Romanian Reports in Physics, Vol. 66, No. 4, P. 929-938, 2014

ATOMIC PHYSICS

SINGLE AND DOUBLE TUNNELING IONIZATION OF THE NOBLE GASES EXPOSED TO A LINEARLY OR CIRCULARY POLARIZED LASER FIELD

V. PETROVIĆ, T. MILADINOVIĆ, V. RISTIĆ

Kragujevac University, Faculty of Science, Department of Physics, Kragujevac, Serbia tanja.miladinovic@gmail.com

Receipt November 22, 2013

Abstract. We theoretically studied different influences like a nonzero initial momentum of ejected electron and the ponderomotive potential on the ionization probabilities and the ion yields for the noble gases atoms in a linearly and circularly polarized electromagnetic field whose intensity varies from 10^{12} to 10^{14} Wcm⁻². The ionization process occurred completely in the tunneling regime. The calculated values are in reasonable agreement with the experimental data.

Key words: tunneling ionization, ionization probability, initial momentum, ponderomotive potential.

1. INTRODUCTION

Rapid advancement of laser technology posed the interaction of intense laser field with atoms and molecules as the subject of many theoretical and experimental investigations. The study of ionization atoms and molecules by an intense laser fields is an important component of understanding light/matter interaction in highly nonlinear regimes and also a fundamental problem in atomic physics. The key features of most phenomena could be described by considering a single active electron which, after being ionized by tunneling (or multiphoton) ionization, is accelerated by the laser field: molecular aliment [1], high harmonic generation [2], above threshold ionization and dissociation [3], laser control and imaging [4]. For understanding of these phenomena the key role has investigated longer than fifty years. In 1964, Keldysh [5] derived the formula for the ionization probability of atom in the field which strength is compared to the atomic field strength. The most important finding is that the adiabaticity parameter γ (also known as Keldysh's parameter) defined as the ratio between laser field frequency, ω , and the

tunneling frequency, ω , $\gamma = \omega/\omega_t$. In atomic units, which will be used in this paper $(e = m = \hbar = 1)$, the Keldysh parameter has the following form:

 $\gamma = \frac{\omega}{\omega_t} = \frac{\omega\sqrt{2E_i}}{F}$, where F is strength of laser field and E_i is the ionization

energy. This parameter allows to distinguish two ionization mechanisms: for $\gamma >> 1$ the multiphoton ionization process is dominant and, in opposite case, $\gamma << 1$ tunneling.

Shortly after the appearance of Keldysh theory, Perelomov, Popov and Terent'ev [6] obtained analytical expression valid for arbitrary values of γ parameter. Ammosov, Delone and Krainov [7] derived the formula for the tunnel ionization probabilities of arbitrary complex atoms and atomic ions. In all of these theories (Keldysh, PPT and ADK) the exponent of the formulas are very similar and the photoionization rates show very similar behaviors. They are different form one another only in pre-exponential factors. Aforementioned theories are asymptotic and assume that, in the some part of space, the external electric field can be neglected. Because of its simplicity the ADK theory is utilized more frequently than others.

In this paper we considered the tunneling ionization probabilities and the ion yields for complex, noble atoms and single and double ionized corresponding atomic ions expressed by the ADK theory without and with correction for initial momentum of ejected photo electron and the ponderomotive potential. We reported obtained results for a linearly and circularly polarized electromagnetic field.

2. THE CORRECTION OF THE IONIZATION PROBABILITY

In this section we give theoretical background.

Depending on the intensity of the laser light, different mechanisms for the ionization process can be distinguished. In this paper, we focus our attention on the tunnel ionization mechanism. We used the ADK model which explains it very successful. We considered the ionization probability of an atom in an alternating electric field.

Here it is convenient start with the formula for the ionization probability in the static electric field [8]:

$$W_{stat} = \frac{4}{F} \operatorname{Exp}\left[-\frac{2}{3F}\right],\tag{1}$$

The ionization probability for the polarized laser field based on the Eq. 1 can be written in the following form [6]:

$$W(F,\omega) \propto W_{stat}$$
 (2)

For a linearly polarized laser field the Eq. 2 obtained well known form [9]:

$$W(F,\omega) = \sqrt{\frac{3F}{\pi (2E_i)^{3/2}}} W_{stat}, \qquad (3)$$

i.e. for a circularly polarized laser field:

$$W(F, \omega, \varepsilon) = \sqrt{\frac{3F}{\pi(1 - \varepsilon^2)(2E_i)^{3/2}}} W_{stat}, \qquad (4)$$

where is $0 < \varepsilon < 1$ [6]. From the Eq. 1, 3 and 4 it can be seen that the ionization probabilities in static and alternating electric field are different only by preexponential factor.

