Anticorrelated electrons from high-intensity nonsequential double ionization of atoms

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The nonsequential double ionization (NSDI) yield and final electron-elecon correlation are examined for a laser wavelength of 800 nm over a wide range of laser intensities, using three-dimensional fully classical models. It is shown that in the transition region between NSDI and sequential ionization, turn-on stage recollisions can lead to having a majority of anticorrelated (back-to-back) electron pairs.

I. INTRODUCTION

It is well known that in nonsequential double ionization (NSDI) of some systems [1,2] the doubly ionized electron pairs are more likely to be detected on the same side of the nucleus than on opposite sides. This correlation in final momenta can be explained in terms of the recollision [3] process that is now accepted as the primary NSDI mechanism: One electron ionizes and departs the core but is impelled back to the core by the oscillating laser field so that it shares energy with the other electron. The process often concludes with the two electrons traveling out together. As discussed in the following (and in Ref. [4]), a complete analysis requires considerations of electron energies, of laser phase at recollision, of whether one or both electrons may be bound to the nucleus for a brief time after the collision [5], and even of whether a free electron backscatters off the nucleus [6]. Regardless, there is a well-established net correlation in the final electron momenta along the laser polarization axis for some systems. This correlation is widely recognized as one signature of NSDI and can be manifest as a doublet [7] in the final net momentum along the laser polarization axis.

At laser intensities below the threshold for recollision impact ionization, experiments [8] revealed the possibility that the final electron momenta could be anticorrelated, indicating back-to-back drift relative to the laser polarization axis. This surprising result was subsequently examined theoretically by some of us using three-dimensional classical models [9]. We found that weak recollisions could lead to doubly excited states, from which the electrons escape one at a time at different phases of the laser field. The delay leads preferentially to oppositely directed electrons.

At intensities above the familiar NSDI "knee" or plateau, NSDI is replaced by sequential ionization, a process in which the electrons ionize independently. The loss of correlation can be manifest by having the familiar doublet in the net momentum spectra collapse into a singlet [10].

II. RESULTS FOR PRIMARY MODEL

A. DI yield and high-intensity anticorrelation

Figure 1 shows the total single and double ionization yields in our model as functions of laser intensity for a laser wavelength of 800 nm. For these plots, the ensemble size

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was fixed at 50 000. We considered laser pulses with electric field $\mathbf{E}(t) = \hat{\mathbf{z}} f(t) E_0 \sin(\omega t)$, where $f(t)$ is a trapezoid with one-cycle turn-on, three-cycle full strength, and one-cycle turn-off. The double ionization (DI) yield shows the familiar plateau. There is also a small dip at $5 \, \text{PW/cm}^2$. We have categorized the DI trajectories based on whether the electron pairs emerge in the same or opposite momentum hemispheres relative to the laser polarization axis and have drawn curves for each. There is an intensity range, beginning at the dip, in which there is a net *anticorrelation*. For example, at an intensity of $7 \, \text{PW/cm}^2$, anticorrelated pairs outnumber correlated pairs by about a factor of 2 (440 versus 217). In the following, we will find that this anticorrelation arises from weak recollisions that occur during laser turn-on.

A second region of very strong anticorrelation is present for intensities of around $5 \times 10^{16} \, \text{W/cm}^2$, above saturation of DI. This anticorrelation is easily explained—a check of individual trajectories reveals that usually one electron spills out of the potential-energy well in the first half cycle of the laser pulse, with the second electron moving in the opposite direction the next half cycle. Thus the ionization is sequential (one at a time) but anticorrelated. At intensities approaching $10^{17} \, \text{W/cm}^2$ the confining well is basically obliterated during the first half cycle, and both electrons spill out on the same side.

Our three-dimensional classical ensemble method was introduced in [5] and discussed in some detail in [4]. Each two-electron atom in the ensemble is given slightly different initial conditions, but all begin with zero angular momentum and energy equal to that of the helium ground state, -2.9035 a.u. (We use atomic units unless stated otherwise.) To prevent autoionization in our fully classical model, we soften the nuclear potential, replacing $-2/r$ with $-2/\sqrt{r^2 + a^2}$, where $a = 0.825$ a.u. For the *e-e* interaction we use softening parameter 0.05 [11]. Each atom is evolved separately using Newton's equations of motion, first with no laser field for abut 100 a.u., then through the pulse. In this completely classical model, all ionizations occur over the barrier.

