Contribution of recollision ionization to the cross-shaped structure in nonsequential double ionization

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Abstract: With the three-dimensional classical ensemble model, we investigate the correlated electron emission in nonsequential double ionization (NSDI) of argon atoms by few-cycle laser pulses. Our calculations well reproduce the experimentally observed cross-shaped structure in the correlated two-electron momentum spectrum [Nature Commun. 3, 813 (2012)]. By tracing these NSDI trajectories, we find that besides the process of recollision-induced excitation with subsequent ionization just before the next field maximum, the recollision ionization also significantly contributes to the cross-shaped structure.

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38. Y. Zhou, C. Huang, and P. Lu, “Coulomb-tail effect of electron-electron interaction on nonsequential double

1. Introduction

As a standard process for studies of dynamical electron correlations, nonsequential double ionization (NSDI) has been a hot topic in the strong field physics since the observation of the dramatically enhanced double ionization yields [1–3]. Differently from sequential double ionization (SDI) where the two electrons are ejected one by one independently [4–6], a great number of experimental works and theoretical studies [7–16] provide strong evidences that the quasiclassical recollision model [17,18] is dominantly responsible for the NSDI process. There, the first released electron is driven back by the oscillating laser field and collides with the parent ion inelastically, leading to the release of the second electron in a direct recollision ionization process (RCI) or indirectly via recollision-induced excitation with subsequent field ionization (RESI) [19].

The quasiclassical recollision model [17,18] provides a general physical picture for NSDI of atoms and molecules in strong laser fields. With the deep study of NSDI, many novel characteristics in the correlated electron momentum spectra were found [20–24]. By means of these novel characteristics many detailed microscopic dynamics processes under the recollision mechanism have been revealed. For example, the high resolution experiments on DI of helium observed a fingerlike (or V-like) structure in the correlated electron momentum distribution for two different laser intensities [20,21]. For the relatively low intensity, both the nuclear attraction at recollision and electron-electron repulsion in the final state play significant roles in forming the fingerlike shape [25,26]. At the relatively high laser intensity, it is demonstrated that the V-like shape structure originates from asymmetric energy sharing at recollision [27]. At the laser intensity below the recollision threshold, Liu et al. experimentally found the dominant back-to-back emission of the correlated electron pairs from NSDI of atoms [24]. The anticorrelation behavior was attributed to the delayed emission of the second electron after recollision [28].

Most of previous NSDI experiments have been carried out using many-cycle laser pulses, where the contribution of multiple recollisions to NSDI could hamper further understanding of the detailed recollision dynamics. By controlling the carrier-envelope phase (CEP), few-cycle laser pulses can achieve only one recollision event contributing to NSDI, which is of great importance to the understanding of correlated electron emissions. Previous experiments have measured the momentum distribution of doubly charged ions from NSDI by few-cycle laser pulses [29], and found the strong CEP dependence of the NSDI process. The correlated electron momentum spectra from NSDI by few-cycle laser pulses have been explored by various theoretical studies [15, 30–33]. Until recently the measurement of correlated two-electron momentum spectra for NSDI in the single-cycle limit is presented by Bergues et al [34]. The measured two-electron correlated spectrum exhibits a cross-shaped structure that qualitatively differs from spectra recorded in all previous experiments using many-cycle pulses. This experiment provides a benchmark data for the theoretical study of NSDI. However, in their semiclassical calculation they assume that for RCI mechanism the transfer energy between the returning electron and the bound electron is right equal to the second ionization potential $I_{p2}$ of argon. With this assumption it is found that only RESI mechanism contributes to the cross-shaped structure and RCI mechanism has no contribution to the cross-shaped structure. In this paper, we carried out the ab initio calculation of the NSDI of argon atoms by few-cycle laser pulses with the 3D classical ensemble model. Our results indicate that the RCI assisted by laser electric...
field significantly contributes to the cross-shaped structure, and its contribution is comparable to that of the RESI mechanism [34]. Here, the calculated correlated electron momentum spectrum including the two mechanisms well reproduce the experimentally observed cross-shaped structure.