We notice that the negligence of some, more or less important, mechanisms and processes influence on the ionization probability. In this paper we took into account the influence of the ponderomotive potential on the ionization probability and a nonzero initial momentum and the ponderomotive potential on the ions yield. Because an interesting aspect of the tunneling ionization concerns the role of circular and linear laser pulses, we considered both cases of a laser field polarization.

The ionization probability in alternating linearly polarized laser field without correction for non-zero initial momentum of ejected electron is given by [10]:

$$W_{lin}^{ADK} = \frac{F_{lin}D_{lin}^2}{8\pi Z} \sqrt{\frac{3n^{*3}F_{lin}}{\pi Z^3}} \times \operatorname{Exp}\left[\frac{-Z}{3n^*F_{lin}E_i}\right],$$
(5)

where F_{lin} is field strength in atomic unit, $F_{lin} = \frac{27,5}{5.1 \times 10^9} \sqrt{Int}$ and

 $D_{lin} \equiv \left(\frac{4Z^3 e}{F_{lin}n^{*4}}\right)^{n^*}$, where $n^* = Z(2E_i)^{1/2}$ is the effective principal quantum

number.

When a free electron is placed in a laser field it possesses, in addition to any translational kinetic energy, quiver energy due to the oscillatory motion imparted on it by the field.

This quiver energy is the so-called ponderomotive potential and for linearly polarized laser field is $U_p^{lin} = F^2 / 4\omega^2$ [10]. The dependence of the photon energy and the ionization energy, kinetic energy and ponderomotive potential is given by following expression [10]:

$$E_k = K\hbar\omega - \left(E_i + \frac{F^2}{4\omega^2}\right). \tag{6}$$

In order to examine the influence of the ponderomotive potential on ionization probability we started from formula with correction for non-zero initial momentum of ejected electron:

$$W_{lin}^{ADK} = \frac{F_{lin}D_{lin}^2}{8\pi Z} \sqrt{\frac{3n^{*3}F_{lin}}{\pi Z^3}} \times \operatorname{Exp}\left[\frac{-Z}{3n^*F_{lin}E_i} - \frac{p^2\gamma^3}{3\omega}\right].$$
 (7)

Using the energy conservation relation (the Eq. 6), the ionization energy can be written as $E_i = K\omega - E_k = K\omega - p^2/2$. First we assumed $U_p = 0$. Here K, defined as $K = E_i/\omega + 1$ is the multiphoton order of the process *i.e.* the minimal required number of photons for the ionization process. By a series expansion, from the formula $p = \sqrt{-\frac{1}{4} + \frac{1}{2\eta} + \frac{1}{4\eta^2} + \frac{1}{4}F_{lin}\eta}$ [11, 12], for the case of outside barrier momentum, p, is obtained in the following form [11]:

$$p = \frac{1}{2} \left(\sqrt{F_{lin} \eta - 1} + \frac{1}{\eta \sqrt{F_{lin} \eta - 1}} \right), \tag{8}$$

where η is parabolic coordinate and for the case when electron is outside of the barrier, $\eta > 1/F_{lin}$ [12].

If a system's total energy is independent of the coordinate η then momentum is conserved along the classical path *i.e.* $p_{\eta} = p$ [13]. For observed intensities of laser field η takes the values from the interval (19–185).

We now modify the ionization potential E_i so that it incorporates the ponderomotive potential. Based on this the probability of ionization is described as:

$$W_{lin,p,Up}^{ADK} = \frac{F_{lin}D_{lin}^{2}}{8\pi Z} \sqrt{\frac{3n^{*3}F_{lin}}{\pi Z^{3}}} \times \operatorname{Exp}\left[\frac{-Z}{3n^{*}F_{lin}(K_{Up}\omega - \frac{p^{2}}{2} - \left(\frac{F_{lin}}{2\omega}\right)^{2})} - \frac{p^{2}\gamma^{3}}{3\omega}\right], (9)$$

where K_{Up} is the number of absorbed photons which takes into account introduced ponderomotive potential U_p^{lin} . For this case K is given by:

$$K_{Up} = \frac{E_i}{\omega} + \frac{\left(\frac{F^2}{4\omega^2}\right)}{\omega}.$$
 (10)

