atomic core, $F_{\rm BSI}$ is the characteristic strength of the above-barrier field, and $E_n(F)$ is the perturbed energy of the initial atomic state with principal quantum number n. By definition, barrier-suppression ionization of an atom occurs under the condition $F \gtrsim F_{\rm BSI}$ if the frequency ω of the field is sufficiently small (see below).

Thus, barrier-suppression ionization is a classical threshold effect. For fields with $F \gtrsim F_{\rm BSI}$ the electron leaves the atom after approximately one orbit around the atomic core (i.e., after a time of order one Kepler period $t_n = 2\pi n^3$):

$$w(F_{\rm BSI}) t_n \sim 1. \tag{3}$$

Here w is the probability per unit time for an atom to be ionized. We have used atomic units $e = m_e = \hbar = 1$ everywhere (for details see Ref. 3, Sec. 4.4).

If the field is turned on adiabatically slowly compared with typical times for Landau–Zener transitions to neighboring states, then in the switching-on process the atom undergoes a transition into states with different quantum numbers many times. However, the electron energy in this process remains practically unchanged. Consequently, the quantity $E_n(F)$ does not depend on F and is related to $F_{\rm BSI}$ by the well-known expression

$$F_{\rm BSI} = E_n^2/4Z[\text{a.u.}] \tag{4}$$

(1 a.u.$=5.14\cdot 10^9$ W/cm). Hence the energy E_n is related to the principal quantum number n by the Rydberg formula (for a hydrogen atom)

$$E_n = Z^2/2n^2[\text{a.u.}] \tag{5}$$

(1 a.u.$=27.2$ eV). For complex atoms the appropriate theory was worked out in Ref. 4 using the Thomas-Fermi approximation.

In the limit $F \ll F_{\rm BSI}$ the atom undergoes tunneling ionization (assuming the frequency of the laser radiation is small, i.e., the adiabaticity parameter γ satisfies[5]

$$\gamma = \frac{\omega\sqrt{2E_n}}{F} \ll 1, \tag{6}$$

and $\omega \ll E_n$). The energy and angular distributions of the electrons associated with tunneling ionization by a low-frequency radiation field have been treated theoretically by Delone and Krainov.[6]

The present work is devoted to deriving a theory of the energy and angular distributions of outgoing electrons due to barrier-suppression ionization of atoms by a strong low-frequency laser radiation field. This theory is based on the Keldysh–Faisal–Reiss approximation.[7]

2. THE KELDYSH–FAISAL–REISS APPROXIMATION

The Keldysh–Faisal–Reiss approximation differs from the exact expression for the amplitude of a transition from the initial state n of an atom to the final state f of the continuum obtained using the S matrix in using the so-called Volkov wave function, i.e., the solution of the Schrödinger equation for electrons in the field of the laser radiation only, without the potential of the atomic core, instead of the exact wave function of the final state f. This approximation imposes no restriction on the frequency ω of the laser radiation. The only restriction on the intensity of the radiation is that it must exceed some lower limit. The stronger the field F of the laser radiation, the smaller is the role of the potential of the atomic core in the final state of the continuous spectrum. Furthermore, the larger the energy of the outgoing photoelectron the more accurate is the use of the Volkov wave function approximation.

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Thus, the amplitude A_{nf} for the transition from the initial bound state n of the atom to the final state f of the continuum is given by an element of the S matrix

$$A_{nf}=-i\int dt\langle\Psi_f^{(V)}|V(\mathbf{r},t)|\Psi_n^{(0)}\rangle. \tag{7}$$

Here $\Psi_n^{(0)}$ is the unperturbed wave function of the initial state of the atom, $\Psi_f^{(V)}$ is the Volkov wave function of the final state of the continuous spectrum, and $V(\mathbf{r},t)$ is the potential of the interaction between the atom and the external electromagnetic field

$$V(\mathbf{r},t)=\mathbf{pA}/c+\mathbf{A}^2/2c^2, \tag{8}$$

here $\mathbf{A}(\mathbf{r},t)$ is the vector potential of the field and $\mathbf{p}$ is the momentum of the outgoing photoelectron.

Here as an example we consider barrier-suppression ionization of the ground state of the hydrogen atom by a circularly polarized radiation field. For this we must introduce a correction coefficient in Eq. (7), which represents the perturbation-theoretic treatment of the Coulomb potential in the wave function of the final state f. Here we proceed in analogy with the treatment of the Coulomb potential in Ref. 8.

