EFFECT OF ELECTRON-ELECTRON CORRELATION ON THE NONSEQUENTIAL IONIZATION PROCESS IN A LINEARLY POLARIZED LASER FIELD

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Abstract. Analytic expression for the nonsequential transition rate in a linearly polarized laser field has been found, based on the adiabatic Landau-Dykhne approach. The formula involves electron-electron correlation through the mechanism of collective tunneling. The formula fails for a “knee” structure, but improves agreement with experimental results in the range of higher laser field intensities. A brief discussion on the dependence of the nonsequential transition rate on the laser pulse shape and wavelength was made.

Key words: tunnel ionization, collective tunneling, nonsequential ionization, laser beam profile.

1. INTRODUCTION

Interactions of strong fields with matter is one of the current topics in atomic, molecular and optical physics. Keldysh was the first who attempted to perform a general analysis of the ionization process of atoms caused by laser radiation [1]. His approach is known as single active electron (SAE), which assumes that the ionization process is stepwise and that there are no nonsequential processes. He introduced the Keldysh adiabaticity parameter, \( \gamma = \omega \sqrt{2 E_i / F} \), where \( E_i \) is the ionization energy, while \( \omega \) and \( F \) are the frequency and the strength of the laser field, respectively. Ionization is usually considered as either a “tunneling” or a “multiphoton” process depending on the value of the Keldysh parameter. Tunneling is the dominant process for \( \gamma \ll 1 \), while multiphoton is dominant for \( \gamma >> 1 \). Practically, a low frequency and strong field make the physical environment for tunneling photoionization. Shortly Keldysh’s work was published, his results were refined in [2, 3] for a short-range potential. Ammosov, Delone and Krainov proposed the ADK theory to predict the tunneling rate of an atom exposed to a laser field [4]. This tunneling formula is applicable for one electron ionization from neutrals, as well as, from multiply charged ions and in many cases has good agreement with experiment [5, 6, 7]. This model uses the previously calculated probability derived in [3].

Strong field double ionization is one of the most important and fundamental processes among various intense laser induced phenomena. At the same time, it is one of the most challenging problems in atomic and optical physics. It has been, and still is the subject of many investigations, both theoretical and experimental [8, 9, 10, 11]. But, in spite of this, the ionization mechanism is not fully understood. Detailed studies of the intensity dependence of single and double ionization rates in the intensity range of a laser field 0.01–10 PW cm\(^{-2}\) showed that for very high intensities there is good agreement with both numerical SAE calculations and the ADK tunneling rate. At lower intensities there is significant enhancement of the yield of doubly charged ions several orders of magnitude above what would be expected for various ionization models such as the ADK theory and SAE model. In graphs of the yield or transition rate as a function of intensity this is observed as a “knee” structure which vanishes if the intense laser light is circularly polarized, indicating that in this case ionization proceeds purely sequentially [12]. The “knee” structure is most notable in helium but it is present in the double ionization ion rates of all rare gases [12, 13, 14, 15, 16, 17]. The first experimentally reported enhancement of doubly charged ions in the form of a prominent “knee” structure in the ionization rate as a function of intensity was observed by Fittinghoff et al. [18]. Soon after, several other groups (Kondo et al. 1993 [19], Walker et al. 1994 [20]) measured the yields of single and double ionized helium with very high accuracy. In order to explain these experimental results, two distinct mechanisms have been proposed: sequential double ionization, where electrons are assumed to be tunneling independently, which means that there is no correlation between ionization events, and nonsequential double ionization, where electrons cannot be treated separately. It was agreed that nonsequential double ionization has to incorporate correlated two-electron dynamics in intense laser fields [21, 22]. Double ionization measurements are the strongest evidence to date of electron correlation effects in strong field physics. A review can be found in [23, 24].

Four mechanisms have been proposed to explain nonsequential double ionization. Fittinghoff et al. [18] suggested a nonsequential ionization process due to a “shake-off” mechanism. Corkum [25] gave an alternative mechanism based on electron–electron inelastic rescattering, directly related to the recollision model developed by Corkum [25], Kulander et al. [26], Schafer et al. [27]. Eichmann et al. [12] proposed collective multi electron tunneling.