In order for recolliding electrons to be able to experience a large nuclear force we reduce the softening parameter a, trajectory by trajectory, once one electron reaches $r = 10$ a.u. [12]. To conserve energy when we change the potential energy we also increase the electrons' radial speeds. We work in the dipole approximation and ignore magnetic field and other relativistic effects, which may start to become important at the highest intensities considered [13]. We consider changing the softening parameter to be a necessary adaptation of our
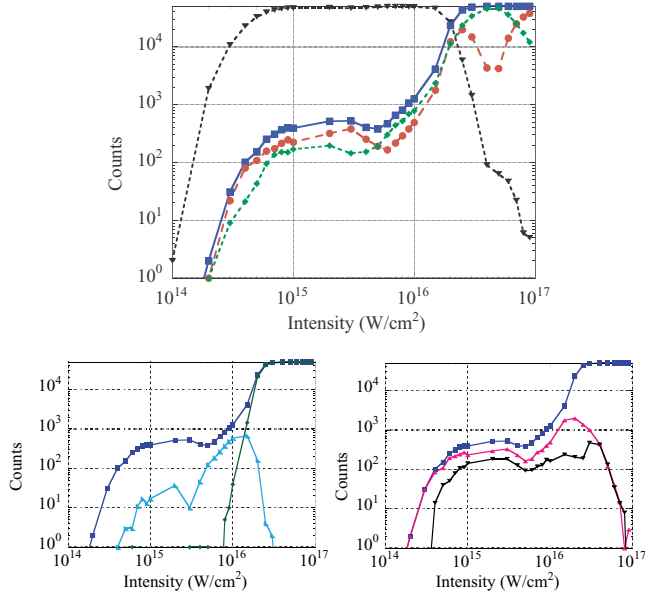
FIG. 1. (Color online) Absolute single and double ionization total yields vs laser intensity for a wavelength of 800 nm for our three-dimensional classical model with an ensemble size of 50 000. The top black dotted curve with inverted triangles shows single ionization yield, and the solid blue curve with squares shows total double ionization yield. The bottom two curves in the top plot include same- and opposite-hemisphere trajectories, denoted by red circles and green diamonds, respectively. The second row shows yields divided by process: sequential (green diamonds) and exit collisions as discussed in text (cyan triangles) on the left panel and RESI (magenta triangles) and recollision with impact ionization (black inverted triangles) on the right panel.

fully classical model to allow for the quantum behavior of real systems. Fortunately, there is also a strength that emerges from being able to change a. By doing successive runs with different softening parameters, we are able to consider the importance of the core potential on specific results. In the present work we consider primarily the final value $a = 0.4$, but then we test the robustness of the results by considering different values of a, including $a = 0.825$ where there is no change at all.

B. Four categories of trajectories

In the second row of Fig. 1 we have backtracked all DI trajectories [14] and sorted them into four categories. Sequential ionization is shown as the green curve with diamonds in the left plot. It dominates at the highest intensities. In this process, the electrons ionize one at a time without energy sharing. An example trajectory is shown in the top left plot of Fig. 2 for a laser intensity of 1×10^{16} W/cm^2. Recollision with impact ionization is shown on the right in Fig. 1 as the black curve with inverted triangles. In this process the returning electron simply knocks the other one free [15]. The maximum energy with which an electron can return is about $3.2U_p$, where U_p denotes the ponderomotive energy [3]. Because the most energetic returns occur at about the time of a laser zero, we can estimate the threshold intensity for impact ionization simply by setting $3.2U_p = |E_g^+|$, where E_g^+ is the ground state energy of the positive ion. For $\lambda = 800$ nm and $E_g^+ = -2$ a.u., we obtain

a threshold of 3×10^{14} W/cm^2. This is consistent with Fig. 1. An example impact ionization trajectory at the much higher laser intensity of 3×10^{15} W/cm^2 is shown in the top right plot of Fig. 2. For this intensity, the ponderomotive energy is 6.6 a.u., and there is plenty of energy at recollision. Thus, recollisions are not limited to times close to a zero of the laser field. In the example trajectory, the recollision occurs against the laser force shortly before a laser extremum. Backscattering of the struck electron and the laser force then lead to backward drift relative to the recollision.

