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Miladinovi, T. B.; Petrovi, V. M.

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Quasiclassical approach to tunnel ionization in the non relativistic and relativistic regimes

T. B. Miladinović and V. M. Petrović

*Department of Physics, Faculty of Science, Kragujevac University, Kragujevac, Serbia,
e-mail: violeta.petrovickg@gmail.com*

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Non relativistic and relativistic transition rates were observed for the deep tunnel regime for the case of a linear polarized laser field. The contribution of the initial momentum of an ejected photoelectron, the ponderomotive potential and the linear Stark shift for the Ar atom and its ions were taken into account. It was shown that, in both regimes, these processes have an influence on the transition rate behavior. The curves obtained for the transition rate show that with increasing laser field intensities and ion charge the influence of relativistic effects increases.

Keywords: Tunnel ionization; transition rate.

PACS: 32.80 - t; 32.80 Fb

1. introduction

Studying of the photoionization of atomic and molecular systems is one of the important ways to investigate the characteristic properties of the interaction of these systems with an electromagnetic field. This provides many fundamental insights into light-matter interactions and because of that it has been the subject of many theoretical and experimental investigations over the years. As a result, many theories were defined as the theoretical framework of these processes. Everything started in 1964 with the Keldysh theory [1] that showed, for the first time, that the tunnel effect and the multiphoton ionization of atoms are two limiting cases of photoionization. The theory is quasi classical meaning that an atom is described quantum-mechanically whereas the laser field is treated classically. Keldysh introduced the Keldysh parameter of adiabaticity, determined through the field strength F , the frequency ω of the external field and the binding energy E_i of the electron in the atom, $\gamma = \omega(2E_i)^{1/2}/F$ (in the atomic units) [1]. Tunneling ionization is the limiting case of a photoionization process, when the dimensionless Keldysh parameter $\gamma \ll 1$.

Tunneling ionization of atoms occurs in an intense laser field when the potential barrier of an atom is deformed in a way that bound electrons can tunnel through this barrier and “escape” from an atom easily. Tunneling plays a central role in the interaction of matter with intense laser pulses. For $\gamma \ll 1$ the multiphoton dynamics dominates. Soon after, Perelomov, Popov and Terent’ev developed the new PPT theory [2] to calculate the ionization rate for hydrogen like atoms in a linearly and circularly polarized laser field. Twenty years later Ammosov, Delone and Krainov derived the ADK theory [3] for the case of tunneling ionization for arbitrary complex atoms and atomic ions. This is still one of the commonly used models for calculating the ionization rate.

The ionization process is directly determined by the electron binding energy. When an atom is placed in an intense

laser field this field influences the electron’s binding potential, perturbs it and makes it much higher than the unperturbed value. There are at least two reasons for this increase: the linear Stark effect and the ponderomotive potential.

If an atom is placed in an external electric field, its energy levels are altered. This phenomenon is essential and known as the Stark effect. As the field increases, the states spread out in energy and therefore have linear Stark effects even in lower intensity fields.

The ponderomotive potential is caused by the quiver motion of an electron in an atom. The ponderomotive potential is directly proportional to the laser field intensity, meaning that its significance increases with increasing laser intensity. Therefore, the description of ionization must take into account both effects.

2. Corrections of the ionization potential

The field free ionization potential of an atom can change under laser irradiation. We observed two effects which change the ionization potential and, at the same time, the tunneling rate in non relativistic and relativistic domains of laser field intensities. In recent years (recently) modern laser systems are able to generate laser pulses with peak intensities up to $\sim 10^{22} \text{ Wcm}^{-2}$. This progress in laser physics and the laser technique enables numerous experiments opening a window to the physical phenomena occurring in the relativistic domain. They became significant for intensities up to $\sim 10^{18} \text{ Wcm}^{-2}$. We considered the deep tunneling regime, where the Keldysh parameter $\gamma \leq 0.2$. According to Reiss [4] the $\gamma \rightarrow 0$ limit also requires relativistic treatment.

