Multiple recollisions in strong-field nonsequential double ionization

Xiaomeng Ma,1 Yueming Zhou,1,* and Peixiang Lu1,2,†

1School of Physics, Huazhong University of Science and Technology, Wuhan 430074, People’s Republic of China
2Laboratory of Optical Information Technology, Wuhan Institute of Technology, Wuhan 430205, People’s Republic of China

(Received 10 September 2015; published 28 January 2016)

With the three-dimensional classical ensemble model, we revisited the recollision dynamics in strong-field nonsequential double ionization (NSDI) by linearly polarized laser pulses. Back analysis of the NSDI trajectories shows that a significant part of the double ionization events undergo a multiple-recollision process. Similar to the previously revealed single recollision induced NSDI, the multiple recollisions induced NSDI also occurs through doubly excited states at low laser intensities. Further, we find that the contribution of multiple recollisions in NSDI decreases with the increasing laser intensity and wavelength.

DOI: 10.1103/PhysRevA.93.013425

I. INTRODUCTION

Nonsequential double ionization (NSDI) is a fundamental process in intense laser-atom interactions. It has been a hot topic since the first observation of the enhanced double ionization (DI) yields [1]. Now it is known that the responsible mechanism for NSDI is the recollision process [2,3]. In this picture, the first electron ionized through tunneling is accelerated by the oscillating laser field, and returns back to the parent ion when the electric field changes its direction, leading to the direct ionization or excitation with subsequent ionization of the second electron by an inelastic recollision [4]. Because of the recollision, the two ionized electrons exhibit a highly correlated behavior [5]. During the past decades, strong-field NSDI has drawn a great number of studies [6–21]. The development of the experimental technic [22–27] as well as the theoretical methods [28–40] has provided deep insight into the details of the correlated electron dynamics in NSDI. For example, at high laser intensities, the prominent V-like [41] and crosslike structures [42] in the correlated electron momentum spectra were experimentally observed. Theoretical studies have shown that asymmetric energy sharing between the two electrons during recollision is responsible for these structures [43,44]. At relatively low laser intensities, the role of the final-state electron repulsion and electron-ion attraction for the V-like structure is identified [35,36,45,46]. At lower laser intensities, it has been shown that NSDI occurs through doubly excited states and the decay dynamics of the doubly excited states has been detailedly explored [47].

In the recollision picture, the tunnelled electron driven by the oscillating electric field of the linearly polarized laser pulse can return many times to the parent ion. Because of the diffusion of the electron wave packet, recollision mainly occurs at the first return. But the truth is more complicated. For example, recent studies have shown that in some situations recollision at later returns has the main contribution to the recollision-induced phenomena, such as the photoelectron holography at the low-momentum part [48] and the zeptosecond temporal structure in high harmonic x-ray bursts [49]. Nevertheless, it is believed that usually there is only one effective recollision even if the electron returns to the ion many times. However, recent experiments on above threshold ionization have shown that the multiple recollisions could occur. It is responsible for the low-energy peaks in the photoelectron energy spectrum [50–53]. In NSDI, multiple recollisions are also speculated. For instance, in a previous experiment, it was discussed that multiple recollisions may be responsible for the observed anticorrelated behavior in the correlated electron momentum spectrum and the high-energy cutoff in the sum-energy spectrum of NSDI at the low laser intensity regime [54]. In a very recent experiment on NSDI by a midinfrared laser pulse, several peak structures in the electron momentum spectrum, though very faint, were observed [55]. Those structures are very similar to the multiple-recollision induced low-energy structures in strong-field above threshold ionization [52], and thus they are believed to be the hint of multiple recollisions in NSDI. However, no convincing theoretical study has been performed to demonstrate the multiple recollisions in NSDI.

In this paper, with the three-dimensional (3D) fully classical ensemble model, we revisit the recollision dynamics in NSDI by linearly polarized laser pulses. Back tracing of the NSDI trajectories shows that multiple recollisions indeed have a significant contribution to NSDI. The laser intensity and wavelength dependence of the contribution of multiple recollisions to NSDI indicate that multiple recollisions are more prevalent at lower laser intensities and shorter wavelengths.

