Sequential double ionization of Ar to Ar2+ over intense laser field pulses

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Electrons ionize from an atom when enough external energy is applied to the system so that they can escape the potential energy barrier. Lasers can produce this external energy, and laser induced ionization is a three-step process with multiple results. One such result is called sequential double ionization, where enough energy is given to the system that two electrons ionize at the same time. This occurs when the laser intensity is strong, and as a result, the sudden loss of screening provided by the first electron causes a rearrangement of electrons and a second electron ionizes. These high intensity laser profiles are often difficult to calibrate and produce, so we built a program that will determine the probability of sequential double ionization given certain initial laser parameters, which will then help physicists to calibrate their laser profiles. We created this program in three steps: (1) initial conversion of laser parameters into electric field to calculate ionization rates, (2) calculation of ionization yield from the ionization rates obtained using both a Runge-Kutta method and a rate equation method, and (3) deduction of the probability of ionization from simple calculations using the ionization yield. This program matched well with experimental data for the rate equation method, but there was some error found when the Runge-Kutta method was used.

Keywords: laser profiles, sequential double ionization, intense laser pulses

I. INTRODUCTION

Typically, an electron is bound to an atom by a potential energy well that "traps" the electron and prevents it from escaping. The electron needs an energy boost from an external source in order to overcome the potential energy barrier and escape. Lasers are able to provide this additional energy by distorting the potential well, which allows the electron to then pass through the barrier. This process where an atom loses an electron (since it escapes) is known as ionization. Laser induced ionization is described by a three-step model: in the first step, a laser of intensity $10^{13} - 10^{16} W/cm^2$ distorts the potential barrier and an electron escapes. In the second step, the electron "rides" the laser out and gains kinetic energy, returning back towards the atom when the laser pulse switches direction. Then, in the final step of this process one of several outcomes can occur, including: (a) electron diffraction, where the electron is quasi-elastically scattered, (b) high harmonic generation, where the electron recombines with the atom and it releases energy as UV light, or (c) multiphoton ionization, where more than one electron ionizes, either independently or assisted, due to various factors.

Multiphoton ionization occurs when two or more electrons escape from an atom, and this can be a sequential or a nonsequential process. Nonsequential double ionization occurs when the laser intensity is weak and the second electron ionizes via rescattering, while sequential double ionization dominates at strong laser intensities where both the first and second ionizations occur independently via a tunneling mechanism, as described by Lin and Yuen [1]. During a nonsequential multiphoton ionization (NMPI) rescattering event, the first electron collisionally ionizes the second, which causes a delay between the initial and secondary ionization. On the other hand, the initial and secondary ionization occur without a delay in the sequential multiphoton ionization (SMPI) process because the sudden loss of screening previously provided by the first electron causes a rapid rearrangement of the remaining electrons, and therefore a second electron is ejected via tunneling almost immediately. There are important differences between these two processes: in SMPI, the electrons are emitted in bursts near maxima in the oscillating electric field, while in the rescattering process the electron excitation is constant. Additionally, the electrons originate at the outer turning point of the potential barrier during tunneling, but during NMPI the electrons appear in continuum near the nucleus, per Walker et al [2]. Although both of these processes are significant for different purposes, this research focuses exclusively on the sequential double ionization (SDI) of Argon, from Ar to Ar+ to Ar2+.

Laser intensity is particularly important in the double ionization process. Sequential double ionization occurs when the laser intensity is high, typically in the range of $10^{14} - 10^{16} W/cm^2$. However, high intensity laser profiles are difficult to calibrate and read. This research examines the phenomena associated with the sequential double ionization process under the influence of an ultrashort high intensity laser pulse with the aim of determining when sequential double ionization will occur under previously specified conditions. Therefore, the goal of this project is to create a program that will determine the probability of ionization given certain laser pulse parameters with the hopes of aiding experimental physicists in calibrating laser beam profiles. This paper will describe the process of creating the program in three steps (and the two comparative methods used), as well as the results generated during this process and how well these results match experimental data.

II. METHODS

In order to obtain the probability of ionization of the Argon electrons, we begin by converting parameters such as the laser frequency and pulse duration ($7 * 10^4 W/cm^2$ and 4 femtoseconds, respectively) into electric field. Once this is done, we can solve for the survival probability, i.e. how many electrons will remain attached to the atom, in two different ways: the Runge-Kutta method and the integral method. Both methods will allow us to determine the survival probability, which in turn determines the probability of ionization. We choose the element Argon as our reference atom as we have plenty of experimental data from Kubel et al [3] to verify that our program works as expected.

