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Abstract

High-order harmonic generation (HHG), attosecond pulse train (APT), and non-sequential double ionization (NSDI) in the He atom under high intense femtosecond laser pulses are calculated by time-dependent Schrodinger equation (TDSE) in one dimension (1D). By considering the mutual electron-electron and electron-nuclei interactions along with calculating the He atom ground state wave function by imaginary time propagation (ITP) method, besides calculating probability density of electrons, dipole acceleration, HHG, and APT, we could generate the well-known "knee structure" in the probability of the He atom ionization against the intensity in an ionization
boundary condition model. The results are in good agreement with the experimental data reported by Walker et al. [B. Walker et al. Phys. Rev. Lett. 73, 1227 (1994)].

Keywords

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Introduction

Laser advances, especially the invention of the chirped pulse amplification (CPA) technique [1], have provided the possibility of generating ultrashort laser pulses down to few femtoseconds with high intensities up to $10^{22}$ W/cm$^2$ [2]. The interaction of ultrashort laser pulses with atoms and solid-state targets can cause amazing phenomena such as laser wakefield acceleration [3, 4], ion beam acceleration [5, 6], laser-driven fusion [7, 8], high-order harmonic generation (HHG) [9, 10], radiation generation [11, 12], nuclear resonance fluorescence [13], vacuum birefringence [14-16], electron-positron pair production from vacuum [17, 18], photon-photon scattering [19, 20], electromagnetic cascades [21, 22], etc.

In HHG, the interaction of high intense laser pulses with atom can be described by a semi-classical model, called the three-step model or the simple man theory [23]. According to this model, in the first step, the electron tunnels through the atomic or molecular potential with the help of the strong field of the laser pulse. In the second step, the electron is accelerated in the laser field, classically. The third step is re-collision and recombination of the electron with the parent's ion. Depending on the type of recombination, various phenomena may occur: (i) if the recombination is elastic, it would lead to HHG; (ii) if it is inelastic, recombination will lead to non-sequential double ionization (NSDI); and finally (iii) if it is dispersed by the nuclei, it will lead to above threshold ionization (ATI) [24].
High-order harmonic generation (HHG) is an extreme and non-perturbative nonlinear optical process, in which at the presence of a gaseous or solid-state target, the driving laser frequency is converted into its integer multiples. If the harmonic phases are locked properly, attosecond pulse trains will be emerged from interaction that can be measured by methods such as streak camera [25].

Non-sequential double ionization (NSDI) is another phenomenon happening in the interaction of high intense laser fields with more-than-one-electron atoms. NSDI was observed for the first time in 1992 [26]. Fittinghoff et al. observed that He atom is ionized doubly and simultaneously at the intensities lower than those predicted by the famous ADK model, where it is assumed that the ionization of He occurs sequentially. They observed a "knee-shaped" curve, that is, average number of doubly-ionized He atoms produced against the laser intensities showed a curve like a knee. Two years later, Walker et al. observed again the same effect, but with different intensities that phenomenon started [27]. This phenomenon was justified by two descriptions: (i) Shake-off, in which the second electron is ejected due to sudden change of the atomic potential after a fast removal of the primary photoelectron [28]; and (ii) recollection scenario, in which the ionized electron is hurled back at the core and ionizes the second electron [29, 30]. The latter is expressed in re-scattering model included in three-step model. To study the NSDI effect, at least three approaches can be followed: (i) classical ensemble, which is based on Newton's equations; (ii) S-matrix; and (iii) time-dependent Schrodinger equation (TDSE), which is one of the strong nonlinear method. The latter, however, is a difficult task in particular in three-dimension (3D), when no approximation is considered. The high volume of calculations in this approach [31, 32] is due to this fact that solving the Schrödinger equation in 3D for He atom requires at least six dimensions; three dimensions for each He electron. Although one-dimensional (1D) solutions cannot express all effects, however,
we will show that this approach, with its own difficulties, can well describe the NSDI for the He atom. In particular, this model can show the "knee structure" with very good agreement with experiment [27]. Truong et al. believe that besides huge computational demands, physical propagation of the ionized electron cannot be traced by TDSE [33]. Therefore, they used a classical ensemble model for Ar atom, and with using a soft parameter for Coulomb potential as $a = 1.5$, they could achieve the knee structure at the intensity range of $I = 4.0 \times 10^{14} \, \text{W/cm}^2$ to $I = 5.0 \times 10^{14} \, \text{W/cm}^2$.

