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TUNNELING IONIZATION STUDY OF LINEAR MOLECULES IN STRONG-FIELD LASER PULSES*

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ABSTRACT We theoretically studied photoionization of atoms and molecules in the frame of Perelomov-Popov-Terent'ev (PPT) and Ammosov-Delone-Krainov (ADK) theories. Strong-field single ionization of two diatomic molecules, N_2 and O_2, are studied and compared to Ar and Xe atoms, using an 800 nm Ti:sapphire laser in the 3×10^{13} to 1×10^{15} Wcm^{-2} intensity range. To eliminate disagreement between theoretical and experimental findings in a low intensity fields ($\sim 6 \times 10^{13}$ Wcm^{-2}), we considered the influence of shifted ionization potential. Including these effects in the ionization rates, we numerically solved rate equations in order to determine an expression for the ionization yields. The use of modified ionization potential showed that the ionization yields will actually decrease below values predicted by original (uncorrected) formulas. This paper will discuss the causes of this discrepancy.

Keywords: *tunneling ionization, ionization rate, ionization yield, molecules.*

INTRODUCTION

In the past decade substantial progress has been made in the understanding of the dynamics of molecules in intense-laser fields (10^{13} – 10^{18} Wcm^{-2}) [1,2]. As a result, experimental, theoretical and computational investigation of this phenomena have demonstrated fundamental processes such as bond softening and hardening, laser induced alignment, and enhanced ionization at critical internuclear distances. Most of these phenomena are based on tunnel ionization of neutral atoms and molecules as the first step of the physical processes [3].

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There is a wealth of theoretical approaches to the atomic photoionization dynamics in a strong field, which can be defined as tunnel, multiphoton, or some combination of both. Keldysh [4] was first who introduced well known parameter to distinguish tunnel and multiphoton photoionization process, $\gamma = \omega\sqrt{2I_p}/F$, where I_p is unperturbed ionization potential, F the amplitude of the electric field and ω the laser frequency. It was widely accepted that for $\gamma \gg 1$ multiphoton ionization is the dominant process, while for $\gamma \ll 1$ tunnel. It is worth noting that according to Reiss [5], the regime when $\gamma \sim 1$ at $\lambda = 800\ \mathrm{nm}$ ionization in a strong laser field can successfully be described as a tunneling process. Here and throughout the paper, all equations are given in atomic units ($e = m_e = \hbar = 1$) [6] unless otherwise stated.

Keldysh theory is extended into the so-called Strong Field Approximation (SFA) [7]. Following the Keldysh formalism of the tunneling ionization rate in a strong electromagnetic field Perelomov, Popov and Terent'ev developed another tunneling model (PPT) [8] that was further extended by Ammosov, Delone and Krainov and is now known as ADK-theory [9]. During the last years, the scope of strong field physics has been extended to the systems more complex than atoms, including molecules, fullerenes and clusters where all abovementioned theories are also being suitably adapted (molecular orbital SFA (MO-SFA) [10], molecular orbital PPT (MO-PPT) [11], and molecular orbital ADK (MO-ADK) [12]). All mentioned theories are based on single active electron approximation (SAE) where only the one electron interacts with the applied laser field. Also, experimental results [13,14] show excellent agreement with these one theoretically predicted by the commonly used ADK, as well MO-ADK in the case of noble gasses and small molecules. Because of additional nuclear degrees of freedom, the motion of electrons and nuclei (there are nuclear rotational and vibrational dynamics) which must be taken into consideration, molecules are much more complex to model theoretically than atoms. Models are still being developed and in generally they are more complex than earlier [15,16].

THEORETICAL FRAMEWORK

The abovementioned atomic tunneling theories [7-12] have been very successful in describing the ionization rates of both atoms and molecules. It is well known that these theories depend primarily on the field intensity, I (relationship between intensity (I) and electric field amplitude (F), is given by: $F \sim \sqrt{I}$), and ionization potential, I_p. If the ionization potential is one of the most important

quantities, one would expect similar ionization rates for molecules and atoms with similar ionization potentials, the so-called companion atoms. Several experiments were performed in order to measure the ratio of ion yields of diatomic molecules and their companion atoms [17,18]. The findings have revealed that some molecules are harder to ionize than their companion atoms. This phenomenon is known as suppressed ionization [19].

