Mechanisms of nonsequential double ionization process of argon by near-single cycle laser pulse

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Abstract. In this paper, the near-single cycle laser pulse is used to trigger the nonsequential double ionization process of argon atom due to its ability to eliminate the contamination from secondary recollisions. The mechanisms and distributions of recollision-ionization channels are thoroughly investigated for two representative laser intensities which provide returning energies well below and above the recollision-ionization threshold of argon. The results indicate that recollision-induced excitation with subsequent ionization and direct ionization mechanisms are dominant for low and high intensity of laser, respectively. In addition, the extraction of cross-shaped structure in the correlated two-electron momentum distribution is presented along with the investigation to understand the origin of this interesting structure.

1. Introduction

In recent decades, the physics of nonlinear processes of atoms and molecules induced by ultrashort intense laser pulses has been drawn many interests of atomic, molecular and optical physicists. The intense laser pulses play an important role to provoke new physical phenomena of current interests such as high-order harmonic generation [1, 2] above-threshold ionization [3], nonsequential double or multiple ionization [4], and the generation of attosecond pulse [5]. These phenomena are the steady bricks for fulfilling the understanding of atomic and molecular structures. Among them, the nonsequential double ionization (NSDI) process is unsuspicious to be the strongest mean for the understanding of the electron-electron ($e$-$e$) correlation dynamics in the atomic or molecular orbitals toward the recollision process [6]. The most well-known and intuitive picture for understanding NSDI is the so-called simple-man theory [7]: one electron is firstly tunneling ionized and accelerated by the laser field, then driven back to revisit its parent ion as the direction of laser field reverses, the second electron is then liberated toward recollision process. The NSDI can occur toward several recollision-ionization channels mostly depending on the energy of returning electrons. In the case of high returning energy, the direct NSDI called ($e$,2$e$) in which the second electron is directly knocked out dominates. Other channels are opened for sufficiently low returning energy: the returning electron has just enough energy to excite the second one, and keep free itself [8] or even be recaptured and form a transition doubly excited state [9]; subsequently they are released one after another. The excitation-ionization channels without and with the formation of doubly excitation states are referred to recollision-excitation with subsequent ionization (RESI) and recollision-doubly-excitation with subsequent ionization
(RDESI), respectively. The intuitive picture for comprehensively understanding the microscopic
dynamics and distributions of these recollision-ionization channels to the NSDI is still vague
due to the contamination of secondary recollisions to the primary one [10]. One of the most
efficient methods to eliminate the contagion of unexpected recollisions to the final NSDI is the
usage of near-single cycle (NSC) intense laser pulse firstly made by Bergues et al. in 2012 [11].
As presented in Refs [11, 12], the correlated two-electron momentum distribution (CTEMD)
induced by NSC laser pulse exhibits an interesting cross-shaped structure which has not been
experimentally observed previously. Note that such structure was theoretically predicted in 2010
by using S-matrix theory for the exploration of the sub-cycle dependence of electron correlation
spectra in which the double-ionization dynamics were confined to a single cycle of a laser pulse
[13]. The root of this peculiar structure has not been comprehensively explained. Hence this
is deserved to provide investigation of the microscopic mechanism leading to NSDI induced by
NSC laser pulse.

In this paper, the classical ensemble model proposed by Eberly et al. in 2001 [14] is utilized
due to the fact that the ionized electrons are evolved solely under the influence of the laser
field. The advantages of the classical approach over the solutions of time-dependent Schrödinger
equation were stated elsewhere [15]. The NSC laser pulse used in our calculation has sin-squared
envelope and 750-nm wavelength in consistence with Ref [11]. The peak intensities of the laser pulse are
chosen as $0.8 \times 10^{14}$W/cm$^2$ and $2.5 \times 10^{14}$W/cm$^2$
that enable to produce the returning electrons whose energies are well below and above the
recollision-ionization threshold of argon, respectively. The recollision-ionization dynamics are
analyzed using back trajectory analysis method to evaluate the impact of the ionization channels
on NSDI for different laser intensities. All intuitive pictures depicting the microscopic dynamics
beneath the NSDI process of argon are thoroughly discussed. The results indicate that for low
intensity laser pulse, the RDESI is dominant. The influence of RDESI reduces while the RESI
and direct ionization emerge as the laser intensity increases. In addition, we figure out that the
cross-shaped structure in CTEMP solely corresponds to a specific range of rescattering angle
defined as the angle between the momenta of returning electron before and after recollision with
its parent ion. This fact coincides with the experiment data in Ref [11]. Moreover, it is the
$(e,2e)$ mechanism that takes responsibility for forming such interesting structure in CTEMP.