II. METHOD

Accurate description of NSDI needs full quantum theory. However, a numerical solution of the time-dependent Schrödinger equation for the multielectron systems in strong laser fields requires a huge computational demand [29,56,57]. In the past decade, numerous studies have resorted to the classical methods [33–38,58–61]. Previous studies have convincingly confirmed that the classical methods are reliable tools in exploring electron dynamics in strong field DI [35,36,43,44,59,61]. The classical methods succeeded not only in explaining the experimental data but also in predicting various phenomena [62–65]. Another important advantage of the classical methods is that the underlying processes can be intuitively revealed by tracing the classical trajectories. Thus, in this paper, we employ the 3D classical ensemble model...
In this classical ensemble model, the evolution of the two-electron system is determined by Newton’s equation of motion (atomic units are used throughout until stated otherwise):

\[
\frac{d^2 \mathbf{r}_i}{dt^2} = -\nabla \left[ V_{ne}(\mathbf{r}_i) + V_{ee}(\mathbf{r}_1, \mathbf{r}_2) \right] - \mathbf{E}(t),
\]

where the subscript \(i\) is the label of the two electrons and \(\mathbf{r}_i\) is the coordinate of the \(i\)th electron, and \(\mathbf{E}(t)\) is the electric field, which is linearly polarized along the \(x\) axis and has a trapezoidal pulse shape with a two-cycle turn on, six cycles at full strength, and a two-cycle turn off. The potential \(V_{ne}(\mathbf{r}_i) = -2/\sqrt{r_i^2 + \alpha^2}\) represents the ion-electron interaction, and the soft parameter \(\alpha = 1.5\) is introduced here to avoid autoionization [35]. \(V_{ee}(\mathbf{r}_1, \mathbf{r}_2) = 1/\sqrt{(\mathbf{r}_1 - \mathbf{r}_2)^2 + b^2}\) is the electron-electron interaction and parameter \(b\) is set to be 0.1.

To obtain the initial conditions for Eq. (1), the ensemble is populated starting from a classically allowed position for the energy of \(-1.59\) a.u., corresponding to the sum of the first and second ionization potentials of Ar. The available kinetic energy is distributed between the two electrons randomly, and the directions of the momentum vectors of both electrons are also randomly assigned. Then, the system is allowed to evolve in the absence of the laser field for a sufficiently long time (100 a.u.) to obtain stable position and momentum distributions, which are the initial conditions for Eq. (1). After that the laser field is turned on. We check the energies of both electrons at the end of the laser pulse, and a DI event is determined if both electrons achieve positive energies. Then we back trace the DI trajectories and find that all of the DI events experienced recollision at least once and thus they are NSDI events. The ensemble size in our calculations is two million which results in tens of thousands of NSDI events when the laser pulse is turned off.

### III. RESULTS AND DISCUSSIONS

Figure 1(a) shows the correlated electron momentum distribution along the laser polarization direction. Here the laser intensity is \(0.8 \times 10^{14}\) W/cm\(^2\) and the wavelength is 780 nm. The distribution exhibits a prominent anticorrelated behavior, i.e., the two electrons emit into the opposite hemispheres. This behavior has been observed in a previous experiment [54]. At this laser intensity the recollision energy of the returning electron is not high enough to ionize the second electron directly. It has been explored that NSDI occurs through doubly excited states [66]. In the classical model, the doubly excited states describe such a picture that the first electron excited the bound electron through the recollision but the recollision electron itself is recaptured, as shown by the classical trajectories in the left column of Fig. 2. The time delay between the final ionizations of the two electrons from the recollision induced doubly excited states determines the correlations of the electrons [66,67]. Figure 1(b) shows the laser phase at the time of final ionization for one electron versus the other, where the final ionization time is defined as the instant when the electron achieves positive energy after recollision (the energy includes the kinetic energy, the ion-electron potential, and half electron-electron energy). In this figure, the events indicated by the green solid circles correspond to the correlated electron distribution and those indicated by the red dashed circles are responsible for the anticorrelated distribution [66,67].