The knee structure has been simulated in numerical works, using classical ensemble [34-38], S-matrix[39, 40] and numerical solution of 1D TDSE [41, 42]. In this work, we present a full quantum mechanical approach for a He atom with two interacting electrons under the radiations of femtosecond laser pulses with various intensities. This approach provides us with the opportunity of calculating dipole acceleration, high-order harmonic spectrum, attosecond pulse train, distribution of electron probability density, and finally the non-sequential double ionization.

For the He atom, the main challenge for solving the TDSE is finding the ground state wave function. He ground state wave function plays a crucial role for solving the TDSE. For a hydrogen atom, the time-independent Schrödinger equation (TISE) presents an exact solution for the ground state, however, for more-than-one-electron atoms, such as He atom, it is not the case. The latter can be found by some approximated methods such as implicitly restarted lanczos [43], spectral method [44], Numerov method [45], symplectic schemes [46], Nikiforov-Uvarov method [47], exact quantization rules [48], etc.

In this work we apply the imaginary time propagation (ITP) method [49] to find the ground state wave function and energy of the He atom. ITP method is based on solving the TDSE, without external fields, in which “$t$” is replace by “$-i * t$”, where $i = \sqrt{-1}$. It is proven that for time long
enough times, the wave function collapses on its ground state. This method is used to be applied for the calculation of ground state energy and ground state wave functions of atoms and molecules [50-59]. To reach a more precise solution, we also apply iteration method along with the ITP method. The iteration method can help to reach as much as an exact solution for the wave function, even for an unsuitable initial guess. Our criteria for recognizing the right result is the experimental value for the He ground state energy which is 2.903385 a.u. [60]. We base our numerical calculations on the Euler method.

In order to avoid divergence in the coordinate origin \((\vec{r} = 0)\), the soft core potential is used which is defined as \(\propto \frac{1}{\sqrt{r^2 + a^2}}\) [61]. We find that the soft-core potential constant “\(a\)” has a very important role in determining the ground state energy. As we will see, “\(a\)” factor has an important role also in the HHG spectrum and the other characteristics extracted for the He atom. The investigation of soft core potential constant has been reported in the fewer works [33]. After making sure about the accuracy of the ground state wave function, we put it as an initial condition in the TDSE and calculate the time-dependent wave function, dipole acceleration, HHG, and NSDI. The result for probability of the He atom ionization against the laser intensity should give the knee-shaped curve, which is observed in experiment [27]. We calculate the knee structure by defining an ionization boundary condition [44]. This boundary in fact is a distance from the nucleus, after which the electrons are assumed to be separated from the atom, and so atom is assumed to be ionized.

All the calculations are performed in the atomic unit system, \(i.e. \hbar = |e| = m_e = 4\pi\varepsilon_0 = 1\). In order to solve the time-dependent and time-independent Schrodinger equations, the Euler method (central derivative) is used, but for ITP method, in which forward difference is used for time derivatives. Also, in order to prevent the reflection of wave functions from the boundaries and
unwanted interferences, the boundary condition of mask function is used. We normalize the wave functions in each step of time loop to avoid divergence in the wave function in the next steps.