In this paper we have aim to explore how corrections on the ionization potential, I_p, influence the ionization rate, $W(F,t)$, of homonuclear diatomic molecules, N_2 and O_2, and their companion atoms Ar and Xe, respectively. As Table 1 illustrates, these atoms and molecules have comparable ionization potentials. To achieve this, we modified I_p by taking into account the Stark shift, I_{st}, and the ponderomotive potential, U_p.

Table 1. Ionization potentials of diatomic molecules, N_2 and O_2, and companion atoms, Ar and Xe [20].

	Ar	N_2	Xe	O_2
I_p [atomic units]	0.57916	0.57255	0.45202	0.44319

If a quantum system is found in a state with energy I_p and is perturbed by an external monochromatic field of amplitude F and frequency ω, then the shift of the ionization energy is well known as a Stark shift which can determined by the following expression: $I_{st} = \alpha_p F^2/4 + \gamma_h F^4/24$ [21]. In this inline equation, α_p represents the dipole polarizability, while γ_h is the dipole hyperpolarizability. In addition, this field also causes the oscillating movement of electron. The ponderomotive potential, i.e. the average oscillation kinetic energy of a free electron in the electric field of the laser with strength F, is then readily calculated as $U_p = F^2/4\omega^2$ [22]. Having both effects in mind, we can write the corrected ionization potential I_p^{corr} in the following form:

$$I_p^{corr} = I_p + I_{st} + U_p = I_p + \frac{\alpha_p F^2}{4} + \frac{\gamma_h F^4}{24} + \frac{F^2}{4\omega^2}. \quad (1)$$

The values of polarizability α_p and hyperpolarizability γ_h for different linear molecules can be found in [21].

In the following we briefly review the basic rate equations of the MO-ADK and the MO-PPT theories.

MO-ADK ionization rate

The MO-ADK theory is extension of the ADK tunneling theory on the more complex system, molecules. Following exactly the same procedure as in [12], the tunneling ionization rate of linear molecules, $\mathrm{W_{MOADK}}$, can be calculated as [12]:

$$\mathrm{W_{MOADK}} = \frac{B^2(m)}{2^{|m|}|m|!}\frac{1}{\kappa^{\frac{2Z_C}{\kappa}-1}}\left(\frac{2\kappa^3}{F}\right)^{2Z_C/\kappa-|m|-1}\mathrm{Exp}\left[-\frac{2\kappa^3}{3F}\right], \quad (2)$$

where m is the magnetic quantum number along the molecular axis, Z_C is the effective Coulomb charge [23], κ is the characteristic momentum of the bound state, $\kappa = \sqrt{2I_p}$, and I_p is already defined and here presents ionization potential for the given valence orbital. The factor $B^2(m)$ in Eq. (2) measures the electron density in the tunneling region along the direction of the electric field and for the case of linear molecules can be expressed as: $B(m) = \sum_l C_l\, Q(l,m)$, where C_l is the structure coefficient of the molecule, l is the angular momentum quantum number and $Q(l,m)$ is the coefficient given by: $Q(l,m) = (-1)^m\left[\left((2l+1)(l+|m|)!\right)/(2(l-|m|)!)\right]^{1/2}$. The values of C_l for a specific linear molecule can be found in [12].

MO-PPT ionization rate

Based on PPT theory [8] and results presented by Tong et al. [12], Benis and his coworkers in [11] improved Eq. (1), developing the MO-PPT model. Based on this model, the ionization rate of linear molecules is given by [11]:

$$\mathrm{W_{MOPPT}} = \frac{B^2(m)}{2^{|m|}|m|!}\frac{A_m(\omega,\gamma)}{\kappa^{2Z_C/\kappa-1}}\left(\frac{2\kappa^3}{F((1+\gamma^2)}\right)^{2Z_C/\kappa-|m|-1} \times \mathrm{Exp}\left[-\left(\frac{2\kappa^3}{3F}\right)\mathrm{g}(\gamma)\right]. \quad (3)$$

