The role of electron-electron repulsion to the nonsequential double ionization mechanisms

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Abstract: In this study the classical three-dimensional ensemble model is utilized for investigating the role of final state electron-electron repulsion in forming the ultimate correlated two-electron momentum distribution. For the first time, a comprehensive analysis has been made to thoroughly understand such repulsive force associating with each microscopic mechanism of nonsequential double ionization. The helium exposed to 800-nm laser with two representative intensities of $3.5 \times 10^{14}$ W/cm$^2$ and $4.5 \times 10^{14}$ W/cm$^2$ is used for illustration. The results indicate the dominance of electron-electron repulsion in direct and recollision-induced excitation with subsequential ionizations. While its contribution in case of exchanging-state mechanism gradually emerges as the laser intensity increases.

1. Introduction

In the framework of strong-field physics for striking understanding of laser-matter interaction, the non-linear phenomena induced by intense laser field such as the generation of high-order harmonic [1,2], the existence of above-threshold electron [3], and double or multiple ionization [4,5] have been considered recently. Among them, nonsequential double ionization (NSDI) process is scrutinized as a tool to comprehensively and purely study the electron-electron (e-e) correlation toward the recollision process [5] (see also [6] for further information). Since the first observation was explored by V. V. Suran and I. P. Zapesochny in 1975 [7], a large number of experimental and theoretical studies have been performed on NSDI to investigate two-
electron correlated dynamics in the atomic or molecular orbitals toward the recollision process [5,8-15]. The most intuitive model for understanding the NSDI process is the so-called simple-man theory [1]: the first electron is tunneling ionized and accelerated by the laser field then is driven back to revisit its parent ion leading to the liberation of the second one when laser field reverses its direction. One of the most simple and intuitive approach to this problem is classical ensemble model proposed by Hann et al. showing good consistency to that using quantum calculations provided that the laser intensity is sufficiently high [16]. The wide use of this classical approach is based on its advantages over the full-quantum consideration: (i) the entire process of double ionization can be easily calculated from beginning to the end of the pulse, and (ii) at any time, individual double ionization trajectories can be back analyzed to extract insight into their dynamic [17].

In recent years, the experimental physics have made great progress in exploring the microscopic dynamics of NSDI [18-20] that continuously challenges our understanding of NSDI. For instance, a fingerlike (or V-like) structure in the correlated two-electron momentum distribution (CTEMD) was experimentally observed, which indicates strong backscattering of the first ionized electron at the nucleus upon recollision [21]. Theoretical studies showed that for sufficiently low laser intensity both the ion-electron interaction at recollision moment and the e-e repulsion in the final state play considerable roles in creating such peculiar pattern [22,23]. While in case of high laser intensity, V-like structure still is observed in two-electron momentum spectra [5], however later study illustrated that the origin of V-like shape is the asymmetric energy sharing process after recollision moment, neither the nuclear Coulombic attraction nor the final state e-e repulsion has responsibility for this structure [24].

As a central role in NSDI, the responsibility of the final e-e repulsion in forming the drift CTEMMD has been revisited several times for more detailed investigation for atom [22,23,25] and molecule [26]. These studies are based on the scrutiny of several physical quantities such as the transverse momenta and ionization moment using overall double ionization (DI) signals while totally neglect the microscopic mechanisms governing the final DI events. Meanwhile, the modern advancement in laser technology has paved an effective way to well control the evolution of the first ionized electrons. One can adjust either the phase different or the intensity of orthogonal two-color laser pulses [27], use two-color circularly polarized laser [28,29], consider near single-cycle laser [30], mid-IR [31] laser, as well as spatially inhomogeneous pulses [32] to control all vital characteristics of the first ionized electron including its returning energy and the momentum of recolliding with its parent ion. According to simple man model, these quantities are essential to resolve the ionization channel from which the DI occurs. Furthermore, previous studies mostly classified the DI mechanism into two categories [30,33,34]: the first mechanism is direct ionization in which the first ionized electron has appropriate returning energy to directly liberate the bounded one, the second channel is recollision-induced excitation
with subsequent ionization (RESI) where the first ionized electron just has adequate returning energy to excite the bounded one while keeping itself freely. Besides there are still other two possibilities have to be taken into account since they are equally essential to the two former mechanisms and can be observed experimentally: the exchange-state ionization (ESI) [34] and the recollision-doubly induced excitation with subsequent ionization (RDESI) including the existence of the doubly excited state [35]. In addition, the critical condition for such classification is mainly based on the delay time defined as the time difference (TD) between recollision and second ionization events [36] which is really a coarse approximation and somehow contrary to the spirit of classical approach based mainly on the energies of the system.

