1-1-2004

Speed-up collisions in strong-field double ionization

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Abstract: We compare quantum and classical models of double ionization (DI) for aligned-electron helium in strong laser fields, considering specifically the role of recollision processes in which the returning electron travels in the direction of the laser force. Quantum studies show that for the knee region in our model a small but persistent portion of the total DI occurs through these speed-up collisions. We show that classical modeling displays similar collisions and reveals that with-the-force doubly ionizing collisions typically involve two-particle trajectories in which both electrons can be said to have been bound or very nearly bound at the zero of the laser field just before the collision. Trajectories leading to the with-the-force doubly ionizing collisions can be classified into two categories–direct excitation, in which there is no unambiguous single ionization before the doubly ionizing collision, and recapture, in which an ionized electron returns to the core and is recaptured prior to the speed-up collision. Comparison of the classical and quantum situations for our laser parameters yields evidence that for our parameters the quantum system favors the direct-excitation pathway over the reattachment pathway.

OCIS codes: (020.4180) Multiphoton processes (270.6620) Strong-field processes (260.3230) Ionization

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References and links

1. D. N. Fittinghof, P. R. Bolton, B. Chang and K. C. Kulander, "Observation of nonsequential double ionization of helium with optical tunneling," Phys. Rev. Lett. **69**, 2642-2645 (1992).
2. B. Walker , B. Sheehy, L. F. DiMauro, P. Agostini, K. J. Schafer and K. C. Kulander, "Precision measurement of strong field double ionization of helium," Phys. Rev. Lett. **73**, 1227-1230 (1994).
3. R. Dörner, Th. Weber, M. Weckenbrock, A. Staudte, M. Hattass, R. Moshammer, J. Ulrich, and H. Schmidt-Böcking, "Multiple Ionization in Strong Laser Fields," Advances in Atomic, Molecular, and Optical Physics **48**, 1-35 (2002).
4. M.V. Ammosov, N.B. Delone, and V.P. Krainov, "Tunnel ionization of complex atoms and of atomic ions in an alternating electromagnetic field," Sov. Phys. JETP **64**, 1191 (1986).
5. Th. Weber *et al.*, "Recoil-Ion Momentum Distributions for Single and Double Ionization of Helium in Strong Laser Fields," Phys. Rev. Lett. **84**, 443 (2000).
6. R. Moshammer *et al.*, "Momentum Distributions of Ne^{n+} Ions Created by an Intense Ultrashort Laser Pulse," Phys. Rev. Lett. **84**, 447 (2000).
7. P. B. Corkum, "Plasma perspective on strong field multiphoton ionization," Phys. Rev. Lett. **71**, 1994-1997 (1993).
8. K. J. Schafer, B. Yang, L. F. DiMauro and K. C. Kulander, "Above threshold ionization beyond the high harmonic cutoff," Phys. Rev. Lett. **70**, 1599-1602 (1993).

#5029 - $15.00 US Received 11 August 2004; revised 16 September 2004; accepted 20 September 2004