It is worth noting that the MO-ADK model is a simplified version of MO-PPT. When Keldysh parameter, $\gamma \to 0$, the Eq. (3) goes back to the MO-ADK rate (Eq. (2)). Regard to MO-ADK, Eq. (4) has the following two correction factors $A_m(\omega,\gamma)$ and $\mathrm{g}(\gamma)$ [24], which for the case of the tunneling ionization can be defined as: $A_m(\omega,\gamma) = \frac{4\gamma^2}{(1+\gamma^2)\sqrt{3\pi}\,|m|!}\sum_{k\geq\nu}^{\infty}\mathrm{Exp}[-\alpha(k'-\nu)]\,w_m\left(\sqrt{\beta(k'-\nu)}\right)$ and $\mathrm{g}(\gamma) = 1 - \frac{1}{10}\gamma^2 + \frac{9}{280}\gamma^4$ [12]. In the above equations the following

coefficients are introduced [24]: $\alpha(k-\nu)=2(k-\nu)^3/3$, $\beta(k-\nu)=2(k-\nu)/\sqrt{1+\gamma^2}$, $\nu=I_p\left(1+\frac{1}{2\gamma^2}\right)/\omega$, $k'=\left\langle\frac{I_p}{\omega}+1\right\rangle$ and $w_m\left(\sqrt{\beta(k-\nu)}\right)=\frac{\left(\sqrt{\beta(k-\nu)}\right)^{2|m|+1}}{2}\int_0^1\frac{\mathrm{Exp}[-t\sqrt{\beta(k-\nu)}]t^m}{\sqrt{1-t}}dt$. The symbol $\langle\,\rangle$ indicates the integer part of the value inside, which refers to the minimum number of photons required to ionize the system.

Corrections of MO-ADK and MO-PPT ionization rates

To analyze how the ionization rates $\mathrm{W_{MOADK}}$ and $\mathrm{W_{MOPPT}}$ are affected by the corrected ionization potential I_p^{corr}, we substituted the unperturbed ionization potential I_p with the shifted one, I_p^{corr}, in the MO-ADK and MO-PPT rate equations. We assume the envelope of the electric field to be a Gaussian beam, $F_G(t)=F_0\exp[-4t^2/\tau^2]$ (τ is full width at half maximum (FWHM) of the laser pulse), in our calculations. The modulation of generally assumed laser beam shape, F, with the Gaussian shaped laser beam, $F_G(t)$, allows us to compare our results with the experimental data [25].

First, we incorporated the Gaussian laser beam shape, $F_G(t)$, and corrected ionization potential, I_p^{corr}, in the formula for the MO-ADK ionization rate, $\mathrm{W_{MOADK}}$, and obtained the following expression:

$$\mathrm{W}^{\mathrm{corr}}_{\mathrm{MOADK}}(t)=\frac{B^2(m)}{2^{|m|}|m|!}\frac{1}{\sqrt{2I_p^{corr}(t)}^{\,2Z_C/\sqrt{2I_p^{corr}(t)}-1}}\left(\frac{2(2I_p^{corr}(t))^{3/2}}{F_G(t)}\right)^{2Z_C/\sqrt{2I_p^{corr}(t)}-|m|-1}$$

$$\times\,\mathrm{Exp}\left[-\frac{2(2I_p^{corr}(t))^{3/2}}{3F_G(t)}\right]. \quad (4)$$

We repeated the same procedure in order to obtain the corrected version of the standard MO-PPT ionization rate, using Eq. (3):

$$\mathrm{W}^{\mathrm{corr}}_{\mathrm{MOPPT}}(t)=\frac{B^2(m)}{2^{|m|}|m|!}\frac{A_m^{corr}(\omega,\gamma^c(t))}{\sqrt{2I_p^{corr}}(t)^{\,2Z_C/\sqrt{2I_p^{corr}(t)}-1}}\times$$

$$\times\left(\frac{2(2I_p^{corr}(t))^{3/2}}{F_G(t)(1+(\gamma^c(t))^2)}\right)^{2Z_C/\sqrt{2I_p^{corr}}(t)-|m|-1}\mathrm{Exp}\left[-\left(\frac{2(2I_p^{corr}(t))^{3/2}}{3F_G(t)}\right)\mathrm{g}\left(\gamma^c(t)\right)\right]. \quad (5)$$

For the sake of optimizing Eq. (5), we introduced the corrected Keldysh parameter, $\gamma^c(t)$, in the following form: $\gamma^c(t) = \omega\sqrt{2I_p^{corr}(t)}/F_G(t)$.