By tracing these NSDI trajectories, we find that there are many multiple-recollision trajectories, where the first electron returns to the parent ion many times and more than once significant energy exchange occurs during the returns. In Figs. 1(c) and 1(d), we separately show the correlated electron momentum distributions for the single-recollision and the multiple-recollision NSDI events, respectively. In our calculations, most of the multiple-recollision trajectories experienced significant energy exchange twice. So we call them double-recollision NSDI. Figures 1(c) and 1(d) show that for the single-recollision events the distribution exhibits a strong anticorrelated behavior while for the double-recollision events it is almost uniform in the four quadrants.

Figure 2 shows four illustrative trajectories to provide an intuitive picture of the recollision process of NSDI. The left column of Fig. 2 displays the energy evolution and the right column shows the time evolution of distance between each electron and parent ion for the corresponding trajectories in the left column. The trajectories in Figs. 2(a) and 2(c) experience only one recollision. The recollision occurs at the first returning for the trajectory in Fig. 2(a) and it occurs at the third returning for the one in Fig. 2(c). The trajectories in Figs. 2(e) and 2(g) undergo two recollisions, where the first recollision occurs at the first returning and the second recollision occurs at the second [Fig. 2(e)] or third [Fig. 2(g)] returning.

In order to understand the dynamics of the double-recollision process more clearly, we trace the classical trajectories and perform statistical analysis. We find out the single
ionization time $t_{SI}$, the recollision time $t_r$, and the double ionization time $t_{DI}$. Here, the single ionization time is defined as the instant when one electron achieves positive energy or is outside the nuclear well [66]. The recollision time is defined as the instant when the returning electron enters the core area so that the distance of the two electrons $r_{12} < 2.0$ a.u. For the double-recollision trajectories, the recollision electron visits this area twice. We record them as the first and second recollisions, respectively. The double ionization time is defined as the instant when both electrons achieve positive energies after recollision. These definitions are also indicated in the illustrative trajectory in Fig. 2(c). Figure 3(a) displays the traveling time distribution of the single-recollision NSDI trajectories, where the traveling time is defined as the time difference between the recollision and the single ionization. It shows that for the single-recollision trajectories, the traveling time distribution exhibits several peaks located at 0.6$T$ ($T$ is the laser period), 1.7$T$, and 2.7$T$, which correspond to the recollisions that occurred at the first, third, and fifth returns, respectively. According to the recollision picture, the recollision electron possesses highest energy at the first return and second (third) highest energy at the third (fifth) return [68]. For the second and fourth returns, the recollision energy is too low to induce DI and thus the corresponding peaks in Fig. 3(a) are suppressed. For the double-recollision trajectories [Fig. 3(c)], the traveling time distribution for the first recollision mainly locates around 0.6$T$, corresponding to the recollision at the first returning. For the second recollision, the distribution is shifted by 0.5$T$ and has a relatively uniform distribution for the later returns. In Fig. 3(d) we show the time delay between the two recollisions. The main peak locates at 0.5$T$, meaning that the second recollision mainly occurs at the next return after the first recollision.

In Fig. 3(b) we show the time delay between double ionization and the last recollision. It is interesting that the time delays for the single-recollision and double-recollision events are significantly different. First, the first peak of the distribution for the double-recollision events locates at the smaller delay time than that of the single-recollision events. Second, the second and later peaks for the double-recollision events are lower. Both features indicate that the double ionization of the double-recollision events occurs more quickly. In order to understand this behavior, we trace the energy transfer during the recollisions. The red squares of Fig. 4(a) show the recollision energy $E_r(t_r - \Delta t)$ for the single-recollision NSDI events. Here the recollision energy is calculated as the energy of the recollision electron at the instant $\Delta t = 3$ a.u. just before recollision $t_r$ [66,67]. At the low laser intensity here, the returning electron was captured after the recollision and formed a doubly excited state [66,67]. Thus, for the single-recollision trajectories the recollision energy is the total energy that was “injected” into the two-electron system. So the energy transferred to the bound system by the recollision electron is $E_{\text{trans}} = E_r(t_r - \Delta t)$. For the double-recollision NSDI events, the returning electron only transfers part of its energy to excite the ion during the first recollision. It passes through the core area with the remaining energy and then returns back to transfer energy to the parent ion during the second recollision, after which the recollision electron is recaptured. Thus in the second recollision the energy of recollision is totally transferred to the system. So the total energy transferred to the bound system in the double-recollision NSDI events is $E_{\text{trans}} = E_r(t_r - \Delta t) + E_r(t_r + \Delta t)$, where $E_{\text{trans}}(1) = E_r(t_r - \Delta t) - E_r(t_r + \Delta t)$ and $E_{\text{trans}}(2) = E_r(t_r + \Delta t) - E_r(t_r - \Delta t)$.
contribution of double recollisions also decreases, as shown in
the wavelength increases (for the fixed laser intensity), the recollisions decreases with the increasing laser intensity. When of 780 nm. It is clearly shown that the contribution of double NSDI events as a function of laser intensity at the wavelength
NSDI. Here only the events corresponding to the first peak of the time delay distributions in Fig. 3(b) are included. For the single-recollision portion of events corresponding to the first peak of time delay recollision and double-recollision NSDI events, where only the trajectories, as shown in Fig. 3(b).