Results and discussion

In this section, we present the results of our numerical simulations consisting of electrons' wave function, dipole acceleration, high-order harmonic generation, attosecond pulse trains, and non-sequential double ionization. Towards this end, as mentioned, computational codes were written in Fortran 95 and run in LINUX operation system with Ubuntu version 20.04 mint. In solving the Schrodinger equation and extracting HHG spectrum, we considered a spatial axis from -50 a.u. to 50 a.u., with spatial meshes of 0.25 a.u. of length, i.e. 400 equidistance meshes were generated for using in FDM codes. The time within which the Schrodinger equation was solved is $5T$, which is equal to pulse duration, where $T = \frac{2\pi}{\omega}$ with $\omega = 0.057$ a.u.. Therefore, the equations were solved in 551 a.u. time interval with time mesh of 1/1600 a.u., so, the total meshes in time interval is 881850. In order to seek for the knee structure, since we need to calculate the Schrodinger equation in a larger spatial scale, the position interval was expanded to −75 a.u. to 75 a.u., with the same 0.25 a.u. as mesh value.

To be sure about the results, we checked the accuracy of the helium ground state wave function in three ways: first, with calculating the ground state energy and providing that it should be close to the experimental values of 2.903385 a.u. [60]; second, with ignorance the external laser field and solving the Schrodinger equation, such that the wave function of ground state does not change with time; third, by checking the cut-off frequency of the harmonic spectrum, providing that it should obey the cut-off law of $\hbar\omega_c = I_p + 3.17U_p$.

A. Precise value for “α” factor
Using ITP method along with iteration, the helium ground state wave function was calculated. Having found this wave function, we put it in the TDSE of equation (2) and then calculated the mean value of Hamiltonian. The correct value of “$a$” giving the expectation value for ground state energy as 2.903517 a.u. is $a = 0.744$. In the Table 1, various “$a$” factors along with the resulted ground state energies are listed. In Ref. [62], by choosing $a = 1$, the value of 2.2 a.u. has been reported for ground state energy which is in agreement with our calculations.

Table 1 The absolute value for He ground state energy calculated by changing "a" factor

<table>
<thead>
<tr>
<th>$a$ factor (a.u.)</th>
<th>0.1</th>
<th>0.3</th>
<th>0.4</th>
<th>0.7</th>
<th>0.74</th>
<th>0.744</th>
<th>0.75</th>
<th>0.8</th>
<th>1.0</th>
</tr>
</thead>
<tbody>
<tr>
<td>$E_0$ (a.u.)</td>
<td>7</td>
<td>6.23</td>
<td>4</td>
<td>3.05</td>
<td>2.91</td>
<td>2.903</td>
<td>2.88</td>
<td>2.72</td>
<td>2.2</td>
</tr>
</tbody>
</table>

B. Helium ground state wave function

As mentioned, the He atom ground state wave function is calculated by ITP method. In this method, after replacing "t" by "i×t", we first allowed the computational time goes to 8000 a.u.. However, after analyzing our results, we realized that after computational time of 3000 a.u., the wave function did not change noticeably. No noticeable change in the wave function distribution means it is very closed to the He atom eigenstate. To verify that, we put it in equation (2) with dropping the external field and let the time passes. According to quantum mechanical principles, “if the acquired eigenstate is the Hamiltonians eigenstate, its probability distribution should not change over the time”. By checking the results, we made sure that wave function is actually the ground state of the He atom. Fig. 2 shows the ground state wave function of the He atom at various
times. Accordingly, variations at various times \((t=0, T, 2T, 3T, 4T, \text{ and } 5T)\) are very small, such that one can safely say that it is just the ground state wave function.