The paper is organized as follows. In section 2, the classical ensemble model used to study the
NSDI process is briefly presented. More details of this model can be referred to our preceding
paper [16]. In section 3, we present and discuss the numerical results for the recollision-ionization
channels primarily determining the NSDI process of argon for two representative values of laser
intensity. Section 4 concludes the paper.

2. Three-dimensional classical ensemble model
From being developed by Eberly and coworkers in 2001 [14], the classical ensemble model has
been widely used for investigating the NSDI process qualitatively and quantitatively. This
approach provides a computational efficient and physically intuitive method to understand
insight into the NSDI process. In the classical model, the evolution of the two-electron system is
determined by the explicitly classical equations of motion (unless otherwise stated, atomic units
are used throughout this paper)

$$\frac{d^2 \mathbf{r}_i}{dt^2} = -\nabla \left( \frac{2}{\sqrt{r_i^2 + a^2}} + \frac{1}{\sqrt{r_{12}^2 + b^2}} \right) - \mathbf{E}(t),$$

where index $i = 1, 2$ stands for two electrons, $a$ and $b$ are the softening parameters for nuclear-
electron and electron-electron interacting potential and are chosen as 1.5 and 0.1, respectively,
in order to avoid autoionization [10]. Here $r_i$ and $r_{12}$ are the distances between each electron to
the parent ion and between two electrons, respectively and $\vec{E}(t)$ is the electric field polarizing along $x$ direction and has a sin-squared envelope of the form

$$\vec{E}(t) = E_0 \sin^2 \left(\frac{\pi t}{\tau}\right) \sin (\omega t + \varphi) \hat{i},$$

in which $\varphi$ is carrier envelope phase (CEP) and $\tau = 0.5NT_0$ is the pulse duration defined as the FWHM (Full Width at Half Maximum) of the full sin-squared pulse. Here the full length of the pulse is $NT_0$ with $T_0$ is the optical cycle and $N = 4$ in our consideration. To simulate the experiment data in [11], for each atom in the ensemble, the CEP is randomly chosen in range of 0 to $2\pi$. The wavelength of considered laser fields is 750 nm, the peak intensities are chosen as $0.8 \times 10^{14} \text{W/cm}^2$ and $2.5 \times 10^{14} \text{W/cm}^2$.

Equation (1) is straightforwardly solved using the four-ordered Runge-Kutta method [17]. To obtain the initial condition, the ensemble is populated starting from a classically allowed position for the argon ground-state energy of -1.59 a.u. The available kinetic energy is distributed between two electrons randomly in momentum space. Then the electrons are allowed to evolve a sufficiently long time (200 a.u.) in the absence of the laser field to obtain stable position clustering around the core and stable momentum distribution [16, 18]. The positions and momenta of the two electrons are able to be traced at each time step during the laser pulse. The atoms are considered to have NSDI only if the energies of both electrons are positive at the end of the pulse. Note that we can obtain both quantities that can be experimentally observed and purely theoretical such as the double ionization probability or end-of-pulse CTEMP, and the released moment of each electron or the recollision moment, respectively. These theoretical quantities are vital for deeper understanding of the physics beneath the whole process. We also note that in the classical framework, only over-the-barrier mechanism of ionization can be considered. For obtaining stable and statistically reliable results, we use ensemble sizes as millions of atoms.

3. Numerical results and discussion

We proceed to discuss the NSDI process of argon induced by NSC laser pulses. Firstly, the CTEMDs along the polarization axis of laser fields are presented in figure 1 for two representative laser intensities. The left and right panels correspond to laser intensities of $0.8 \times 10^{14} \text{W/cm}^2$ and $2.5 \times 10^{14} \text{W/cm}^2$, respectively. The results specify that the CTEMDs robustly depend on the laser intensity. In the situation of low laser intensity, the spectrum exhibits a strong anticorrelated pattern with the signals clustering along the secondary diagonal $p_1x = -p_2x$ as shown in figure 1a. This fact indicates that two ionized electrons obtain equivalent momenta with opposite directions, and this is consistent with the previous result [19]. Meanwhile for the case of high intensity in figure 1b, the CTEMD exhibits an interesting feature which is neither correlated nor anticorrelated behavior. The double ionization events mainly locate in the first and third quadrants and form somehow V-like structure as observed in the case of helium [6] or the intriguing cross-shaped structure observed experimentally in Ref [11]. This structure implies that there exists a large deviation in momenta of ionized electrons as they are liberated from parent ions and driven by the laser pulse. Note that our result for $2.5 \times 10^{14} \text{W/cm}^2$ does not perfectly mimic the cross-shaped structure in the experiment [11]. The extraction of cross-shaped structure and its origin are discussed in the next section. We also note that the first and second ionized electrons are referred to returning and bounded electrons, respectively.