The recollision excitation with subsequent ionization (RESI) yield is shown on the right in Fig. 1 as magenta triangles [15]. In our model RESI is consistently more important than impact ionization. At very low intensities, one or more recollisions can lead to a doubly excited state from which electrons ionize one at a time, often in opposite directions [9]. Unfortunately, the ensemble size we used for Fig. 1 was too small for the anticorrelation we discussed in [9] to be detected. At intensities somewhat above the threshold for impact ionization, we found that unequal energy sharing at recollision would typically leave one electron in a highly excited bound state [16]. This electron would often ionize within one-half laser cycle after recollision (during the first barrier suppression) into the direction opposite from the recollision [5]. Since the electron that is free after the collision is likely to be swept into the backward direction (relative to recollision) by the laser field, the two RESI electrons can emerge as a correlated pair [5,17]. A more complete discussion of final directions from our model in this region for various wavelengths can be found in [4]. An example RESI trajectory for the much higher laser intensity of 3×10^{15} W/cm^2 is shown in the bottom left plot of Fig. 2. In this example, the recolliding electron misses the core on its first two returns, but on its third return it transfers sufficient energy to the inner electron for it to escape at the next laser extremum. Energy versus time for this trajectory is shown as the bottom right plot of Fig. 2. The energy transfer just before $t = 2.6$ cycles is clearly visible. This trajectory also suggests a reason why RESI can be more important than impact ionization even at high intensities. It suggests the importance of impact parameter. It is not enough for the returning electron to have sufficient energy for double ionization—there must be an effective transfer of energy to the second electron. We will return to this point in the following.

The curves for RESI and impact ionization only include trajectories for which one electron goes out past $r = 10$ before returning to the core and recolliding. The curve with cyan triangles in the bottom left plot of Fig. 1 denotes trajectories in which an electron travels out past $r = 3$ but not $r = 10$ before returning and colliding with the second electron. We will call these trajectories exit collisions. An example is shown in Fig. 3. After a laser field maximum, one electron begins moving outward in the direction of the diminishing laser force. The electron remains bound and the nucleus pulls it back, reversing its direction of motion. If the direction change occurs at about the time of a laser zero, then the laser force does positive work both before and after the turnaround. Some of this energy can be transferred to the second electron as the first one passes it on its way out. The energy versus time plot [18] in the lower right corner of the figure clearly shows
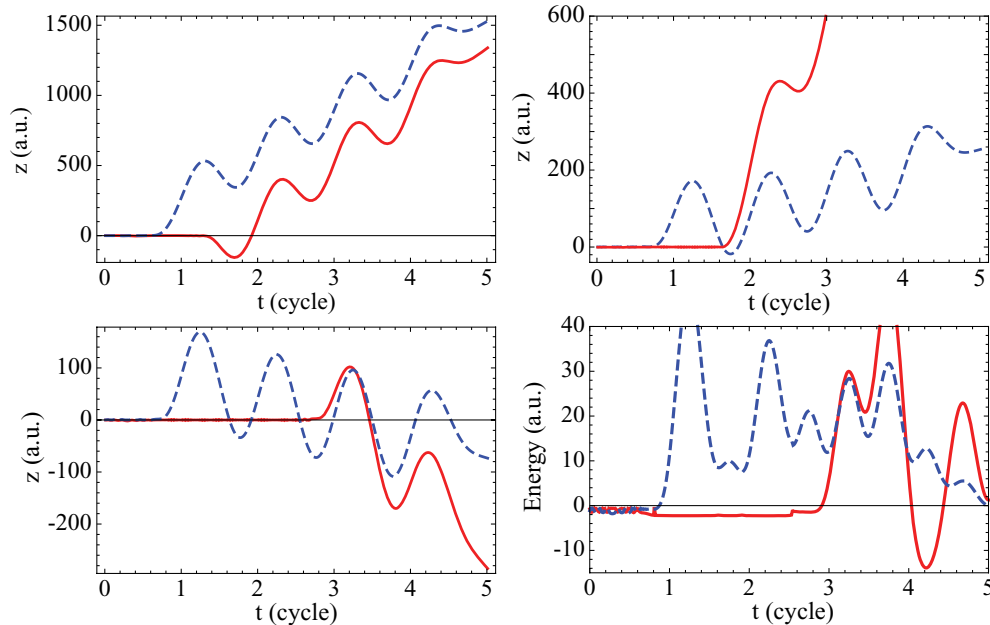
FIG. 2. (Color online) Sample trajectories, showing longitudinal coordinate z (parallel to laser polarization) vs time for each electron. Top left: Sequential ionization for $I = 1 \times 10^{16}$ W/cm^2. Top right: Impact ionization for $I = 3 \times 10^{15}$ W/cm^2. Bottom left: RESI for $I = 3 \times 10^{15}$ W/cm^2. Bottom right: Electron energy vs time for the RESI trajectory (including potential energy from interaction with the laser). The energy transfer at recollision is visible near $t = 2.6$ cycles.