An electron is always oscillating around its nucleus, but we did not take into account this motion in our analysis. We observed only electron motion in an oscillating electric field. The physical picture of this motion is mathematically described by the ponderomotive potential which represents the time average kinetic energy of the electron oscillating

in a laser field. We have an electron in a varying external laser field and with a kinetic energy large enough so Newtonian equations of classical physics $\ddot{\vec{r}} = \vec{\mathcal{F}}(\vec{r}, t)$ could be used (here and below, the atomic system of units is used $e = m = \hbar = 1$). $\vec{\mathcal{F}}(\vec{r}, t)$ is the electric field vector at the position of the electron. The field intensity should be large along the beam axis, and should fall smoothly to zero at some distance from the axis. $\vec{\mathcal{F}}(\vec{r}, t)$ can be decomposed into a fast oscillation term modulated by a slowly varying amplitude $\vec{F}(\vec{r})$. For a linearly polarized field:

$$\ddot{\vec{r}} = \vec{F}(\vec{r}) \cos \omega t. \quad (1)$$

As the light frequency from the optical range is very big, we can picture our electron wiggling slightly around the central position at the optical frequency. This picture is mathematically described if one adds a slow drift term, and a slow amplitude modulation, to the standard rapidly oscillating sinusoidal solution of this problem:

$$\vec{r}(t) = \vec{\alpha}(t) + \vec{\beta}(t) \cos \omega t, \quad (2)$$

where $\vec{\alpha}(t)$ is the coordinate of the position of the electron and $\vec{\beta}(t)$ the amplitude of the wiggling motion. The temporal variations of α, β take place in a time interval much longer than one optical cycle. That means we can expand $\vec{F}(\vec{r})$ in a series in β and keep the first two terms in the expansion:

$$\begin{aligned} \vec{F}(\vec{\alpha}(t) + \vec{\beta}(t) \cos \omega t) \\ = \vec{F}(\vec{\alpha}) + (\vec{\beta} \cdot \nabla) \vec{F}(\vec{\alpha}) \cos \omega t + \dots \end{aligned} \quad (3)$$

Substituting Eq. (2) and Eq. (3) into Eq. (1) we obtain:

$$\begin{aligned} \ddot{\vec{\alpha}} - \omega^2 \vec{\beta}(t) \cos \omega t = [\vec{F}(\vec{\alpha}(t)) \\ + (\vec{\beta} \cdot \nabla) \vec{F}(\vec{\alpha}(t)) \cos \omega t] \cos \omega t. \end{aligned} \quad (4)$$

On the right hand side of Eq. 4 we have the term which includes $\cos^2 \omega t$, a factor very rapidly oscillating at frequency ω . After averaging over the rapid oscillation at frequency ω will be equivalent in gain to the term on the left hand side which does not contain the $\cos \omega t$ factor, so we obtain:

$$\langle \ddot{\vec{\alpha}} \rangle = \frac{1}{2T} \int_0^T (\vec{\beta} \cdot \nabla) \vec{F}(\vec{\alpha}) dt. \quad (5)$$

There is also a correspondence between the remaining two terms multiplied by $\cos \omega t$:

$$\langle \vec{\beta} \rangle = -\frac{1}{\omega^2} \langle \vec{F}(\vec{\alpha}) \rangle. \quad (6)$$

Since α, β and F are slowly varying quantities compared to the optical frequency, their average values over an optical cycle do not change, thus we can remove the average sign in Eq. (5) and Eq. (6):

$$\ddot{\vec{\alpha}} = \frac{1}{4\omega^2} \nabla(\vec{F}^2). \quad (7)$$

Equation 7 indicates that under the influence of the laser field, ignoring the rapid oscillatory term, the movement of the slow-varying term is driven by an effective force:

$$\vec{f} = -\frac{1}{4\omega^2} \nabla(\vec{F}^2) \quad (8)$$

\vec{f} is normally referred to as the ponderomotive force which points along the gradient of the intensity. If the laser field is non-time-varying, this force is a conservative one, and can be associated with a potential given by:

$$U_p = \frac{F^2}{4\omega^2}, \quad (9)$$

or, in a standard system of units:

$$U_p = \frac{e^2 F^2}{4m_e \omega^2}. \quad (10)$$

From Eq. 9 follows that with increasing of the laser field intensity the influence of the ponderomotive potential on the field free ionization potential E_i becomes larger and it becomes more significant. Increase of the laser field intensity also leads to the relativistic domain and for these intensities the relativistic ponderomotive potential may be written in the following form $U_p^{\text{rel}} = \sqrt{c^4 + 2c^2 U_p} - c^2$ [5], where c is the speed of light and U_p is the nonrelativistic ponderomotive potential (see the Eq. 9). To account for the ponderomotive potential we replaced the ground bound state E_i with the shifted energy $E_{\text{ief}} = E_i + U_p = E_i + F^2/4\omega$ for the nonrelativistic and $E_{\text{ief}}^{\text{rel}} = E_i + U_p^{\text{rel}} = E_i + (\sqrt{c^4 + 2c^2 U_p} - c^2)$ for the relativistic domain.

As we already mentioned when an atom is placed in an external electrical field its energy levels undergo a splitting proportional to the field intensity. This effect is known as the linear Stark effect and unlike the weak fields where it may be neglected in a strong laser field that is not the case. Using the perturbation theory approximation the shifted ionization potential can be represented in the following form $E_{\text{ief}} = E_i + E_{St} = E_i + \alpha F^2/4$ [6], where E_{St} is the shift caused by the linear Stark effect and α is the static polarizability of the atom. It is convenient to use the perturbation theory for describing the aforementioned effects because of its simplicity and the availability of experimental data for polarizabilities for atoms and ions [7].

Having both effects in mind *i.e.* the shift caused by the ponderomotive potential and the linear Stark effect, the effective ionization potential E_{ief} , for the nonrelativistic (Eq. 11) and relativistic (Eq. 12) laser field intensity, can be expressed as:

$$E_{\text{ief}} = E_i + U_p + E_{St} = E_i + F^2/4\omega + \alpha F^2/4, \quad (11)$$

$$\begin{aligned} E_{\text{ief}}^{\text{rel}} &= E_i + U_p^{\text{rel}} + E_{St} \\ &= E_i + \sqrt{c^4 + 2c^2 U_p} - c^2 + \alpha F^2/4. \end{aligned} \quad (12)$$

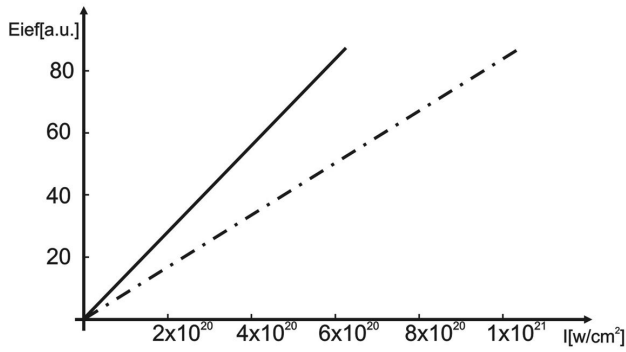


FIGURE 1. The effective ionization potential, $E_{\text{ief}}^{\text{rel}}$ (dot-dashed line represents the ionization potential corrected with the ponderomotive shift and the solid line when corrected with the ponderomotive and the Stark shift).

The presence of additional terms leads to increasing of the ionization potential (Fig. 1) which means that more photons or a stronger laser field is necessary for the photoionization process. For the intensity $\sim 10^{20} \text{ Wcm}^{-2}$ this increase is approximately 13%. Figure 1 shows that with increase of the laser field intensity the Stark shifted potential grows and this is in accordance with theoretical predictions.