The model of collective tunneling assumes simultaneous tunneling of several bound electrons through the potential barrier lowered by a laser field. This process is possible only if the electrons are on the same distance from the core. For very short pulse durations or large differences in the 1st and 2nd ionization potential, the collective tunneling process plays a significant role (one-two particle tunneling
process at an effective $E_i$ of the sum of the $1^{\text{st}}$ and $2^{\text{nd}}$ $E_i$ should have a higher probability than the independent sequential tunneling of the outermost and $2^{\text{nd}}$ electron through their individual tunneling barriers).

Almost a decade after the first reported enhancement of doubly charged ionic, Eichmann et al. [12] dealt with the question whether the process of simultaneous tunneling of electrons in external fields exists and how it can help to explain experimental double ionization results. Discussions based on the results where double ionization is dominantly mediated by electron-electron correlation through the collective tunneling process have been the subject of many papers. Kornev et al. show that the two-electron collective tunneling effect becomes essential, changing the total probability of the ion formation more than by an order [28].

In this work, an analytical formula for the nonsequential transition rate, in the presence of an intense linearly polarized laser via the Landau-Dykhne approach is obtained. Based on the obtained formula, we investigated double ionization of the helium atom over a wide range of Keldysh adiabaticity parameters, $\gamma$. Atomic units were used ($m = e = \hbar = (4\pi\epsilon_0) = a_0 = 1$) [29]. The paper is structured as follows: in section 2 we obtained an expression for the nonsequential transition rate. In section 3, we applied it on the helium atom and discussed the obtained results. Finally, conclusions are summarized in section 4.

2. THEORETICAL FRAMEWORK

Nonsequential double ionization significantly enhances the production of doubly charged ions over yields expected for the sequential tunneling mechanism. In order to explain the discrepancy between experimental data and theoretical prediction, based on the sequential approach to the ionization rate, we derived an analytical expression for the nonsequential transition rate which includes the effect of collective tunneling of electrons from the valence shell. We used the Landau-Dykhne approach applied to the ADK model, supposing the independent electron model assuming that each electron is moving independently in the combined field of the nucleus and average distribution of other electrons. But electron-electron correlation plays an important role in a significant number of processes. The reason to modify the ADK formula is its impossibility to describe the double transition rate. The performed calculation can predict the double tunneling rate of an atom exposed to a linearly polarized laser field.

The transition rate $W$ 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 $E_f$ is given within the frame of an adiabatic approximation by the Landau-Dykhne formula with exponential accuracy [30, 31]:

$$W_{gf} \propto \exp[-2\text{Im}S(\tau)]. \quad (1)$$

It is seen that the transition rate is proportional to the imaginary part of action $S(\tau)$:

$$S(\tau) = \int_0^\tau [E_f(t) - E_i(t)] dt, \quad (2)$$

where $\tau$ is the classical turning point in the complex plane. For this approximation to be valid the photon energy of the laser radiation needs to be small compared to the ionization potential, $E_i$. The single electron ionization rate $W$ in the tunneling regime of a laser field with amplitude $F$ can be described well by a sequential ADK ionization rate tunneling formula, proportional to $W_{SI}^{ADK} \propto \exp\left(-2/3\tilde{F}\right)$

where $\tilde{F}=(F/2E_i)^{3/2}$ is a dimensionless parameter. However, electron correlation is essential for the description of many important physical phenomena. It was believed that the “knee” structure could be due to some kind of electron correlation. In this work we observed this correlation through the process of collective tunneling. In order to estimate the rate of this process, for the general case of $N$ electrons it is convenient to treat them like one particle with $N$ times the charge and the mass and to interpret the binding energy as the binding energy $E_{tot}$ of all $N$ electrons. In the case of the helium atom the total binding energy is $E_{tot} = E_1 + E_2$ where $E_1$ and $E_2$ are the particular binding energies for the first and second electron from the valence shell.

Now, the initial bound state energy is obtained in the following form:

$$E_i = -E_{tot} + \frac{5Z}{(4\pi\epsilon_0)^{1/2}a_0}. \quad (3)$$

The second term in Eq. 3 presents the contribution of the electron correlation [32]. Here we omit the Stark shift of the initial binding state following the usual Keldysh approach [1].