One of the most important quantities determining the mechanism of NSDI is the energy of returning electron. Classically, this energy can be in range from zero to well-known cut-off value $3.17U_p$ [7] where $U_p = I/4\omega^2$ is the ponderomotive energy of a classical charged particle moving in a monochromatic field with $I$ and $\omega$ are the intensity and angular frequency of laser field, respectively. It is obvious that the returning energy governing the mechanism of NSDI process strongly depends on the laser intensity. In figure 2, the distribution of returning energy
Figure 1. (color online) Correlated electron momentum distributions along the polarization axis of laser fields for two representative intensities $0.8 \times 10^{14}$ W/cm$^2$ (a) and $2.5 \times 10^{14}$ W/cm$^2$ (b).

of the first ionized electron just before the recollision moment is presented for two representative laser intensities as in figure 1. The energy spectra exhibit clear cut-off behaviors which are well consistent with the classical approximation of 13.5 eV and 42 eV for $I = 0.8 \times 10^{14}$ W/cm$^2$ (figure 2a) and $I = 2.5 \times 10^{14}$ W/cm$^2$ (figure 2b), respectively. These results confirm the reliability of our calculations. We also indicate in figure 2 the recollision-ionization threshold energy of argon as the dash-dotted vertical green line. Obviously, the returning energy in case of $I = 0.8 \times 10^{14}$ W/cm$^2$ is much lower than the threshold, thus there is only RDESI possible. We also show the resonant energy of 13.48 eV which is associated with the required energy to excite electron from the ground to the first excited state of $\text{Ar}^+$ as the dashed blue vertical line. That is a firm evidence of RDESI process through the first excited state of the second electron. While for high intensity laser field, the energy spectrum extends to higher cut-off value of about 42 eV which is much larger than the threshold, hence both RESI and direct ionization mechanisms are important. They compete to each other to be the key dynamic determining the NSDI process.

We also use back trajectory analysis to trace the energy evolutions of both ionized electrons from the turn-on to the end of the laser pulse. Figure 2c corresponds to the case of $I = 0.8 \times 10^{14}$ W/cm$^2$. Figure 2d and 2e associated with the laser intensity $I = 2.5 \times 10^{14}$ W/cm$^2$ correspond to two different mechanisms in which the energies of the returning electrons just after recollision are negative and positive, respectively. These results are thoroughly and unsuspiciously demonstrate all possible mechanisms leading to NSDI process for argon exposed to NSC laser pulse. In case of low intensity, after recollision the returning electron (solid red curve) leaves its energy to the bounded electron and is captured by the parent ion as expected. However this amount of energy is not enough to kick out the bounded electron (dashed blue curve) but is sufficient to make the transition of this electron from the ground to the first excited state. In addition, it is obvious that two electrons have equivalent energies; this is the distinct signature of the existence of doubly excited state [9] and RDESI (see figure 2c). For high laser intensity, the NSDI through RESI channel is taken over from two other different mechanisms which interplay to affect the drift CTEMD. One possibility is the exchange of the statuses of the returning and bounded electrons, means the returning electron kicks out the bounded one at the expense of being captured by parent ion (figure 2d). Note that the state at which the returning electron temporarily settles before subsequently being ionized is not ground but an
excited state; this is the RESI mechanism. Such recollision-ionization channel is popular for moderate intensity but rare for high intensity of laser. Another situation for high laser intensity is the direct recollision-ionization through \((e,2e)\) mechanism. Here the returning electron has enough energy to directly liberate the bounded one while keeping free itself (figure 2e).

To reconfirm our investigation, we proceed to analyze the time delay between recollision and DI moments as shown in figure 3. Where the recollision time is defined as the instant of closest approach to the core after the ionization of the first electron [14] and DI time is defined as soon as the energies of two electrons achieve positive values [18]. For low laser intensity, the time delay peaks at around \(0.5T_0\) and \(T_0\) with \(T_0\) equals to one cycle of laser. In this case, the doubly excited state is formed upon recollision, then two electrons enter the continuum one after another with the time difference around \(0.5T_0\) (see figure 2c), thus create the anticorrelated pattern in drift CTEMP (see figure 1a). For high laser intensity, the time delay only extends to \(0.5T_0\) with the peaks separating by around \(0.2T_0\). By back trajectory analysis, we figure out that the first two peaks are associating with the \((e,2e)\) mechanism, while the last peak corresponds to RESI mechanism. The presented feature supports our expectation of the emergence of \((e,2e)\) mechanism as the laser intensity increases.