\( \Delta t \) are the energy transfers during the first and the second recollisions, respectively. In Fig. 4(b) we separately show the energy transfer during the first recollision and the second recollision for the double-recollision trajectories. The energy transfer during the first recollision is significantly lower than the returning energy [the red squares in Fig. 4(a)], confirming that only part of the recollision energy is transferred to the bound system. The total energy transfer during the two recollisions, which is the sum of the two parts in Fig. 4(b), is shown by the blue triangles in Fig. 4(a). Obviously, the total energy transfer during these two recollisions is higher than that in the single-recollision trajectories, implying that the system is staying at the higher excited states after the second recollision. Thus the double ionization of the doubly excited states occurs more quickly for the double-recollision trajectories, as shown in Fig. 3(b).

The consequence of the different DI rates is the different correlated electron momentum distributions. Figures 5(a) and 5(b), respectively, show the distributions for the single-recollision and double-recollision NSDI events, where only the proportion of events corresponding to the first peak of time delay distributions in Fig. 3(b) is included. For the single-recollision case, the distribution exhibits an antecorrelated behavior while it exhibits a correlated behavior for the double-recollision NSDI.

Finally, we investigated the intensity and wavelength dependence of multiple recollisions in NSDI. Figure 6(a) shows the proportion of double-recollision trajectories in total NSDI events as a function of laser intensity at the wavelength of 780 nm. It is clearly shown that the contribution of double recollisions decreases with the increasing laser intensity. When the wavelength increases (for the fixed laser intensity), the contribution of double recollisions also decreases, as shown in

\[ \text{Fig. 6(b).} \] These behaviors can be easily understood as the following: When the laser intensity and/or wavelength increases, the recollision energy increases and thus the bound system can be excited to higher states or ionized directly with just one recollision. Consequently, more NSDI events can occur without a second recollision. We can deduce that multiple recollisions will be more prevalent at lower laser intensities and shorter wavelengths.

**IV. CONCLUSION**

In conclusion, we have investigated the multiple recollisions in strong-field NSDI. The multiple recollision process has a significant contribution to NSDI at low laser intensities. We have shown the intuitive classical trajectories of the multiple recollisions in NSDI as well as their statistical characteristics. At the low laser intensities, the recollision energy of the first electron is very low. Thus, it can be easily understood that more recollisions are needed for NSDI at low laser intensities. Because of the multiple recollisions, the atoms gained more energies from the laser field through the recollisions, forming higher doubly excited states. Consequently, the double ionization of the doubly excited states by multiple recollisions occurs more quickly than that in the single-recollision events, resulting in the different correlated electron momentum distributions for these two cases. From the laser intensity and wavelength dependence of the multiple recollisions, it can be deduced that multiple recollisions are more prevalent at low laser intensities, and thus theories in
treating NSDI at that region should incorporate the multiple recollisions.

Note added in proof. Recently, a paper concerning the similar issue was published [69].

ACKNOWLEDGMENT

This work was supported by the National Natural Science Foundation of China under Grants No. 61405064 and No. 11234004.