Recollision Excitation, Electron Correlation, and the Production of High-Momentum Electrons in Double Ionization

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The production of high-momentum electrons in double ionization of helium by near-infrared lasers is investigated using three-dimensional classical ensembles. The nucleus' role is examined by systematically adjusting the nuclear potential. The primary source of the high-energy electrons is found to be backscattering off the nucleus at recollision. Recollision excitation with backscattering of the unbound electron is found to be especially important. It is shown that recollision excitation with ionization before the next field maximum can lead to a correlated electron pair.

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Electron correlation plays a very important role in double ionization (DI) of atoms by strong-field, near-infrared lasers. For intensities of order 10^{14} to 10^{15} W/cm^2, orders of magnitude more double ionization (DI) occurs than would be expected for independent, sequential ionization of electrons [1], with electron pairs often emerging in the same momentum hemisphere [2]. The explanation lies in the very rich dynamics of recollision [3], a process in which one electron moves out from the nucleus, but then is propelled back by the oscillating (linearly polarized) laser field and collides with the other electron. There may be an $(e, 2e)$ impact ionization similar to the binary ionizing collisions of electron scattering [4,5]. Alternatively, there may be recollision excitation [6] with subsequent ionization (RESI) of the excited electron, with time delay as short as a small fraction of a cycle [7] or as long as several laser cycles. The recollision may be accompanied by scattering off the nucleus, as in recoil collisions of electron scattering [4,5]. In complex atoms there can be multiphoton excitation effects [8]. Finally, the oscillating laser field can influence the electrons' motions after ionization and change their drift directions.

Experiments [2] have shown that the net longitudinal momentum of DI electron pairs rarely exceeds about $4\sqrt{U_p}$ (here the longitudinal direction denotes the laser polarization axis, U_p is the ponderomotive energy, and we use atomic units). One thus might expect that each electron separately would have longitudinal momentum of at most $2\sqrt{U_p}$. However, two recent experiments [4,9] with helium at 780 or 800 nm revealed that one electron of a correlated DI pair often achieves longitudinal momentum above $2\sqrt{U_p}$. Reference [4] presented evidence that the high-momentum electrons are produced through recollision ionization with backscattering of one of the electrons.

A 2006 study [10] of helium DI at wavelength 390 nm has also reported the production of significant numbers of electrons with momenta greater than $2\sqrt{Up}$, and thus energies above $2U_p$. That work presented a model which suggested that the high-energy electrons could be produced through backscattering of one of the electrons at the time of recollision. In [11] we found that a classical ensemble model for wavelength 390 nm also yielded electrons with energy above $2U_p$. Significant yield was obtained not only through backscattering of an unbound electron at recollision but also through a process we called the nuclear boomerang: a collisionally excited electron could be pulled back by the nucleus so as to begin traveling in the backward direction and escape over a suppressed barrier before the next field maximum. Such over-the-barrier escape can lead to energy above $2U_p$.

For the 390 nm studies of [10] the returning electron did not have sufficient energy for impact ionization at recollision. However, for the longer-wavelength studies of Refs. [4,9] and for the present work it does. Experiments have shown [12] there is no sharp threshold for DI at the minimum energy for impact ionization.

In the present work we use 3D classical ensembles to investigate the role of the nucleus in the production of high-energy DI electrons for laser wavelength 780 nm. We find that backscattering off the nucleus at recollision is indeed important in their production. We also find that recollision *excitation* plays a particularly key role, especially when accompanied by backscattering, and can lead to a longitudinally *correlated* electron pair.

The use of 3D classical ensembles was described in [7]. Each atom in an ensemble of at least 400 000 is prepared separately, with energy equal to the helium ground state and zero net angular momentum [13]. Each atom is then exposed to the same ten-cycle trapezoidal $(2 + 6 + 2)$ pulse.

A feature of 3D classical atoms is that they can autoionize if an unshielded nuclear potential $-2/r$ is used. Thus, we replace the Coulomb potential with $-2/\sqrt{r^2 + a^2}$, in analogy to the familiar soft core of 1D analysis (e.g., [14]). The screening parameter a is initially set to 0.825. The first ionization then occurs over a laser-suppressed barrier, not through autoionization or tunneling.

To investigate the importance of the nucleus in recollision, we change the nuclear screening parameter trajectory by trajectory as soon as one electron reaches $r = 10$. We offset the resulting decrease in potential energy of each electron with an appropriate kinetic energy boost for its radial motion. This "toggle switch" change in shielding is *ad hoc*, but allows us to explore the importance of the nuclear potential at recollision. In the present work we maintain constant *e-e* shielding 0.05.

