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Recollision of excited electron in below-threshold nonsequential double ionization

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Abstract

Consensus has been reached that recollision, as the most important post-tunneling process, is responsible for nonsequential double ionization process in intense infrared laser field, however, its effect has been restricted to interaction between the first ionized electron and the residual ion so far. Here we identify the key role of recollision of the second ionized electron, which is enhanced by the stronger Coulomb potential of the higher valence residual ion, in the below-threshold nonsequential double ionization process by introducing a Coulomb-corrected quantum-trajectories method, which enables us to well reproduce the experimentally observed cross-shaped and anti-correlated patterns in correlated two-electron momentum distributions, and also the transition between these two patterns. Being significantly enhanced relatively by the recapture process which is also attributed to the stronger Coulomb potential of the residual ion, recolliding trajectories of the second electron excited by the first- or third-return recolliding trajectories of the first electron produce the cross-shaped or anti-correlated distributions, respectively. And the transition is induced by the increasing contribution of the third return with increasing pulse duration. Our work provides a new insight into atomic ionization dynamics and paves the new way to imaging of ultrafast dynamics of atoms and molecules in intense laser field.

Introduction

Post-ionization process has been the focus of strong-field atomic and molecular physics in the past thirty years. A semiclassical perspective, in which the recollision process plays a key role, is established with great effort. It can well explain many intriguing strong-field phenomena, such as high-order above-threshold ionization (HATI), high harmonics generation (HHG), and nonsequential double ionization (NSDI), and also serves as the foundation of attosecond physics (see, e.g., Refs. (1-4) for reviews and references therein). In the recollision picture (5, 6), an electron is liberated from the neutral atom or molecule through tunneling, then is driven back by the laser field to collide with the parent ion elastically or inelastically, or recombine with the ion, resulting in HATI, NSDI and HHG, respectively. Since the electron strongly interacts with the ion, the products upon recollision carry information of the parent ion, and can be used to probe its structure and dynamics. Based on the recollision process, different methods, such as laser-induced electron diffraction (LIED) (7) and laser-induced electron inelastic diffraction (LIID) (8), are proposed and successfully applied in imaging of atomic and molecular ultrafast dynamics and
structure with unprecedented spatial-temporal resolution (8-15). However, the recollision in the above-mentioned strong-field processes and ultrafast imaging methods is limited to interaction between the first ionized electron and the residual ion.

In the NSDI process, one electron \(e_1\) firstly experiences a recollision with the parent univalent ion and deliver energy to the bounded electron \(e_2\). In the below-threshold regime, the maximal kinetic energy of \(e_1\) upon recollision is smaller than the ionization potential of \(e_2\), so \(e_2\) can be only pumped to an excited state, as illustrated in Fig. 1. Then \(e_2\) is ionized from the excited state by the laser field at a later time, dubbed as recollision excitation with subsequent ionization (RESI) process. Usually, it is believed that \(e_2\) will travel directly to the detector (16-19), i.e., the post-tunneling process of \(e_2\) has been largely ignored. In fact, after tunneling ionization, \(e_2\) may be driven back to recollide with the divalent ion or be recaptured into a Rydberg state of ion as illustrated in Fig. 1. Due to the strong Coulomb field of the divalent ion, these post-tunneling dynamics may be prominent. It has been recently reported experimentally and theoretically that the probability of recapture in double ionization, dubbed as frustrated double ionization (FDI), is much higher than expectation (20, 21).

In this work, by introducing a Coulomb-corrected quantum-trajectories (CCQT) method, we identify the key role played by the recollision between the second ionized electron and the divalent ion in the below-threshold NSDI process. We find that, only when this recollision is included, the experimentally observed cross-shaped (22, 23) and anti-correlated (24) patterns of correlated electron momentum distribution (CEMD), and also the transition between them (25), can be well reproduced.

**Results**

**Comparison with experimental results**

Fig. 2 displays the calculated results for Ar under different pulse durations to compare with the experimental results in Ref. (25). Intensities higher than the measured ones by \(0.25 \times 10^{14} \text{ W/cm}^2\) are used in the present calculations (see Supplementary Materials for details of the fitting procedure). As shown in Fig. 2, for shorter pulse durations (4 fs and 8 fs), the distributions show a cross shape with the maxima lying at the origin. While for longer pulses (16 fs and 30 fs), the electrons are more homogeneously distributed over the four quadrants, actually, prefer the second and fourth quadrants, which indicates an anti-correlation. This transition of CEMD from cross-shaped to anti-correlated patterns is in agreement with the measured results reported in Ref. (25), although there is some discrepancy in details. In the measurement, the transition occurs when pulse duration increases from 4 fs to 8 fs, whereas in Fig. 2 it occurs when pulse duration increases from 8 fs to 16 fs. This discrepancy may be due to that the pulse shape and duration employed in our calculations are not exactly the same as that in the measurements.