When an atom is “exposed” to the external laser field its Coulomb potential is deformed and as a result the barrier through which the electron can tunnel is formed. The

resulting potential is the so-called effective potential. The correction of the ionization potential changes the value of the effective potential U_{eff} (see The Fig. 2). In the parabolic coordinates, as a function of variable η and the corrected ionization potential E_i (see the Eq. 11 and 12) this potential can be expressed in the form

$$U_{\text{eff}} = -\frac{(2n_2 + |m| + 1)(2(E_i + F^2/4\omega + \alpha F^2/4))^{1/2}}{\eta} - \frac{F\eta}{2}$$

for the nonrelativistic case and for the relativistic case in the form

$$U_{\text{eff}}^{\text{rel}} = -\frac{(2n_2 + |m| + 1)(2(E_i + \sqrt{c^4 + 2c^2 U_p - c^2} + \alpha F^2/4))^{1/2}}{\eta} - \frac{F\eta}{2}$$

where n_2 is the parabolic and m is the magnetic quantum number of the initial state. From all possible values of parabolic quantum number n_2 only $n_{2\text{max}}$ is interesting to us because the contribution of all others is negligible; $n_{2\text{max}} = n^* - 1$ where n^* is the effective quantum number, $n^* = Z/(2E_i)$.

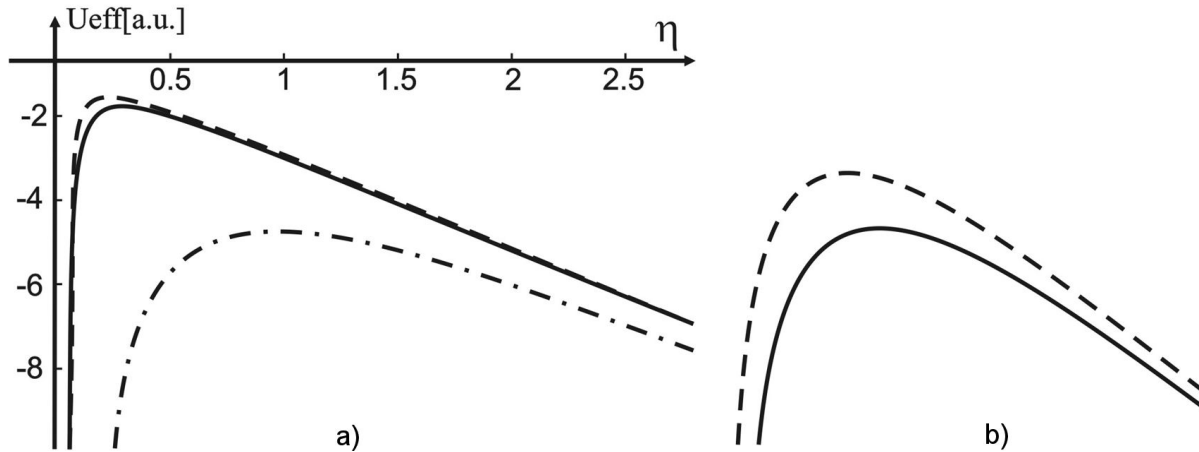


FIGURE 2. (a) The effective potential $U_{\text{eff}}^{\text{rel}}$ for the fixed laser field intensity at the value of $I = 10^{18} \text{ Wcm}^{-2}$ and $\eta(0, 2.5)$, (b) Shown maximum from (a). For both graphs the following notation is used: dot-dashed for the ionization potential without corrections, solid line for the ionization potential corrected with the ponderomotive shift and dashed line with the ponderomotive and the Stark shift, respectively.

TABLE I. The tunneling distances.

