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Abstract

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The tunneling photoionization rate for a two color (bichromatic) field consisting of coherent superposition of the fundamental laser field frequency ω and its second harmonic with frequency 2ω , was studied theoretically within the framework of the adiabatic Landau–Dykhne approach. Analytical expressions were derived for the case of fixed specified values of relative phase shift, $\varphi = 0$ between the harmonics of the incident bichromatic field.

Keywords: transition rate, tunneling, two color

(Some figures may appear in colour only in the online journal)

Introduction

With the availability of powerful laser systems, the study of atomic processes in a strong laser field has become the subject of many theoretical and experimental investigations. Almost everything that we know about structure, we know from investigations of these processes. Atom and molecule ionization are such processes and as such an important component of understanding light/matter interactions in highly nonlinear regimes and also a fundamental problem in atomic physics. There are two types of ionization mechanisms, multiphoton and tunneling. To distinguish these types Keldysh [1] introduced the Keldysh adiabatic parameter: $\gamma = \sqrt{2E_i}/(F/\omega)$, where E_i is the ionization potential, ω is the frequency, and F is the strength of the laser field. In the regime of the Keldysh parameter $\gamma \gg 1$ ionization in a strong laser field can successfully be described as multiphoton, while for $\gamma \ll 1$ as a tunneling process. Tunneling ionization occurs when the laser pulse is intense enough to perturb the bound electron so that its atomic orbit is modified, and the binding potential between the electron and positively charged nucleus is lowered to a point where the probability of an initially bound electron tunneling through the potential well to become free becomes large.

There are several analytical tunneling models [2, 3]. One of the most used, the Amosov–Delone–Krainov (ADK), gives good agreement with the time dependent Schrodinger equation [4, 5] and excellent agreement with experimental results [6]. This model for a linearly polarized laser field and the non zero initial momentum, p, of ejected photoelectrons describes the transition ionization rate as an exponential dependence on the field strength, F, the ionization potential of state, E_i , the effective principle quantum number, n^* [7]:

$$W^{\text{ADK}} = \frac{FD^2}{8\pi Z} \sqrt{\frac{3n^*F}{\pi Z^3}} \times \exp\left[\frac{-Z}{3n^*FE_i} - \frac{p^2\gamma^3}{3\omega}\right]$$
(1)

where *F* is the field strength in atomic units, $F = \frac{27.5}{5.1 \times 10^9} \sqrt{I}$, *I* is the field intensity, $D \equiv \left(\frac{4Z^3 e}{Fn^{*4}}\right)^{n^*}$ and $n^* = Z/(2E_i)^{1/2}$ is the effective principal quantum number [8], with *Z* being the ionization state level. In equation (1) the initial momentum of the ejected electron is taken as $p = \frac{1}{2} \left(\sqrt{F\eta - 1} + \frac{1}{\eta \sqrt{F\eta - 1}} \right)$ [9]. It is expressed via the field strength, *F* and parabolic coordinate η , and for the case when the electron is outside the barrier, it can acquire any value $\eta > 1/F$ [10]. If a system's total energy is independent

of the parabolic coordinate η then the momentum is conserved along the classical path, $p_{\eta} = p$ [10]. The rate W was derived based on the assumption that the laser frequency is low, excited states play no role, and the Keldysh parameter is small compared with unity. In this case we consider only direct photoelectrons ($E_e < 2U_p$) which dominate in the total photoionization yield.

In this paper we present a theoretical extension of the ADK theory rate formula for two color laser pulses. These results were compared with the experimental results obtained based on the quasi-static model for ionization and also theoretical prediction of the ADK theory.

Theory

Lately, dual-wavelength (two color, bichromatic) lasers have been a hot international research topic [11-13]. They are natural tools in atomic control research as they offer practical control parameters such as polarization, amplitudes and phases [14-16].

Depending on the relative phase between consisting waves there are three different polarizations. If they are in phase, $\varphi = 0$, it is linear polarized light. If $\varphi = \pi/2$ or an odd multiple of $\pi/2$, the polarization is circular. For everything else, the light is elliptically polarized. Experiments with the fundamental and the second harmonic of a laser field showed that the relative phase significantly influences the ionization yield, photoelectron spectra, and also angular distributions [17].