9. K.C. Kulander, J. Cooper, and K.H. Schafer, "Laser-assisted inelastic rescattering during above-threshold ionization," Phys. Rev. A **51**, 561 (1995).
10. F. H. M. Faisal and A. Becker, "Nonsequential double ionization: mechanism and model formula," Laser Phys. 7, 684 (1997).
11. A. Becker and F.H.M. Faisal, "Interpretation of momentum distribution of recoil ions from laser induced nonsequential double ionization," Phys. Rev. Lett. **84**, 3546 (2000).
12. C. Figueira de Morisson Faria, H. Schomerus, X. Liu, and W. Becker "Electron-electron dynamics in laser-induced nonsequential double ionization," Phys. Rev. A **69**, 043405 (2004).
13. H.W. van der Hart and K. Burnett, "Recollision model for double ionization of atoms in strong lasser fields," Phys. Rev. A **62**, 013407 (2000).
14. E. Eremina *et al.* "Laser-induced non-sequential double ionization investigated at and below the threshold for electron impact ionization," J. Phys. B: At. Mol. Opt. Phys. **36**, 3269-3280 (2003).
15. S. L. Haan, P. S. Wheeler, R. Panfili, and J. H. Eberly, "Origin of correlated electron emission in double ionization of atoms," Phys. Rev. A **66**, 061402(R) (2002).
16. J. Javanainen, J.H. Eberly, and Qichang Su, "Numerical simulations of multiphoton ionization and above-threshold electron spectra," Phys. Rev. A **38**, 3430-3446 (1988).
17. R. Grobe and J.H. Eberly, "Photoelectron spectra for a two-electron system in a strong laser field," Phys. Rev. Lett. **68**, 2905-2908 (1992).
18. D. Bauer, "Two-dimensional, two-electron model atom in a laser pulse: Exact treatment, single-activie-electron analysis, time-dependent density-functional theory, classical calculations, and nonsequential ionization," Phys. Rev. A **56**, 3028-3039 (1997).
19. J.B. Watson, A. Sanpera, D.G. Lappas, P.L. Knight, and K. Burnett, "Nonsequentiall Double Ionization of Helium," Phys. Rev. Lett. **78**, 1884-1887 (1997).
20. W. -C. Liu, J. H. Eberly, S. L. Haan and R. Grobe, "Correlation Effects in Two-Electron Model Atoms in Intense Laser Fields," Phys. Rev. Lett. **83**, 520-523 (1999).
21. A.M. Popov, O.V. Tikhonova, and E.A. Volkova, "Mechanisms of double-electron ionization of atomic systems in a strong laser field," Opt. Express **8**, 441-446 (2001).
22. J. S. Parker, B.J.S. Doherty, K.J. Meharg, and K.T. Taylor, "Time delay between singly and doubly ionizing wavepackets in laser-driven helium," *J. Phys. B* **36**, L393 (2003).
23. R. Heather and H. Metiu, "An efficient procedure for calculating the evolution of the wave function by fast Fourier transform methods for systems with spatially extended wave function and localized potential," J. Chem. Phys. **86**, 5009-5017 (1987).
24. S. L. Haan, K. Hoekema, S. Poniatowski, W.-C. Liu, and J. H. Eberly, "Directional correlation in direct and sequential double ionization of model atoms," Opt. Express **7**, 29-38 (2000). http://www.opticsexpress.org/oearchive/21863.htm
25. S. L. Haan, N. Hoekema, R. Panfili, J. H. Eberly, "Exploration of Double Ionization Using Wavefunction Masking," manuscript in preparation.
26. R. Panfili, J. H. Eberly and S. Haan, "Comparing classical and quantum simulations of strong-field double ionization," Opt. Express **8**, 431-435 (2001). http://www.opticsexpress.org/oearchive/source/31132.htm
27. R. Panfili, S. L. Haan, and J. H. Eberly, "Dynamics of classical slow-down collisions in non-sequential double ionization," Phys. Rev. Lett. **89**, 113001 (2002).

1. Introduction

Atoms exposed to strong laser fields have been shown [1]- [3] to undergo more double ionization (DI) than would be expected for independently ionizing electrons [4]. Subsequent experimental [5]-[6] and theoretical [7]–[12] studies using a variety of approaches have lent support to an explanation of this excess double ionization in terms of recollision, a process in which one electron is detached and then moves in the oscillating laser field so as to return to the core where a collision with the inner electron leads to double ionization. In the simplest picture, the recolliding electron simply knocks the other electron out of the atom [7], [8]. However, the energy of the returning electron is limited, and in some situations it has insufficient energy for a simple "knock-out" collision. Then the combined effects of collisional excitation [13]-[14] and a lowering of the nuclear potential-energy barrier by the oscillating laser field can lead to double ionization, often with a time lag between the recollision and emergence of the electrons. Thus, the dynamics of the recollision process is very rich, and many aspects of it remain to be explored.

In the present manuscript we perform numerical studies within a model system to consider electron collisions that occur in the direction of the laser force, and thus while the confining potential-energy barrier is suppressed. We show that these speed-up collisions exist in classical-ensemble studies of the model as well as in the quantum model. We then examine specific trajectories from the classical ensemble to learn the full sequence of events leading to double ionization from speed-up collisions. We find that in some cases there is no unambiguous single ionization event preceding the doubly ionizing collision, while in other cases there is recapture of a previously ionized electron. We then consider again the quantum model, and use the technique of quantum masking [15] in an effort to determine if quantum trajectories leading to speed-up collisions and double ionization are similar to classical trajectories.