![Figure 2](image-url)

**Figure 2.** (color online) Upper panels are returning energy spectra of first ionized electrons for two representative laser intensities: \(I = 0.8 \times 10^{14} \text{W/cm}^2\) (a) and \(I = 2.5 \times 10^{14} \text{W/cm}^2\) (b). In which, the dashed-dotted vertical green line represents for the recollision-ionization threshold of \(\text{Ar}\) (27.63 eV), the dashed blue vertical line in left panel shows the required energy to excite the bounded electron from ground to the first excited state of \(\text{Ar}^+\) (13.48 eV). Lower panels are the evolution of the energies of the returning (solid red line) and bounded (dashed blue line) electrons during the interaction with laser field for two representative laser intensity: \(I = 0.8 \times 10^{14} \text{W/cm}^2\) (c) and \(I = 2.5 \times 10^{14} \text{W/cm}^2\) (d,e).

In the following, the cross-shaped structure is extracted from the total CTEMD signals. Firstly, the distribution of \(\text{Ar}^{2+}\) yield as a function of scattering angle \(\beta\) is portrayed in figure...
4a. Figure 4a shows that the angular distribution peaks around 20° regardless the intensity of the laser field. This result is completely consistent with the semiclassical consideration in [11]. The behavior of the angular distribution here is interesting and we believe that it is the intrinsic property of the inelastic scattering between two charged particles. This behavior is still an open problem due to its complexity since the first ionized electron may or may not be recaptured by its parent ion upon recollision. A deep investigation is going to be done in our next project for comprehensively understanding the nature of inelastic scattering between two charged particles in the scenario of NSDI process. We then apply an addition condition for considering CTEMD associating with the scattering angle $\beta$. The filtered CTEMDs are plotted in the lower panels of figure 4 for $I = 2.5 \times 10^{14}$W/cm$^2$ for four representative $\beta$ around 10°, 20°, 30°, 40°. It is now transparently seen that the cross-shaped structure observed in the experiment of [11] can be extracted from total CTEMD for specific condition of $\beta \approx 20°$. For other scattering angles $\beta$, the NSDI event is lower than that of $\beta \approx 20°$ and the filtered CTEMDs exhibit totally different structures. Thus we expect that the experimental data of cross-shaped structure in [11] was measured for the highest angular distribution of NSDI events due to $\beta \approx 20°$.

![Figure 3](image.png)

**Figure 3.** (color online) The distribution of delay time between recollision and DI moments for two representative laser intensities $I = 0.8 \times 10^{14}$W/cm$^2$ (solid red curve) and $I = 2.5 \times 10^{14}$W/cm$^2$ (dashed blue curve).

We proceed to understand the physics governing the cross-shaped structure in CTEMD. We note that a similar structure for helium atom known as finger-like shape was also presented both experimentally [6] and theoretically [18, 20, 21]. The root of finger-like structure is electron-ion attraction, final electron-electron repulsion [20, 21] or asymmetric energy sharing [18] for low or high laser intensity, respectively. However, the finger-like structure in case of helium atom was induced by regular laser field consisting of many cycles which is totally different from the situation considered in this study. In case of argon atom induced by NSC laser field, we figure out that neither of these mechanisms corresponds to the cross-shaped structure. We then classify the NSDI events from the filtered CTEMD for $\beta = 20°$ in figure 4c into two primary recollision-ionization channels: $(e,2e)$ and RESI. Obviously, the cross-shaped structure only retains in the case of $(e,2e)$ mechanism and is faded for RESI situation. The analyses indicate that the combination of the appropriate scattering angle $\beta$ and $(e,2e)$ mechanism is the origin of the cross-shaped structure.
Figure 4. (color online) (a) The scattering angle distribution for two representative laser intensities $I = 0.8 \times 10^{14}$ W/cm$^2$ and $I = 2.5 \times 10^{14}$ W/cm$^2$. Filtered CTEMDs along the polarization axis of laser field for four representative scattering angles $\beta = 10^0$, $20^0$, $30^0$, $40^0$ for laser intensity $I = 2.5 \times 10^{14}$ W/cm$^2$ (b-e).

Figure 5. (color online) Correlated electron momentum distributions along the polarization axis of laser field with the scattering angle $\beta = 20^0$ for $I = 2.5 \times 10^{14}$ W/cm$^2$ in two recollision-ionization channels: $(e,2e)$ (a) and RESI channels (b).
4. Conclusion
In this paper, by using the three-dimensional classical ensemble model, we have provided deep insight into the evolution of the electrons in laser field as well as the physical mechanisms essentially contributing to NSDI process. The results show that RDESI is dominant for low laser intensity, while RESI and $(e,2e)$ emerge as laser intensity increases. We also focus on extracting the cross-shaped structure observed in [11] and figure out that the combination of $(e,2e)$ mechanism and scattering angle $\beta = 20^\circ$ is the root of such interesting feature. This finding simplifies the study of inelastic scattering between two charged particles in the framework of NSDI process since there is no recapture required for consideration. A comprehensive study of this issue is postponed to our next project.

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