We consider an atom in the single active electron approximation interacting with a linearly polarized two colors laser field. The aim is to take into account simultaneous contribution of both waves to the transition rate. Within the framework of the adiabatic Landau–Dykhne approach [18], we calculated the transition rate for tunneling photoionization for the case of the aforementioned field consisting of the coherent superposition of a fundamental field and its second harmonic which, in the interaction region, can be described as: $F(t) = F_1 \cos(\omega_1 t) + F_2 \cos(\omega_2 t + \varphi)$. In this case F_i , ω_i (i = 1, 2) are the respective amplitude and the angular frequencies of two pulses. For the purpose of this paper, we selected, $\omega_1 = \omega$, $\omega_2 = 2\omega$ and $F_1 = F_2 = F_0$. Accordingly, the inline equation for F(t) becomes:

$$F(t) = F_0(\cos(\omega t) + \cos(2\omega t + \varphi)), \qquad (2)$$

where F(t) presents the coherently combining two laser waves with equal strength F_0 . We assumed a two color input field without any spatial dependencies. In what follows, we employ a system in which F_1 and F_2 lie in the z direction. Since all motion takes place along the polarization axis, the problem can be treated one dimensionally. Here and throughout the paper atomic units, $e = m = \hbar = 1$, are used unless otherwise stated.

The exponential accuracy of the transition rate between the initial bound state i with the ionization potential E_i of the considered atomic ion and final continuum state f with the energy E_f from the imaginary part of the action $S(\tau)$ is starting point [19]:

$$W \propto \exp\left[-2\mathrm{Im}S(\tau)\right] \propto \exp\left[-2\mathrm{Im}\int_0^\tau (E_{\rm f}(t) - E_{\rm i}(t))\mathrm{d}t\right]$$
(3)

where E_i is the initial and E_f the final state energy, and τ is the complex turning point in the time plane. In equation (4), $\text{Im}S(\tau)$ is given by $\text{Im}S(\tau) = \text{Im} \int_0^{\tau} (E_f(t) - E_i(t)) dt$. The Landau–Dykhne adiabatic approximation is based on the fact that the slowness of perturbation causes a long duration of transition processes, and therefore part of the timedependent effects of $S(\tau)$ have large values leadings to quasi classical understanding of the problem and in that sense the problem is semi-classical and the condition $E_i(\tau) = E_f(\tau)$, must be satisfied [20].

In order to calculate equation (3), we started from the expressions for initial, $E_i(t)$ and final, $E_f(t)$ states:

$$E_{\rm i}(t) = -E_{\rm i} \tag{4a}$$

$$E_{\rm f}(t) = \frac{1}{2} \left(\vec{p} - \frac{\vec{A}}{c} \right)^2$$
$$= \frac{1}{2} \left(p - \frac{1}{c} \left(-\frac{c}{\omega} F_0 \sin \omega t - \frac{c}{2\omega} F_0 \sin(2\omega t + \varphi) \right) \right)^2 (4b)$$

where $\overline{A}(t)$ in $E_{\rm f}(t)$ denotes the vector potential. Based on $\overline{F}(t) = -\frac{1}{c} \frac{d\overline{A}(t)}{dt}$ follows: $A(t) = -\frac{c}{\omega} F_0 \sin \omega t - \frac{c}{2\omega} F_0 \sin(2\omega t + \varphi)$. We assumed that pulses are short enough to pass over the electron before it has a chance to experience the transverse spatial gradient of the focused pulse. In this event, the spatial dependence of the vector potential A(t) can be neglected [21]. Also in $E_{\rm f}(t)$ we take into account the initial electron's momentum: $p \neq 0$.

From the condition $E_i(\tau) = E_f(\tau)$, where τ is the classical turning point in the complex plane, follows $ik = p + \frac{F_0}{\omega} \sin \omega \tau + \frac{F_0}{2\omega} \sin(2\omega \tau + \varphi)$, $k = \sqrt{2E_i}$. After a few simple transformations and using the Maclaurin series $\arcsin x = \sum_{n \ge 0} \frac{(2n-1)!! x^{2n+1}}{2n!! (2n+1)}$, we obtained the expression for the turning point, τ in the form:

$$\tau = \frac{1}{2\omega} \left(\alpha + \frac{1}{6} \alpha^3 - \frac{\varphi}{2} \right),\tag{5}$$

where α is the parameter defined as: $\alpha = \frac{\omega(ik-p)}{F_0 \cos \frac{\varphi}{2}}$. The obtained values correspond to 'turning points' from classical mechanics, in which, this transition is forbidden. Because of that the solution of equation (5) is complex.