The treatment of the Coulomb interaction between an electron and the nucleus using perturbation theory contributes a factor

$$\exp\left(-i\int U dt\right)=\exp\left(i\int\frac{dt}{r}\right)=\exp\left(i\int_r^{1/2F}\frac{dr}{rp(r)}\right), \tag{9}$$

to the Volkov wave function, where $p(r) = i\sqrt{1-2Fr}$ is the imaginary electron momentum in the above-barrier region. The integral in Eq. (9) is easily evaluated in the limit $F/p\ll 1$ and is found to be $2/(Fr)$. Note that this procedure is justified in both the tunneling region $F\ll 1$ and in the barrier-suppression region $F\sim 1$, since as we will see the typical values satisfy $p\sim F/\omega$, and hence this condition reduces to a requirement that the frequency of the field be small, $\omega\ll 1$. The factor $1/r$ changes the wave function of the initial state into $(1/r)\exp(-r)$, i.e., into the wave function of a particle in a potential of zero radius. This procedure is completely analogous to that in the derivation of the exact coefficient in the expression for the ionization probability of an atom by a constant electric field.[9]

Hence it follows from (7) that the probability for ionization of the ground state of the hydrogen atom into the solid angle $d\Omega$ by a circularly polarized field is equal to

$$\frac{dw}{d\Omega}=\frac{\sqrt{32\omega}}{\pi F^2}\sum_{N_0}\sqrt{N-z-\frac{1}{2\omega}}J_N^2(x). \tag{10}$$

Here N is the number of absorbed photons, $J_N(x)$ is the Bessel function, and N_0 is the minimum number of absorbed photons, equal to

$$N_0=\left\{\frac{1}{2\omega}+z\right\}=\left\{\frac{1}{\omega}\left(\frac{1}{2}+\frac{F^2}{2\omega^2}\right)\right\}, \tag{11}$$

(the braces {...} indicate the integer part of a number). The interaction between the atom and the radiation field is treated in the dipole approximation. In accordance with (11) the electron has a kinetic energy measured with respect to the boundary of the continuum which is shifted upward by $F^2/(2\omega^2)$ due to the dynamic Stark effect.

The intensity parameter z which appears in (11) is defined as

$$z=F^2/2\omega^3, \tag{12}$$

and we have written

$$x=\frac{F}{\omega}\sqrt{2N-\frac{F^2}{\omega^3}-\frac{1}{\omega}}\sin\theta, \tag{13}$$

where θ is the angle between the direction of the outgoing electron and the direction in which the electromagnetic wave propagates.

Expression (10) determines both the energy and the angular distributions of the outgoing photoelectrons in the most general case, i.e., for arbitrary values of the strength F and frequency ω of the electromagnetic radiation.

Corkum *et al.*[10] used the asymptotic properties of the Bessel function $J_N(x)$ in expression (10) to obtain the tunneling limit which is realized for $F\ll 1$ a.u. under condition (6):

$$dw/d\Omega=A\exp(-2/3F-F\psi^2/\omega^2)\times\sum_{\delta N}\exp[-\omega^4(\delta N)^2/F^3] \tag{14}$$

where

$$A=2\omega/(\pi F)^2, \tag{15}$$

and $\psi=\pi/2-\theta\ll 1$ is the small angle between the direction of the outgoing electron and the plane of polarization of the circularly polarized radiation,

$$\delta N=N-2z-1/2\omega. \tag{16}$$

Thus, the energy spectrum has a maximum for $N\approx 2z$, which corresponds to the kinetic energy $E_e=F^2/2\omega^2$ of the outgoing electron if we take into account the upward shift in the boundary of the continuum by the same amount $F^2/2\omega^2$ due to the dynamic Stark effect.

Note that the results of Ref. 6 in the part related to circularly polarized fields agree with the result (14) of Ref. 10, although they are obtained by completely different techniques.

Expression (14) determines both the angular and the energetic distribution of the electrons in the tunneling regime. Integrating over all angles of the outgoing electron trajectory and summing over all numbers δN of absorbed photons (this summation is also transformed into an integral), we find from (14) as is to be expected the familiar expression[9] for the probability of tunneling ionization by a constant electric field with strength F:

$$w=\frac{4}{F}e^{-2/3F}. \tag{17}$$

3. BARRIER-SUPPRESSION IONIZATION OF ATOMS BY A LOW-FREQUENCY FIELD

For above-barrier fields we can use the well-known asymptotic expansion

$$J_N(N+N^{1/3}t)=(2/N)^{1/3}\mathrm{Ai}(-2^{1/3}t), \qquad (18)$$

for the Bessel function appearing in the general expression (10) for the ionization probability of a hydrogen atom per unit time by the field of a circularly polarized wave. Quantities $t \gg 1$ correspond to tunneling ionization and $t \lesssim 1$ to barrier-suppression ionization. Here Ai is the Airy function.

