Electron energy spectra in tunneling ionization of atoms and ions by a strong low-frequency electromagnetic field

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A simple analytical representation of the electron energy spectrum in tunneling ionization of atoms and ions by a strong low-frequency electromagnetic field is suggested. It is demonstrated that allowing for a finite initial kinetic energy of an electron at the moment of ejection by an atom improves the theoretical description of the phenomenon.

1. INTRODUCTION

Recent years have seen a revival of interest in multiphoton ionization of atoms of rarefied gases by intense laser pulses. Many experiments realize the so-called tunneling ionization mode in which what is known as the Keldysh parameter[1] (see below) is smaller or much smaller than unity. For instance, tunneling ionization of atoms by radiation with $\lambda = 248$ nm has been observed by Gibson *et al.*[2] The energy spectra of electrons in the tunneling ionization of atoms and ions of xenon and potassium by the radiation of a high-power carbon-dioxide laser has been studied both experimentally and theoretically by Xiong and Chin.[3] In this paper we focus on their results. We believe that the theoretical aspect deserves further analysis. The theoretical part of Ref. 3 is based on the assumption that at the moment of its ejection from an atom and electron carries a zero kinetic energy, although actually the ejected electrons are characterized by a certain kinetic-energy distribution, which in general form was obtained in Ref. 4. Here this distribution is used to analyze the experimental energy spectra obtained by Xiong and Chin.[3]

2. THE KINETIC-ENERGY DISTRIBUTION OF EJECTED ELECTRONS

We will briefly examine the main results of Ref. 4. The atomic system of units ($e = \hbar = M_e = 1$) is used throughout the paper.

The probability of tunneling ionization by an alternating electromagnetic field per unit time in the quantum-mechanical adiabatic Landau–Dykhne approximation[5] has the following general form (to within a pre-exponential factor of the order of unity):

$$w=\exp\{-2\,\mathrm{Im}\,S(t_0)\}. \qquad (1)$$

The time-dependent part of the classical action integral, $S(t_0)$, is given by the formula

$$S(t_0)=\int_0^{t_0}[E_f(t)-E_i(t)]\,dt,$$

where $E_i(t)$ and $E_f(t)$ are, respectively, the energies of the initial and final states of the system and depend on t as a parameter. The complex-valued time t_0 is the quasiclassical turning point determined by the equation

$$E_i(t_0)=E_f(t_0). \qquad (2)$$

We assume that the strength F of the electric field in the external electromagnetic wave is low compared to the atomic field strength. The perturbation inflicted by the field on the final state f in the continuous spectrum is, therefore, much stronger than the field perturbation of the initial bound state i. Hence, Eq. (2) can be written in a simpler form $E_f(t_0) = -E_i$, where E_i is the unperturbed ionization potential of the atom or ion considered.

The adiabatic approximation holds true when the frequency ω of the external electromagnetic field is much lower than $|E_i - E_{f0}|$, where the unperturbed value E_{f0} of the final-state energy can be taken as equal to the boundary of the continuous spectrum, or simply set at zero. The above condition can, therefore, be written as $\omega \ll E_i$.

For the sake of definiteness we take the case of linearly polarized electromagnetic radiation. This means that

$$E_f(t)={}^1\!/_2[\mathbf{p}_0+(1/c)\mathbf{A}(t)]^2={}^1\!/_2[\mathbf{p}_0-(\mathbf{F}/\omega)\sin\omega t]^2,$$

where $\mathbf{p}_0$ is the momentum of an electron ejected by an atom (the so-called canonical, or generalized, momentum).