C. Helium high-order harmonic generation and attosecond pulse train

By solving Equation (2) through the symmetric Euler method in the spatial range of \(-50\) to \(50\) a.u. and using the He ground state wave function obtained by ITP method, then using equation (3)-(5), the high-order harmonics of the He atom were obtained. For comparison, we have also calculated the high-order harmonics for the case of non-interacting electron model for the He atom. Fig. 3 shows the HHG of the He atom for (a) non-interacting electrons, (b) interacting electrons with ITP method without iteration, (c) interacting electrons with ITP and one iteration, and (d) interacting electrons with ITP and two iterations. As the figure shows vividly, considering the interaction potential of the electrons, changes overall the shape of HHG spectrum. Also, iteration has been able to improve the HHG spectrum. Nevertheless, considering the iteration two times, does not lead to noticeable improvement in the HHG spectrum. Therefore, we proceeded the rest of the calculations with only one iteration. In Fig. 3, the simulations have been done for \(I = 1 \times 10^{14} \text{ W/cm}^2\) according to field amplitude of \(E_0 = 0.05338 \text{ a.u.}\). For this parameter, the pondermotive energy becomes \(U_p = 0.219 \text{ a.u.}\) and according to cut-off law, the cut-off frequency becomes \(\omega_c = 63 \text{ a.u.}\). Fig. 3(c, d) shows the cut-off frequency of HHG as \(67\omega_0\) which is in good agreement with cut-off law.

To make another test for proper choosing of "\(a\)" factor, we calculated the HHG for three values of "\(a\)" as 0.3, 0.744, and 1. For all cases, the ground state wave function was calculated by ITP. The results are depicted in Fig. 4. As one can see, this parameter significantly affects the HHG spectrum. Results do not give proper cut-off frequency for "\(a\)" parameter rather than 0.744.
We have also examined the effects of other laser intensities on the HHG spectrum. The cut-off frequency for $I = 5 \times 10^{14} \, W/cm^2$, according to cut-off law, is $111 \, \omega_0$. Our HHG spectrum shows exactly $111 \, \omega_0$ (Fig. 5 (a)). Also for $I = 1 \times 10^{15} \, W/cm^2$, the cut-off law gives a cut-off frequency of $171 \, \omega_0$, while our HHG gives it as $159 \, \omega_0$ (Fig. 5 (b)). For intensities higher than $1 \times 10^{15} \, W/cm^2$, the shape of the HHG spectrum is disturbed and do not show a common spectrum (c and d). We believe that this distortion comes from this fact that two electrons are engaged in NSDI process, and therefore the situation is more complicated than singel active electron case.

To be insured about the interpretation given above, regarding the statement that "why at higher intensities than $1 \times 10^{15} \, W/cm^2$, the cut-off frequency law does not consistent with that given by a full quantum mechanical calculation", we calculated the probability of the presence of electrons around the nucleus for two intensities of $1 \times 10^{15} \, W/cm^2$ (Fig. 6) and and $1 \times 10^{16} \, W/cm^2$ (Fig. 7). Fig. 6 confirms that for intensity of $1 \times 10^{15} \, W/cm^2$, two-electron wave function distributes in the vicinity of the nucleus. However, for intensity of $1 \times 10^{16} \, W/cm^2$, the electrons move away from the nucleus, even in a short simulation time, and actually double ionization occurs. Therefore, for intensities higher than $1 \times 10^{15} \, W/cm^2$, the dominant ionization process would be the NSDI. As a result, HHG spectrum should not necessarily give a cut-off frequency according to cut-off law. Note that the length scales are different for figures 6 and 7, to show how far the electrons are from the nucleus.

To generate a clean attosecond pulse train, one has to use a frequency window. By choosing a window from $30 \omega_0$ to $60 \omega_0$ and using Fourier summation, we calculated the HHG spectrum for various intensities below and above the intensity of $I = 1 \times 10^{15} \, W/cm^2$. The results are shown
in Fig. 8 for $I = 1 \times 10^{14} $ W/cm$^2$ (Fig. 8(a)), $I = 5 \times 10^{14} $ W/cm$^2$ (Fig. 8(b)), $I = 1 \times 10^{15} $ W/cm$^2$ (Fig. 8(c)), and $I = 1 \times 10^{16} $ W/cm$^2$ (Fig. 8(d)). With increasing the laser intensity, the height of APT peak increases, but for the intensity of $1 \times 10^{16} $ W/cm$^2$, which is decreased. This effect provides another evidence for NSDI, which happens at intensities higher than $1 \times 10^{15} $ W/cm$^2$. At this intensities, the electrons contribu mainly in NSDI, rather than attosecond pulse generation.