In this study, we propose an alternative method to clarify the DI mechanisms based on the energies of two electrons just after the recollision process. This clarification contains real physical issues as well as provides more intuitive picture to the DI mechanisms. Then the role of final e-e repulsion is deeply analyzed with respect to each DI mechanisms based on the ionization TD between two electrons after the occurrence of recollision. The goal of this analysis is to provide a comprehensive scenario of the final e-e repulsion and its contribution associating with individual DI mechanism for the purpose of experimentally controlling the e-e correlation pattern via the manipulation of returning electron.

2. Method

In this study the classical three-dimensional ensemble model which widely used in exploring the dynamics in strong-field NSDI [37-39] is employed. Here, the motion of two-electron system is determined by Newton’s equation (atomic units are used throughout the paper until stated otherwise)

\[ \frac{d^2 \mathbf{r}_i}{dt^2} = -\nabla \left[ V_{ne}(\mathbf{r}_i) + V_{ee}(\mathbf{r}_i, \mathbf{r}_j) \right] - \mathbf{E}(t), \]  

where subscript \( i \) is the label of the two electrons, \( \mathbf{r}_i \) is position of \( i \)-th electron and \( E(t) \) is the electric field. In our calculation, we use the linearly polarized laser along \( x \) axis having a trapezoidal pulse shape including ten optical cycles, in which two-cycle turn on, six-cycle at full strength, and two-cycle turn off. The peak intensities chosen are \( I = 3.5 \times 10^{14} \text{ W/cm}^2 \) and \( I = 4.5 \times 10^{14} \text{ W/cm}^2 \) for the purpose of providing the maximum value of returning energy of the first ionized electron just and well above the second ionization potential, respectively. Here \( V_{ne}(\mathbf{r}_i) = -2/\sqrt{\mathbf{r}_i^2 + a} \) and \( V_{ee}(\mathbf{r}_i, \mathbf{r}_j) = 1/\sqrt{(\mathbf{r}_i - \mathbf{r}_j)^2 + b} \) are the electron-ion attractive and the e-e repulsive potentials, respectively. We note that in the classical model, there exists a possibility that one electron can drop deep into the Coulomb well and transfer enough energy to ionize the second, this is the autoionization process. To prevent autoionization of Coulomb interaction, we consider the soft-core Coulomb potential, in which the parameter \( a \) in principle can take any value from \( a_{\text{min}} = 0.476 \) to \( a_{\text{max}} = 0.8 \) for the
ground state of helium atom [40]. In this paper, we choose $a = 0.75$ for the sake of consistency to previous study [24,25]. In addition, to avoid the singularity in $e-e$ repulsive potential, again the softening parameter $b$ has to be used. The higher $b$ is (for instance $b = 1$), the lower DI yield occurs due to weaker recollisions. Hence appropriate value of $b$ should be empirical chosen as 0.01 [24,25].