The production of high-momentum electrons in our classical ensembles is clearly evident in Fig. 1, where for each DI pair we plot final longitudinal momentum for the recolliding electron vs the struck electron. (All our DI trajectories feature recollision.) For this figure, we have defined directions so that positive indicates final drift in the forward hemisphere, matching the longitudinal motion of the recolliding electron just before recollision. All the plots show population primarily in the third quadrant, indicating that the two electrons most often drift out in the backward hemisphere regardless of the details of the nuclear force. The squares indicate $p_{iz} = \pm 2\sqrt{U_p}$. As the shielding parameter a is decreased and the nucleus increasingly exposed, the number of individual electrons with longitudinal momentum exceeding $2\sqrt{U_p}$ increases, indicating that the nucleus plays an important role in the production of high-energy electrons. Also, the distinction between the struck and recolliding electron diminishes.

Spectra for transverse momenta are shown in Fig. 2. The spectra remain narrow regardless of nuclear shielding. Thus, high momenta are primarily in the longitudinal direction. For example, at shielding $a = 0.4$, 82% of the electrons with energy above $2U_p$ have longitudinal momentum above $2\sqrt{U_p}$, while only 1% have transverse momentum above that value. Figure 2(a) is in good agreement with experiment [15].

To investigate how electrons achieve large momentum, we have monitored each trajectory that leads to an electron with final $|p_z| > 2\sqrt{U_p}$, checking every 0.02 cycles for the recollision and the time of final ionization [16]. From recollision onward, we classify each trajectory based on whether electrons are bound or free. We present results in Fig. 3. Magenta curves with diamonds plot the percentage of trajectories that have both electrons ionized vs the time since recollision. As in Ref. [7], most trajectories have at least one electron bound for a portion of a laser cycle after recollision. The green curve with triangles in Fig. 3 shows the percentage of trajectories in which the electron that will achieve high energy is unbound but the other bound. This is the most important category about 0.1 cycle after recollision for all but the largest shielding value. These curves establish the importance of recollision excitation and show that an electron is more likely to achieve high energy if it remains unbound after the recollision. The blue curve with inverted triangles in Fig. 3 plots the percentage of high-energy trajectories in which the electron that finishes with high energy remains bound, with the other free, at the specified time since recollision. This category will include trajectories that achieve high energy through the nuclear boomerang [11]. This category is not very important when the nuclear screening is small. Another possible result of recollision is a doubly excited state which subsequently decays. This scenario is indicated by the red curve with square markers in Fig. 3. This category also becomes less important as the shielding is decreased.

In Fig. 4, we plot final energy spectra of all electrons that achieve energy greater than $2U_p$, for trajectory categories

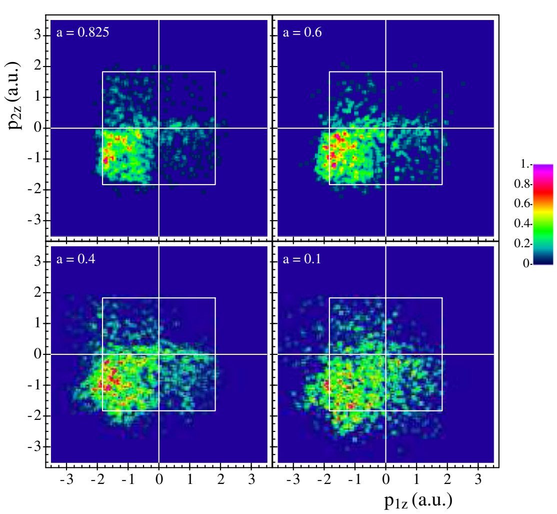

FIG. 1 (color online). Final longitudinal momenta of the recolliding electron (p_{2z}) vs struck electron (p_{1z}), for laser intensity 4×10^{14} W/cm^2 and wavelength 780 nm ($U_p = 0.838$). Signs are defined so that positive indicates final drift in the forward longitudinal direction. Values of the nuclear shielding parameter a after first ionization are shown. White boxes indicate $\pm 2\sqrt{U_p} = \pm 1.83$. Each plot is scaled separately for color coding.

