Investigation of the dependence of multiple recollision on laser wavelength in the nonsequential double ionization process

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Abstract. This paper investigates the recollision dynamics of the nonsequential double ionization process induced by linearly polarized laser pulses with the three-dimensional classical ensemble model. The results show that the correlated two-electron momentum distribution is contaminated by the double recollision events for sufficiently short laser wavelength. When the laser wavelength increases from near-infrared to mid-infrared, the single-recollision events are more prominent than the double-recollision one. Moreover, the mechanisms governing the nonsequential double ionization process are also thoroughly studied in the case of double-recollision.

1. Introduction
Over the past three decades, laser technology development helped scientists create extremely short laser pulses of high intensity [1, 2]. The interaction between these laser pulses with atoms and molecules has resulted in a series of nonlinear effects such as high order harmonic generation [3, 4] (HHG), above-threshold ionization (ATI) [5, 6], nonsequential double ionization (NSDI) [7, 8]. These nonlinear phenomena have been intensively studied since they provide insight into the laser-matter interactions, typically using high-order harmonic waves to obtain information about the inter-nuclear structure [9]; using the momentum spectrum of ionized electrons to extract information about the structure and symmetry of the atom/molecular [10, 11]; using the correlated momentum spectrum of two electrons in NSDI to investigate the interaction of two electrons in the atom/molecular as well as the properties of the three objects process through the recollision between the first ionized electron and the parent ion [12, 13].

The strong-field physics relating to the interaction between atoms/molecules and the external electric field has drawn much attention theoretically and experimentally. In 2007, A. Rudenko experimentally found the “V-like” structure in two-electron correlated momentum spectrum of helium atom when using a laser pulse with a pulse length of 25 fs, wavelength 800 nm, and intensity $1.5 \times 10^{15}$ W/cm² [8]. For relatively low-intensity lasers, theoretical studies indicate that the nuclear interaction [14] or the final electron-electron repulsion [15] is the root of the
“V-like” structure. However, for high-intensity lasers, Yueming Zhou [16] has shown that the origin of the “V-like” structure is the asymmetric energy sharing between the returning and bounded electrons upon the recollision process. In 2012, B. Bergue et al. [13] experimentally observed the cross-shape structure in the correlated two-electron momentum spectrum of argon atom. Especially in this work [13], the authors used a near-single cycle laser pulse. Note that in NSDI, the recollision process can occur many times for an atom or molecule and interfere with the drift information imprinted in the correlated two-electron momentum distribution. Thus the use of a near-single cycle laser pulse enables us to eliminate the contamination from multiple-recollision effects. However, such laser technology is not available worldwide. Thus most of the experimentists use an alternative way to eliminating the multiple recollision is increasing the laser wavelength. For instance, X. Ma investigated the multiple recollisions in the NSDI process of argon atom using 10-cycles trapezoidal laser pulses [12] in 2016 and concluded that as the laser field intensity and wavelength increases, the multiple-recollision events decrease. However, the mechanisms that govern the ionization of electrons during single and second recollisions were not comprehensively discussed in this work. In addition, the use of near-single cycle laser pulse, long wavelength to investigate the multiple-recollision trajectories in NSDI process has not been specifically investigated.

In the framework of the recollision model, the returning energy and dynamics of the ionized electrons in NSDI significantly depend on the wavelength of the laser pulse and exhibit many special effects. For example, it is interesting that the trajectories dominating the NSDI process induced by the mid-infrared laser fields are no longer the short first-returning ones. Besides, longer second-returning trajectories may be more efficient in producing NSDI, although they usually have lower returning energies than the first-returning trajectories [17]. Moreover, when the laser wavelength increases, the contribution of the recollision impact ionization pathway is greatly enhanced [18, 19]. Furthermore, for the correlated two-electron momentum spectrum of xenon atom NSDI induced by the MIR laser pulses, most of the electrons cluster is in the first and third quadrants and form a cross-like structure [20] as in the work of B. Bergues for argon atom [13]. These experimental and theoretical studies have provided a specific physical picture of the NSDI process. However, the physics governing the NSDI in the scenario of multiple collision is still vague and deserves a comprehensive investigation.