A. Step 1

Initially, we convert the laser parameters mentioned above into electric field over time, and then we evaluate the ionization rates of these electrons using an analytical expression based on tunneling ionization with the Ammosov-Delone-Krainov (ADK) theory, rather than solving the Schrodinger equation. In this project, we use the ADK theory as follows:

$$W_{TI}(F) = \frac{C_l^2}{2^{|m|}|m|!} \frac{(2l+1)(l+|m|)!}{2(l-|m|)!} \frac{1}{K^{2Z_c/K-1}} \left(\frac{2K^3}{F}\right)^{\frac{2Z_c}{K-|m|-1}} e^{\frac{-2K^3}{3F}} \quad (1)$$

This expression requires an input of electric field as a function of time (where C_l is the amplitude of the field free bound electron wavefunction in the asymptotic region, l and m are the corresponding orbital angular momentum and magnetic quantum numbers, Z_c is the charge seen by the active electron asymptotically, and $K = \sqrt{2I_p}$ where I_p is the ionization energy), and outputs the ionization rate. However, short pulses at higher intensities cause ionization in the barrier suppression regime (at or above the potential energy barrier height), and this results in an overestimation of the ionization rates when using the ADK method as noted by Tong and Lin [4]. While the ADK theory fails in this high intensity regime, it is sufficient for our purposes when modified. In order to correct our calculations and remove this overestimation, we use the following modified form of the ADK method with an empirical correction factor as given by Tong and Lin [4]:

$$W_{TBI}(F) = W_{TI} e^{-\left(\frac{Z_c^2}{I_p}\right)\left(\frac{F}{K^3}\right)} \quad (2)$$

Here, alpha is a fitting parameter, and W_{TI} is the expression (1) given above. Therefore, W_{TBI} gives us a more correct ionization rate and can be used in the next steps of building our program.

B. Step 2

Next, we use the ionization rate to calculate the expected yield of Ar+ and Ar2+. We use two separate methods to verify our results in this step. Method 1 involves a standard integration of several rate equations, while Method 2 uses the Runge-Kutta technique to solve three linear first order differential equations. Both methods are outlined below.

1. Method 1: Integral Method

We begin by using the following expression, which is a rate equation for population transfer for the first ionization from Ar to Ar+, to calculate survival probability. Here $p_0(t)$ is the survival probability and $W(t)$ is Eqn. (2) from Step 1.

$$\frac{dp_0}{dt} = -p_0(t)W(t) \quad (3)$$

We can solve this equation for $p_0(t)$ in the following manner:

$$\int \frac{dp_0}{p_0(t)} = -\int W_0(t)dt \quad (4)$$

To obtain:

$$p_0(t) = e^{-\int W_0(t)dt} \quad (5)$$

Once we have this expression, we can use Python to solve for $p(t)$. For example, when we use a laser intensity of $7 * 10^{14} W/cm^2$ with a pulse length of $4fs$ (or $164 a.u.$) and a wavelength of $750nm$, we get:

$$p_0(t)Ar = 0.5314800911592326$$

$$p_1(t)Ar+ = 0.469$$

where p_0 and p_1 are the survival of Ar and Ar+ accordingly.

2. Method 2: Runge-Kutta Method

We use the Runge-Kutta method to solve the following first order linear differential equations given by Lin and Yuen [1]. Here $p_0(t)$ and $p_1(t)$ are the survival probabilities of Ar and Ar+, respectively, and $W_0(t)$ and

$W_1(t)$ are the ADK expressions with respective factors for each ionization.

$$\frac{dp_0}{dt} = -p_0(t)W(t) \tag{6}$$

$$\frac{dp_1}{dt} = p_0 W_0(t) - p_1 W_1(t) \tag{7}$$

$$\frac{dp_2}{dt} = p_1 W_1(t) \tag{8}$$

The Runge-Kutta method is an iterative method that takes an initial value problem with some initial condition. In our case, the initial conditions are $p_0(0) = 1$ and $p_1(0) = 0$ since there are no Ar+ ions at the beginning of the laser pulse. The rate of change $\frac{dp}{dt}$ is a function of t and p itself. A maximum step size of 1 is chosen, and each iterated value is determined by the previous value plus the weighted average of four increments. Each increment is the product of the size of the interval and its height, or slope, as explained by Sureshkumar [5].