The model that we use is the familiar aligned-electron model [16]-[21] for helium, which considers only electron motion in the direction of the laser polarization, thereby avoiding the complexity of the full three-dimensional problem [22]. In order to avoid unphysical Coulomb singularities, the potential is softened with a shielding term. In atomic units, the two-particle Hamiltonian in the presence of the laser field $E_0 f(t)\sin\omega t$ is

$$H(x_1,x_2) = \frac{p_1^2}{2} + \frac{p_2^2}{2} - \frac{2}{\sqrt{x_1^2+1}} - \frac{2}{\sqrt{x_2^2+1}} + \frac{1}{\sqrt{(x_1-x_2)^2+1}} + (x_1+x_2)E_0 f(t)\sin\omega t \quad (1)$$

where x_1 and x_2 are the positions of the two particles, and p_1 and p_2 their momenta.

As in previous works, we solve the time-dependent Schrödinger equation on a numerical grid using split-operator techniques [23]. We have used grids extending to plus or minus 200 and 400 $a.u.$, with absorbing boundaries. We start the system in the quantum ground state, and apply a 6-cycle trapezoidal (2+2+2) laser pulse. Also, in order to connect most directly with our previous works, we work here with laser intensity 6.5x10$^{14}W/cm^2$ and frequency ω = 0.0584 $a.u.$, corresponding to 13-photon single ionization and 39-photon double ionization. These parameters place the DI yield in the familiar knee region.

2. Quantum masking

Solutions (e.g.,[24]) of the time-dependent Schrödinger equation on numerical grids have revealed pulses of double ionization occurring at times when the the laser field is maximal and the confining nuclear potential-energy barrier is most suppressed. To investigate the formation of these pulses, or jets, we have employed a wave-function masking technique [15],[25]. This technique separates the full wave function at a particular time into component parts based on spatial regions, and then evolves each component independently on the full grid according to the Hamiltonian. If desired, the full wave function can be reconstituted by summing the components.

The masking technique has revealed that for our model and parameter choices the majority of the non-sequential DI arises from recollison events that occur against the laser force, i.e., while the laser force is slowing down the returning electron [15]. These slow-down collisions involve an efficient energy exchange between the electrons, a reversal in direction of motion, and escape over a suppressed potential-energy barrier. Thus, the collision occurs shortly after a zero of the laser field, but the actual escape is delayed until the barrier is suppressed.

In addition to these slowdown collisions, our results [15, 25] also show the persistent existence of double-ionization recollision events that occur with the laser force. They are visible, for example, in the third plot of Fig. 3 of Ref. [15], and in our present Fig. 1. One can expect such with-the-laser-force recollisions to be highly efficient, since the recolliding electron need simply push the inner electron over a suppressed barrier. However, a classical particle oscillating in a sinusoidal electric field will (in the absence of other forces) have a 90-degree phase

#5029 - $15.00 US Received 11 August 2004; revised 16 September 2004; accepted 20 September 2004