RESULTS AND DISCUSSION

In this section results of a theoretical investigation of the modified ionization rates, $\mathrm{W}^{\mathrm{corr}}_{\mathrm{MOADK}}(t)$ and $\mathrm{W}^{\mathrm{corr}}_{\mathrm{MOPPT}}(t)$, have been presented and compared with experimental results (taken from [25]). We considered the case of single ionized diatomic molecules, N_2 and O_2, and noble atoms, Ar and Xe, which are the most commonly used targets in strong-field studies. This was accomplished by considering a $\lambda = 800$ nm, central wavelength pulse with a $20 - 30$ fs duration. Field intensities, I, have been varied within the range: $I = 3 \times 10^{13} - 1 \times 10^{15}$ Wcm^{-2}. These parameters limited the value of the Keldysh parameter in the range which is characteristic for the tunnel ionization. We assumed the Gaussian beam profile with, step by step, included fully corrected ionization potential. The yields presented in this paper are normalized to the maximum value which is $\sim 1.237 \times 10^9$, while the rates are normalized at the saturation intensity, so that the maximum yield denotes $W_{max} = 1$ and $\log_{10}(W_{max}) = 0$.

In Fig.1 we presented comparative review of the rates for O_2 molecule, obtained by the $W_{MOADK}(t)$ and the $W_{MOPPT}(t)$, as well as by the $W^{corr}_{MOADK}(t)$ and the $W^{corr}_{MOPPT}(t)$. It is obvious that the rates are underestimated in the MO-ADK compared with those from the MO-PPT until the field intensities $I < 6 \times 10^{13}\ Wcm^{-2}$. This completely follows the findings in [13,26]. For the higher filed intensities, the $W_{MOADK}(t)$ overestimates the $W_{MOPPT}(t)$. Also, from Fig. 1 is obvious that the inclusion of both, the ponderomotive and the Stark shift, in the ionization potential causes the rate decrease. Graph shows cumulative decrease causes by both effects. This follows observed behavior in atom's systems [27]. Similar conclusions can be drawn for N_2 molecule when field intensity varies in the range $I = 2 \times 10^{13} - 2 \times 10^{14}\ Wcm^{-2}$. It is important to note that the rates presented in Fig. 1 are very sensitive to the ponderomotive potential change. Under the same conditions, the Stark shift has a significantly smaller influence. Such result is completely in accordance with the theoretical predictions [9,10].

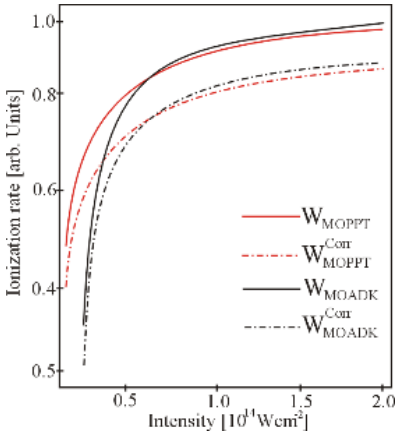

Figure 1. Comparative review of the ionization rates for O_2 molecule as a function of the laser intensity. The following notation is used: solid line for uncorrected ionization potential, I_p, and dot-dashed for fully corrected ionization potential, I_p^{corr}. Field intensity varies in the range $I = 2 \times 10^{13} - 2 \times 10^{14}\ Wcm^{-2}$. Red lines are from the corrected MO-PPT and black from the MO-ADK model.

As we already mentioned, there are molecules with similar or almost same ionization potential with atoms. In this case, one can expects the similar ionization rates for molecule and its so-called companion atom, such as N_2 molecule and Ar atom or O_2 molecule and Xe atom. Many papers deal with this phenomenon [17,28,29]. Because of the similar ionization potential, one can expect similar ionization rate. In Table 2, the quantitative measure of the match is expressed thought the ratio between belonging rates N_2: Ar and O_2: Xe. Results are given for the assumed general form of laser beam shape. Our analysis clearly indicated that inclusion of the Gauss form of beam with the temporal evolution without spatial one, increases this ratio out of expected values.