Based on equations (4a) and (4b) we expressed the imaginary part of the action $S(\tau)$ which, in general, depends on the electric field frequency and strength, as well as the phase φ . We integrated it: ImS(τ) = Im $\int_0^{\tau} \left(\frac{1}{2} \left(p + \frac{F_0}{\omega} \sin \omega t + \frac{F_0}{2\omega} \sin(2\omega t + \varphi)\right)^2 + E_i\right) dt$, and as a result, the expression as a sum of eight terms was obtained. We transformed each using expansion of the trigonometric functions and τ degrees and grouped obtained items according to the degree of the introduced parameter α . Taking into account the assumption for a linearly polarized monochromatic field, $\varphi = 0$, we separated real and imaginary parts. Only the imagine part of α^n , $n \leq 5$, interest us. Also, because of the very strong field, the contribution of terms with $\frac{1}{F_{\alpha}^{n}}$, n > 3 are neglected. According to the all mentioned, the transition rate can be done through the following formula:



Figure 1. Comparative review of the transition rate of the resulting two color field (dashed black line), W_{TC}^{ADK} , and the ADK rate, W^{ADK} (dashed red line), as a function of the laser field intensity, $5 \times 10^{13} < I < 5 \times 10^{16}$ W cm⁻². The initial momentum is set to be zero, p = 0.



Figure 2. Comparative review of the transition rate of the resulting two color field (solid black line), W_{TC}^{ADK} , and the ADK rate, W^{ADK} (solid red line), as a function of the laser field intensity, $5 \times 10^{14} < I < 5 \times 10^{16}$ W cm⁻². The initial momentum, $p \neq 0$.

$$W_{\rm TC}^{\rm ADK}(p, E_{\rm i}, F_0, \omega) \propto \exp\left[-2\left(\frac{1}{2}\frac{Z}{n^*}\left(\frac{1}{2}\frac{F_0}{\omega^2} + \frac{E_{\rm i}}{F_0}\right) - \frac{1}{12}\frac{Z^3}{n^{*3}F_0} + \gamma^3 \frac{1}{144}\frac{F_0^2}{\omega^8} - p^2\left(\gamma \frac{1-\omega}{4\omega^2} - \gamma^3 \frac{1+\omega}{8\omega^2}\right)\right)\right]$$
(6)

where the superscript ADK denotes the ADK basic theory, and the subscript TC denotes two colors pulses. It is obvious that we introduced the Keldysh parameter, γ , and effective quantum number, n^* . The derived formulas are applicable as long as $\gamma \rightarrow 0$. The last term in the exponential part on the right side of the above equation shows that the initial momentum will contribute additionally to the transition rate in the laser field direction.

Let us now consider obtained resulting expression. It consists of two parts: the first independent from the initial momentum of ejected photoelectrons and the other momentum dependent. From equation (6) follows that the momentum independent contribution have two parts: one determined by the effective quantum number, n^* and the ion charge, Z (the first and the second term on the right side of the above equation) and the second (the third) by the Keldysh parameter, γ . This is significantly different from the case of the one color laser (see equation (1)). The term $p^2 \left(\gamma \frac{1-\omega}{4\omega^2} - \gamma \frac{31+\omega}{8\omega^2} \right)$ is contribution caused by the initial momentum of ejected photoelectrons. It is important to note that in the limiting case, for a low electron momentum, we supposed that p^2 affect most of the order of p^4



Figure 3. Comparative review of the single, Z = 1 and double ionization, Z = 2 rate's curve. The green solid line corresponds to the single ionization rate, while black is the double. $\gamma = 0.003$ and $p \neq 0$ for both curves.

and that, for a strong field, the terms $\left|\frac{1}{F_0}\right|$ have larger contribution than $\left|\frac{1}{F_0^3}\right|$, $\left|\frac{1}{F_0}\right| \gg \left|\frac{1}{F_0^3}\right|$. Bearing all this in mind follows that the second degree of the initial momentum, determines the momentum dependent part of the transition rate.