As we know, APT generation occurs when the electrons return to their parrent and recolide with it. For hydrogen atoms, this effect has been inspected wastly. However, for He atom, fewer theoretical works can be found [63, 64]. We calculated the expectaion value for electron's position, $\langle x \rangle$, and show it along with APT in a same curve, where the horezontal axis is time. Fig. 9 shows the expectaion value of electron's position and APT for the intensity of $2 \times 10^{14} $ W/cm$^2$. The figuer clearly illustrates that the peaks of APT occur where the expectation value of electron 's position vanishes, i.e. when the electron re-collides with nucleus. This means that the attosecond explosion occurs when one of the electrons re-collides with the nucleus. Since we ignore the electron spin in our calculations, the calculation has been doen only for one electron.

D. Knee- structure for NSDI

At the last part of this work, we show the results of NSDI of the He atom. In the previous sections, we obtained the He atom ground state wave function and tested it with some criteria. Using this wave function as the initial wave function of equation (2) and applying the ionization boundary condition (Fig.1), we calculated the NSDI for the He atom. According to the experimental results [27], the probability of ionization has been measured for an intensity interval of $\sim 1 \times 10^{14}$ to $\sim 1 \times 10^{16}$ W/cm$^2$. We carried out our simulations for sixteen points between these two
intensities. Since at the beginning, the exact value of \( R \) is not known, we performed calculations for various values of \( R \) as 10, 20, 30, 40, 50 and 60 a.u.. The results are given in Fig. 10. As the figure shows, the main features of NSDI of the He atom, which is known as "knee structure", is quite clear for most of the curves.

According to Fig. 10, almost all curves show knee structures, although \( R=10 \) a.u., has not given a proper curve. For \( R=20, 30, \ldots 60 \) a.u., however the simulations results show very good knee structure. This shows that for the He atom, the double ionization occurs for distances of the order of \( 20 \) a.u. and farther. In Ref. [29], the authors have chosen \( R=30 \) a.u. to show NSDI. We showed, however, that the NSDI occurs from shorter distances. We attribute this result to the TDSE, that we believe that it give better results compared to classical approaches [29].

Compression with experimental data [27], our simulation shows a very good agreement, especially the starting intensity for the curve, and the intensity of the knee point.

**Conclusion**

In conclusion, in this work we investigated the interaction of a high intense femtosecond laser pulses with the He atom using the time-dependent Schrodinger equation. The ground state of the He atom was achieved by imaginary time propagation method. By choosing proper spatial and time meshes, along with proper boundary condition of mask function, we could solve the Schrodinger equations by Euler method and obtain accurate solutions for ground state wave function, high-order harmonics, attosecond pulse train, and non-sequential double ionization. We observed that soft core potential constant, \( \alpha \) factor, has a crucial role in obtaining accurate solutions for the ground state wave function and energy of the He atom. We tried to include the least approximations in our modeling. For high-order harmonics, our calculations agreed with cut-off law, for intensities for which NSDI has not occurred. For laser intensity exceeding
\[ 1 \times 10^{15} \text{ W/cm}^2 \text{ the HHG spectrum exit from its common shape and dose not match with cut-off law. Using a full quantum mechanical treatment, we showed that this discrepancy occurs when electrons move away from the nucleus, which happens for intensity higher than } 1 \times 10^{15} \text{ W/cm}^2. \]

We could also generate the well-known knee structure for non-sequential double ionization, which has been observed in the experiment. We attributed this effect to correlated ionization (NSDI) happening for high intensities.

**Theory and method of solution**

In this section, we describe the theory and the methods that are used for finding the ground state wave function and the time-dependent wave function of the He atom, and calculating the HHG, APT, and NSDI for the He atom.