Substituting (18) in (10) we find

$$\frac{dw}{d\Omega}=\frac{2^{8/3}\omega}{\pi F^{7/3}}\sum_{\delta N}\mathrm{Ai}^2\left(\frac{1+F^2\psi^2/\omega^2+\omega^4\delta N^2/F^2}{(2F)^{2/3}}\right). \qquad (19)$$

Here the quantity δN is defined by (16); the angle ψ is also defined above.

Equation (19) describes the angular and energy distribution of the outgoing electrons in the barrier-suppression regime for a hydrogen atom. For ionization of other atoms only the overall factor in front of the summation over δN in Eq. (19) changes, but the behavior of the functional dependence on the angle ψ and the number of absorbed photons N remain the same. Thus, Eq. (19) is quite general for barrier-suppression ionization of atoms by a circularly polarized electromagnetic field.

To be sure, in the weak-field case [$t \gg 1$ in Eq. (18)] expression (19) goes over to the result (14) obtained above, corresponding to the tunneling mechanism of ionization. This limit is found using the familiar asymptotic expansion of the Airy function for large argument.

From (19) it follows that just as in the case of tunneling ionization, the peak of the energy distribution of the outgoing photoelectrons is found for an electron kinetic energy $E_e=F^2/2\omega^2$, since the boundary of the continuum is shifted upward by the same amount $F^2/2\omega^2$ due to the dynamic Stark effect. The difference between this and the tunneling limit is that the maximum is considerably broader in the case of barrier-suppression ionization than in that of tunneling ionization (for further details see below).

4. ANGULAR DISTRIBUTION OF ELECTRONS FOR BARRIER-SUPPRESSION IONIZATION

By summing the ionization probability per unit time (19) over all numbers N of absorbed photons (actually, by integrating with respect to δN) we find the angular distribution of the outgoing electrons due to ionization by a circularly polarized electromagnetic field. The result can be written in the form

$$F\omega\frac{dw}{d\psi}=32\int_0^\infty dx\,\mathrm{Ai}^2(x^2+t). \qquad (20)$$

Here we have introduced the parameter t in place of the angle ψ through the definition

$$t=(2F)^{-2/3}+(F/2)^{4/3}(\psi/\omega)^2. \qquad (21)$$

Thus, the dependence of the product $F\omega\, dw/d\psi$ on the parameter t has a universal behavior, and it can be used in

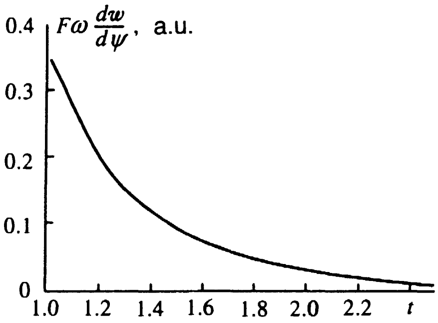

FIG. 1. Universal angular distribution for photoelectrons from ionization by a circularly polarized field as a function of the dimensionless variable t defined by Eq. (21).

calculations for different values of the field strength F and frequency ω in the barrier-suppression regime. Note that in the case of the hydrogen ground state the quantity F_{BSI} for which the energy level is the same as the peak of the effective potential barrier in the direction of the outgoing electron we have $F_{\mathrm{BSI}}=0.208$ (Ref. 11).

The universal function (20) is shown in Fig. 1. As one would expect, the angular distribution is always centered on $\psi=0$, i.e., for electrons leaving in the plane of polarization of the circularly polarized radiation. The same picture holds as well in the tunneling limit (see Refs. 6 and 10). The peak shifts toward $\psi\neq 0$ only when relativistic effects are taken into account.[12]

If we specify a particular value of the field strength F, then from (20) the ionization probability $dw/d\phi$ is a function only of the ratio $\phi=\psi/\omega$, i.e., it is a universal function for arbitrary frequency ω. As an example, in Fig. 2 we plot $dw/d\phi$ versus ϕ for $F=0.3$ a.u., i.e., for a value of the field greater than the barrier-suppression value. Since in atomic units we have $\omega \ll 1$ a.u., it is clear that the angular distribution is concentrated in the region of small angles ψ (for small values of the frequency ω). Just as in the tunneling limit [cf. Eq. (14)], the angular distribution as a function of the field strength F becomes more peaked in the plane of polarization of the electromagnetic field as F increases.