Result and discussion

In this section we present theoretical results using our analytical formula (equation (6)) for an atomic system and compare them with experimental results and the ADK theory. For



Figure 4. Comparative review of the transition rate of the resulting two color field (dashed black line), W_{TC}^{ADK} , and the ADK rate, W^{ADK} (dashed red line), as a function of the laser field intensity, $10^{13} < I < 10^{16}$ W cm⁻². The initial momentum, p = 0.



Figure 5. Comparative review of the transition rate of the resulting two color field (solid black line), W_{TC}^{ADK} , and the ADK rate, W^{ADK} (solid red line), as a function of the laser field intensity, $10^{14} < I < 10^{16}$ W cm⁻². The initial momentum, $p \neq 0$.

the purpose of this work, the intensity of the low frequency field was varied within the characteristic interval for tunneling, $I = 10^{13} \,\mathrm{W \, cm^{-2}}$ to $I = 10^{16} \,\mathrm{W \, cm^{-2}}$ and the Keldysh parameter had values $\gamma \rightarrow 0$. The wavelength of $\lambda = 800$ nm, Ti: sapphire short, with the corresponding laser photon energy ($\omega = 0.05696$ a.u.) is used. Noble and alkali atoms, single and double ionized were considered.

The discussion of results will be separated into two main parts, one devoted to tunneling ionization with zero initial momentum of the ejected photoelectrons and the other with non zero initial momentum. Each part will be give corresponding graphs and a brief discussion.

First, a single ionized atom argon, Ar, is observed.

In figure 1 we show the transition rate W_{TC}^{ADK} as a function of the field intensity, calculated using equation (6) (dashed black line) with the assumption of a zero initial momentum of ejected photoelectrons. For this purpose we set p = 0 and isolated the momentum independent part in W_{TC}^{ADK} . The corresponding curve is shown as a solid line in figure 1. The position of the theoretical ADK curve is also denoted in the figure.

Figure 1 shows a significant deviation in shape occurring between the curves. The W_{TC}^{ADK} curve has an asymmetric shape and seems to approach the field intensity axis, unlike W^{ADK} . This behavior clearly shows the stabilization effect which is here defined to be that property of atoms in strong laser fields in which continued increases in field intensity will lead to a decrease in the photoionization transition rate, unlike W^{ADK} .

 $W_{\text{TC}}^{\text{ADK}}$ has a maximum for $I = 8.6 \times 10^{13} \text{ W cm}^{-2}$. There is also an obvious shift of the $W_{\text{TC}}^{\text{ADK}}$ curve to the right indicating the possibility of tunneling at lower field intensities. This noticeable difference could be explained through a simple analysis of the laser field that results from the sum of the two laser fields.

One of the questions that arise is how the initial momentum affects the transition rate. In other words does the initial momentum of the ejected photoelectron influence the transition rate and if it does, how much? From [22] we know that for a linearly polarized monochromatic field the answer is positive. In order to examine this, we assumed non zero initial momentum of the ejected photoelectrons in equation (6), $p \neq 0$. As a result the graphs in figure 2 are obtained.

It is obvious that W^{ADK} is more sensitive to the initial momentum than W^{ADK}_{TC} under the same conditions. In this case curves have a more similar behavior. Both first grow almost exponentially, reaching a maximum and then decrease. The W^{ADK}_{TC} curve has a much sharper slope. The W^{ADK}_{TC} curve is again shifted to lower values of field intensity and the rate intensity, W^{ADK}_{TC} is much lower than for W^{ADK} . However, although in the higher region the W^{ADK}_{TC} dependence is a decreasing function of field intensity, nonetheless, this dependence does not follow the W^{ADK}_{TC} prediction and noticeable deviation is noted and strongly related to the considerably altered incident bichromatic field.



Figure 6. 3D graphs for the potassioum atom, $\mathbf{Z} = 2$, (a) for the parabolic coordinate $\eta = 9 - 9.3$, (b) for the Keldysh parameter $\gamma = 0.002 - 0.003$.

We then applied our formula to a double ionized Ar atom. The curve behaves in a similar way as the one for a single ionization. In figure 3 a comparative review of the probability of a single and double ionized Ar atom is shown in a contour plot against the field intensity.