**A. Hamiltonian of the He atom and the boundary conditions**

We commence with the time-dependent Hamiltonian of the He atom. The Hamiltonian of helium atom with considering mutual interaction of electrons as well as the electron-nucleus interactions in a.u. unite system is given by:

\[
H = \frac{1}{2} \frac{\partial^2}{\partial x_1^2} + \frac{1}{2} \frac{\partial^2}{\partial x_2^2} - \frac{2}{\sqrt{x_1^2 + a^2}} - \frac{2}{\sqrt{x_2^2 + a^2}} + \frac{1}{\sqrt{(x_1 - x_2)^2 + a^2}} - E(t)(x_1 + x_2) \tag{1}
\]

where \( x_{1(2)} \) is the position of electron 1(2). The soft-core potential constant “\( a \)” is used to avoid divergence in calculations in the origin \((x_1, x_2 = 0)\). In equation (1), \( E(t) \) is the pulsed laser field, which is assumed to be \( E(t) = E_0 \sin(\omega t) f(t) \), with \( E_0 \) as the amplitude of the field. The envelope function of the pulse is assumed to be \( f(t) = \sin^5(\pi + \omega t/10) \). Although using the Gaussian, \( \cos^2 \), and \( \sech \) functions are common for pulsed laser envelops, however, since \( \sin^5 \) function shows a
better fit on the Gaussian pulse, and that it intercepts the horizontal axis at finite values on t-axis, it can dramatically reduce the time meshes and so save the run time noticeably. In \( \sin^5 \) function, \( \omega \) is the center frequency of the driving pulse, which is assumed to be 0.057 a.u., according to the center wavelength of 800 nm for Ti:sapphire laser. In the equation (1), the fifth term is the mutual potential of electrons.

In solving equation (1) by Euler method, a mask function of the form of \( \cos^{1/8} \) is applied on the wave function at the far boundaries. We multiply the He atom wave function by \( \cos^{1/8}[x - x_0/X - x_0] \pi/2 \) from \( x_0 \)-point and after that, where \( x_0 = 0.8 X \), with \( X \) being the end point of the spatial interval in the simulation frame. This function prevents unwanted reflections of the wave function from the boundaries and random interferences, that leads divergence of solution.

**B. Helium atom ground state wave function and energy**

In this section, we explain the details of ITP method towards obtaining the He atom ground state wave function and energy. The time-dependent Schrodinger equation is given by:

\[
i \frac{\partial \psi(x_1, x_2, t)}{\partial t} = H \psi(x_1, x_2, t)
\]  

As mentioned, to calculate the ground state wave function with ITP method, we first replace "t" by "\(-i \times t\)" as:
\[ \frac{\partial \psi(x_1, x_2, t)}{\partial t} = \left[ \frac{1}{2} \frac{\partial^2}{\partial x_1^2} + \frac{1}{2} \frac{\partial^2}{\partial x_2^2} + \frac{2}{\sqrt{x_1^2 + a^2}} + \frac{2}{\sqrt{x_2^2 + a^2}} - \frac{1}{\sqrt{(x_1 - x_2)^2 + a^2}} \right] \psi(x_1, x_2, t) \]  

(3)

Then, so solve it, we use the finite difference method. Implicit Euler method for time derivative and symmetric Euler method for spatial derivatives are used. Codes are written in Intel Fortran and run in Linux operating system. Since we do not consider the electron spin, it is predictable that the parity of the He ground state wave function is even. Therefore, the initial function which is considered as the initial conditions of numerical calculations should be a symmetric function. We consider \( \psi_0(x_1, x_2, t = 0) = 4x_1x_2e^{-2(|x_1| + |x_2|)} \) [65] as the initial guess for ground state wave function, which is actually a multiplier of two non-interacting electron wave functions. We then let the time goes to large values. To obtain a precise result, the procedure is iterated few times, that is, the solution is return as the new initial solution and the procedure is repeated. The number of adequate iterations is determined by the energy eigenvalue that is returned after each run. The desirable value for ground state energy is 2.903517 a.u. [60]. We should note here a crucial point that is to achieve accurate solutions for quantities such as HHG spectrum, APT, and NSDI probability, the boundary conditions as well as the spatial interval, time interval, and time and position meshes, all should be the same for all codes.