The laser field intensity Wcm^{-2}	The tunneling distance [a.u.]		
	$R_d = \frac{E_i}{F}$	$R_d = \frac{(E_i + F^2/4\omega)}{F}$	$R_d = \frac{(E_i + F^2/4\omega + \alpha F^2/4)}{F}$
10^{14}	10.7397	10.9763	11.126
10^{18}	$R_d = \frac{E_i}{F}$ 0.107397	$R_d = \frac{(E_i + F^2/4\omega)}{F}$ 23.7738	$R_d^{\text{rel}} = \frac{(E_i + (\sqrt{c^4 + 2c^2 U_p - c^2} + \alpha F^2/4))}{F}$ 38.7371

As we can see from Fig. 2 the potential barrier is increasing making the tunneling process harder and at the same time the transition rate decreases.

Here it is convenient to show how increasing of the laser field intensity influences the tunneling distance, $R_d = E_i/F$ [8]. It is assumed that an electron in the classical Coulomb well moves back and forwards along one chosen axis. With the aforementioned corrections of the ionization potential the tunneling distance can be written the following form as $R_d = (E_i + F^2/4\omega + \alpha F^2/4)/F$ for the nonrelativistic, and

$$R_d^{\text{rel}} = \frac{(E_i + (\sqrt{c^4 + 2c^2 U_p} - c^2) + \alpha F^2/4)}{F}$$

for the relativistic domain. The results obtained for the two chosen laser field intensities, one from the nonrelativistic (10^{14} Wcm^{-2}) and one from the relativistic domain (10^{18} Wcm^{-2}) are shown in Table I.

As can be seen from Table I, the influences of additional processes which certainly occur in an atom are more significant in the relativistic domain. But, no matter how small, they also exist in the nonrelativistic domain.

3. Correction of the transition rates

From all aforementioned it follows that these corrections of the ionization potential must be incorporated into an expression for the tunneling rate in order to obtain a more precise picture of the tunnel ionization process.

Krainov's expression for the relativistic transition rate [9] is given by formula:

$$W_{\text{rel}} = W_{\text{nonrel}} \text{Exp} \left[-\frac{2E_e \gamma^3}{3\omega} - \frac{E_e^2 \gamma}{c^2 \omega} \right], \quad (13)$$

where E_e is the kinetic energy of ejected electrons. This equation is obtained based on the Landau-Dykhne formula [9] (with exponential accuracy):

$$W_{if} \text{Exp}[-2Im \int_0^{t_0} (E_f(t) + E_i) dt], \quad (14)$$

where $E_f(t)$ is the relativistic energy for a free electron in the laser field [10] and E_i is the ionization energy for the initial state. This formula is also valid for the relativistic case when the aforementioned condition concerning the photon energy should be exchanged for a milder requirement that it must be small compared to the kinetic energy of the ejected electron. The classical relativistic motion of charged particles in a laser field is assumed. According to Eq. 13 the non relativistic part of the expression has an important influence on the general rate behavior. We first considered non relativistic ionization probability, W_{nonrel} in the frame of a widely used ADK theory with the correction for non zero initial momentum of the photoelectron

$$W_{\text{nonrel}} = \left(\frac{4z^3 e}{Fn^4} \right)^{2n-1} \text{Exp} \left[-\frac{2Z^3}{3Fn^3} - \frac{p_0^2 \gamma^3}{3\omega} \right] \quad (15)$$