2. The classical ensemble model

Here, we employ the classical ensemble model [35,36], which is widely recognized as a useful approach in studying strong field double ionization [37–39]. In this model the evolution of the three-particle system is determined by the Newton’s equations of motion (atomic units are used throughout until stated otherwise):

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

where the subscript \(i\) is the label of the two electrons, and \(E(t)\) is the electric field of a 750 nm linearly polarized laser pulse with a 4-cycle \(\sin^2\)-shaped envelope (corresponding to a full-width at half-maximum of 3.64 fs). The laser intensity is \(3 \times 10^{14}\) W/cm\(^2\). The potentials \(V_{ne}(\mathbf{r}_i) = -2/\sqrt{\mathbf{r}_i^2 + a^2}\), \(V_{ee}(\mathbf{r}_1, \mathbf{r}_2) = 1/\sqrt{(\mathbf{r}_1 - \mathbf{r}_2)^2 + b^2}\) represent the ion-electron and electron-electron interactions respectively. To avoid autoionization, we set the screening parameter \(a\) to be 1.5. \(b\) is set to be 0.05. To obtain the initial values, the ensemble is populated starting from a classically allowed position for the Ar ground-state energy of -1.59 a.u. The available kinetic energy is distributed between the two electrons randomly in momentum space. Then the electrons are allowed to evolve a sufficient long time (600 a.u.) in the absence of the laser field to obtain stable position and momentum distribution. Once the initial ensemble is obtained, the laser field is turned on and all trajectories are evolved in the combined Coulomb and laser fields.

3. Results and discussions

Figure 1 shows correlated two-electron momentum distributions along the laser polarization direction for NSDI of Ar by 750 nm, 4-cycle linearly polarized laser pulses at an intensity of \(3 \times 10^{14}\) W/cm\(^2\). The CEP-averaged correlation spectrum shown in Fig. 1(a) reveals a cross-shaped structure, which indicates that the momentum of one electron is always close to zero whereas the momentum of the other electron varies within a finite range [-1.5 ∼ 1.5 a.u. in Fig. 1(a)]. This result is well in agreement with the experimental data [34]. Previous studies have indicated that the CEP-resolved correlated two-electron spectra are asymmetric with respect to the diagonal \(p_{||e1} = -p_{||e2}\), and this asymmetry strongly depend on laser CEP. The asymmetry can be represented by the parameter \(A = (N_+ - N_-)/(N_+ + N_-)\), where \(N_+\) and \(N_-\) are the number of double ionization events above and below the minor diagonal, respectively. The asymmetry parameter as a function of the CEP \(\phi\) is shown in Fig. 2. The asymmetry exhibits a sin-like behavior with the CEP increasing. Figures 1(b) and 1(d) show the CEP-resolved correlation spectra for the CEPs of -0.4π and 0.6π respectively, which manifest maximum asymmetry. The correlated momentum spectrum for the CEP of 0.1π corresponds to near-zero asymmetry [Fig. 1(c)].

In order to explore the origin of the cross-shaped structure in the CEP-averaged correlated two-electron momentum distribution [Fig. 1(a)], we trace classical NSDI trajectories and carefully examine their histories. In this way, we can easily obtain the recollision time and ionization times of two electrons. Here, we define an electron to be ionized if its energy turns positive [38], where the energy of each electron contains the kinetic energy, potential energy of the electron-ion interaction and half electron-electron repulsion. We scan the time interval from the time
when one electron first ionizes until final ionization of both electrons, and we define the time of closest approach as the recollision time, which is marked by $t_r$ [28,36,38]. $t_{DI}$ represents the double ionization time.

For simplicity but without loss of generality, next we discuss the case of $\phi=0.6\pi$. In Fig. 3(a) we present the final longitudinal momentum distribution of the first electron versus the second electron for $\phi=0.6\pi$. Here, the two electrons are distinguished based on the order in which they achieve final ionization after recollision. The electron ionized first after recollision is defined as the first electron and the other electron is defined as the second electron. $p_{||e1}$ and $p_{||e2}$ represent the final longitudinal momenta of the first and the second electrons, respectively. It is clear that the first electron often escapes with a near-zero final momentum in the polarization direction, whereas the second electron most likely drifts out with a non-zero final longitudinal momentum. This is consistent with the result in [34]. Figure 3(b) shows the double ionization time $t_{DI}$ versus the recollision time $t_r$ for $\phi=0.6\pi$. It is clearly seen that NSDI events mainly cluster

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Fig. 1. The correlated two-electron momentum distributions for NSDI of Ar by 750 nm, 4-cycle linearly polarized laser pulses at an intensity of $3 \times 10^{14}$ W/cm$^2$. (a) all CEPs averaged, (b) $\phi=-0.4\pi$, (c) $\phi=0.1\pi$, (d) $\phi=0.6\pi$. The two electrons are not distinguished.