Tunnel ionization occurs close to a maximum of the field, $F_0$, because the barrier is the narrowest, since small fields give rise to a potential barrier and the probability for tunneling is extremely low. In the final state we included the term for the Coulomb interaction given in the parabolic coordinate [31]. Then the energy of the final state of two-electron system is:
\[
E_j(t) = \left( p - \frac{F_0}{\omega} \sin \omega t \right)^2 - \frac{(2n_2 + |m| + 1)\sqrt{4E_{\text{tot}}}}{\eta}.
\]  

(4)

Here \( p \) is the momentum of the ejected electron. By a series expansion, from the formula \( p = \sqrt{\frac{1}{4} + \frac{1}{2\eta} + \frac{1}{4\eta^2} + \frac{1}{4} F_\eta \) \[31, 33\], for the case of outside barrier momentum, \( p \), is obtained in the following form \[33\]
\[
p = \frac{1}{2} \left( \sqrt{F_\eta \eta - 1} + \frac{1}{\eta \sqrt{F_\eta \eta - 1}} \right)
\]
where \( \eta \) is parabolic coordinate and for the case when electron is outside of the barrier, \( \eta > 1/F_0 \) \[34\]. If a system’s total energy is independent of the coordinate \( \eta \) then momentum is conserved along the classical path \( i.e. \ p_\eta = p \) \[31\]. The next \( n_2 \) is parabolic, \( m \) magnetic quantum number successively, and \( \eta \) is the parabolic coordinate. From all possible values of \( n_2 \) only \( n_{2\text{max}} \) is interesting because all others contribute significantly less, \( n_{2\text{max}} = n^* - 1 \). The quantity \( n^* \) is related to the energy \( E_i \) of the considered state by relation: \( n^* = Z\sqrt{2E_i} \).

According to the Landau-Dykhne expression (Eq. 1) we should now evaluate the integral:
\[
\text{Im} S(\tau) = \text{Im} \left[ \int_0^\tau \left( E_j(t) - E_i(t) \right) dt \right] = \text{Im} \left[ \int_0^\tau \left( \left( p - \frac{F_0}{\omega} \sin \omega t \right)^2 - \frac{(2n_2 + |m| + 1)\sqrt{4E_{\text{tot}}}}{\eta} + E_{\text{tot}} - \frac{5Z}{(4\pi e_0)8a_0} \right) dt \right].
\]

Calculating this temporal integral and after some simple manipulations, for the imaginary part of the action, we obtained:
\[
\text{Im} S(\tau) = \text{Im} \left[ \left( \alpha + \frac{F_0^2}{2\omega^2} \right) \tau + \frac{2pF_0}{\omega} (\cos \omega \tau - 1) - \frac{F_0^2}{4\omega^3} \sin 2\omega \tau \right].
\]  

(5)

The parameter \( \alpha \) which appears in Eq. 5 is defined as
\[
\alpha = p^2 - \frac{(2n_2 + |m| + 1)\sqrt{4E_{\text{tot}}}}{\eta} + E_{\text{tot}} - \frac{5Z}{(4\pi e_0)8a_0}.
\]

To simplify Eq. 5 we employed well known series expansions:
\[
\cos \omega \tau = 1 - \left( \omega^2 \tau^2 / 2 \right) + \left( \omega^4 \tau^4 / 4 \right) - \ldots
\]
\[ \sin 2\omega \tau = 2\omega \tau - \left(4\omega^3 \tau^3 / 3\right) + \left(4\omega^5 \tau^5 / 15\right) - \ldots \]

Now, Eq. 5 becomes:

\[ \text{Im} S(\tau) = \text{Im} \left[ \alpha \tau + pF_0 \tau^2 + \frac{F_0^3}{3} \tau^3 \right]. \]  (6)