Using this method and the same laser profile, we obtain the following:

$$p_0(t)Ar : 0.53124674$$

$$p_1(t)Ar+ : 0.46$$

It is important to note that the population of the state is also the probability of finding the system in that state.

Additionally, we consider that the curve produced from this method has waves due to the fact that tunneling electrons are emitted in bursts near the maxima and minima in the oscillating electric field. The time is converted to atomic units from a 4 femtosecond pulse, with the maximum strength of the laser at time 0. Figure 1 was obtained using Method 2.

While previously in Figure 1, it looks as if there is no survival probability for Ar2+, that is not the case. The pink line in Figure 2 comes directly from using the Runge-Kutta method to solve for p_1 in Eqn. (8), while the black dotted line is obtained by solving for the survival probabilities of p_0 and p_1 in Eqns. (6) and (7) and then subtracting them from 1. All three survival probabilities (for Ar, Ar+, and Ar2+) add up to a complete ionization probability of 1. There is a slight error of less than $5x10^{-3}$ in the calculated survival probability from the Runge-Kutta method, which is expected.

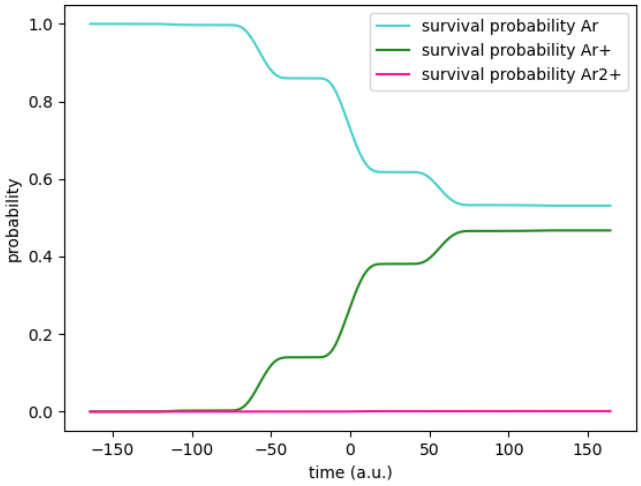

FIG. 1. Survival Probability of Ar, Ar+, and Ar2+ Over a 4fs Pulse

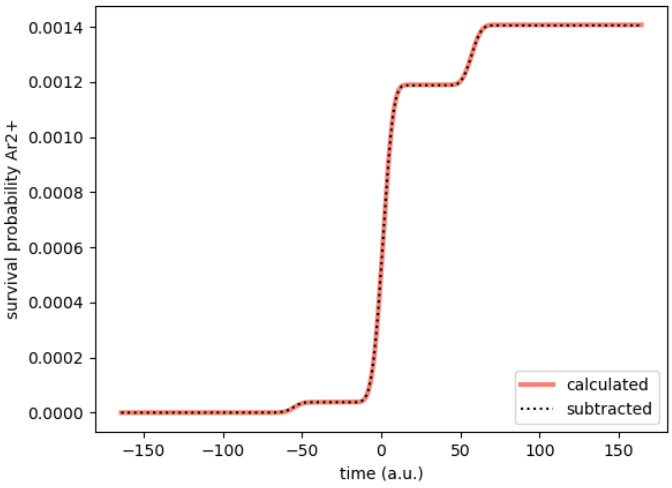

FIG. 2. Survival Probability of Ar2+ Over a 4fs Pulse

C. Step 3

Finally, using both of these methods, we can obtain the probability of the second ionization. For Method 1, we subtract each survival probability from the total ionization probability of 1, or

$$Ionization = 1 - p(t).$$

Subtracting the factors given by Method 1 from 1, the ionization yield is given by $1 - p(t)$. Here $p(t)$ is the sum of $p_0 = 0.5314800911592326$ and $p_1 = 0.469$ and equals 0.9947161003161279. Thus the Ar2+ yield is 1 - 0.9947161003161279, which gives $p_2(t)$ to be 0.00528389968387.