To quantitatively characterize the CEMD, in Fig. 2c we plot the ratio \(Y_{2&4}/Y_{1&3}\) for different pulse durations and different intensities. \(Y_{1&3}\) (\(Y_{2&4}\)) denotes the integrated yield in the first and third (the second and fourth) quadrants. We also present the measured results (25) in Fig. 2f for comparison. In general, the simulation reproduces most of the features in the measured results. The ratio increases with pulse duration and becomes saturated at 16 fs when the intensity is fixed, and it decreases with laser intensity both for pulse durations of 8 fs and 16 fs. However, compared with the measured results, the simulation obviously overestimates the ratio for the highest intensity. This discrepancy can be attributed to that the contribution of the process that \(e_2\) is directly knocked out by \(e_1\), whose distribution mainly locates in the first and third quadrants, becomes more significant with increasing intensity, but is not included here.

**Recolliding trajectories of \(e_2\)**

In Fig. 3, we present CEMDs corresponding to recolliding trajectories and direct trajectories of \(e_2\) at 4 fs and 30 fs, respectively. Here, we define it as the recolliding trajectory if the minimal distance of \(e_2\) from the residual ion is less than the tunnel exit. Otherwise, it is the direct trajectory.
Since momenta of direct trajectories of e2 are much smaller than that of recolliding trajectories, CEMDs for direct trajectories are localized around the origin for both 4 fs and 30 fs pulses, as shown in Figs. 3A and 3D. Whereas the CEMD for recolliding trajectories exhibits a cross structure at 4 fs (Fig. 3b), and exhibits an anti-correlated pattern at 30 fs (Fig. 3e). Meanwhile, recolliding trajectories of e2 have dominant contributions for all pulse durations as depicted by the ratio $Y_{\text{res}}/Y_{\text{dir}}$ ($Y_{\text{res}}$ and $Y_{\text{dir}}$ denote the yields of recolliding and direct trajectories, respectively) for double ionization (DI) events in Fig. 3c, as a consequence, the total CEMDs also shows a cross or an anti-correlated pattern at 4 fs or 30 fs, respectively.

But why the relative contribution of the recolliding trajectories of e2 is so high? Intuitively, the Coulomb focusing effect imposed on e2 by the divalent cation, which is much stronger than that of the univalent cation in ATI process, will effectively enhance the probability of recollision. We can indeed see this clearly from Fig. 3c in which the ratio $Y_{\text{res}}/Y_{\text{dir}}$ with all events included is greater than 1. But it is still much smaller than the ratio considering only DI events. This deviation is the result of the important contribution of recapture or FDI process. More than two-thirds of direct e2 are recaptured into the Rydberg states of Ar$^+$ at 4fs, and the probability of FDI for direct e2 decreases quickly with increasing pulse duration, as shown in Fig. 3f. Compared with recolliding trajectory of e2, direct e2 cannot move far away from Ar$^{2+}$ at the end of the pulse due to its much lower momentum, especially in shorter laser pulse, therefore is easier to be recaptured by the strong Coulomb field of the divalent ion. More direct e2 being recaptured means fewer of them contribute to DI, resulting in larger relative contribution of recolliding trajectories of e2 to DI. In brief, the enhanced FDI probability significantly enlarges the relative contribution of recolliding trajectories of e2 to DI, and eventually induces the experimentally observed cross-shaped and anti-correlated patterns. In addition, this point is strongly supported by the fact that when only the direct trajectories of e2 are considered, the calculated $Y_{2\&4}/Y_{1\&3}$ is significantly different from the experimental result (see Fig. 2c).