The ionization probability for the circular polarized light differs from the probability for linear polarized laser field by pre-exponential factor [10]:

$$W_{cir}^{ADK} = \frac{F_{cir}D_{cir}^2}{8\pi Z} \times \operatorname{Exp}\left[\frac{-Z}{3n^*F_{cir}E_i}\right],\tag{11}$$

٦

where are $D_{cir} = \left(\frac{4Z^3 e}{F_{cir} n^{*4}}\right)^{n^*}$ and $F_{cir} = \frac{19}{5.1 \times 10^9} \sqrt{Int}$ (in atomic units).

Repeating the procedure (derivation) it is easy to show that:

$$W_{cir,p}^{ADK} = \frac{F_{cir}D_{cir}^2}{8\pi Z} \times \operatorname{Exp}\left[\frac{-Z}{3n^*F_{cir}\left(K\omega - \frac{p^2}{2}\right)} - \frac{p^2\gamma^3}{3\omega}\right],$$
(12)

where is the expression for momentum given by the Eq. 8 (for F_{cir}) and $\eta > 1/F_{cir}$. For considered radiation intensities η takes the values from the interval (3-265). Finally we obtain:

$$W_{cir,p,Up}^{ADK} = \frac{F_{cir}D_{cir}^2}{8\pi Z} \times \operatorname{Exp}\left[\frac{-Z}{3n^*F_{cir}\left(K_{Up}\omega - \frac{p^2}{2} - \frac{1}{2}\left(\frac{F_{cir}}{\omega}\right)^2\right)} - \frac{p^2\gamma^3}{3\omega}\right].$$
 (13)

The ponderomotive potential in circular polarized laser field is given by $U_p^{cir} = \frac{1}{2} \left(\frac{F}{\omega}\right)^2 [10].$

In summary, we derived an expression for the ionization probabilities including both the dependence on a nonzero initial momentum and the ponderomotive potential. All above described cases have been considered in single active electron approximation which neglects the dynamics of the remaining bound electrons.

Usually, the measured value in experiments is the ion yield. So we have also analyzed it. The ionization yield can be calculated by integrating the probability over the pulse time:

$$Y = \int_0^\tau W \mathrm{d}t \;, \tag{14}$$

Here τ is the duration of a laser pulse.

3. DISCUSSION

In this section, we represent the results by using the improved analytical formulas for the ionization probabilities and the ion yields for the atomic system in a linearly and circularly polarized laser field. As has been noted above, we have studied the influence of the ponderomotive potential on the ionization probabilities on single and double ionized atoms and a non zero initial momentum and the ponderomotive potential the ion yields. We compared these results with experimental data.

Our calculations were performed for the laser photon energy $\omega = 0.004298$ a.u. and the laser field intensities, $I = 10^{12} - 10^{14}$ Wcm⁻². The Keldysh parameter was chosen as $\gamma = 0.5$. Noble, single and double ionized atoms are observed.

Figure 1 displays theoretical curves for the ionization probabilities for (a) Ar^+ and (b) Ar^{++} , calculated according to Eq. 7 and Eq. 9. Our numerical calculation predicts a deviation of the ionization probabilities compared to the standard ADK theory [14]. In our opinion at least two factors are responsible for the observed difference. The standard ADK formula assumes that the ejected electrons leave the atom with a zero initial momentum and that the ponderomotive potential is too small compared to the ionization energy, E_i . It would be natural to expect that these assumptions are not completely exact and we clearly show that.



Fig. 1 – The ionization probabilities, $W_{lin,p}^{ADK}$ and W_{lin,p,U_p}^{ADK} versus the laser field intensity, I, for: a) Z = 1, $W_{lin,p}^{ADK}$ max is for $I = 2 \times 10^{13}$ Wcm⁻² and W_{lin,p,U_p}^{ADK} max is for $I = 2.2 \times 10^{13}$ Wcm⁻²; b) Z = 2, $W_{lin,p}^{ADK}$ max is for $I = 1 \times 10^{13}$ Wcm⁻² and W_{lin,p,U_p}^{ADK} max is for $I = 1.75 \times 10^{13}$ Wcm⁻². The value of η is fixed at 190.