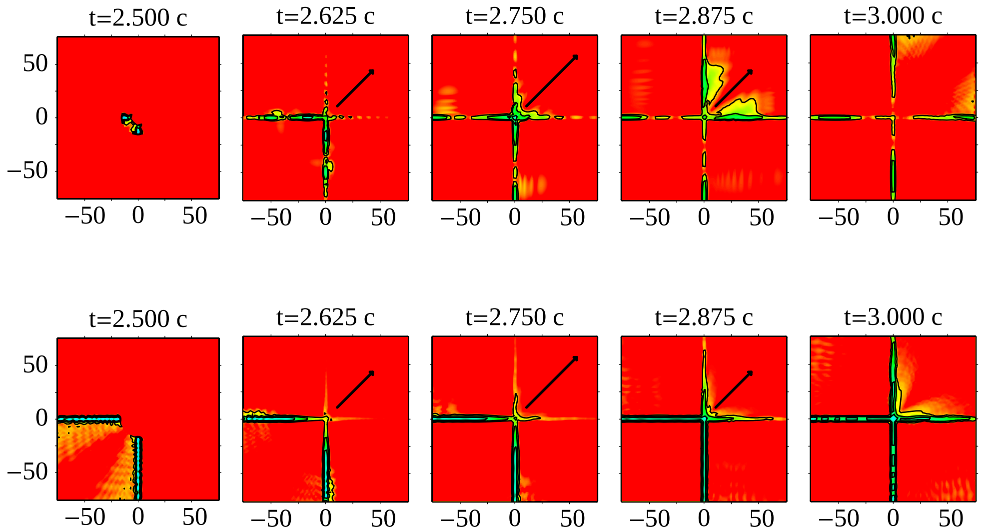

Fig. 1. Logarithmic plots of the modulus squared of the spatial wavefunction vs. positions x_1 and x_2 for the indicated times (in laser cycles). A spatial mask is applied at t = 2.5 c. In the top row the retained population lies 6 to 16 $a.u.$ from the origin and in the half-plane where $x_1 + x_2 < 0$. The bottom row keeps population that lies further an 16 $a.u.$ from the origin at t=2.5c. The laser frequency is 0.0584 $a.u.$, and the intensity 6.5x10^{14} W/cm^2. The wedge-shaped jets of DI are typical of any half-cycle during which the laser pulse is at full intensity. Contours are drawn beginning at population density 10^{-6}, with color gradations beginning at 10^{-7}.

lag between velocity and force. Thus when the force is maximal the velocity is small, hardly a good scenario for an effective recollision. The purpose of the present paper is to investigate the dynamics and origins of these “speed-up collisions.”

In Fig. 1 we show the formation of jets from speed-up collisions. The first plot in each sequence shows the logarithm of the modulus squared of the wavefunction upon application of a “ring-cut” quantum mask at t=2.5 cycles. (We use a smooth cutting function in order to reduce introduction of unwanted Fourier components.) In the upper plot, we keep all population in the $x_1 + x_2 < 0$ half plane, lying from 6 to 16 $a.u.$ from the origin. In the lower plot we show remaining population in that half plane with distance more than 16 $a.u.$ from the origin. The top row dominates the DI from speed-up collisions. At t=2.75 c, population in the region $5a.u. < x_1, x_2 < 75a.u.$ in the upper plot is 1.93x10^{-3} and the lower is 0.22x10^{-3}. The jets, most visible at t=2.875 c, propagate close to the axes, indicating one electron traveling faster than the other.

The masking technique allows for a semiclassical tracking of quantum populations, and classical studies [26] have verified that major quantum features have classical analogues. The classical studies have also provided rich insights into the dynamics of the slow-down collisions. We thus turn to a classical model for more detailed analysis of individual speed-up collisions.

Received 11 August 2004; revised 16 September 2004; accepted 20 September 2004

3. Classical filtering

The classical model uses the same aligned-electron model as the quantum study. Initial conditions are obtained [15],[26] by assigning quantum ground-state energy $E = -2.238$ $a.u.$ to a pair of pilot electrons at the origin, randomly allocating the kinetic energy between them, and then allowing them to propagate without the laser field throughout the allowed phase space. Initial conditions are determined by sampling their state at randomly chosen particular times over long time intervals. Our primary ensemble contains one million such two-particle trajectories, of which $28,004$ doubly ionize by the end of the six-cycle laser pulse. In the present work a double ionization is defined to have occurred at the end of the pulse if each electron has positive total energy, excluding the electron-electron interaction.

One advantage of classical modeling is that we can filter the ensemble for individual two-particle trajectories possessing a desired characteristic or exclude those with undesirable characteristics. We have filtered the DI trajectories for those that doubly ionize from speed-up collisions between t=2.5 c and 3.0 c. Each such trajectory is displayed as a dot in the plots of Fig. 2. The classical trajectories show strong clustering at t=2.50 c along the negative axes, and also show jets that are similar to the wedge-shaped jets observed for the quantum system, with trajectories lying fairly close to the positive axes.