To solve equation (1), the fourth-order Runge-Kutta method [41] is considered for individual atom in the ensemble. Note that in classical model, the electron is set to continuum solely via over-the-barrier ionization mechanism, the tunneling ionization does not exist. Thus the systems are characterized only by ionization energy. To obtain the initial conditions, the ensemble is set to populate from a classical allowed position for helium ground-state energy of $-2.9035$ a.u. The available kinetic energy is randomly distributed between two electrons in momentum space, means each electron in an atom has arbitrary velocity. Then the electrons are allowed to evolve in sufficiently long time (200 a.u.) in the absence of the laser field to attain the stable positions and momentum distributions, this is the demanded initial values [24]. The DI event is determined only if both electrons have positive energies including the kinetic, the ion-electron and half $e-e$ potential energies at the end of the pulse. We also note that by using classical method in combination to back trajectory analysis technique, all crucial quantities associating with individual electron such as its energy, the recolliding moment, the moment of ionization can be traced at each time step. Thus the microscopically physical process can be straightforwardly investigated and understandable.

In other to survey the role of Coulomb repulsion in CTEMD, an additional treatment is scrutinized in which the long-range Coulomb potential $V_{ee}(r_i, r_j) = 1/\sqrt{(r_i - r_j)^2 + b}$ is suddenly replaced by Yukawa screening potential

$$V_{ee}'(r_i, r_j) = \left(1/\sqrt{(r_i - r_j)^2 + b}\right) \exp\left(-\lambda\sqrt{(r_i - r_j)^2 + b}\right)$$

where the screening parameter is chosen as $\lambda = 5$ to sufficiently cut off the final repulsion between two ionized electrons [22, 24].

3. Results and discussion

Figure 1 displays the CTEMD along the polarization axis of laser field at two representative intensities $I = 3.5 \times 10^{14}$ W/cm$^2$ and $I = 4.5 \times 10^{14}$ W/cm$^2$ in the left and right columns, respectively. The upper panels represent for the situation where the Coulomb potential is maintained after two electrons ionized, while the results when blocking final $e-e$ repulsion by using Yukawa potential are shown in the lower panels. Let firstly discuss the original results in the upper panels. In case of $I = 3.5 \times 10^{14}$ W/cm$^2$ as shown in figure 1a, the spectrum exhibits a strong correlated pattern with the signal clustering around the main diagonal $p_{1x} = p_{2x}$. This result indicates the side-by-side DI in which two electrons escape the parent ion with similar velocity either parallel or opposite to the direction of electric field. As the laser intensity increases to
\[ I = 4.5 \times 10^{14} \text{ W/cm}^2 \] in figure 1b, although the DI events still concentrate on the first and third quadrants implicating the side-by-side DI, the spectrum exhibits strong spreading out of the main diagonal as V-like structure which was observed experimentally [5,21]. Now the Yukawa potential is immediately turned on upon the ionization of the second electron. The lower panels display the gradual shrinking of the DI signals into the main diagonal at both laser intensities as in figure 1c and 1d. In addition, the concentration in case of \( I = 4.5 \times 10^{14} \text{ W/cm}^2 \) is much significant than that of \( I = 3.5 \times 10^{14} \text{ W/cm}^2 \) implicating the stronger \( e-e \) repulsive effect for higher laser intensity. We again note that the root of such V-like structure is assigned to the final repulsive interaction between two ionized electrons and asymmetric energy sharing upon recollision process in cases of low [22,23] and high laser intensity [24], respectively.

Figure 1. Correlated two-electron momentum distributions along the laser polarization axis at two representative intensities \( I = 3.5 \times 10^{14} \text{ W/cm}^2 \) and \( I = 4.5 \times 10^{14} \text{ W/cm}^2 \) in left and right panels, respectively. In the upper panels (figures 1a and 1b) the Coulomb potential is maintained while in the lower panels (figures 1c and 1d) the Yukawa is used to eliminate the final repulsive interaction between two ionized electrons.