In Fig. 1a, we assume first that the ejected photoelectron has non zero initial momentum (blue solid line). This results the curve which deviates from the characteristic feature of the tunneling ionization probability. It is obviously that the

curves obtained by our improved formulas has the Gaussian shape, decreases very rapidly (with decreasing of the laser intensity) and asymptotically approaches to the x axis. The full width at half maximum (FWHM) is lower, which means that the tunnel ionization process can be occur for a more specific range of a laser field intensities. Second, we incorporate the ponderomotive potential in the ionization probability (green dashed line). It can be seen that the maximal values of the corrected ionization probability downshift and move to the right *i.e.* to the higher laser field intensities. The reason is that during the ionization process, a part of photons energy is "spent" on the initial momentum and the quiver energy of the ionized outgoing electrons (not only on the ionization energy, E_i). Because of that, the ionization probabilities are reduced. The similar behavior it can be observed for Ar⁺⁺, Fig. 1b. But, the maximal values of the ionization probabilities are greater and on lower field intensities. This is because the ionization of first electron disturbs stabile closed shell structure and second electron is ionized from an open shell.

The same approach is used for a circularly polarized laser field. In Fig. 2 we plot values of the ionization probabilities based on Eq. 12 and Eq. 13.



Fig. 2 – The ionization probabilities, $W_{cir,p}^{ADK}$ and W_{cir,p,U_p}^{ADK} versus the laser field intensity, I, for: a) Z = 1, $W_{cir,p}^{ADK}$ max is for $I = 7 \times 10^{12}$ Wcm⁻² and W_{cir,p,U_p}^{ADK} max is for $I = 1.4 \times 10^{13}$ Wcm⁻²; b) Z = 2, $W_{cir,p}^{ADK}$ max is for $I = 4 \times 10^{12}$ Wcm⁻² and W_{cir,p,U_p}^{ADK} max is for $I = 1 \times 10^{13}$ Wcm⁻². The value of η is fixed at 270.

We note that in the circularly polarized laser field the maximums of the probabilities are moved to the lower intensities. From Fig. 2, it is clear that the probability curves with the ponderomotive correction have significant spreading of FWHM *i.e.* of the intensity range on which the tunnel ionization can be occurred. As in the case of a linearly polarized laser field, the probabilities are greater for ionization of a second electron, Z = 2.

Similar results were obtained for the others noble atoms.

Next we present the results for the ion yields calculated by Eq. 14 and the appropriate formulas for the ionization probabilities, Eqs. 5, 7, 9, 11, 12 and 13.

Figure 3 illustrates the results for the ion yields as a function of laser intensity, (a) Ar^+ and (b) Ar^{++} , predicted by the ADK theory in a linearly polarized laser field.



Fig. 3 – The ionization yields, Y_{lin}^{ADK} , $Y_{lin,p}^{ADK}$, and Y_{lin,p,U_p}^{ADK} versus the laser field intensity, *I*, for: a) Z = 1; b) Z = 2. The value of η is fixed at 190.

The plotted yields demonstrate that the curves taking into account corrections for a non zero initial momentum and the ponderomotive potential are shifted to the lower intensities in comparison with the pure ADK theory. This is more obviously for the doubly charged ion yields, Z = 2. This behavior is completely in the accordance with the conclusions about corresponding probabilities. For the circularly polarized radiation the yield curves are shown in the Fig. 4.



Fig. 4 – The ionization yields, $Y_{cir,p}^{ADK}$, $Y_{cir,p,U}^{ADK}$, and Y_{cir,p,U_p}^{ADK} versus the laser field intensity, *I*, for: a) Z = 1; b) Z = 2. The value of η is fixed at 270.

The results are given comparatively, with and without corrections. From the Fig. 4, we can see that, in a circularly polarized laser field, the influence of additional factors is more significant. Especially we note the ponderomotive potential. It is obviously that in a circularly polarized laser field the movement of electron which as a consequence has the ponderomotive potential becomes more interesting for considered processes.