In this paper, with a three-dimension classical ensemble model, we investigate the recollision dynamics in the NSDI of argon atom induce by near-single cycle laser pulse for a wide range of wavelength. By tracing the NSDI trajectories, we deeply study the dependence of the single and double recollisions on the laser wavelength, as well as the dynamics of the recollisions process. Moreover, ionization mechanisms also investigate clearly in this paper.

2. Three-dimension classical ensemble model
In this paper, the classical model is used to describe the NSDI process of argon. This model has been successfully used for understanding NSDI in high intensity regime [14, 16, 21] and low intensity [22] since being proposed in 2001 [23]. In this model, the most loosely bounded electron is assumed to release into the continuum by tunneling through the potential barrier created by the superposition of the atomic Coulomb potential and the laser electric field. The motion of two ionization electrons are governed by this Newton’s equation:

$$\frac{d^2 \vec{r}_i}{dt^2} = -\nabla \left( \frac{-2}{\sqrt{r_i^2 + a^2}} + \frac{1}{\sqrt{r_{i2}^2 + b^2}} \right) - \vec{E}(t),$$

in which $i = 1, 2$ is the index of the first and the second electrons. To avoid autoionization, softening parameters $a$ and $b$ are set up at 1.5 and 0.1, respectively [12]. $\vec{E}(t)$ is the electric
field polarizing along the $x$ direction and has a sin-squared envelope of the form

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

where $\tau = NT_0$, $\varphi$ are the pulse duration, carrier-envelope-phase (CEP), respectively, and $T_0 = 2\pi/\omega$. In this paper, we choose $N = 4$ and the CEP is randomly chosen in the range of 0 to $2\pi$. The peak intensity is chosen as $3.0 \times 10^{14}$ W/cm$^2$ and the wavelength of considered laser fields is 750 nm, 1200 nm, and 1600 nm which are available in the experiment [13, 24].

This classical model is based on the Monte Carlo method. We used ensembles as large as millions of atoms to simulate the laser-atom interaction. To solve equation (1), we need the initial conditions for the differential equation, such as the initial position and momentum of two electrons. These initial conditions correspond to the ground state of the atom. For argon, we set the energy of the classical model atom to be the summation of the first and second ionization potentials of argon, i.e., -1.59 a.u. To obtain the initial condition, the electrons are allowed to move a sufficient long time (100 a.u.) in the absence of the laser field to obtain stable position clustering around the core and stable momentum distribution [16, 22]. When the initial condition is determined, we solve equation (1) for each atom independently. After the laser is turned off, we analyze the energy of the two electrons in each atom (including kinetic energy, electron-ion potential and half electron-electron energy). If the energies of both electrons are positive, we say that the atom is doubly ionized.

3. Numerical results and discussion

We proceed to discuss the NSDI process of argon by laser pulses whose parameters are introduced in Section 2. Figure 1 shows the correlated two-electron momentum distribution (CTEMD) along the laser’s polarization axis driven by wavelengths $\lambda = 750$ nm (figures 1a,d,g), $\lambda = 1200$ nm (figures 1b,e,h), and $\lambda = 1600$ nm (figures 1c,f,i) with four-cycle sin-squared-shaped laser pulses. In which the first, second, and third rows show the CTEMD for total, single-recollision, and double-recollision trajectories, respectively. Note that the spectra for all cases were obtained by averaging over different CEPs from 0 to $2\pi$, corresponding to the CEP-unlocked experiments [13]. The results show that the momentum distribution exhibits a prominently correlated behavior when changing wavelength. At the wavelength of $\lambda = 750$ nm, as shown in Fig. 1(a), the spectrum appears a cross-shaped structure, which is in good agreement with the experimental and theoretical results [13, 25]. When the laser wavelength increases, the double ionization (DI) events gradually decrease and momentum distribution exhibits the double line-shaped structure along the main diagonal in the first and third quadrants of the plot, which agrees with the experimental data at low intensity [24]. Moreover, the distribution in the second and fourth quadrants grows, showing that two electrons are liberated from the parent ion with the similar momentum and opposite direction, as shown in Fig. 1(b). For the CTEMD driven by the 1600 nm laser pulse, NSDI events wider distributes, i.e., two electrons ionize with a large velocity at the end of the laser pulse.