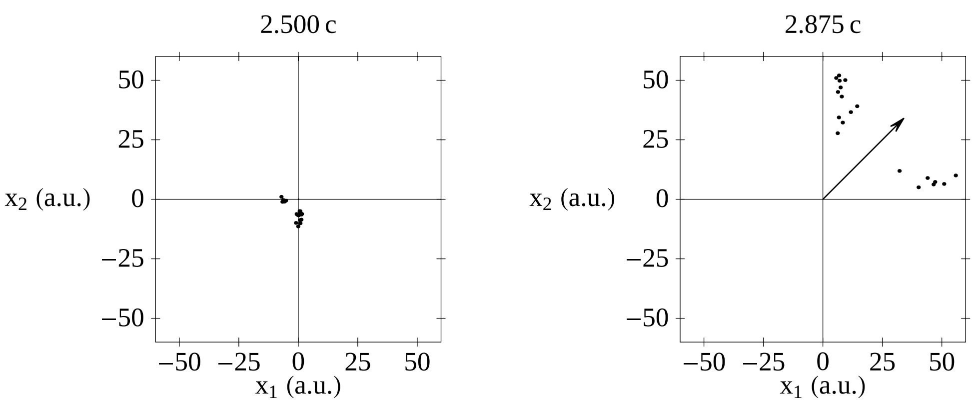

Fig. 2. Two-electron classical trajectories that doubly ionize by with-the-force collisions between $t = 2.5c$ and $t = 3.0c$. The figure on the left shows their positions at the start of the half-cycle ($t = 2.5c$). The figure on the right shows the jets of DI at t=2.875 c. These can be compared with the quantum results of Fig. 1.

Further examination of the trajectories shown in Fig. 2 reveals that most of the outer electrons (87%) have slightly negative energies as the laser field passes through zero and thus can be considered bound. We have found that the above-described clustering of trajectories in the half cycle before recollision is typical, with no DI occurring if the outer electron is more than 20 $a.u.$ from the nucleus or has energy more than 0.3 $a.u.$. Electrons that are too far from the nucleus or which have too much outward momentum simply do not return in time for an effective speed-up collision.

The discovery that the majority of the recolliding electrons in the DI speed-up collisions are bound at the previous zero of the laser cycle perhaps raises the question of what should be called "recollision." There certainly is a collision between the two electrons (as will be demonstrated more fully below), but the sequence that we are discussing is very different from the original simpleman picture [7], [8] that involves the return of an ionized electron.

Of the 28,004 total double ionizations during the pulse, only 154, just 0.55%, result from collisions with the laser force. However, with-the-force DI begins much sooner in the laser pulse than the majority of double ionization. In fact, collisions with the force contribute to more than a third of all double ionization occurring during laser turn-on, as shown in Fig. 3.

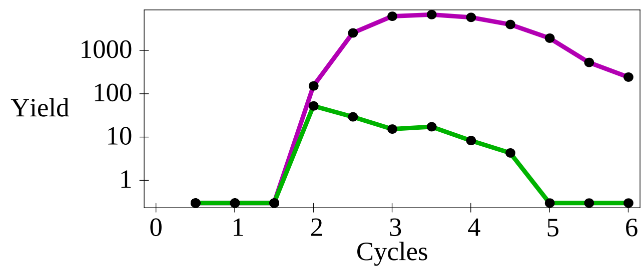

Fig. 3. Absolute yields of all DI (purple, and shown above) for each half cycle and of DI from speed-up collisions (green) below. The laser turns on from t=0 to 2c, is at full strength from t=2c to t=4c, and turns off t=4c to t=6c. The vertical scale is logarithmic, and the quantity actually graphed is yield+0.3 so that zero-yield data points can be included.

4. Analyzing jet formation

Classical analysis allows us not only to filter the trajectories but also to study them individually or collectively over the duration of the pulse. We have done that for the trajectories shown in Fig. 2 and for speed-up DI trajectories occurring in other half cycles, in an effort to determine the pathways that lead to effective speed-up collisions. We have found that there are two distinct histories that can be distinguished. These are described below, and illustrated in the animations of Fig. 4 for two trajectories that doubly ionize between t=2 c and 2.5 c. The still images in these figures are very similar–one electron is tightly bound in the nuclear well, and the other is about 6 *a.u.* away. In these plots, the electrons are shown as colored dots. The solid curves represent effective potential energies for each electron [27], and the vertical position of each dot is determined as the sum of the electron's effective potential energy and kinetic energy. Arrows in the animations show the direction and magnitude of the force from the laser.