Now we must calculate the imaginary part of the integral

$$S(t_0)=\int_0^{t_0}\{{}^1\!/_2[\mathbf{p}_0-(\mathbf{F}/\omega)\sin\omega t]^2+E_i\}\,dt.$$

Let us assume that $\mathbf{p}_0$ is aligned with the electric field vector $\mathbf{F}$. The probability of ionization in the direction perpendicular to the electromagnetic-field polarization is extremely low [see below the remark to Eq. (6) concerning the initial transverse momentum $p_{0\perp}$]. Bearing in mind that the lower limit of integration is a real quantity, we find that

$$\mathrm{Im}\,S=\mathrm{Im}\,\{[{}^1\!/_2p_0^2+E_i+(F^2/4\omega^2)]t_0 + (p_0F/\omega^2)\cos\omega t_0-(F^2/8\omega^3)\sin 2\omega t_0\}.$$

Since in the tunneling mode Re $\omega t_0 \ll 1$ [t_0 is determined by Eq. (2)], we expand the sines and cosines in power series. For that part of the result that is inversely proportional to the first-order term in F we obtain (after introducing the notation $k^2 = 2E_i$)

$$(k/F)[p_0^2/2 + E_i - p_0^2 + p_0^2/2 - k^2/6] = k^3/3F = (2E_i)^{3/2}/3F. \qquad (3)$$

This yields the well-known tunneling exponential in the ionization probability (see, e.g., Ref. 6).

The terms in the classical action integral that are inversely proportional to the cube of the electric field strength F in the wave have the following form:

$$(k\omega^2/3F^3)[p_0^2E_i-2E_i^2/5]. \qquad (4)$$

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The second term in (4) combines with (3) to produce

$$(k^3/3F)\,[1-2\omega^2E_i^2/5F^2k^2]=(k^3/3F)\,[1-\gamma^2/10].$$

Thus, we have obtained the well-known tunneling correction term for a linearly polarized field. The smallness of this term is due to the smallness of the Keldysh parameter, $\gamma=\omega k/F\ll 1$, in tunneling.

Substituting the first term in (4) into the general expression for the ionization probability, $w=\exp(-2\,\mathrm{Im}\,S)$, we find that it contributes the following factor to the probability:

$$\exp(-2k\omega^2p_0^2E_i/3F^3)=\exp(-p_0^2\gamma^3/3\omega). \tag{5}$$

This factor determines the kinetic-energy distribution of the ejected electrons in tunneling ionization.

3. THE ENERGY SPECTRUM

To match the above result with the well-known formula of Ammosov, Delone, and Kraĭnov[7] (the ADK theory), we multiply the electron kinetic-energy distribution specified by (5) by the corresponding expression for the ionization rate (ionization probability per unit time) in the ADK theory integrated over all the momenta of an ejected electron. Here, if we are interested only in longitudinal momenta, Eq. (5) must be normalized to unity. Bearing in mind that the ground states of the atoms and ions can for all practical purposes be assumed to have an orbital quantum number $l=0$ or $l=1$ and a magnetic quantum number $m=0$, we get

$$dw(l=1,\ m=0)=3dw(l=0)=(3\pi\omega/\gamma^3)^{1/2}(3e/\pi)^{3/2}Z^2n^{-4,5}\times(16eE_i^2/ZF)^{2n-1,5}\exp\{-2(2E_i)^{3/2}/3F-p_0^2\gamma^3/3\omega\}\,dp_0 \tag{6}$$

(here we have corrected an error of Ref. 7). In Eq. (6) Z is the ion charge, the effective principal quantum number n is defined as $n=z/(2E_i)^{1/2}$, and p_0 is the initial momentum of an electron ejected along the polarization axis. For real atoms the orbital angular momentum l is an approximate quantum number, since we assume that the interaction of an electron with the radiation field is weak compared to the energy separation from neighboring levels (say, compared to the scale of the fine structure of levels). In this case there is no degeneracy in the levels.

The term $p_0^2\gamma^3/3\omega$ in (6) brings a new element into the ADK theory: it determines how the ionization probability depends on the initial kinetic energy $p_0^2/2$ of an ejected electron.

The final electron-energy distribution, achieved after the electron has left the laser focus, differs greatly from the initial distribution in the case of long laser pulses because of the ponderomotive-force effect.[3,8] The ponderomotive force shifts the peak in the energy distribution away from the zero value according to Xiong and Chin,[3] or away from $p_0^2/2$ according to our research, to a certain value of the order of the electron ponderomotive vibrational energy (see also the theoretical paper of Goreslavskiĭ, Narozhnyĭ, and Yakovlev[9]).