this energy transfer just after $t = 0.6$ cycle. The energy gained in the collision can allow the second electron to escape without further interaction with the first electron.

There are three features of the trajectory in Fig. 3 to which we call special attention. First, the exit collision occurs during laser turn on. Second, the exiting electron is bound at the time of the laser zero ($t = 0.5$ cycle) before the collision. Third, the two electrons ionize in different half cycles and into opposite directions, thus giving a back-to-back pair. In
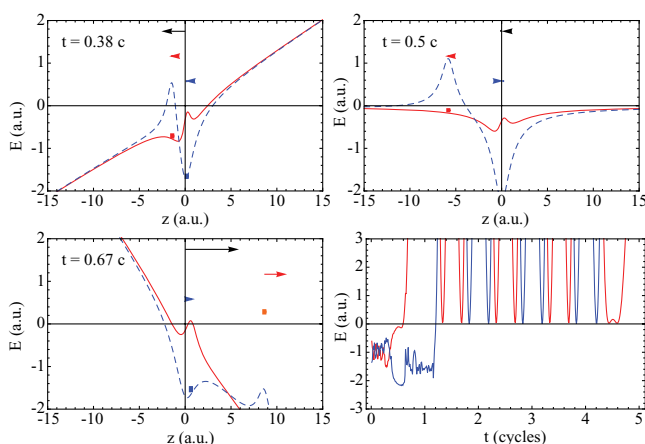

FIG. 3. (Color online) An example of an exit collision during laser turn-on. One electron, coded here in red, moves out from the core but is pulled back by the nucleus and shares energy with the other electron on the way out, allowing subsequent escape. The top arrows show the laser force, and other arrows show v_z. The bottom right plot shows electron energies vs time excluding potential energy from interaction with the laser. Neither electron achieves positive energy until after the collision.

Ref. [19], it was noted that there were some short-excursion one-dimensional DI trajectories that had no unambiguous first ionization before recollision. Such trajectories, referred to there as "direct-excitation trajectories," were found to have recollision occur most often during the laser turn-on. They were evident in both quantum and classical one-dimensional systems. Final momenta were not investigated in [19]. Exit collisions were also noted in Ref. [9], but at very low intensities where multiple collisions are needed to free the second electron. There, an exit collision could provide an initial energy transfer, so that less energy would be needed from subsequent recollision(s).

Having the laser do positive work on a bound electron both before and after the electron changes direction has also been noted in the context of what was called the boomerang in Ref. [12]. For laser wavelengths such as 390 nm [12] or 483 nm [9], recollision excitation could push an electron away from the nucleus in the direction of a weakening laser field. The bound electron would then switch directions (boomerang) so that the laser could continue to do positive work as the electron traverses the nuclear well and escapes out the other side over the laser-suppressed barrier. At the higher frequencies this process allows an electron to finish with final energy above $2U_p$, the maximum drift energy of an electron that starts from rest in a simple oscillating field.