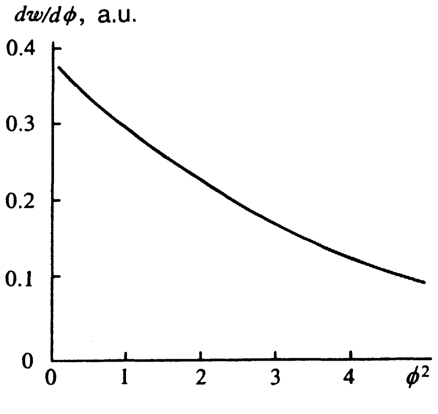

FIG. 2. Angular distribution of photoelectrons as a function of $\phi=\psi/\omega$ for field strength $F=0.3$ a.u.

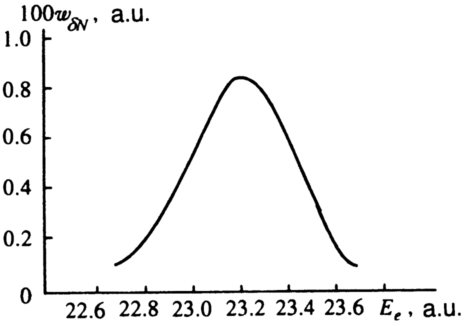

FIG. 3. Energy spectrum of the outgoing photoelectrons for ionization by a circularly polarized radiation field with amplitude $F=0.3$ a.u. and frequency $\omega=1.2$ eV=0.044 a.u. as a function of the photoelectron kinetic energy E_e.

To summarize, we can say that the smaller the frequency ω and the larger the amplitude F, the narrower the angular distribution becomes with respect to the plane of polarization of the radiation. In the tunneling limit the width of the angular distribution is equal to $\omega/\sqrt{F}$ (Ref. 10), whereas in the case of barrier-suppression ionization it is on the order of ω (as a function of the angle ψ).

5. ELECTRON ENERGY DISTRIBUTION FOR BARRIER-SUPPRESSION IONIZATION

In order to find the energy distribution of the outgoing photoelectrons we must identify the term with a specific number N of absorbed photons in the general expression (19) and integrate it over the solid angle $d\Omega$. As a result we find

$$(F^3/\omega^2)w_{\delta N}=32\int_0^\infty \mathrm{Ai}^2(x^2+t')dx. \tag{22}$$

Comparing (22) and (20) we see that the universal functional dependence remains unchanged, but the expression for t becomes

$$t'=(2F)^{-2/3}+\omega^4\frac{(\delta N)^2}{(2F^4)^{2/3}} \tag{23}$$

in place of Eq. (21).

Figure 3 shows the energy spectrum of the outgoing photoelectrons for ionization by a circularly polarized field with amplitude $F=0.3$ a.u. and frequency $\omega=1.2$ eV=0.044 a.u. It can be seen that the distribution of the outgoing photoelectrons in kinetic energy,

$$E_e=\frac{F^2}{2\omega^2}+\omega\delta N, \tag{24}$$

is centered about the value equal to the oscillation energy $F^2/2\omega^2$ of an electron in the field of the circularly polarized electromagnetic wave (as in the tunneling limit; cf. Refs. 6 and 10). The stronger the field and the smaller its frequency the broader the spectrum of the outgoing photoelectrons.

In summation we can say that whereas in the tunneling limit the energy spectrum has a width of order $F^{3/2}/\omega$, for barrier-suppression ionization it is of order $1/\omega$, i.e., quite large in atomic units.

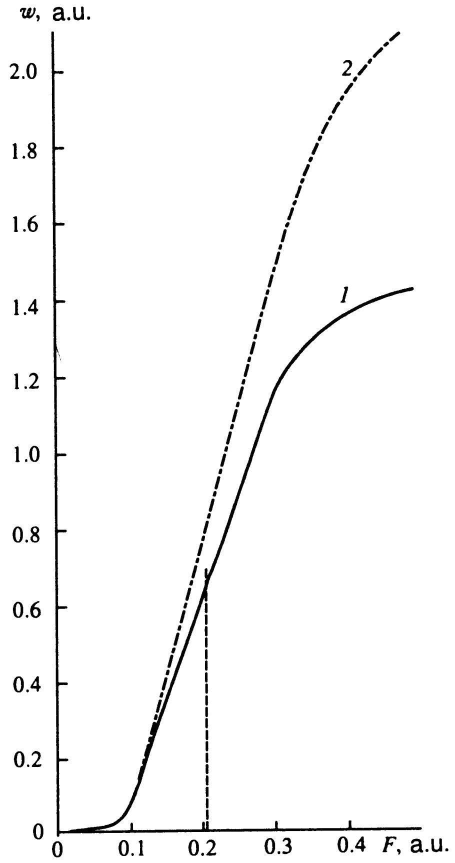

FIG. 4. Ionization probability per unit time due to a strong low-frequency field from the ground state of a hydrogen atom: 1) calculated from Eq. (25) for barrier-suppression ionization; 2) calculated from Eq. (17) for tunneling ionization. The broken vertical line corresponds to the field strength F_{BSI} for which the energy level equals the peak of the effective potential barrier, i.e., for which above-threshold decay of the atom is possible classically.