C. High-order harmonic generation and attosecond pulse train

Having found the right ground state wave function of the He atom, the next step is to calculate the He atom wave function under the interaction with laser pulse in next times. The time-dependent wave function of the He atom allows us to calculate the HHG spectrum, that is carried out through calculating the dipole acceleration of the atom. Using Ehrenfest' theorem, the dipole acceleration is given by [61]:

14
\[
\ddot{x}(t) = \dot{i}[H(t), [H(t), x_1]] + \dot{i}[H(t), [H(t), x_2]]
\]  

(4)

Substituting Hamiltonian of equation (1) into equation (4), we obtain:

\[
\ddot{x}(t) = \left(\psi(x_1, x_2, t) \frac{x_1}{(x_1^2 + \alpha^2)^{3/2}} + \frac{x_2}{(x_2^2 + \alpha^2)^{3/2}} \right) \psi(x_1, x_2, t) + 2E(t)
\]  

(5)

where \(\psi(x_1, x_2, t)\) is the solution of equation (2). The HHG spectrum is calculated by taking a fast Fourier transform (FFT) of the dipole acceleration, given by:

\[
\ddot{x}[\omega] = FFT(\ddot{x}[t]) = \sum_{t=0}^{N-1} \ddot{x}[t] \left(\exp\left(\frac{2\pi i t}{N}\right)\right)^{t\omega}, \text{ for } \omega = 0, 1, 2, ..., N - 1
\]  

(6)

where \(N\) is the number of \(\omega\) points. To do FFT well, a command code in Scilab software is used. Attosecond pulse train is achieved by a coherent superposition of harmonics calculated by equation (6), first by filtering out the extra frequencies (through defining a frequency window), and then by doing a Fourier summation of frequency components. In our numerical calculations, frequency window is defined as a frequency range between \(n\omega_0\) and \(m\omega_0\), with \(n\) and \(m\) as two integers. HHG spectrum has a universal shape: it consists of some primary high intense harmonics, followed by a plateau region, and finally a cutoff at the end. The cutoff frequency of HHG should follow the cut-off law: \(\hbar \omega_c = I_p + 3.17U_p\) [23], with \(\omega_c\) being the cutoff frequency, \(I_p\) the ionization potential, which is equal to the negative of the He ground state energy, and \(U_p\) the pondermotive energy, defined by \(U_p = \frac{e^2E_0^2}{4m_e\omega^2}\).

D. Non-sequential double ionization
Non-sequential double ionization (NSDI) in more-than-one electron atoms occurs when an intense laser pulse removes simultaneously atomic electrons. In the He atom, this effect starts from laser intensities near $2 \times 10^{14} \, W/cm^2$ [27]. The famous "knee" on the curve happens near the $1 \times 10^{15} \, W/cm^2$. For lower intensities than $2 \times 10^{14} \, W/cm^2$, one expects single ionization. The knee diagram is generated when probability of ionization is plotted against the laser pulse intensity. The probability of ionization is calculated through the integration of squared wave function obtained by solving equation (2) in outer space of the ionization boundary. The ionization boundary condition is plotted in Fig. 1, where “$R$” is the ionization boundary, after which the electrons are assumed to be separated completely from nucleus, and so the atom is ionized. In fact, $R$ is unknown at the beginning, but it should not be too short that overlaps with bound states, shouldn’t be too large that ionized part of wave function reaches the end point of simulation frame. If $|x_1| > R$ and $|x_2| < R$ or $|x_1| < R$ and $|x_2| > R$, single ionization will occur; If $|x_1| > R$ and $|x_2| > R$, double ionization will occur. We calculate the double ionization probability ($P_{di}$) as:

$$
P_{di} = \int_0^{t_f} \int_{R,R}^{x_{1f},x_{2f}} \psi^*(x_1,x_2,t) \psi(x_1,x_2,t) \, dx_1 dx_2 \, dt \tag{7}
$$

where $x_{1f}$ ($x_{2f}$) are the end points of simulation frame for 1(2) electron and $t_f$ is the total time of simulation. The ionization probability is calculated for various intensities in the range of $1 \times 10^{13} \, W/cm^2$ to $1 \times 10^{16} \, W/cm^2$.

Reference


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Declarations

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this manuscript.

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All authors agree to the ethics and consent to participate in this article and declare that this submission follows the policies of *Scientific Reports*. Accordingly, the material is the authors’ original work, which has not been previously published elsewhere. The paper is not being considered for publication elsewhere. All authors have been personally and actively involved in substantial work leading to the paper and will take public responsibility for its content.

**Consent for publication**

Not applicable.

**Availability of data and materials**

The datasets used and/or analyzed during the current study are available from the corresponding author on reasonable request.

**Competing of interests**

The authors declare that they have no conflict of interest.

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**Authors’ contributions**

Mohammad Sabaeian: Supervising the project, Conceptualization, Editing the manuscript, Revising. Marjan Zakavi: Making computational codes, Conducting computations, Data curation, Preparation figures, Methodology, Data validation, Writing original draft.

**Figure legends**

Figure. 1: The ionization boundary condition: $x_1$ and $x_2$ are the coordinates of electrons, $x_0 = 0.8 \times x_{1f}$ ($x_{1f}$) with $x_{1f}$ ($x_{2f}$) as the end points of simulation frame, and $R = 10, 20, ..., 60$ a. u.
Figure 2: The helium atom ground state wave function obtained by TDSE without external field. Total run time is $5T$ where $T = 110.2$ a.u.. Figure 3: High-order harmonic spectrum of the He atom for $\lambda = 800$ nm and $I = 1 \times 10^{14} W/cm^2$: (a) non-interacting electrons model, (b) interacting electrons model with ITP without iteration, (c) interacting electrons model with ITP and one iteration, and (d) interacting electrons model with ITP and two iterations. Figure 4: High-order harmonic spectrum for $\lambda = 800$ nm and $I = 1 \times 10^{14} W/cm^2$. The helium atom ground state energy has been obtained by ITP method with one iteration. The soft-core potential constant of: (a) $a=0.3$, (b) $a=0.744$, and (c) $a=1$ have been considered. Figure 5: High-order harmonic spectrum for $\lambda = 800$ nm and intensities: (a) $I = 5 \times 10^{14} W/cm^2$, (b) $I = 1 \times 10^{15} W/cm^2$, (c) $I = 8 \times 10^{15} W/cm^2$, and (d) $I = 1 \times 10^{16} W/cm^2$. Figure 6: Probability of existence electrons for intensity $1 \times 10^{15} W/cm^2$ that $T$ is time period of $\omega = 0.057$ a.u.. all of the simulation time was $5T$. Figure 7: Probability of the presence of electrons for the laser intensity of $1 \times 10^{16} W/cm^2$. $T$ is the time period of $\omega = 0.057$ a.u., and the total simulation time is $5T$. Figure 8: ATP for $\lambda = 800$ nm and intensity of: (a) $I = 1 \times 10^{14} W/cm^2$, (b) $I = 5 \times 10^{14} W/cm^2$, (c) $I = 1 \times 10^{15} W/cm^2$, (d) $I = 1 \times 10^{16} W/cm^2$. Figure 9: Attosecond pulse train (black curve) and expectation value of helium electron's position (blue curve) for $I = 5 \times 10^{14} W/cm^2$. Figure 10: The NSDI for the helium atom showing knee structure for ionization boundary of $R = 10.20.30.40.50.60$ a.u.
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Figure 3

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