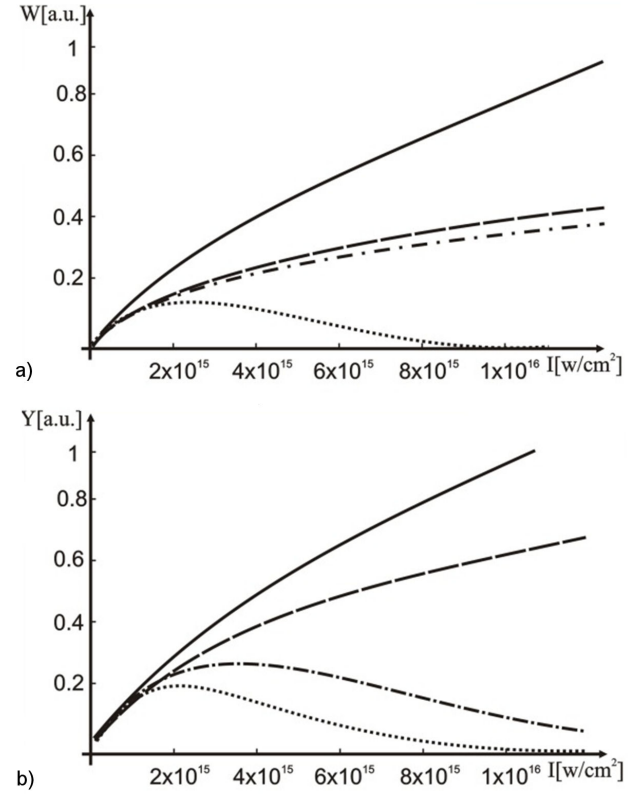


FIGURE 3. (a) Non relativistic tunnel transition rates, W_{nonrel} (b) Non relativistic tunnel yields, Y_{nonrel} . The laser field intensity = $10^{14} - 10^{16} \text{ Wcm}^{-2}$, the fixed parabolic coordinate at value $\eta = 150$ and $Z = 1$. For both graphs the following notation is used: solid line without any correction of the tunnel rate (yield), dashed line with a correction for the initial momentum of the ejected electron, dot-dashed line with a correction for the ponderomotive shift and dotted line with a correction for the ponderomotive and the Stark shift.

[11] (see Fig. 3). Here p_0 denotes the longitudinal component of the initial momentum, Z is the ion charge and n the principal quantum number. Selection of the ADK theory is in accordance with the fact that this theory for rare gas atoms such as Ar, Kr and Xe just for $\gamma < 0.5$ fits experimental results [12]. We incorporated corrections of the ionization potential in the expression for the momentum of the ejected photoelectron

$$p_0 = \frac{1}{2} \sqrt{\frac{\omega \sqrt{2(E_i + F^2/4\omega + \alpha F^2/4)}}{\gamma} \eta - 1} - \frac{1}{\eta \sqrt{\frac{\omega \sqrt{2(E_i + F^2/4\omega + \alpha F^2/4)}}{\gamma} \eta - 1}}, \quad (16)$$

where ω is the frequency of the electromagnetic laser field and F is the electric field strength. It is found that the observed physical picture can be best described using parabolic coordinates [13]. The parabolic coordinates are defined as

$\xi = r+z$, $\eta = r-z$ and $\phi = \arctan(y/x)$ where $\xi, \eta \in [0, \infty]$ and $\phi \in [0, 2\pi]$.

The obtained tunnel rates and appropriate yields,

$$Y = \int_{\tau} W dt,$$

are shown in Fig 3.

It can be seen that the ponderomotive potential has a more significant influence on the photoionization yield (see Fig. 3(b)).

Now it was interesting to observe how the tunnel rate behaves for larger laser field intensities, *i.e.* for values belonging to the relativistic domain. So we returned to Eq. 2. It is assumed that the major number of photoelectrons is focused along the polarization axis and that the electrons have moderate values of kinetic energies $E_e = \sqrt{p_0^2 c^2 + c^4} - c^2$ [9], $c = 137.02$ is the speed of light in atomic units. We transformed this expression into the following form:

$$E_e = \sqrt{\left(\frac{1}{2} \sqrt{\frac{\omega \sqrt{2(E_i + \sqrt{c^4 + 2c^2 U_p - c^2} + \alpha F^2/4)}}{\gamma}} \eta - 1 - \frac{1}{\eta \sqrt{\frac{\omega \sqrt{2(E_i + \sqrt{c^4 + 2c^2 U_p - c^2} + \alpha F^2/4)}}{\gamma}} \eta - 1} \right)^2 c^2 + c^4 - c^2}. \quad (17)$$