As can be seen from Eq. 6, the quantity \( S(\tau) \) is a function of the complex time, \( \tau \), which is determined from the following condition \( E_i(\tau) = E_i(\tau) \):

\[ -E_{tot} + \frac{5Z}{(4\pi e_0)8a_0} = \left( p - \frac{F_0}{\omega} \sin \omega \tau \right)^2 - \frac{\left(2n_2 + |m| + 1\right)\sqrt{4E_{tot}}}{\eta}. \]  (7)

Based on Eq. 7 the classical turning point is of the form:

\[ \tau = \frac{1}{\omega} \arcsin \frac{\omega}{F_0} \left( p - i\sqrt{E_i^*} \right), \]  (8)

where \( E_i^* = E_{tot} - \frac{5Z}{(4\pi e_0)8a_0} - \frac{\left(2n_2 + |m| + 1\right)\sqrt{4E_{tot}}}{\eta} \).

To evaluate \( \tau \) we took the first two terms from the series

\[ \arcsin x = \sum_{n=0}^{\infty} \left( \frac{2n+1}{2^n (n!)^2} \right) \frac{x^{2n+1}}{2n+1} \]

and obtained for the turning point:

\[ \tau = \frac{1}{\omega} \left( \frac{\omega}{F_0} \left( p - i\sqrt{E_i^*} \right) + \frac{\omega^3}{6F_0^3} \left( p - i\sqrt{E_i^*} \right)^3 \right). \]

The previous equation could be rewritten to become:

\[ \tau = \frac{p}{F_0} + \frac{p^3 \omega^2}{6F_0^3} - \frac{p \omega^2 E_i^*}{2F_0^3} - \frac{\sqrt{2E_i^*}}{F_0} \left( 1 + \frac{p^2 \omega^2}{2F_0^2} - \frac{\omega^2 E_i^*}{6F_0^4} \right). \]  (9)

When \( \tau \) is known in the analytical form, based on Eq. 9, by exponentiation, we found corresponding expressions for \( \tau^2 \) and \( \tau^3 \):

\[ \tau^2 = -2i \frac{\sqrt{E_i^*}}{F_0} \left( \frac{p}{F_0} + \frac{p^3 \omega^2}{6F_0^3} - \frac{2p \omega^2 E_i^*}{3F_0^3} \right), \]  (10)

\[ \tau^3 = -2i \frac{\sqrt{E_i^*}}{F_0} \left( \frac{p^2}{F_0^2} + \frac{p^4 \omega^2}{3F_0^4} - \frac{5p^2 \omega^2 E_i^*}{6F_0^4} \right). \]  (11)
It should be noted that, because of simplicity, in $\tau^2$ and $\tau^3$ we kept only those terms which, after substituting in Eq. 6, give the third-order terms $-\frac{1}{F_0^2}$.

Replacing Eq. 9, Eq. 10 and Eq. 11 into Eq. 6 we obtained:

$$\text{Im} S(\tau) = \alpha \left( -\frac{\sqrt{E_i^*}}{F_0} - \frac{1}{2} \frac{p^2 \omega^2 \sqrt{E_i^*}}{F_0^3} + \frac{1}{6} \frac{\omega^2 (\sqrt{E_i^*})^3}{F_0^3} \right)$$

$$+ \frac{8}{3} \frac{p^2 \sqrt{E_i^*}}{F_0} - \frac{5}{9} \frac{p^4 \omega^2 \sqrt{E_i^*}}{F_0^3} - \frac{17}{9} \frac{p^2 \omega^3 (\sqrt{E_i^*})^2}{F_0^3}$$

Here it is convenient to introduce the following notation: $\gamma^* = \frac{\omega}{F_0} \sqrt{E_i^*}$.

Then we have:

$$\text{Im} S(\tau) = \alpha \left( -\frac{\gamma^*}{\omega} - \frac{1}{2} \frac{p^2 \omega \gamma^*}{F_0^3} + \frac{1}{6} \frac{\gamma^*^3}{\omega} \right) + \frac{8}{3} \frac{p^2 \omega \gamma^*}{\omega} - \frac{5}{9} \frac{p^4 \omega \gamma^*}{F_0^3} - \frac{17}{9} \frac{p^2 \gamma^*^3}{\omega} \right). \quad (12)$$