Table 2. Ratios of single-ionization rates W_{MOADK}^{corr,N_2}: $W_{ADK}^{corr,Ar}$ and $W_{MOADK}^{N_2}$: W_{ADK}^{Ar} for N_2 molecule with its companion Ar atom and W_{MOADK}^{corr,O_2}: $W_{ADK}^{corr,Xe}$ and $W_{MOADK}^{O_2}$: W_{ADK}^{Xe} for O_2 molecule with its companion Xe atom. The ionization rate for ADK, W_{ADK}, is obtained by Eq. (1) of [9].

	I [10^{13} Wcm^{-2}]				
	5	8	11	14	17
$W_{MOADK}^{N_2}$: W_{ADK}^{Ar}	1.31	1.18	1.10	1.06	1.02
W_{MOADK}^{corr,N_2}: $W_{ADK}^{corr,Ar}$	0.26	0.17	0.08	0.05	0.03
$W_{MOADK}^{O_2}$: W_{ADK}^{Xe}	0.029	0.018	0.010	0.005	0.001
W_{MOADK}^{corr,O_2}: $W_{ADK}^{corr,Xe}$	0.0058	0.0036	0.0019	0.0009	0.0001

From Table 2 one can observe that there is a significant difference between ratios for the same fixed laser field values, with and without included ionization potential's corrections. The corrective effects on the ionization potential suppress the ratio by a factor approximately five for the lower field intensities, with the intent to grow up with field increasing. In papers, it can be found significantly different results for N_2 and O_2 molecules and their companion atoms Ar and Xe. Tong et al. [12] suggested that if the N_2 molecule is aligned along the field direction, its ionization rate would be identical to Ar, and the ratio should be near 1, and for $I \sim 10^{14}\ \mathrm{Wcm^{-2}}$ ratio was calculated to be 0.98. In contrary, Liang et al. [30] suggested lower value, 0.2, or 0.7 by [31], and our results are in good agreement with them for the lower fields. Suppressed ionization ratio of O_2 molecule and Xe atom has been observed numerous of times [18,32]. Our results are in accordance with these findings. Kjeldsen and his group [17] predicted strongest suppression with the ratio below 0.01. In addition, their results clearly indicated that at lower intensities $I < 5 \times 10^{13}\ \mathrm{Wcm^{-2}}$, the experimental ratios are scattered between 0.02 and 0.2. The reason for disagreement between experimental and our result can be found in the fact that the experimentally obtained yield is always larger than those obtained by using theoretical models. That is why Hoang et al. [33] corrected the standard MO-ADK theory including the influence of permanent dipole and dynamic core-electron polarization on tunneling ionization. Such correction provided overall fairly good agreements with numerical solutions of the time-dependent Schrodinger equation, although a satisfactory agreement with experimental data in a wide laser intensity range was not achieved.

Next, we compared the theoretically predicted yields of molecule N_2 and its companion atom Ar, from the MO-ADK [12] and ADK [9] theory respectively, with experimental results. In order to achieve this, we integrated observed rates by the following expression [34]:

$$Y(t) = \int W(t)dt. \tag{6}$$

Mentioned comparison directly reveals the role of the electronic structure played in the tunneling ionization of molecules. According to [35] molecular ionization is known to depend on the structure and electron density of a molecule. Concretely, [36] found that the electronic structure influenced the ionization mechanisms for O_2 and F_2 molecules. Based on obtained results, they concluded that such trend can be expected for the other diatomic molecules. Earlier [37] stated the same for N_2. The suppression of transition rate intensity of N_2 compared to Ar is in accordance with [12] where it can be found that the tunneling ionization rate is determined by

the suppressed barrier that occurs at a large distance from the atom and that the binding energy determines the tunneling rate each time the electron reaches the barrier. To obtain yield, we applied Eq. (6) on the molecule rate equations, Eqs. (2) and (4), and atom rate equation (Eq. (1)) from [9] over some definite time.