The transition rate is usually numerically calculated value, since the ion yield is more often measured in experiments. We compared our theoretically obtained results with experimental data. In Fig. 3 is shown the ion yields for the case of linearly polarized laser field and the main result of this work can be summarized as follows: the line which presents the ion yield without any correction doesn't fit in the area with larger ion yield with experiment, but the yield with corrections $Y_{lin,p}^{ADK}$ and $Y_{lin,p,Up}^{ADK}$ fitted well [15]. We also found agreement with other experimental data [16, 17]. There are fewer available experimental results for the circularly polarized laser field. For us was interesting comparison with them given in Kornev's work [18]. In this paper we found characteristic shape of the ion yield which we obtained when we included both corrections in expression for the ion yields, the Fig. 4.

It is important to explore this aspect in more details and this will be the subject of our future researches.

4. FINAL REMARKS

In summary, improved calculations of the tunneling ionization probabilities and the corresponding ion yields of the noble atoms exposed to two different polarized laser fields were discussed. We show that the ionization probability (and corresponding ion yields) clearly depends from additionally incorporate influences over the entire intensity range under study. We find that the results according to these formulas agree closely with the experimental data.

The photoionization probabilities for the circular polarization were found to be uniformly lower than for the linearly polarized case in the intensity range under consideration. On the other hand, the ionization process in the case of a circular polarized laser radiation occurs on lower intensities of laser field.

The ionization curves for the yield of single and doubly charged ions are described well by the ADK expressions both for linear and circular polarization. Good agreement with the measurement ion yields, permit us to conclude that the ADK expressions are applicable for single and double atomic tunnel ionization.

In future calculations our expressions may have to be modified with to allow for the possibility of the other influences on the ionization probabilities especially for the ionization of second electron.

10

Acknowledgements. We are grateful to the Serbian Ministry of Education and Science for financial support through Projects 171020 and 171021.

REFERENCES

- 1. S. Kaziannis, C. Kosmidis, A. Lyras, J. Phys. Chem. A, 112, 4754–4764 (2008).
- 2. X. X. Zhou, M. X. Tong, Z. X. Zhao et C. D. Lin, Phys. Rev. A, 72, 033412-033418 (2005).
- 3. A. Zavriyev, P. H. Bucksbaum, H.G. Muller, D.W. Schumacher, Phys. Rev. A, 42, 5500-5513 (1990).
- 4. S. X. Hu, L. A. Collins, Phys. Rev. Lett., 94, 073004-073007 (2005).
- 5. L. V. Keldysh, Sov. Phys. JETP, 20, 1307-1314 (1965).
- 6. A. M. Perelomov, V. S. Popov, M. V. Terent'ev, Sov. Phys. JETR, 23, 924-934, (1966).
- 7. V. M. Ammosov, N. B. Delone, V. P. Krainov, Sov. Phys. JETP, 64, 1191-1194 (1986).
- L. D. Landau, E. M. Lifshitz, *Quantum Mechanics: Non-Relativistic Theory*, 3rd ed. Pergamon, Oxford, 1991, 296.
- 9. C. Z. Bisgaard, L. B. Madsen, Am. J. Phys., 72, 2, 249-254, (2004).
- 10. N. B. Delone, V. P. Krainov, Physics Uspekhi, 41, 5, 469–485, (1998).
- 11. D. Bauer, *Theory of intense laser-matter interaction*, Max-Planck Institute, Heidelberg, Germany, 2006, 58.
- 12. L. D. Landau, E. M. Lifshitz, *Quantum Mechanics: Non-Relativistic Theory*, 3rd ed. Pergamon, Oxford, 1991, 295.
- 13. L. D. Landau, E. M. Lifshitz, *Quantum Mechanics: Non-Relativistic Theory*, 3rd ed. Pergamon, Oxford, 1991, 298.
- 14. N. B. Delone, V. P. Krainov, Physics Uspekhi, 38, 11, 1247-1269, 1995.
- 15. A. Wirth, Attosecond transient absorption spectroscopy, PhD Thesis, Ludwig-Maximilians-Universität, München, 2011.
- 16. C. M. Maharjan, *Momentum imaging studies of electron and ion dynamics in a strong laser field*, PhD Thesis Kansas State University, Manhattan, Kansas, 2007.
- 17. U. Eichmann, M. Dörr, H. Maeda, W. Becker, W. Sandner, Phys. Rev. Lett., 84, 3550–3553, (2000).
- 18. A. S. Kornev, E. B. Tulenko, B. A. Zon, Phys. Rev. A, 68, 0434141-0434149 (2003).