Naturally, the initial distribution (6) affects the shift of the peak, but the dependence of (6) for the probability of ionization on the electron energy $p_0^2/2$ has not such a strong effect on this shift because the width $3\omega/\gamma^3$ of the energy distribution is small compared to the ponderomotive force. This dependence, however, introduces an additional broadening into the energy distribution, making it asymmetric.

Our objective in this paper is to obtain a simple analytical description of the energy spectrum that allows for a nonzero value of the electron's initial kinetic energy.

We are interested, of course, in the longitudinal initial electron momentum p_0 when the electron is ejected along the direction of the electric field vector **F**. Then the electron turns in the direction of the gradient of the ponderomotive force, that is, in the axial direction. The dependence on the electron's initial transverse momentum $p_{0\perp}$ can be obtained by substituting $E_i+p_{0\perp}^2/2$ for E_i in Eq. (6). However, since the electric field strength F is low, nonzero transverse momenta are unlikely to appear, that is, electrons are ejected primarily along the field's polarization.

Let us consider the spatial-temporal distribution of the laser radiation in the form of the Gaussian distribution (see, e.g., Ref. 6):

$$F=F_0\exp(-\rho^2/2R^2-t^2/2\tau^2). \tag{7}$$

Here τ is the length of the laser pulse, R the radius of the laser focus, F_0 the peak value of the electric field strength, and ρ the axial cylindrical coordinate in the direction perpendicular to that of the propagation of the laser radiation.

In a long laser pulse approximation the final energy E of the electron can be linked to the electron's initial energy $p_0^2/2$ through the following relation:

$$E=p_0^2/2+(F_0^2/4\omega^2)\exp(-\rho_0^2/R^2-\theta_0^2/\tau^2), \tag{8}$$

where ρ_0 and θ_0 are the axial coordinae and the time of emergence of the electron, respectively. As earlier, we are assuming that the radiation is linearly polarized.

Substituting Eqs. (7) and (8) into (6) and using the inequalities $\rho_0\ll R$ and $\theta_0\ll\tau$ [these values are important to (6)], we get

$$w=w_0\exp\{-[(2E_i)^{3/2}/2F_0-(2n-1,5)/2](\rho_0^2/R^2+\theta_0^2/\tau^2)-(2\gamma^3/3\omega)(E-F_0^2/4\omega^2)\}. \tag{9}$$

We have substituted F_0 for F in the Keldysh parameter because of the smallness of the parameter.

The electron yield is determined by averaging the ionization rate w over space and time in the absence of ionization saturation:

$$N_e=n_0h\iint w2\pi\rho_0\,d\rho_0\,d\theta_0. \tag{10}$$

Here n_0 is the initial concentration of atoms (or ions), and h the size of the focal volume along the laser beam. We introduce new coordinates r and ψ in the following manner: $\rho_0=Rr\cos\psi$ and $\theta_0=\tau r\sin\psi$. Integration with respect to ψ from $-\pi/2$ to $\pi/2$ in (10) yields

$$N_e=4\pi n_0hR^2\tau\int_{r_0}^{\infty}w(r)r^2\,dr. \tag{11}$$

According to Eq. (8), we have

$$E=\tfrac{1}{2}p_0^2+(F_0^2/4\omega^2)\exp(-r^2). \tag{12}$$

If $E>F_0^2/4\omega^2$, the domain of integration with respect to r in (11) is $[0,\infty]$, which corresponds to the range $[E-F_0^2/4\omega^2, E]$ of the electron's initial kinetic energies $p_0^2/2$.