Fig. 2. The asymmetry of the correlated electron momentum spectrum as a function of the CEP $\phi$ for the intensity of $3 \times 10^{14}$ W/cm$^2$. 

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two regions, which are marked by two ellipses $G_1$ and $G_2$. For NSDI events in region $G_2$, the double ionizations occur just before the next field maximum after recollision. This corresponds to RESI mechanism, which has been demonstrated responsible for the crossed-shape structure in the correlated electron momentum distribution in [34]. Besides, there is another group of NSDI trajectories indicated by the ellipse $G_1$ in Fig. 3(b). For this group of NSDI trajectories, the recollision times are mostly near 1.71T ($T$ is laser cycle) and the final double ionization occurs around 1.80T. The time interval between the final double ionization and the recollision is only 0.09T. Thus this group of NSDI trajectories corresponds to RCI mechanism. Furthermore, Figs. 4(a) and 4(b) show the correlated electron momentum distributions of NSDI events in regions $G_1$ and $G_2$ of Fig. 3(b), respectively. It is obvious that NSDI events from RCI and RESI mechanisms both mainly distribute near the two axes. The only difference is that RCI events contribute to the inner part of the cross-shaped structure whereas RESI events are responsible for the outer part of the cross-shaped structure.

Figure 5 shows two sample trajectories selected from groups $G_1$ (the left column) and $G_2$ (the right column) of Fig. 3(b). We examine the time evolutions of the longitudinal momenta, coordinate $z$ in polarization direction, energies of two electrons and the momenta in $y$ direction. For the NSDI trajectory in the left column, the first emitted electron is firstly driven to the positive direction by a negative electric field and then is pulled back to the parent ion when the electric field reverses [Fig. 5(b)]. Just after the maximum of the laser electric field, the returning
Fig. 5. Two sample trajectories selected from groups G1 (the left column) and G2 (the right column) of Fig. 3(b). The panels from top to bottom show the longitudinal momenta, coordinate $z$ in polarization direction, energies of two electrons and the momenta in $y$ direction versus the time, respectively. The arrows indicate the recollision time ($t_r$) and the vertical lines indicate the final double ionization time ($t_{DI}$). The black dashed curve marks the laser electric field in arbitrary units.

electron recollides with the second electron (green arrows). From Fig. 5(c) we can find that during recollision the returning electron transfers some energy to the second electron and is still free, but the transfer energy is not enough to promote the second electron to the positive energy. At the instant of recollision, the laser electric field is very large and pronouncedly lowers down the Coulomb barrier of the ion along the z-axis. This results in the second electron well located above the suppressed barrier. Immediately the large electric field force drives the second electron to escape over the suppressed barrier, and meantime the second electron quickly obtains enough energy from the electric field to escape away from the ion. In this process the time interval between the emission of the second electron and the recollision is very short (shorter than 0.1T), and thus it is regarded as RCI mechanism. As is well known, the electron emitted in the laser field will obtain a drift momentum from the subsequent electric field. Here, the first electron is forward rescattered with a negative initial momentum and then it obtains a positive drift momentum from the subsequent electric field. The two options cancel each other, resulting in a near-zero final longitudinal momentum of the first electron [see red curve of Fig. 5(a)]. Due to a near-zero initial momentum after recollision and the acceleration from the subsequent electric field, the second electron has a non-zero final momentum [see blue curve of Fig. 5(a)].