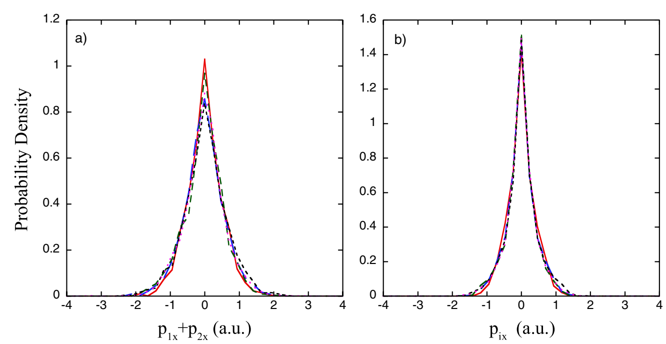

FIG. 2 (color online). Final momentum distributions for a transverse direction: (a) Sum momentum $p_{1x} + p_{2x}$, and (b) individual momenta p_{1x} and p_{2x}, for several values of nuclear shielding ($a = 0.01$, 0.1, 0.4, and 0.825 for small dashed black, solid red, dashed green, and large-dashed blue lines, respectively, though individual plots are difficult to discern). Plots are normalized so the area under each curve is 1. Plots for y components would look the same.

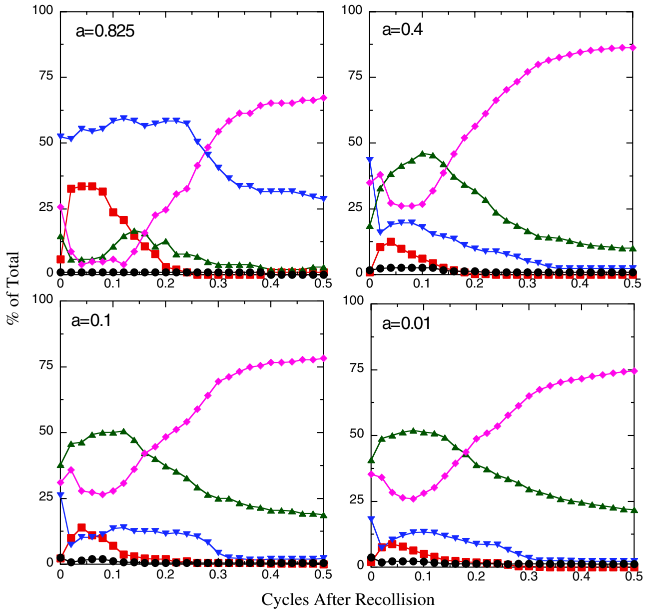

FIG. 3 (color online). For trajectories that achieve $|p_z| > 2\sqrt{U_p}$, the percentage of trajectories vs time since recollision for various post-recollision scenarios. Trajectory classifications: Magenta diamonds, both electrons free; green triangles, electron that achieves high final energy free and the other bound; blue inverse triangles, the inverse of green; red squares, both electrons bound; black circles, both electrons finish with high final energy, but one electron not yet ionized. The number of trajectories sorted are 101, 294, 351, and 1164. Ensemble size for $a = 0.01$ is 1.2×10^6, triple the others.

based on whether the electrons are bound or free 0.06 cycles after recollision. On the left we show results for screening parameter $a = 0.4$, on the right 0.01. The plots indicate that the highest energies are achieved by electrons that remain unbound after collisional *excitation* of the other electron. Figures 3 and 4 establish the importance of recollision excitation, and Fig. 1 established that the

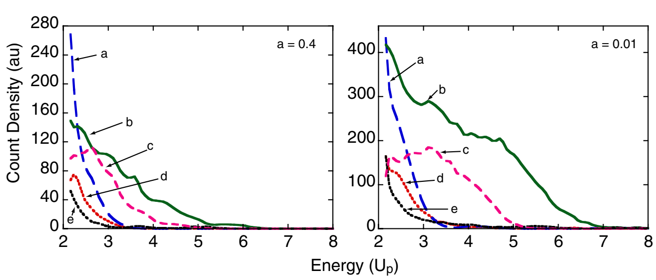

FIG. 4 (color online). Final electron energy spectra for the trajectory categories of Fig. 3, sorted based on whether each electron is free or bound 0.06 cycles after recollision. (a) high-energy bound, low-energy free; (b) high-energy free, low-energy bound; (c) both free; (d) both bound; (e) one bound, and both achieve high energy. The results have been smoothed; $a = 0.4$ on left (ensemble size 400 000) and 0.01 on right (ensemble size 1.2×10^6). All electrons with final energy above $2U_p$ are included.

electron drifts out in the backward direction. Since (as discussed below) the laser field will not result in backward traveling electrons with momentum greater than $2\sqrt{U_p}$, we infer that the direction change must be from nuclear scattering. We also note that an unshielded nucleus can provide the large force needed for backscattering of an unbound electron. Finally, a manual check of individual trajectories confirms backscattering off the nucleus either just before or just after recollision. Because of electron energy exchange, it can be either the recolliding or struck electron that is unbound and achieves high energy.