C. Turn-on stage recollisions and anticorrelation

Figure 1 shows that the exit collisions are very important at intensities of around 10^{16} W/cm^2. However, at higher intensities the number of exit collisions drops dramatically, while the amount of RESI and impact ionization climbs. We have found that this change is in part due to how we define
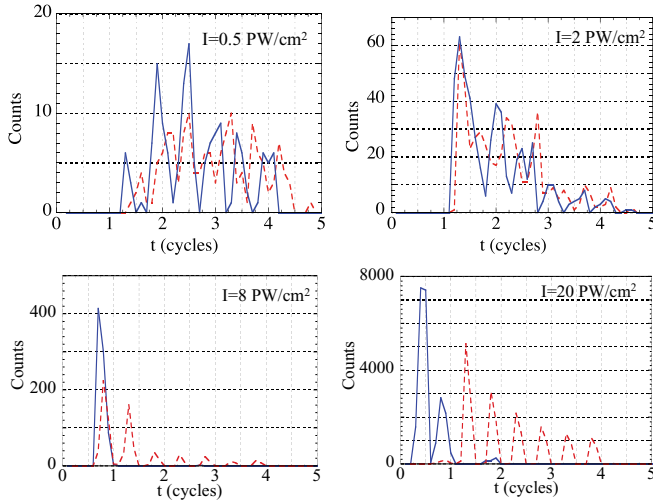
FIG. 4. (Color online) Ensemble recollision times in a $1 + 3 + 1$ trapezoidal pulse (solid blue curve) and final ionization times (dashed red curves). Times were binned to every 0.1 cycle, and the number of counts in each bin was tallied. Laser intensities were 0.5, 2, 8, and $20 \, \mathrm{PW/cm^2}$. Adjustable parameters are all as in Fig. 1.

the boundaries between these processes—if the first electron goes beyond $r = 10$ our classification scheme moves it out of the exit collision category. The trajectories may still feature what Ref. [20] has referred to as "turn-on stage recollision." A quick check of the RESI and impact ionization curves in this region above $10^{16} \, \mathrm{W/cm^2}$ also reveals that they can be associated primarily with turn-on stage recollisions. We thus find that exit-collision trajectories are part of a larger category of trajectories in which recollision occurs during laser turn-on.

The shift toward turn-on stage recollisions at higher intensities is evident in Fig. 4. The solid blue curves indicate recollision times (defined as the time of closest approach of the two electrons after first ionization), and the red dashed curves the final ionization times. For intensities $I = 0.5$ and $2 \, \mathrm{PW/cm^2}$, recollisions and final ionizations are spread throughout the full-strength part of the pulse. However, by $I = 8 \, \mathrm{PW/cm^2}$ recollisions occur primarily during the one-cycle turn-on. This is the intensity range in which Fig. 1 shows exit collisions to be very important. The final plot of Fig. 4 considers an intensity of $20 \, \mathrm{PW/cm^2}$, an intensity for which Fig. 1 indicates that sequential ionization and RESI are about equally important. Because of how we have defined the recollision time, the solid blue curve indicates either the recollision time or—if there is no recollision—the time of first ionization. The figure clearly indicates that, as intensity increases, the first ionization time and the recollision times—for the trajectories that have recollision—shift to the turn-on stage of the pulse.

The final-ionization curves for $I = 8 \, \mathrm{PW/cm^2}$ in Fig. 4 indicate a delay between turn-on stage recollision and final ionization. This delay can lead to anticorrelated electrons. This scenario is similar to the case of Ref. [9], where weak recollisions with time-delayed final ionization also led to anticorrelated electrons. If one electron ionizes early in a laser half cycle, but the other is delayed until late in that half cycle or early in the next, then the two electrons are likely to drift
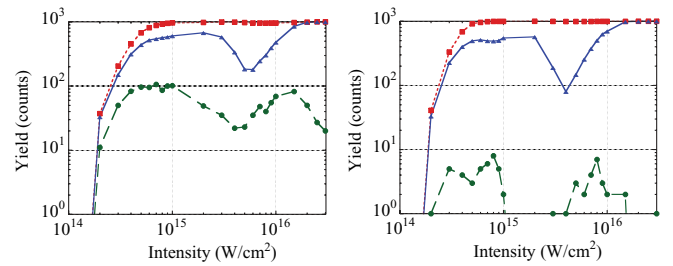