For some NSDI events, because of the asymmetric energy sharing during recollision [27], the energy transfer during recollision is not enough to directly emit the second electron. In this situation the second electron is emitted by subsequent field ionization. An example of such a trajectory (RESI) is shown in the right column of Fig. 5. For this NSDI trajectory, although the returning electron carries very large energy back to the parent ion, only a small part of returning energy is transferred to the second electron. After recollision the second electron is obviously promoted to an excited state and keeps for a certain time [see Fig. 5(g)]. Just before the next
field maximum the second electron achieves the positive energy and is ionized [see the vertical lines in Fig. 5(g)]. It is also clearly illustrated by the time evolution of the electron transverse momentum $p_y$. Once the second electron is excited, the oscillation of its transverse momentum $p_y$ becomes intense. After it is ionized its transverse momentum stops oscillating and keeps unchanged [the red curve see Fig. 5(h)]. Finally, as a result of the acceleration of the laser field the second electron obtains a non-zero momentum. For the first electron, the large initial momentum offsets the acceleration of the laser field and finally it has a near-zero longitudinal momentum [see Fig. 5(e)].

The discussion above has indicated that for $\phi=0.6\pi$ both RCI and RESI mechanisms contribute significantly to NSDI and are responsible for the cross-shaped structure in the correlated electron momentum spectrum. Besides the case of $\phi=0.6\pi$, we also trace and carefully back examine the classical NSDI trajectories from other CEPs. Our analysis finds that for all CEPs both RCI and RESI mechanisms contribute significantly to NSDI, and their contributions are comparable. The only difference is that for some CEPs there is only one recollision event contribute to NSDI, whereas for other CEPs two recollision events are involved.

For a realistic laser focus, the existence of an out-of-phase electric field component in the laser propagation direction produces an effective ellipticity. The work by Paquette et al [40] has demonstrated that the effective ellipticity can result in a significant reduction in doubly charged ion yields for tight focusing condition (such as $1\mu m$ beam waist). We have calculated NSDI for different effective ellipticities. The results show that for the effective ellipticities of 0.05 and 0.10, the correlated electron momenta are uniformly distributed in the four quadrants and the cross-shaped structure disappears. However, for the experiment of Bergues et al [34], the beam waist is about $40\mu m$. In this condition, the effective ellipticity is less than 0.006 within the $1/e^2$ intensity volume. For such a small ellipticity, its influence on the electron momentum distribution is negligible and the crossed-shaped structure is still clear. Thus under this experimental condition of Bergues et al [34], it is valid to neglect the effect of the effective ellipticity of the realistic laser focus. In addition, we have calculated NSDI of argon atoms by 750 nm, $3\times10^{14}$ W/cm² laser pulses with the full width of 4, 5 and 6 cycles. The ratios of RCI events to RESI events are 0.90, 0.62 and 0.49, respectively. This indicates that compared with RESI mechanism, the contribution of RCI mechanism to the observed cross-shaped structure decreases with the pulse width increasing.

In the semiclassical model of [34] the scattering angle $\beta$ is a very important free parameter. Its value significantly influences the shape of the correlated electron momentum distribution. In the calculation of [34], the cross-shaped structure is well reproduced only when the scattering angle $\beta$ for $\phi=0.6\pi$. 

Fig. 6. Counts of NSDI trajectories versus the scattering angle $\beta$ for $\phi=0.6\pi$. 

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angle $\beta$ is set to 20°. In our classical simulation, we also examine the scattering angles of NSDI trajectories. Here, the scattering angle $\beta$ is defined as the angle between the momentum vectors at the instants 0.03T before and 0.03T after the recollision. Figure 6 shows counts of NSDI trajectories versus the scattering angle $\beta$ for the CEP of 0.6$\pi$. Obviously, for the classical NSDI trajectories the scattering angles $\beta$ are also around 20°, which is consistent with the fitting value in [34].

4. Conclusion

In conclusion, with the 3D classical ensemble model we have investigated the correlated electron emission in NSDI of argon atoms by few-cycle laser pulses. The calculated correlated electron momentum spectra well reproduce the experimentally observed cross-shaped structure. By tracing those NSDI trajectories, we identify two categories of trajectories contributing to the cross-shaped structure. One category of NSDI trajectories corresponds to the dynamics process of recollision-induced excitation with subsequent field ionization just before the next field maximum, which has been proposed in [34]. The other category of NSDI trajectories is triggered by recollision ionization. Back analysis indicates that the two mechanisms have the comparable contributions to the cross-shaped structure in the correlated two-electron momentum spectrum.

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