**Transition of CEMD with increasing pulse duration**

The specific pattern of CEMD also requires the appropriate momentum of e1 which is determined by the microscopic dynamics of the recollision process for e1. According to our calculations, the first- and third-return recolliding trajectories of e1 are dominant for the laser parameters interested here. For other returns, either the return energy is too small to excite e2, or the collision probability is negligible due to the spreading of the wave packet. In Figs. 4A and 4B, we present the CEMDs corresponding to the first- and third-return trajectories of e1, respectively, in 1.25×$10^{14}$ W/cm$^2$, 30 fs laser pulse. Note that all trajectories of e2 (direct and recolliding trajectories) are included. The CEMD for the first-return trajectories of e1 (Fig. 4a) shows a cross-shaped pattern, whereas that for the third-return trajectories (Fig. 4b) exhibits an anti-correlated pattern. As shown in Fig. 4c, the ratio of the integrated yield of the third-return trajectories to that of the first-return increases quickly with increasing pulse duration. Correspondingly, the CEMD changes from a cross-shaped to an anti-correlated pattern. Therefore, the transition between the two patterns of CEMD with increasing pulse duration is the result of increasing contribution of the third-return trajectories of e1. The significant contribution of the third-return trajectories can be attributed to the Coulomb focusing effect from the univalent cation. The similar effect has also been reported for high-order ATI process (32).

**Cross-shaped and anti-correlated CEMDs**

Next, we will explain how the cross-shaped and anti-correlated patterns of CEMDs are formed by the recolliding trajectories of the two electrons. Without indistinguishability symmetrization, the first-return trajectories of e1 will show a band-like distribution along the $p_{1z}$=0 axis with the maxima away from the origin, i.e., vanishing momentum of e1 but much higher momentum of e2 (Fig. 5b). Whereas the CEMD for the third-return consists of two bands and the maximum of the left (right) band lies in the up (low) part, giving rise to an anti-correlation (Fig. 5c). These band-like distributions can be understood as follows. The final momentum of e1 is determined by the
residual momentum after exciting \( e_2 \) and the drift momentum it obtains from the laser field. Since forward scattering is favored in this inelastic scattering process, the residual momentum and the drift momentum are in opposite directions and will cancel with each other. At the present intensity (\( 1.25 \times 10^{14} \text{ W/cm}^2 \)), the magnitudes of them for the first-return trajectories of \( e_1 \) are nearly equal, resulting in a vanishing momentum of \( e_1 \). When the laser intensity increases, the band will become tilted towards the main diagonal (23) due to the faster-increasing residual momentum. For the third-return, its return energy is smaller than that of the first-return, so the residual momentum is not enough to compensate the drift momentum, resulting in a non-vanishing momentum of \( e_1 \). Since electrons ionized at times separated by a half optical cycle will have opposite momenta, there is one band on each side of \( p_{t_2}=0 \) axis. Actually, there are also two bands for the first return, but they merge together.

The anti-correlation between the two electrons for the third-return trajectories of \( e_1 \) is illustrated in Fig. 5a. The recollision of \( e_1 \) most probably occurs around the crossing of the electric field at \( t_{r_1} \) or \( t_{r_1}' \). Since the magnitude of the drift momentum after recollision, which is equal to \( -A(t_{r_1}) \) (vector potential at the recollision time), is larger than the residual momentum for the third-return recollisions trajectories of \( e_1 \), its final momentum is in the direction of the drift momentum. If the recollision of \( e_1 \) occurs at \( t_{r_1}' \), the final momentum of \( e_1 \) will be positive, corresponding to the right band in Fig. 5c. Upon recollision, \( e_2 \) is pumped to the first excited state, then it is most probably ionized at the subsequent electric field peak at \( t_{2i} \). If the Coulomb attraction of the ion is not considered and no recollision occurs, \( e_2 \) will have vanishing final momentum. This can be seen clearly in Figs. 5D and 5E, in which the CEMDs are obtained by calculating \( M_3^{(5)} \) in Eq. (1) (see the Materials and Methods) with the standard SFA. But if the ionic Coulomb potential is taken into account, momenta of \( e_2 \) for recolliding trajectories (trajectory I) shift to the negative direction, opposite to the direction of the final momentum of \( e_1 \) (see Fig. 5a). This is exactly the situation of the right-band distribution in Fig. 5c. The left band corresponds to the situation that \( e_1 \) recollides with the ion at \( t_{r_1} \) and \( e_2 \) is ionized at \( t_{2i} \). As a consequence, the two electrons are emitted back-to-back and the CEMD exhibits an anti-correlated pattern. In addition, it is also possible that the recollision of \( e_1 \) occurs at \( t_{r_1} \) while \( e_2 \) is ionized at \( t_{2i} \), which will produce a correlated CEMD. But since its contribution is smaller due to the depletion effect of the excited state, the total CEMD will still exhibit an anti-correlated pattern.