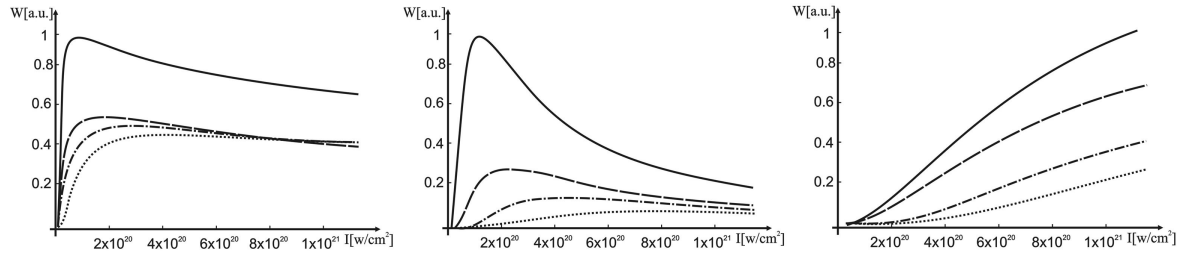


FIGURE 4. Relativistic transition rate W_{rel} for the ion charge $Z = 1, 5, 10$ respectively, the laser field intensity $I = 10^{18} - 10^{21} \text{ Wcm}^{-2}$ and the fixed parabolic coordinate at value of $\eta = 0.25$. For all graphs the following notation is used: solid line without any correction of the tunnel rate (yield), dashed line with a correction for the initial momentum of ejected electron, dot-dashed line with a correction for the ponderomotive shift and dotted line with a correction for the ponderomotive and the Stark shift.

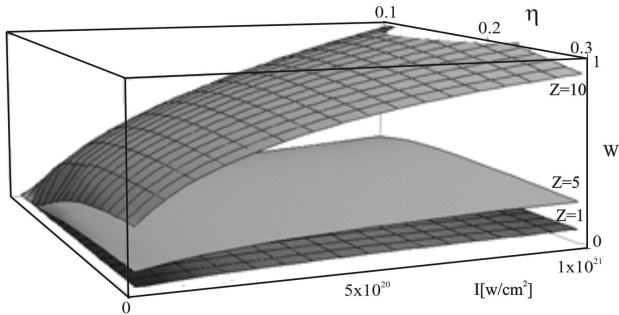


FIGURE 5. The relativistic tunnel transition rate, W_{rel} , for the ion charge, $Z = 1, 5, 10$, the laser field intensity $I = 10^{18} - 10^{21} \text{ Wcm}^{-2}$ and $\eta(0.1, 0.3)$.

In Eq. 17 we expressed the initial momentum taking into account the correction for the ionization potential (see Eq. 16).

We observed the relativistic transition rate based on Eq. 13 for the argon atom which is irradiated with a Ti-sapphire laser of $\lambda = 800 \text{ nm}$. Intensities are up to 10^{18} Wcm^{-2} . Figure 4 shows the dependence between the transition rate and the laser field intensities for $Z = 1, 5, 10$.

From Fig. 4 it is obvious that for all values of ion charges, $Z = 1, 5, 10$ incorporation of the momentum, ponderomo-

tive potential and the linear Stark shift leads to decreasing of the transition rate. Part of the laser pulse energy is used for increasing the momentum, ponderomotive potential and the Stark shift of ejected electrons living smaller amounts of light quanta available for ionization of the remaining electrons. But for $Z = 10$ the relativistic effects become more significant and behavior of the transition rate is different. The transition rate increases along the whole observed interval of laser field intensities. This is completely in accordance with the theoretical predictions. As one more illustration we give the dependence of the relativistic tunnel transition rate from the laser intensities and parabolic coordinate η for the ionic charge states $Z = 1, 5, 10$.

4. Conclusion

In this paper we analyzed tunnel ionization of atoms and ions. The ponderomotive potential and the Stark shift were taken into account. Nonrelativistic and relativistic treatment of the transition rate was used. The results of our theoretical analysis show that some usually neglected effects have an influence on the nonrelativistic as well as on the relativistic tunnel ionization rate.

Acknowledgments

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