In the observed range of intensities a significant variability of probabilities is obvious. More precisely in this region the level of single ionization rate is higher than double, which is in accordance with [23]. It can be seen that both graphs first show a rapid rate increase to a maximum value and then decrease and approach the field intensity axis for a monotonic increase in laser field intensity. The double rate curve's maximum is shifted towards higher field intensity and reaches a maximal value at $I = 1.6 \times 10^{14}$ W cm⁻², while the single rate curve maximum is at $I = 1.1 \times 10^{14}$ W cm⁻².

Similar results were obtained for other noble atoms.

There are not many available experimental results. The comparison given in [24, 25] was interesting for us, as it shows shifting of ionization curves to lower values of field intensities compared to those predicted by the ADK formula (because of the different of parameters we were not able to perform a complete qualitative and quantitative comparison). Also in [26], based on [27] it can be found that our curves are in agreement with experimental predictions.

In summary for noble atoms we note a significant impact of the initial momentum of ejected photoelectrons, as well as the ionization degree level.

In order to provide more insight into the applicability of our formula, we repeated the mentioned observations for alkali atoms. Concretely we observed the potassium, K, atom, single and double ionized.

Based on equation (6), figure 4 is obtained for single ionized potassium atom and zero initial momentum, p = 0. A two color field is assumed. For better illustration the W^{ADK} graph (solid red line) is also presented.

It is obvius that our theoretical calculation predicts a deviation of ionization probabilities compared to the standard ADK theory. The intensity range is almost the same but the curve's behavior is different. Figure 4 shows the changes in the slope of the transition rate at different values of field intensity. The sharp drop in the W_{TC}^{ADK} rate is clearly in evidence for the stabilization. Also, the W_{TC}^{ADK} curve has an asymetric form which is sharper than W^{ADK} . The maximum is reached for $I = 2 \times 10^{13} \text{ W cm}^{-2}$.

It is obvious that for alkali atoms the ADK formula has better agreement with our result.

For given conditions, we investigated the influence of the initial momentum on the transition rate.

As shown in figure 5 the transition rate varies with inclusion of p. Although the variation is small, it indeed indicates that the momentum p plays a role in both corresponding equations. For both graphs it can be seen that the rate curves approach the intensity axis with increasing of field intensity, but $W_{\text{TC}}^{\text{ADK}}$ is much faster.

For a double ionized potassium atom, Z = 2, the behavior is almost the same and the curve has shifted slightly to higher values of the laser field intensity.

Similar results were obtained for other alkali atoms.

As seen from figures all curves demonstrate a similar qualitative behavior in the similar laser field intensity range and, as follows from our calculations presented here, the ionization process in an ionizing bichromatic field is generally asymmetric. Additionally, all graphs show the presence of the known effect of atom stabilization in an ultrastrong two color field, i.e. the reduction in the atom ionization rate as the amplitude of the external field grows.

Next we examined how the transition rate W_{TC}^{ADK} depends on the Keldysh parameter, γ and the parabolic coordinate, η . We concluded that it is very sensitive to both mentioned variables.

Figure 6 shows a narrow interval of γ and η . This confirms the high sensitivity of the tunneling ionization probability to the mentioned parameter in the two color field.

Unfortunately the lack of data of other investigations does not allow us to make a more comparative analysis with experimental data.

Conclusion

To summarize, in this paper we report new computations for the tunneling transition rate of an atom in a linearly polarized bichromatic field consisting of the coherent superposition of a fundamental field and its second harmonic. The Landau– Dykhne approach was used. The formula clearly indicated a strong dependence of the ionization rate on the intensity of the driving field. Strictly speaking our approach is valid for a weak slow variant field and deep tunneling photoionization regime. The obtained results were compared to the prediction of the ADK tunneling theory. It is found that the ADK formula give larger values for the ionization rates compared to our formula for the case of non zero initial momentum. It should be noted that the atomic stabilisation effect which is often associated with the superintense strength of the applied field, can be seen on all curves. Additionaly, the new interesting peculiarity of our theoretical formula is prediction of the existence of the atomic stabilization effect for the case of noble atom and assumption of zero initial momentum. Obtained results are in agreement with the experimental predictions.

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