**Prediction for higher valence ions**

It is expected that the effect of the Coulomb field for higher valence ion will be stronger. This can be demonstrated in the DI process of \( \text{Ar}^+ \). As shown in Fig. 6a, the recollision probability of \( e_2 \) in the presence of the Coulomb field of \( \text{Ar}^{3+} \) is higher than that for \( \text{Ar}^{2+} \) shown in Fig. 2e. The CEMD also exhibits a strong dependence on the laser intensity and pulse duration. A similar to Fig. 2a but more obvious cross structure appears in the CEMD for laser field of 400 nm with pulse duration of 4fs and intensity of \( 4 \times 10^{14} \text{ W/cm}^2 \) (Fig. 6b) - the arms get thinner and longer. When the laser intensity increases to \( 8 \times 10^{14} \text{ W/cm}^2 \) which is still lower than the threshold intensity of \( 8.6 \times 10^{14} \text{ W/cm}^2 \), the CEMD transits to a correlated pattern (Fig. 6c). If increasing the pulse duration to 16 fs, the CEMD then transits back to the cross structure (Fig. 6d). It is the result of increasing contribution of the third-return trajectories of \( e_1 \) which just meets the requirements of cross structure. It is noteworthy that, for convenience of experimental observation, we employ 400 nm laser pulses in the above calculations for NSDI of \( \text{Ar}^+ \) which enable us to apply higher laser intensity to obtain higher ionization probability but remains in the below-threshold region. The additional complexity in experimental aspect comes from preparing \( \text{Ar}^+ \) instead of \( \text{Ar} \) atoms as targets, but it should not be an impossible task under current experimental conditions (33).
Discussion

We propose a Coulomb-corrected quantum-trajectories (CCQT) method to describe the below-threshold NSDI process both coherently and quantitatively. It enables us to well reproduce different kinds of CEMDs observed in experiments, and uncover the rich underlying physics induced by the Coulomb field of univalent, divalent and higher valence ions, including the multi-return trajectories of the first ionized electron $e_1$, the recollision and recapture processes of the second ionized electron $e_2$. Especially, recollision process of $e_2$, which is enhanced relatively by the recapture process of $e_2$, is found to play an important role in electron-electron correlation. We expect that the recollision process of $e_2$ can be applied to develop a new scheme to image the ultrafast evolution of the molecular structure and dynamics induced by the strong laser field.

Materials and Methods

To describe the below-threshold NDSI process both coherently and quantitatively, it has to incorporate both the quantum effect and the Coulomb interaction between the residual ion and the ionized electrons in a uniform theory. To achieve this, we introduce a Coulomb-corrected quantum-trajectories (CCQT) method by taking advantage of the well-developed Coulomb-corrected methods dealing with single-electron dynamics. The transition magnitude is expressed as (atomic units $m = \hbar = e = 1$ are used)

$$ M\left(\tilde{P}_1, \tilde{P}_2\right) = \sum_s M^{(1)}_{P_i}(t^s_{i,}, t^s_{f,}) M^{(2)}_{P_i}(t^s_{f,}) M^{(3)}_{P_2}(t^s_{f,}, t^s_{i,}) $$

in which different trajectories labelled with $s$ are summed coherently. $M^{(1)}_{P_i}(t^s_{i,}, t^s_{f,})$, describing the tunneling ionization of $e_1$ at $t^s_{i,}$ and its subsequent propagation in the laser field until colliding with the parent ion at time $t^s_{f,}$, is calculated using the quantum-trajectory Monte Carlo (QTMC) method (26,27) which is efficient to obtain large amount of hard-collision trajectories. Trajectories with minimum distance from the ion less than 1 a.u. are selected to consider the hard collision for the subsequent calculation. Upon collision, $e_1$ will excite $e_2$ and then move to the detector. This excitation process is described by $M^{(2)}_{P_i}(t^s_{f,})$ which is calculated with conventional S-matrix theory. Finally, $e_2$ is ionized through tunneling at $t^s_{f,}$ from the excited state, and then propagates in the laser field until the end of the pulse, which is described by $M^{(3)}_{P_2}(t^s_{f,}, t^s_{i,})$ calculated with the Coulomb-corrected strong field approximation (CCSFA) method (28). The sin-squared pulse shape is employed in our calculation. A model potential (29) is applied to mimic the Coulomb field of Ar$^{2+}$ felt by $e_2$ in its propagation. Only the first excited state 3s3p6 with zero magnetic quantum number (30) is included in the present calculations. The depletion of the excited state is also taken into account in calculating $M^{(3)}_{P_2}(t^s_{f,}, t^s_{i,})$ (18) (see the method in Supplementary Materials).