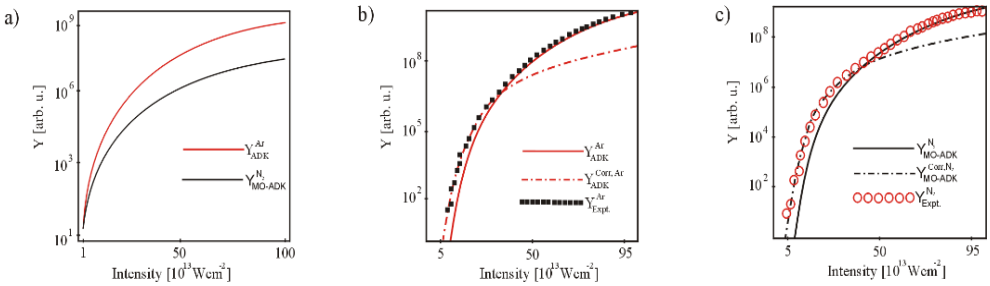

Figure 2. *Yield as a function of laser field intensity. In all panels, the pulse duration is 30 fs and the laser wavelength is at 800 nm. We used the ADK and MO-ADK theory. The experimental data are from [25]. For all graphs the following notation is used: red line for Ar atom, black line for N_2 molecule. Experimental results are shown as black squares for Ar atom and as red open circles for N_2 molecule.*

In Fig, 2(a), we compared the ionization yields of Ar atom and N_2 molecule, using the ADK [9] and the MO-ADK theory, without correction of ionization potential and with assumed Gauss laser beam shape. For intensities higher than $I{\sim}5 \times 10^{13}\ \mathrm{Wcm^{-2}}$, the yields show difference in spite of the similar ionization potential between them. The ionization yield of N_2 is, about two order of magnitude suppressed, in comparison with Ar. This is in accordance with [17] and the fact that the ionization potential of Ar has a slightly higher ionization potential and the corresponding yield lies at bit higher. Next, in Fig. 2, panels (b) and (c), we presented the yields for Ar atom and N_2 molecule, obtained by using Eqs. (2) and (4) and compared them with experimental results taken from [25]. In both graphs, our yield signals, $\mathrm{Y_{ADK}^{corr,Ar}}$ and $\mathrm{Y_{MOADK}^{corr,N_2}}$, fit the experimental perfect in the lower range of field intensity. After the saturation intensity close to $I \sim 2 \times 10^{15}\ \mathrm{Wcm^{-2}}$ the corrected yields decrease much faster than measured. Such behaviour is expected since Guo's group in [25] stated that experimental conditions are chosen so that Stark shift and ponderomotive potential are strongly diminished. This fact could explain the discrepancy between experimental and theoretical results at higher field intensities presented in Figs. 2(a) and 2(b).

Finally, in Fig. 3, we analyzed the laser intensity dependent ionization rate, $\mathrm{W}_{\mathrm{MOPPT}}^{\mathrm{corr}}(t)$, of the N_2 molecule at four different laser central wavelengths of $\lambda = (600, 800, 1000, 1200)$ nm. We would like to mention that the standard MO-ADK theory fails to give the wavelength dependence of ionization rates.

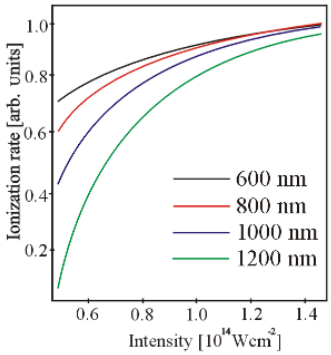

Figure 3. Comparison of the ionization rates predicted by the MO-PPT model of molecule N_2 as a function of laser filed intensity at four different central wavelengths, λ, of: (black line) 600 nm, (red line) 800 nm, (blue line) 1000 nm and (green line) 1200 nm. The laser field is taken to be a Gaussian pulse with a pulse duration (full width at half maximum) of $\tau = 20\ fs$.

From Fig. 3 one can observe that the increase of laser intensity leads to a logarithmic-like growth in the ionization rate for each value of wavelength. Our observations follow a similar trend to those reported by Zhao et al. [13,14]. Additionally, it is obvious that for lower wavelengths the ionization rates are more sensitive to changes in laser intensity. After $\sim 1.2 \times 10^{14}\ \mathrm{Wcm}^{-2}$ a saturation behaviour is observed up to the end of the laser pulse for the four curves. The results of experiments presented in [13] confirm this statement.

CONCLUSIONS

In conclusion, we have analyzed the influence of the ionization potential correction on the tunneling ionization rate for diatomic molecular system. Comparisons are made among the different versions of strong-field approximation. Our results clearly indicated that the correction of ionization potential effects the rate, by decreasing it. Also, some results show that the beam shape significantly influences the observed quantities.

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