The main purpose of this study is to deeply understand the physics beneath the macroscopic behavior of DI spectrum in figure 1 in terms of the role of final \( e-e \) repulsion tightly associating with each microscopic DI mechanism. Here, the recollision moment is defined as the instant when the distance between the first ionized electron and its parent ion approaches to minimum. For the sake of avoiding confusion, in the following the first ionized electron which is driven to recolliding with its parent ion and the second electron ionized by this recollision are simply referred to first and second electrons, respectively. We now classify four possible mechanisms leading to DI in term of the energies of two electrons. Such criteria using in this study is more accurate than previous classification based on the delay time [30,33,36], it also
provides more intuitive picture for understanding the DI mechanisms. We note that which of them being dominant over the others strongly depends on the returning energy of the first electron. Figure 2a and 2b exhibit the returning energy of the first electron driven by lasers whose intensities are \( I = 3.5 \times 10^{14} \text{W/cm}^2 \) and \( I = 4.5 \times 10^{14} \text{W/cm}^2 \), respectively. The dashed vertical blue lines represent for the second ionization potential of helium (means the required energy to ionize the second electron from the He\(^+\) ion which is equal to 54.42 eV). In classical model, the returning energy of first electron exposed to a laser field extends from 0 to 3.17\(U_p\) [1], where \( U_p = I_p / 4\omega^2 \) is the ponderomotive energy of a classical charge particle moving in a monochromatic field with \( I_p \) and \( \omega \) are the ionization potential and laser frequency, respectively. It is obvious that the possibly maximum returning energies are just and well higher than the second ionization potential for lower and higher laser intensity, respectively. We then use the trajectory analysis for DI events at lower intensity of \( I = 3.5 \times 10^{14} \text{W/cm}^2 \) to illustrate four possible DI mechanisms. The first one is direct ionization and shown in figure 2c in which the first electron has enough returning energy to directly promote the second one into the continuum via electron impact ionization [30]. The second mechanism indicated in figure 2d is RESI. Here the first electron still remains in the continuum after recollision and shares part of its energy to excite the second one following by the ionization of this second electron after a delay time. The third mechanism is ESI shown in figure 2e. These events occur when the first electron knocks out the second one with the cost of being recaptured by the parent ion upon recollision process to populate in excited state for a while before subsequent ionization. The fourth mechanism is RDESI, where the doubly excited state is formed due to the sharing energy between two electrons [35]. In our calculation for \( I = 3.5 \times 10^{14} \text{W/cm}^2 \), about 9.23%, 41.03%, 24.16%, and 25.58% DI events associate to direct, RESI, ESI, and RDESI, respectively. While for \( I = 4.5 \times 10^{14} \text{W/cm}^2 \), these respective percentages are 18.36%, 43%, 18.61%, and 20.03%. The emergence of direct ionization as the laser intensity increases well supports our expectation due to more electrons having greater returning energy than the second ionization potential as seen in figures 2a and 2b.
Figure 2. Returning energy spectra of first electrons at two laser intensities as in figure 1 for the top left and top right panels, respectively. The dashed blue lines represent for the second ionization potential. The evolution of the energies of two electrons during the interaction with laser field representing for four DI mechanisms: (c) direct ionization, (d) recollision-induced excitation with subsequent ionization, (e) exchanging-state ionization and (f) recollision-doubly induced excitation with subsequent ionization.