Data availability: All data are available in the main text or the supplementary materials.
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**Author contributions**

X.H., W.Y., and J.C. designed the research; X.H. and Y.B. performed all the simulations; X.H., Y.B., C.L., and J.Z. analyzed data; X.H., W.L., W.Y., and J.C. discussed the results; X.H., W.L.,
W.Y., and J.C. wrote the paper.

Competing interests
The authors declare no competing interests.

Figures

**Fig. 1.** Sketch map to illustrate the below-threshold NSDI process. At time $t_{1i}$, $e_1$ is first ionized by the laser field, then it is driven back to collide the parent univalent ion and excites $e_2$ at time $t_{1r}$. $e_2$ is ionized from the excited state by the laser field at a later time $t_{2i}$. After that, $e_2$ may travel directly to the detector, or it may be driven back to recollide with the divalent ion similar to $e_1$, or it may also be recaptured into a Rydberg state of ion.

**Fig. 2.** Simulated results for below-threshold NSDI of Ar. (a), (b), (d) and (e) Simulated correlated electron momentum distributions (CEMDs) of Ar for different laser pulse durations at...
the intensity of $1.25 \times 10^{14}$ W/cm$^2$. The CEPs are averaged. Each CEMD is normalized to itself. (c)
Simulated yield ratio $Y_{2&4}/Y_{1&3}$ for different pulse durations and different intensities. $Y_{1&3}$ ($Y_{2&4}$) denotes the integrated yield in the first and third (the second and fourth) quadrants in the CEMD. The numbers given in the legends denote peak laser intensities with units of $10^{14}$ W/cm$^2$. The open circles are calculated by only considering direct trajectories of $e_2$ (see text for details). (f) Measured results extracted from Ref.[25]. The black short-dashed lines in (c) and (f) serve as indications of the intensity dependence.

Fig. 3. Distributions corresponding to recolliding trajectories and direct trajectories of $e_2$. (a) and (d) CEMDs corresponding to direct trajectories of $e_2$. (b) and (e) CEMDs corresponding to recolliding trajectories of $e_2$. (c) Pulse-duration dependence of $Y_{rec}/Y_{dir}$, the ratio between the integrated yields of recolliding and direct trajectories for $e_2$ for all events or only double ionization (DI) events. (f) Pulse-duration dependence of $Y_{FDI}/Y_{DI}$, the ratio between the probabilities of FDI and DI when $e_2$ is confined to direct trajectories. The laser intensity is $1.25 \times 10^{14}$ W/cm$^2$. Each CEMD is normalized to itself. The CEPs are averaged.
Fig. 4. Distributions corresponding to the first- and third-return trajectories of $e_1$. (a) CEMDs corresponding to the first return. (b) CEMDs corresponding to the third return. (c) Pulse-duration dependence of $Y_{3rd}/Y_{1st}$, the ratio of the integrated yield of the third-return to that of the first-return trajectories of $e_1$. The laser intensity is $1.25 \times 10^{14}$ W/cm$^2$. Each CEMD is normalized to itself. The CEPs are averaged.
Fig. 5. Effect of Coulomb field of Ar$^{2+}$ on CEMDs. (a) Schematic representation of the laser electric field $E(t)$ and the corresponding vector potential $A(t)$ for pulse duration of 30 fs. $e_1$ collides with the ion most probably at the crossing of $E(t)$ at $t_{1r}$ or $t'_{1r}$. Upon the collision, $e_2$ is excited, and then is ionized most probably at the peak of the laser field at $t_{2i}$ or $t'_{2i}$. The subsequent evolution of the canonical momentum $p_{2z} - A(t)$ for the recolliding trajectories of $e_2$, denoted as I and II, are presented to illustrate the Coulomb-field effect of Ar$^{2+}$. (b) and (c) CEMDs without performing electron indistinguishability symmetrization, and only recolliding trajectories of $e_2$ with ionization time nearest the collision time of $e_1$ are included. (d) and (e) CEMDs calculated by replacing the ionization amplitude $M^{(3)}_{p_2}$ in Eq. (1) with the standard SFA. Trajectories of $e_1$ are confined to the first return in (b) and (d), and the third return in (c) and (e). Each CEMD is normalized to itself. The CEPs are averaged.
Fig. 6. Simulated results for below-threshold NSDI of Ar$^+$ which finally becomes Ar$^{3+}$ in 400 nm laser pulse. (a) The ratio between the integrated yields of recolliding and direct trajectories for e$_2$. (b)-(d) Normalized CEMDs. The CEPs are averaged.
Supplementary Files

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