The left animation in Fig. 4 illustrates the pathway to a doubly ionizing speed-up collision that we shall refer to as direct excitation. Both electrons stay near the core until just before the collision. They jostle each other, and their energies fluctuate. Whether single ionization occurs depends on how one defines ionization in an oscillating field. By our working definition of ionization–positive energy at a time in which the laser field is zero–single ionization does not occur. In the half cycle before double ionization, one electron almost escapes over the suppressed barrier. It hovers near the cusp of the suppressed barrier, with the laser pushing it outward but the core pulling it back. As the field weakens the electron gains potential energy, and can be described as nearly "riding the cusp" of the increasing potential-energy function. When the laser field switches direction, the electron is propelled back toward the core for a collision with the inner electron. Once it passes the inner electron it begins pushing the inner electron back the other direction. In this particular example, the inner electron does one additional oscillation in the nuclear well before escaping over the suppressed barrier. The two electrons emerge with very different speeds.

The second pathway to DI is through single ionization followed by electron reattachment. This process is illustrated in the animation on the right side of Fig. 4. An electron returns to the

#5029 - $15.00 US Received 11 August 2004; revised 16 September 2004; accepted 20 September 2004

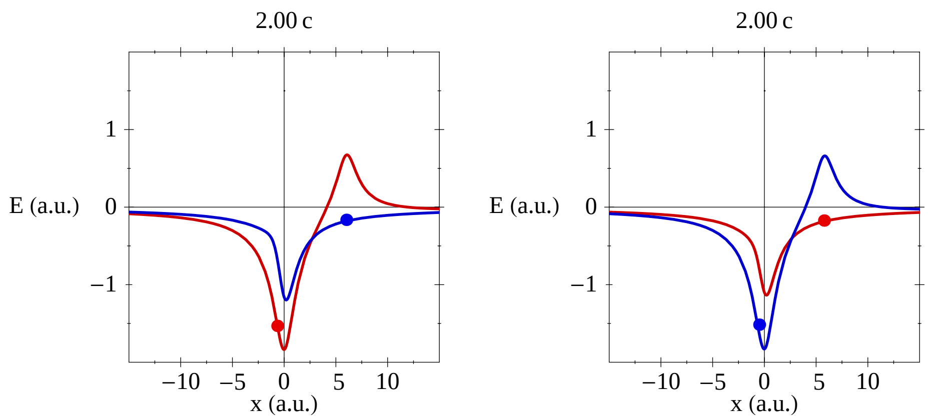

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Fig. 4. Animations of energy vs. position showing the two electrons and their effective potentials. The still images show the excited bound state at $t = 2.0c$ that leads to a with-the-field collision. Although the two plots are very similar at $t = 2.0c$, their prior evolutions are very different. One animation illustrates the direct-excitation pathway, and the other shows single ionization and recapture.

nucleus while traveling slowly (at a time when the laser field is near its maximum), loses energy to the inner electron, and reattaches. In this particular animation, the electron that just gained energy almost escapes in the direction of the laser force, and “rides the cusp” of the increasing potential-energy function to set up a speed-up collision. Again in this case one electron does one extra oscillation in the nuclear well after the final collision (although in this case it is the opposite electron). In the aligned-helium model electron-electron repulsion acts to keep the electrons from coming out together.

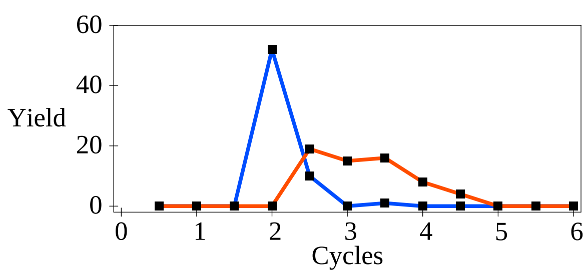

Fig. 5. This figure shows a comparison of absolute yields of with-the-force DI occurring during each half cycle via direct excitation (blue), which peaks in the half cycle ending at $t = 2.0c$, and via prior single ionization and reattachment (red).