It is straightforward to estimate an upper limit to the final energy. The most energetic recollisions can be expected to occur just before a laser zero [3], with the returning electron having kinetic energy up to $3.2U_p$. If a collision at that time results in an electron that has velocity v_0 at the laser zero, and subsequent effects of all forces except the laser are ignored, the drift velocity of the electron would be $v_0 - 2\sqrt{U_p}$. Here we treat the forward direction as positive, and the second term has a minus sign because after the laser zero the laser force will be in the backward direction. If the electron gives up energy $E_B = 2$ a.u. in a collision just before the laser zero, the maximum $|v_o|$ for our laser parameters ($U_p = 0.838$) is about $\sqrt{2(3.2U_p - E_B)} = 1.17$, whereas $2\sqrt{U_p} = 1.83$. If $v_0 > 0$, the drift will be in the backward direction, at speed less than $2\sqrt{U_p}$. However, if interaction with the nucleus changes the sign of v_0, the drift speed could be as high as 3.0 a.u., which corresponds to energy $5.3U_p$. The significant number of trajectories we find exceeding these values in Fig. 4 suggests the importance of recollision ionization. If the electron gives up only sufficient energy for subsequent over-the-barrier escape of the other electron, the maximum drift speed for our laser parameters increases to 3.6 a.u. and the energy to $7.8U_p$. Our ensemble results stay within these bounds. (There remains the possibility of post-DI rescattering off the bare helium ion, which could give energies to $10U_p$ but did not occur in our ensemble.)

A familiar signature of correlation is the doublet in the net longitudinal momentum ($p_{1z} + p_{2z}$) spectrum [2]. Figure 5 shows that trajectories with short time delays between recollision and final ionization lead to a doublet, which fills in as progressively longer delay trajectories are included. We emphasize that the doublet does not arise solely from direct recollision ionization.

Recollision excitation with ionization at the next barrier suppression is not an artifact of classical models. Several quantum studies have noted bursts of double ionization that occur on alternating sides of the nucleus each half cycle of the laser, as the laser field grows in strength. These bursts have been described as "rapid sequential ionization" [17] or "sequential ionization," [18–20]. They have even been noted in 3D quantum studies [20] at higher frequencies. It

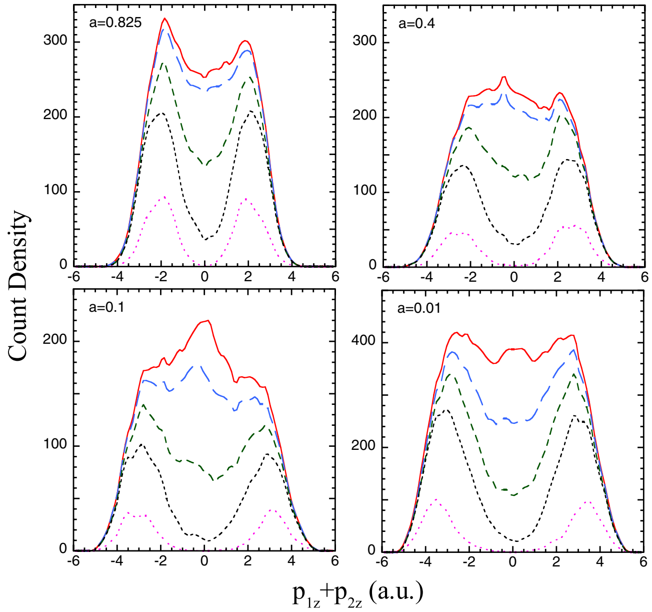

FIG. 5 (color online). Spectra of final net longitudinal momentum $P_z = p_{1z} + p_{2z}$ for the nuclear shielding values indicated and for various maximum time delays between recollision and final ionization. Maximum time delays from lowest to highest curves are 0.06, 0.26, 0.5, 2 cycles, and no limit. For our laser parameters, $4\sqrt{U_p} = 3.66$ a.u.. The curves have been smoothed. (Ensemble size for $a = 0.01$ is 1.2×10^6).

has been shown in 1D quantum models that the bursts occur just after recollision, which has allowed association of these bursts with recollision excitation [21]. They were described for classical 1D models in [14] and cited as evidence of RESI in Ref. [22].

In conclusion, we have employed a 3D classical ensemble model to investigate how electrons can achieve longitudinal momentum greater than $2\sqrt{U_p}$. The primary process is recollision accompanied by backscattering of a free electron. Of particular importance is recollision excitation with backscattering, since it allows the unbound electron additional energy while still being able to produce a correlated electron pair.

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