Substituting (9) into (11) and calculating the integral, we find that

$$N_e = N_{max} \exp\{-(2\gamma^3/3\omega)(E - F_0^2/4\omega^2)\}, \quad E > F_0^2/4\omega^2. \qquad (13)$$

But if $E < F_0^2/4\omega^2$, the range of the electron energies is $0 \leqslant p_0^2/2 \leqslant E$, which corresponds to the interval $r_0 \leqslant r \leqslant \infty$, where r_0 can be found by solving Eq. (12), that is,

$$r_0^2 = (4\omega^2/F_0^2)(F_0^2/4\omega^2 - E). \qquad (14)$$

Calculating the integral in (11) with respect to r, we get

$$N_e = N_{max} \exp\{-(4\gamma^3/3\omega)(F_0^2/4\omega^2 - E)\}, \quad E < F_0^2/4\omega^2. \qquad (15)$$

Here we have ignored the $2n - 1.5$ in (9) owing to its smallness in comparison to $(2E_i)^{3/2}/F_0$. Note that Eqs. (13) and (15) were obtained in Ref. 9 for a zero-radius potential by expressing them in terms of the number of absorbed photons.

The position of the peak in the energy spectrum is determined by the value of the average vibrational energy of an electron in the field of an electromagnetic wave, $E = F_0^2/4\omega^2$. This is true only in the absence of saturation, when formula (10) can be used. As for the ADK theory, it affects only the value of w_0 in (13) and (15), strongly increasing it in comparison to the predictions of the zero-radius potential model. In the next section we will see, however, that when there is saturation the peak in the energy distribution shifts significantly toward lower energies, and the size of this shift depends on the value w_0 of the ionization rate.

From (13) and (15) it follows that the broadening of the peak in the energy spectrum is asymmetric. For $E > F_0^2/4\omega^2$ this broadening proves to be twice as large as for $E < F_0^2/4\omega^2$. In accordance with (12), the broadening can be qualitatively explained by the fact that the initial energy $p_0^2/2$ of an electron ejected by an atom at the moment of ionization is finite and by the presence of a spatial-temporal distribution in the intensity of laser radiation.

Figure 1 depicts the experimental data and theoretical curve for a potassium atom in tunneling-ionization conditions. The experimental data has been taken from the work of Xiong and Chin[3], with $F_0 = 7.29 \times 10^7$ V·cm^{-1}, $R = 100$ μm, and $\tau = 2$ ns. The theoretical curve was obtained from (13) and (15).

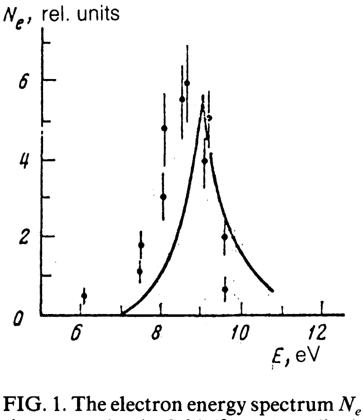

FIG. 1. The electron energy spectrum N_e in tunneling ionization of potassium atoms by the field of a carbon-dioxide laser with a radiation intensity of 10^{12} W·cm^{-2}. The experimental data has been taken from Ref. 3, and the solid curve corresponds to the results of calculations based on (13) and (15).

4. THE ENERGY PEAK

Although the above method provides a good description of the broadening of the energy spectrum, it does not determine the position of the energy peak exactly. For instance, for the data depicted in Fig. 1 (the frequency ω corresponds to a photon energy of 0.1 eV) the energy corresponding to the peak in the energy distribution is $E = F_0^2/4\omega^2 = 10.2$ eV, while in experiments the peak was found to be at $E = 8.5$ eV. The discrepancy is about 20%, and is still greater for atoms and singly ionized ions of xenon.[3]

The discrepancy appears because we have not allowed for the saturation effect in the electron yield in the above formulas. If we include saturation, formula (10) for the electron yield is replaced by a more complicated one,

$$N_e = n_0 h \int \left[1 - \exp\left(-\int_{-\infty}^{\infty} w\, d\theta_0\right)\right] 2\pi\rho_0\, d\rho_0. \qquad (16)$$