FIG. 5. (Color online) Number of trajectories vs laser intensity for 800 nm (left) and 2017 nm (right). The top (dotted red) curve counts trajectories in which at least one electron travels past $|z| = 10$. The middle (solid blue) curve tallies those that subsequently return to $z = 0$. The bottom (dashed green) curve includes only the trajectories that have $\sqrt{x^2 + y^2} < 3$ at return, and thus have at least the possibility of recollision. The ensemble size was 1000.

into opposite directions. At the highest intensity shown, most first ionizations occur in the very first half cycle of the pulse, but most final ionizations are delayed until the laser reaches full strength. Such long delays can destroy the anticorrelation.

D. Shutdown of full-laser-strength NSDI at high intensities

The shift of recollision to the turn-on of the laser field does not explain the dip in the DI yield evident in Fig. 1 around $5 \, \mathrm{PW/cm^2}$. To investigate it, we have taken an ensemble of just 1000 atoms and examined what happens to the first electron that ionizes [16]. The uppermost, dotted red curve in Fig. 5 shows the number of atoms in which at least one electron ionizes. The solid blue middle curve shows the total number of trajectories in which the first electron to ionize subsequently returns to $z = 0$. The dashed green bottom curve with circles counts only the subset of these trajectories for which the electron returns to $z = 0$ with $\sqrt{x^2 + y^2} < 3$. These trajectories have the possibility of recollision. At lower intensities, the number of these trajectories loosely follows the single ionization curve, so that when the latter saturates, the former enters a plateau. However, as the intensity is increased above $10^{15} \, \mathrm{W/cm^2}$, the number of recollision trajectories begins to drop [21]. We interpret these results in terms of increased transverse velocity. Thus we find that full-laser-strength NSDI begins to *shut down* with increasing intensity, producing the dip. A quick check reveals that the curves rise again due to the turn-on stage recollisions. The right-hand plot of Fig. 5 shows a similar plot for a laser wavelength of 2017 nm. This plot shows clearly that at high laser wavelength only a small fraction of electrons return for recollision. Of course, at high wavelengths the laser period is increased, and consequently the effects of transverse velocity become more important.

III. ANTICORRELATION FOR OTHER MODELS

This discussion has shown the possibility of finding a net anticorrelation in DI electrons in the transition zone between the NSDI plateau and the sequential-ionization domain. It is valid to ask, though, whether this result is unique to the particular model we were using. Thus, we have made various adjustments to the model and examined whether the

anticorrelation could be destroyed. We emphasize that our goal here is not to model any one specific physical system, but to explore what can happen and why. Thus we adjust parameters freely. We have adjusted the value of the softening parameter after first ionization. We have found that for $a = 0.825$ and $a = 0.1$, the anticorrelated region is still present. (Note that the $a = 0.825$ case leaves a unchanged throughout the entire pulse). However, for $a = 0.01$ we have found that the plateau ends at lower intensities, and the anticorrelated regime is lost. A full discussion of this shortening of the plateau will need to wait until a subsequent paper.

We have also considered whether the anticorrelation might be an artifact of our pulse shape. We have found the anticorrelation to be present for a three-cycle pulse with $\mathbf{E}(t) = \hat{\mathbf{z}} \sin^2[\omega t/(6\pi)] \cos(\omega t)$. However, we can destroy the anticorrelation by increasing the turn-on time. For example, it is lost for a $2 + 6 + 2$ trapezoidal pulse and for longer $\sin^2$ pulses. For the anticorrelation to be present, it is desirable for the final ionization to occur late in the half cycle of a turn-on stage recollision or before the next laser maximum.

To investigate whether the anticorrelation is unique to our one laser wavelength we have investigated the behavior for different wavelengths (390, 1314, and 2017 nm) and found that the anticorrelation is present there as well.