By tracing these NSDI trajectories, we figure out that several multiple recollisions trajectories exist, i.e., the first electron returns and collides with the parent ion more than one time and transfers significant energy to the second electron during the recollisions. It is noted that in our calculation, the peak intensity is chosen as $3.0 \times 10^{14}$ W/cm$^2$ and the wavelength of considered laser fields is 750 nm, 1200 nm, and 1600 nm, so only the double recollision is indispensable for the total NSDI events. Figures 1d-1g, 1e-1h and 1f-1i correspond to the momentum spectra for the single-recollision and double-recollision NSDI events in the cases of $\lambda = 750$ nm, $\lambda = 1200$ nm, and $\lambda = 1600$ nm, respectively. Our investigation indicates that as the laser wavelength increases from $\lambda = 750$ nm to $\lambda = 1200$ nm, and $\lambda = 1600$ nm, the double-recollision event significantly decreases from 8.9%, 4.5%, and 2.1%, respectively. This is clear evidence of the advantage of using the long wavelengths to eliminate the secondary recollisions, thus enables of
Figure 1. (color online) The correlated two-electron momentum distribution along the laser’s polarization axis at the different wavelengths: \( \lambda = 750 \text{nm} \) (figures 1a,d,g), \( \lambda = 1200 \text{ nm} \) (figures 1b,e,h), and \( \lambda = 1600 \text{ nm} \) (figures 1c,f,i). In which, the top, middle, and bottom rows show the correlated two-electron momentum distribution for all trajectories, single- and double-recollision trajectories, respectively.

In order to further explore NSDI events, we use the trajectory analysis technique [16] to analyze the traveling time of the single- and double-recollision trajectories for three wavelengths: \( \lambda = 750 \text{ nm} \), \( \lambda = 1200 \text{ nm} \), and \( \lambda = 1600 \text{ nm} \). Figure 2 shows the distribution of the traveling time between the first ionization moment \( t_{SI} \) and the recollision moment with the single-recollision events, and the double-recollision events in the top and below rows, respectively. Here, the recollision time is defined as the moment of the closest approach of the two electrons after the first ionization.
Figure 2. (color online) The traveling time distribution between the first ionization and the recollision moments at the different wavelengths: $\lambda = 750$ nm (a,d), $\lambda = 1200$ nm (b,e), and $\lambda = 1600$ nm (c,f). The solid black curve corresponds to the single-recollision events; the solid red line and dashed blue line correspond to the first recollision and the second recollision in the cases of double-recollision events, respectively.

In the case of $\lambda = 750$ nm, in the single-recollision trajectories, the traveling time focuses around $0.5T_0$, with $T_0$ is the laser period, corresponding to the recollisions occurring just after the electric field of the laser reverses its direction for the first time. When increasing the laser wavelength, the second and third returns emerge. According to the recollision picture, returning electrons recollide with the parent ion with the highest possibility at the first return and the second-highest possibility at the third return [26]. This result also indicates that the DI events gradually decrease when increasing the laser wavelength. Our result is in good consistency with the model in [12]. Meanwhile, for the double-recollision events, the traveling time distribution in the case of $\lambda = 750$ nm mainly locates around $0.5T_0$ for the first recollision, and the second recollision is shifted further $0.5T_0$, corresponding to the first and the second returns, respectively. As the laser wavelength increases, the traveling time spectra shift to the right globally. Thus the two peaks are still apart $0.5T_0$ from each other. Those results show that the returning electron propagates longer in the laser field for a long wavelength and the second recollision mainly occurs in the next return after the first recollision.

Figure 3 displays the time delay distribution between the last recollision and the double ionization moment, in which the solid red and dashed blue curves indicate the time delay spectra for the single-recollision and double-recollision trajectories, respectively. Here, the double ionization time is defined as the moment when the energies (including the kinetic energy, the attractive potential energy of the electron-ion interaction, and half of the electron-electron repulsive potential energy) of the two electrons turn positive and keep positive until the end of the laser pulse [16, 27]. For $\lambda = 750$ nm laser, the first peaks of the distribution for both the single- and double-recollision events coincide, but the second and other peaks for the double-recollision events are lower. While in the cases of longer wavelength, the time delay spectra of the single-recollision and double-recollision events are more alike and shift to cluster around zero. This feature indicates that the double ionization of the single- and double-recollision events occurs more quickly as increasing wavelength.
According to the Simpleman model [28], the electron dynamics in NSDI significantly depend on the returning energy. Hence, we proceed to investigate the returning energy distribution to explore the causes of the differences in NSDI for different laser wavelengths. Figure 4 presents the returning energy in the cases of the single- and double-recollision NSDI events for three representative laser wavelengths in the top and bottom panels, respectively. The dashed green vertical line presents the second ionization potential of argon ($I_{P2} = 27.6$ eV). In the classical model, the returning energy of the first ionized electron induced by the laser field extends from 0 to $3.17 U_P$ [4], where $U_P = I/4 \omega^2$ is the ponderomotive energy, with $I$ and $\omega$ are the intensity and angular frequency of laser field, respectively. We classify the NSDI process into two main mechanisms based on the energy of two electrons after the recollision moment [4, 29]. The first mechanism is direct ionization ($e_2e$), in which the returning electron directly causes the second ionization upon the recollision process. The second mechanism is recollision-induced excitation with subsequent ionization (RESI), where the double ionization occurs after a certain time delay after the recollision [13, 22, 30].