By integrating (22) over all energies of the outgoing electrons we find the ionization probability from the ground state of a hydrogen atom in the barrier-suppression regime (the frequency ω drops out and the result is the same as that obtained for ionization by a constant electric field with amplitude F):

$$w(F)=\frac{2^{13/3}\pi}{F^{5/3}}\left\{\mathrm{Ai}'^2\left(\frac{1}{(2F)^{2/3}}\right)-\frac{1}{(2F)^{2/3}}\mathrm{Ai}^2\left(\frac{1}{(2F)^{2/3}}\right)\right\}. \tag{25}$$

Here the field strength F is measured in atomic units. In the limit $F\ll 1$ a.u. expression (25) goes over to the tunneling limit (17).

The dependence of w on F according to (25) is shown in Fig. 4. The tunneling dependence (17) extrapolated to the region of fields corresponding to barrier-suppression ionization is shown in the same figure (F_{BSI} is indicated by the broken line). It is clear that this extrapolation overestimates the ionization probability in the above-barrier region. This should be kept in mind in connection with the extrapolation of the tunneling formulas of Ref. 13 into the barrier-suppression region. In Fig. 4 the extrapolated part of the tunneling probability (17) is shown by the chain curve.

It should also be noted that the probability of barrier-suppression ionization per unit time as a function of the field strength F is approximately linear, according to Fig. 4. Note that this linearity also is found in numerical calculations of the ionization probability of Rydberg states of the hydrogen atom by a constant electric field in the same region.[11]

6. CONCLUSION

Barrier-suppression ionization was first studied experimentally by Augst *et al.*,[1] who used a neodymium laser with wavelength 1.053 μm. The strength of the focused radiation reached values several times 10^{16} W/cm^2. Depending on the location in the focus the ionization is sometimes tunneling and sometimes barrier-suppression. Hence for a detailed comparison of the experimental results with those predicted by theory we should integrate the results of the latter over the intensity distribution of the laser radiation in the focus. This point was first discussed by Kiyan and Krainov.[14]

Furthermore, the theory calculates the ionization probability per unit time, whereas in the experiments the ionization saturates in the central part of the focus. For this reason we should use the well-known Wigner–Weisskopf formulas for the absolute ionization probabilities, which are simply related to the time and the ionization probability per unit time. Here it is also necessary to consider the time dependence of the intensity in the interaction of a short laser pulse with atoms.

These complications were successfully overcome previously for the case of tunneling ionization[15] (see also the analytical approach of Ref. 16). They can be extended without any modification into the barrier-suppression range. However, they do not change the nature of the claims made above regarding the dependence of the angular and energy distributions of the outgoing photoelectrons for barrier-suppression ionization by a low-frequency circularly polarized laser radiation field.

These conclusions imply that the extrapolation of tunneling formulas into the barrier-suppression regime overestimates the value of the ionization probability. The angular distribution of the photoelectrons is concentrated in the plane of polarization of the radiation. The typical width of this distribution in the tunneling case is $\omega/\sqrt{F}$, and in the barrier-suppression case ω. The energy spectrum has a maximum for photoelectron energy equal to the oscillation energy $F^2/2\omega^2$ in a circularly polarized field. The spectral width in the tunneling case is $F^{3/2}/\omega$ and in the case of barrier-suppression ionization is $1/\omega$.

Note that in accordance with the result of Ref. 17 the inclusion of the Coulomb potential in the time-dependent part of the wave function of the final state via the Keldysh–Faisal–Reiss approximation implies that the maximum in the photoelectron energy spectrum shifts toward lower energies by an amount ω^2/F. For above-barrier fields this shift is of order ω^2. It is small in comparison with the oscillation energy $F^2/2\omega^2$.

The analogous results in the case of linear polarization require more work, since in the Keldysh–Faisal–Reiss approximation for the ionization probability sums of products of Bessel functions (the so-called generalized Bessel functions[18]) occur. Consequently, we first need to develop the asymptotic representations of these functions and derive formulas analogous to Eq. (18) of the present work for ordinary Bessel functions. We are currently engaged in this task.

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