Having the comprehensive demonstration for all DI mechanisms, we proceed to deeply investigate the role of final state e-e repulsion associating with each mechanism to understand the microscopic distribution of this effect to entire DI events shown in figure 1. In figure 3, the distributions of ionization TD of two electrons are shown in the top row corresponding to four DI mechanisms mentioned in figure 2. The CTEMs are presented for long-range Coulomb (middle row) and Yukawa potentials (bottom row) associating with each DI mechanism to those in first row. The laser intensity in this case is \( I = 3.5 \times 10^{14} \) W/cm\(^2\). Obviously, the TDs in cases of direct ionization and RDESI peak at almost 0 indicating that two electrons escape from the parent ion simultaneously. Thus one expects that the repulsive force between them has significant magnitude to further accelerate one and decelerate the other resulting in DI signals which spread out of the main diagonal. This fact indeed can be observed from the evolution from figures 3e and 3h to figures 3i and 3l, respectively. When taking over from Coulomb potential to Yukawa one for neglecting the final repulsion, the DI
signals tend to concentrate towards the main diagonal. Note that this tendency is not significant for direct ionization as for RDESI simply due to the lack of signals. Meanwhile for the ESI and RESI, the TDs concentrate at $0.25T$ with $T$ is the optical period of the laser field. This difference in time is sufficiently large so that the first ionized electron driven by the laser field has adequate distance from the second one. Thus these two electrons feel quite weak repulsive interaction. This straightforwardly makes clear of the invariant behavior of the spectra associating with ESI and RESI mechanisms (see pairs of figures 3f – 3j, figures 3g – 3k) when neglecting the final repulsive effect.

![Figure 3](image)

Figure 3. The distribution of ionization time difference of two electrons (top row). The correlated two-electron momentum distributions, in which the interacting potential between two electrons after recollision process is Coulomb potential (middle row) and Yukawa potential (bottom row) for four mechanisms at laser intensity $I = 3.5 \times 10^{14} \text{W/cm}^2$.

Analogous investigation is performed for higher laser intensity of $I = 4.5 \times 10^{14} \text{W/cm}^2$. Similar results also can be drawn in cases of direct ionization, RESI, and RDESI mechanisms. The final repulsive effect is vital for direct ionization and RDESI due to the simultaneous ionization of two electrons while plays no role for RESI by virtue of large ionization lag time. An addition interesting feature can be observed in this situation is the emergence of the contribution of repulsive effect in case of ESI mechanism as laser intensity grows. This fact is straightforwardly understandable.
since the higher laser intensity is, the more electrons with higher returning energy appear. Then after kicking out the second electron, it is recaptured by parent ion but in higher excited state due to its high remaining energy. Thus it takes extremely short time for a large portion of first electrons to stay in the excited state before being ionized almost simultaneously with the second one. This results in a sharp and large peak of TD around 0 in figure 4c. We come to the conclusion that for weak or moderate laser intensity, the final e-e repulsive effect is vital in forming the off-diagonal DI signals such as experiment observed V-like structure [5,21]. Our investigations also unambiguously indicate the main distribution of the repulsive effect in cases of direct ionization, RDESI, and the rise of repulsive effect for ESI as the laser intensity increases. Note that for higher laser intensity such as around $10^{15}$ W/cm$^2$, the final repulsive effect plays no responsibility in the appearance of V-like structure; instead the asymmetric energy sharing is the dominantly physical mechanism [24].

Figure 4. Same as figure 3 but at laser intensity $I = 4.5 \times 10^{14}$ W/cm$^2$.

4. Conclusion

In conclusion, we have proposed an alternative classification for DI mechanisms directly according to the TD between two ionization moments of the ionized electrons succeeding the recollision process. Such classification well suits the framework of classical model which is mainly characterized by the system energy. Four adequate DI mechanisms are unambiguously illustrated including direct ionization, RESI, ESI, and
RDESI at two representative laser intensities of $I = 3.5 \times 10^{14}$ W/cm$^2$ and $I = 4.5 \times 10^{14}$ W/cm$^2$ corresponding to the situations where the maximum value of returning energies are just and well above the second ionization potential, respectively. We also thoroughly investigate the role of final $e$-$e$ repulsive effect with respect to each DI mechanism to microscopically understand the physics governing the drift CTEMĐ. Our calculation shows that for low and moderate laser intensities, the repulsive effect has vital distribution in cases of direct and RDESI due to simultaneous liberation of two ionized electrons while plays no role for RESI mechanism. In the situation where the DI occurs via ESI, the contribution of final $e$-$e$ repulsive effect emerges as the laser intensity increases.

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