In the case of the single-recollision trajectories, it is clear that most of the returning energy is less than $I_{P2}$ for $\lambda = 750$ nm (see figure 4a). When the laser wavelength increases, the returning energy distribution extends which shows that the ($e_2e$) NSDI mechanism emerges to be dominant. In our calculation, for the single-recollision events, there are about 21.4%, 33.5%, and 42.4% DI events belong to direct ionization for $\lambda = 750$ nm, $\lambda = 1200$ nm, and $\lambda = 1600$ nm, respectively. This result is in good agreement which that presented in Fig. 3, where time delay shifts to zero as the laser wavelength grows.

For the double-recollision events, most of the returning energy in the first recollision is larger than $I_{P2}$, which means the returning electrons have enormous energies and pass through the parent ion so fast [16]. Thus instead of directly leaving the core, the bounded electrons gain enough energies to jump to excited states. Moreover, the energies of the second-recollision events are less than those of the first recollision, which is in good consistency with the scenario proposed by Simpleman’s theory [28]. When increasing the laser wavelength, the returning energies for the first- and the second-recollision events increase, so the returning electron moves so fast through the vicinity of the ion core in the first recollision and effectively transfers a small portion of its energy to the bounded one. Note that in this scenario, after the first recollision, some of the bounded electrons move to excited states. Then at the second recollision, the returning electron transfers its energy more sufficiently in the case the value of its energy is sufficient. Here, for
Figure 4. (color online) The returning energy distributions for single-recollision (a-c) and double-recollision trajectories (d-f). The dashed green line presents the second ionization potential of argon ($I_{P2} = 27.6$ eV).

longer wavelengths (see figures 4e-4f), the energies of returning electrons at the second return are again too large. Thus they pass through the core too fast, and the possibility to have interaction and kick out the bounded electrons decreases. This feature was also discussed in [16] and is in good agreement with the result in Fig. 1.

Figure 5. (color online) The energies of the two electrons during the interaction with the laser field for wavelengths $\lambda = 750$ nm, $\lambda = 1200$ nm, and $\lambda = 1600$ nm in the cases of double-recollision trajectories.

Finally, the evolutions of the energies of two electrons during the interaction with the laser field are detailed investigated and showed in Fig. 5 for $\lambda = 750$ nm, $\lambda = 1200$ nm, and $\lambda = 1600$ nm for the cases of double-recollision NSDI events. Noted that the longer the wavelength is, the larger the first returning energy is. Therefore, for the first recollision, the energy exchange between the returning and bounded electrons decreases when the laser wavelength increases, thus the bounded electron transfer to an intermediate excited state, as shown in Figs. 5a-c. However, for the second recollision, the returning energy increases and creates direct ionization for bounded
electron. In our calculation, about 31%, 41%, and 51% DI events for \((e,2e)\) mechanism in double-recollision trajectories for \(\lambda = 750\) nm, \(\lambda = 1200\) nm, and \(\lambda = 1600\) nm, respectively.

4. Conclusion
In conclusion, we have theoretically investigated the electron dynamics in NSDI of argon atom and the role of double recollision in the two-electron correlated momentum spectrum by a few-cycle linearly polarized laser field with different wavelengths. We found that as the laser wavelength increases, the single-recollision events are more prominent than double-recollision ones. Moreover, the results show that the \((e,2e)\) ionization mechanism dominates in the single-recollision trajectories and the double-recollision trajectories for long laser wavelength. Additionally, several details of double recollision are also clearly analyzed in this paper.

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