A remaining question is whether the anticorrelation might be unique to classical models. Thus, we have also returned to the one-dimensional quantum model [19,22] and investigated whether it shows the anticorrelation for $\lambda = 800$ nm. We have found that if the e-e and e-p softening are both set to 1—so that e-e collisions are relatively weak—there is no anticorrelated region. However, if the e-e softening is reduced to 0.4, then there is a net anticorrelation for intensities between the plateau and sequential ionization. Decreased e-e softening increases the efficacy of e-e collisions, thus making collisions that occur on the way out more important.

An indicator of the turn-on stage double ionization can be electrons with quite different final momenta, since the two electrons escape through different mechanisms. This is illustrated in Fig. 6 for an intensity of 1×10^{16} W/cm², for a trapezoidal, random-phase pulse [$\mathbf{E}(t) = \hat{\mathbf{z}} f(t) E_0 \sin(\omega t + \delta)$, where $f(t)$ is a $1 + 3 + 1$ trapezoid and δ is random in $[0, 2\pi)$]. The scatter plot shows the final momentum of the second electron to ionize versus the first, with the direction of the collision defined as the positive direction. The first electron to ionize—usually the recolliding electron in a turn-on stage recollision—most often travels out into the forward direction relative to the recollision. The second electron has greater range of available momenta (with boundaries of the plot being at $\pm 2\sqrt{U_p}$) and is most likely to drift into the backward direction. Anticorrelation is clearly evident in the plot. The right plot of Fig. 6 shows the net momentum parallel to the laser pulse, without defining the direction of recollision to be positive, and is a singlet.
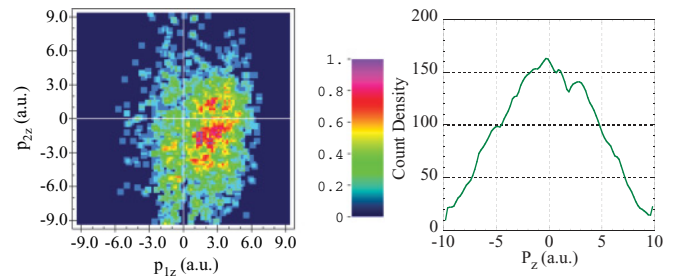

FIG. 6. (Color online) Left: The final momentum of second electron to ionize vs first for $I = 1 \times 10^{16}$ W/cm², with direction of recollision defined as the positive direction, for the classical model with a random-phase $1 + 3 + 1$ trapezoidal pulse. Right: The net electron momentum distribution, $p_{1z} + p_{2z}$, for the same ensemble but without defining the recollision direction as positive.

We conclude that the anticorrelation is not unique to one particular model, or even to classical modeling. However, it does require a fairly rapid laser turn-on, and for the anticorrelation to be visible the turn-on stage recollisions must not be completely outnumbered by subsequent sequential ionization. Thus, short pulses are better for finding the effect.

IV. CONCLUDING COMMENTS

The exit collisions are in some ways analogous to the "way-out" collisions that have been discussed in the context of high-frequency, one-photon ionization [23], but they occur in a very different frequency range. Exit collision excitation is also related to "shake up" as discussed in [24], where experimental evidence was found for an ionic core being left in an excited state if the first ionization occurred under tunneling conditions. That case is different from ours, however, because Ref. [24] considered circular polarization, and we have found that we do not have DI from exit collisions when the laser has circular polarization.

In summary, we have demonstrated the importance of turn-on stage ionization and of exit collisions in NSDI, finding the possibility of collisional excitation producing oppositely directed ("anticorrelated") electrons at laser intensities between the plateau and the onset of sequential ionization. We have also noted the possibility of anticorrelation at higher intensities, due to oppositely directed sequential ionization.

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[16] As in previous work with the model, we define an electron to be ionized if it achieves, and then maintains for at least 0.2 cycle, any of $E > 0$, where E is its energy, inclusive of electron-nucleus and *e-e* interactions but not the laser interaction; $|z| > 10$; or $zF_z > 0$ with $z^2 > 5$, where F_z is the longitudinal component of the net force.

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