This transforms into (10) when saturation is absent, or

$$\int_{-\infty}^{\infty} w\, d\theta_0 \ll 1. \qquad (17)$$

Here, however, we turn to the opposite limiting case, where in a large part of the laser focus

$$\int_{-\infty}^{\infty} w\, d\theta_0 \gg 1. \qquad (18)$$

In this case the expression in brackets in (16) grows very rapidly from zero to unity at a certain value ρ_0 which can be determined to a high degree of accuracy from the condition

$$\int_{-\infty}^{\infty} w(\rho_0)\, d\theta_0 = 1. \qquad (19)$$

Then, in accordance with (16), for the electron yield we can write the following:

$$N_e = \pi\rho_0^2 h n_0. \qquad (20)$$

Thus, ρ_0 has the meaning of the radius of the region where saturation of the ionization probability occurs, that is, inside this region all atoms are ionized while outside the ionization probability is infinitesimal.

Substituting (9) into (19) and integrating with respect to θ_0, we find that

$$\rho_0^2 = R^2 \frac{2F_0}{(2E_i)^{3/2}} \ln\left\{ w_0\tau \left(\frac{2F_0}{(2E_i)^{3/2}}\right)^{1/2} \right\}. \qquad (21)$$

Here we have ignored the energy dependence of the ionization probability and the factor $2n - 1.5$ in (9) (the reason for the latter has been mentioned above). From (21) it follows that ρ_0 can be of the order of, or greater than, R since although $F_0 \ll (2E_i)^{3/2}$ because the strength of the laser field is low compared to the atomic field, the factor $\omega_0\tau$ may be so high, especially when the laser pulse is long, that its logarithm compensates for the low strength of the laser field. Note that even in a moderate field the value of w_0 determined by (6) proves to be large because of the pre-exponential factor inherent in the Coulomb problem. Thus, saturation is achieved much more easily for a potential of the Coulomb type than for a zero-radius potential.

In accordance with (12), the peak in the energy distribution occurs at

$$E = \frac{F_0^2}{4\omega^2} \exp\left\{-\frac{\rho_0^2}{R^2} - \frac{\theta_0^2}{\tau^2}\right\}. \tag{22}$$

Here we have ignored the electron's small initial kinetic energy $p_0^2/2$ and have allowed for the fact that the peak occurs precisely at the value of ρ_0 determined by condition (19). Indeed, at values greater than ρ_0 the ionization probability is infinitesimal, and at values smaller than ρ_0, in accordance with (20), the fractional size of the ionizing volume of the region is small.

In accordance with (9), the characteristic times θ_0 at which ionization occurs must obey the following condition:

$$\theta_0 \sim \left[\frac{2F_0}{(2E_i)^{3/2}}\right]^{1/2} \tau \ll \tau. \tag{23}$$

Thus, in (22) we can ignore θ_0^2/τ^2. Substituting the value of ρ_0 determined by (21) into (22), we arrive at the final expression for the position of the energy peak that allows for saturation of the ionization probability:

$$E = \frac{F_0^2}{4\omega^2} \left\{w_0\tau \left[\frac{2F_0}{(2E_i)^{3/2}}\right]^{1/2}\right\}^{-2F_0/(2E_i)^{3/2}} \tag{24}$$

Since, according to what has been said, ρ_0 can be of the order of or greater than R, the energy value given by (24) can be substantially lower than the value $F_0^2/4\omega^2$ in the absence of saturation. Thus, saturation leads to a marked decrease in the energy of the electrons being ejected.

5. CONCLUSION

Here we focus on Eq. (6), which plays an important part in our work. The first term in the exponent in this equation constitutes a well-known expression valid for both long-range and short-range potentials (see, e.g., Ref. 6). The pre-exponential factor is also known,[7] and is absent in the case of a short-range potential. Finally, the second term in the exponent is also known and in combination with the Gaussian distribution determines the possibility of somewhat improving the theoretical explanation of the experimental data of Xiong and Chin.[3]

It must be emphasized that the explanation of the electron energy spectrum in tunneling ionization given in the present paper is not restricted to the case of a potassium atom. All the results can be applied to other cases, say for the Xe and Xe^+ atoms mentioned earlier, provided that the saturation of ionization is allowed for in the calculations.

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