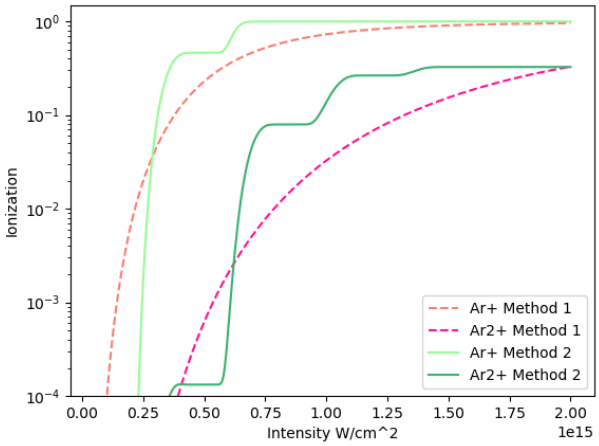

FIG. 3. Ionization Probabilities Over a Range of Intensities

For Method 2, we use our previous $p_0(t)$ and $p_1(t)$ values (0.53124674 and 0.46, correspondingly) as we solve equation (8) to obtain $p_2(t)$, which is determined to be 0.0014.

Finally, we use our program to calculate the probability of both initial (Ar+) and secondary (Ar2+) ionization over a range of strong intensities found in the sequential double ionization regime, from approximately $0.5 * 10^{14}$ to $2 * 10^{15} W/cm^2$. Our program uses both Method 1 and Method 2 to calculate the predicted values, and the comparative results are displayed in Figure 3, where the pink dotted lines are obtained using Method 1 and the green solid lines are obtained from Method 2.

III. RESULTS AND DISCUSSION

The aim of this project was to create a program that uses laser parameters, such as peak intensity and laser frequency, as inputs to predict the probability of nonsequential double ionization. This goal was completed; however, we obtained data that matched experimentally given values much better with one method than with the other.

Using our program, the survival probabilities of $p_0(t)$ and $p_1(t)$ for both Method 1 and Method 2 agree to two or three decimal places. Method 1 and 2 both give $p_0(t) = 0.531$ and $p_1(t) = 0.46$. However, the values found for $p_2(t)$ differ quite significantly between Method 1 and Method 2. Method 1 predicts $p_2(t) = 0.0052$ while Method 2 gives the value $p_2(t) = 0.0014$. Since these values differ by a margin of 0.0038, there is clearly an error. This error is likely to be in Method 2 since this value does not match quite as well with previously obtained experimental data from Kubel et al [3]. It is possible that the step size chosen for this method is too large, and a smaller step size value should be tested in future corrections of this program.

As mentioned previously, the pink dotted lines in Figure 3 are obtained from Method 1, while the green solid lines are obtained from Method 2. These green solid lines seem to underestimate the ionization until about $0.25 * 10^{15} W/cm^2$ and $0.65 * 10^{15} W/cm^2$ for Ar+ and Ar2+, respectively. After that, the predicted ionization probability given by Method 2 overestimates the ionization for the rest of the intensity range and then jumps unexpectedly around $0.5 * 10^{15} W/cm^2$ for Ar+ as well as $0.9 * 10^{15} W/cm^2$ and again at approximately $1.25 * 10^{15} W/cm^2$ for Ar2+. Since this does not match the results obtained from Method 1, more consideration should be given to this method in future work to better calibrate the program.

When compared to experimental data of sequential double ionization of Argon, our results are somewhat mixed; Method 1 generates expected results and appears to match the data more closely than Method 2, likely indicating more error when using the Runge-Kutta method. Despite this, our program is an excellent starting point for calibration of laser profiles with high intensities and predictions of sequential double ionization probabilities over a range of intensities.

IV. SUMMARY

In summary, laser-induced sequential double ionization occurs when a high intensity laser is used, yet the range of these intensities make the subsequent laser profiles difficult to utilize. In order to rectify this, the goal and end result of this project is the program that we built to create Figure 3, which can take in laser parameters and predict nonsequential double ionization probability over a range of different intensities, allowing for better calibration of laser profiles in the barrier suppression regime. This program still needs a few adjustments to properly calculate ion yields and survival probabilities using the Runge-Kutta method; however, overall it is still effective in taking in laser pulse parameters and converting these to electric field data, which can be used to find the ionization rate, and subsequent probability of survival and ionization, when using the integral method. Future work should include fine-tuning this program to fix the error mentioned above, as well as testing other intensity ranges to be sure that it functions correctly before expanding to generate ionization probabilities for other atoms.

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