Finally, taking into account the obtained expression for the imaginary part of the action, the exponential factor in Eq. 1 becomes:

$$w_f \propto \exp \left[ -2 \left( \frac{-\gamma^*}{\omega} - \frac{1}{2} \frac{p^2 \omega \gamma^*}{F_0^3} + \frac{1}{6} \frac{\gamma^*^3}{\omega} \right) + \frac{8}{3} \frac{p^2 \omega \gamma^*}{\omega} - \frac{5}{9} \frac{p^4 \omega \gamma^*}{F_0^3} - \frac{17}{9} \frac{p^2 \gamma^*^3}{\omega} \right]. \quad (13)$$

We obtained the final formula for the nonsequential ionization rate by recalling the single ionization rate in the tunneling regime as given by the ADK formula, $W_{\text{ADK}}$ [4]:

$$W_{\text{NSI}} = \left( \frac{4Z^3 e}{F_0 \omega} \right)^{2n-1} \times$$

$$\times \exp \left[ -2 \left( \frac{-\gamma^*}{\omega} - \frac{1}{2} \frac{p^2 \omega \gamma^*}{F_0^3} + \frac{1}{6} \frac{\gamma^*^3}{\omega} \right) + \frac{8}{3} \frac{p^2 \omega \gamma^*}{\omega} - \frac{5}{9} \frac{p^4 \omega \gamma^*}{F_0^3} - \frac{17}{9} \frac{p^2 \gamma^*^3}{\omega} \right]. \quad (14)$$

Eq. 14 presents our modification of the standard ADK formula. Some additional terms can be seen in the formula structure and they are directly related to the electron-electron correlation.

Our formula is also be valid for the case with N-electron tunneling.
The quality of a laser beam has been a topic of interest to both designers and users of optical systems. Different aspects of beam quality may have higher priority in various optical systems. Lorentzian is one of the mathematical models suitable for different beam shapes. In the general case, the electric field strength of an electromagnetic wave is a function of spatial coordinates and time. Often, such a functional dependence can be factorized and the electric field can be represented as a product of two functions depending on spatial coordinates and time, respectively. In this case, we are interested in the influence of the spatial profile in the nonsequential transition rate. From several profiles which can be found, in this paper we used the following [34]:

\[ F_0(\rho) = F_0 \left( \left( \rho + R \right)^2 \right), \]

where \( \rho \) is the axial coordinate that is normal to the light ray, \( R = R \left( 1 + \frac{\lambda z}{\pi R^2} \right) \) [35] \( R \) is the radius and \( z \) is direction of the laser beam.

With the Lorentzian spatial profile Eq. 14 may be expressed as:

\[ W_{NSI} = \left( \frac{4Z^e}{F_0} \right)^{2 \omega - 1} \times \]

\[ \exp \left[ -2 \left( \frac{\gamma^*}{\omega} - \frac{1}{2} \frac{P^* \omega}{\left[ F_0 / \left( 1 + \left( \rho / R \right)^2 \right) \right]} \gamma^* + \frac{1}{6} \frac{\omega^* \gamma^*}{\omega} + \frac{1}{3} \frac{\gamma^* \gamma^*}{\omega^*} - \frac{5}{9} \left( \frac{F_0 / \left( 1 + \left( \rho / R \right)^2 \right)}{\omega} \right) \gamma^* \frac{17}{9} \frac{P^* \omega}{\omega^*} \right]. \]

This gives us additional possibility to analyze the behavior of the nonsequential transition rate.

3. RESULTS AND DISCUSSION

Based on Eq. 14, within the frame of the nonsequential collective tunneling mechanism, we analyzed the double ionization transition rate in a strong external laser field. Our calculations were performed for a linearly polarized laser field with the following parameters: \( I = 0.01–10 \text{ [PWcm}^{-2}] \) and \( \omega = 0.05696 \text{ a.u.} \) (\( \lambda = 800 \text{ [nm]} \)). We chose a helium atom, \( He \) as the target. We considered first and double tunneling ionization with corresponding ionization potentials \( E_{i1} = 0.905 \text{ a.u.} \) (24.59 [eV]) and \( E_{i2} = 2.004 \text{ a.u.} \) (54.468 [eV]).

In order to demonstrate the difference between sequential ionization of a laser field with two-electron and a nonsequential process of double ionization mediated by collective tunneling (electron-electron correlation) we observed the ionization rate of doubly charged \( He^{2+} \) ions created in the laser field. In Fig. 1, we
show the rate of $\text{He}^{2+}$ as a function of the laser field intensity. Both studies were performed at intensities where the sequential and nonsequential processes are expected:

![Graph showing nonsequential ionization rate (solid line, $W_{\text{NSI}}$) and sequential (dotted line, $W_{\text{SIW}}^{\text{ADK}}$) for double charged ions helium, $\text{He}^{2+}$, as a function of the laser field intensity.]

The solid curve is plotted based on Eq. 14 and dotted by the standard ADK formula [4]. The laser intensity of $I = 2 - 8 \text{ PWcm}^{-2}$ corresponds to the Keldysh parameter $\gamma = 0.28 - 0.56$ indicating that the ionization process mainly occurs via tunnel ionization. In this case most ions are created with NSI. Figure 1 shows that two-electron tunnelling requires field values lower than two-electron tunnelling sequential [36]. We compared the obtained results with experimental data [12, 37, 24]. Figure 1 shows that our result is in good agreement with experimental observations in a high intensity field region. The curve has the characteristic “flow”, but the characteristic “knee” structure can’t be obtained by Eq. 14.

However Eq. 14 is applicable in the high intensity range. For lower intensity it should be improved by including more effects. Thus, the collective tunneling ionization mechanism can be responsible for the two-electron photoionization of atoms, primarily in the case of intensity of above $I = 10 \text{ PWcm}^{-2}$ and in the presence of a low-frequency field.

In the following, we present simple but important examples of the studied transition rate dependence of wavelength, $\lambda$ (Fig. 2). According to Eq. 14 and taking into account that $\omega = 2\pi c / \lambda$, we should obtained this dependence, where $c = 137$. The wavelength varied within the range $\lambda = 300 - 1200 \text{ nm}$. 
Figure 2 shows that the rate first increases, reaches a peak at the value of $\lambda = 1058.4 \text{ [nm]}$, and then drops with increasing wavelength (decreasing frequency). A longer wavelength “makes” lower tunneling probability. This is completely in accordance with the Keldysh conditions, defined through the Keldysh parameter, $\gamma$, for the tunneling regime.

In Fig. 3 (3D graph) we show the dependence of the nonsequential transition rate, $W_{NSI}$, on the laser field intensity, $I$, and wavelength, $\lambda$.

In the regime of laser intensities where the nonsequential process dominates, an increase of laser intensity leads to an increase of the transition rate. On the other
hand, we can observe the same behavior of the transition rate as a function of the wavelength, $\lambda$.

Figure 4 demonstrates the dependence of the nonsequential transition rate on the laser beam shape.

![Graph showing the comparison of nonsequential transition rates for general NSIW and Lorentzian pulse shapes, NSILW. The laser intensity is within the range $I = 5–20 \text{ [PWcm}^2\text{]}$.

In Fig. 4 the results are plotted vs the laser field intensity. The solid curve was calculated by applying the $W_{\text{NSI}}$ rate (Eq. 14), while the dashed one by $W_{\text{NSIL}}$, for a Lorentzian pulse shape (Eq. 15). Different symbols were used for general and Lorentzian pulse shapes. The Lorentz beam shape, at the same conditions, shifts the nonsequential ionization process toward to higher laser field intensities. This delivers useful insight into how important the selection of the laser beam shape might be.

4. CONCLUSION

We obtained a modified ADK formula for the nonsequential transition rate and found that the nonsequential process leads to a different transition rate for doubly charged ions if compared with the sequential process. The main message is that the inclusion electron-electron interaction through the mechanism of collective tunneling in the standard ADK formula for sequential ionization improves agreement with experimental results. It is obvious that nonsequential double ionization represents one striking example of the importance of correlated electron dynamics occurring in laser fields. We also studied the influence of a specified
spatial laser profile and found that our formula is sensitive to the pulse shape as well as the laser frequencies and field intensities.

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