The two pathways do not contribute equally to DI. The with-the-force DI that occurs late in the laser turn-on is dominated by direct-excitation trajectories, as shown in Fig. 5. The double ionization that occurs during full laser intensity is dominated by the reattachment trajectories. The start up of reattachment relative to direct excitation is delayed by the absence of single ionization early in laser turn-on.

Received 11 August 2004; revised 16 September 2004; accepted 20 September 2004

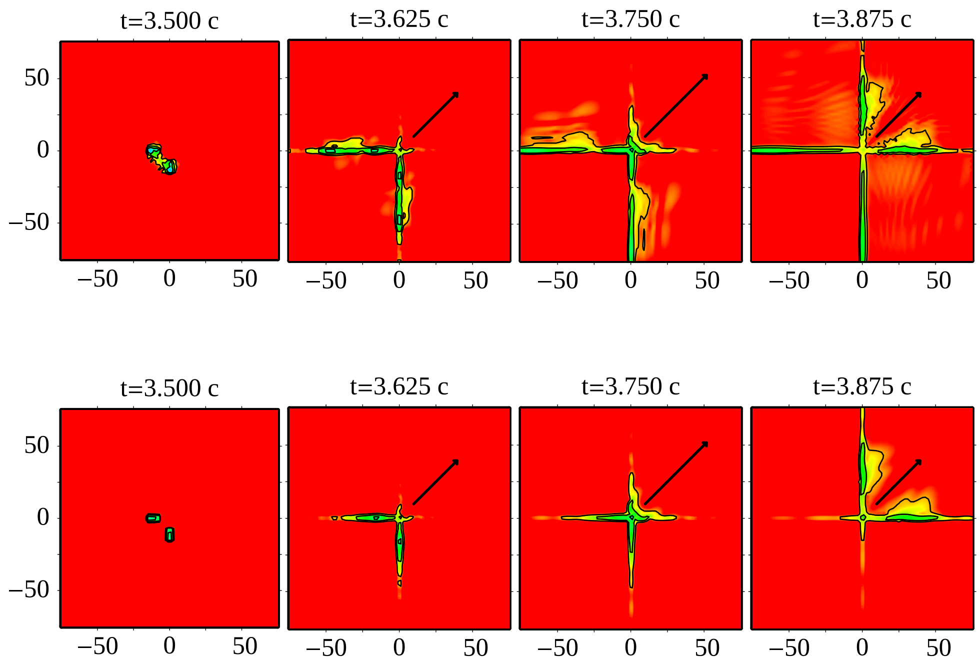

Fig. 6. Moduli squared of the spatial wavefunction vs. positions x_1 and x_2 for indicated times. In the top sequence a ring mask similar to that of Fig.1 is applied to the full wavefunction at t=3.50 c. For the second row, a series of earlier masks was also applied, so as to keep only the regions where both electrons remain bound. Clearly the full wedge jets at $t = 3.875c$ result primarily from this bound population.

5. Quantum comparison

Now that we understand these two methods of achieving DI by collisions with the laser force, we are interested in testing their relevance in the quantum case. In the direct excitation scenario, neither electron makes a large excursion from the atom. We can test the importance of large excursions prior to the formation of the quantum wedge-shaped jets by repeatedly masking any population that gets beyond a chosen distance from the origin. The effects of multiple masks can be seen by comparing the two rows of Fig. 6. In the first row, the only mask that is applied is a "ring mask" similar to that used in Fig. 1, except that it is one cycle later in the pulse. In the second row all population further than 6 $a.u.$ from the origin is masked out at t=1.0, 1.5, 2.0, 2.5, and 3.0 cycles. Strong jets are still present, despite all these masks. The total population in the region $5a.u. < x1, x2 < 75a.u.$ at t=3.875 c is $1.10\text{x}10^{-3}$, compared with $1.23\text{x}10^{-3}$ in the top row. Our conclusion is that large excursions are not important for the preponderance of the with-the-force DI.

We consider next reattachment. A possible reattachement scenario is as follows: one electron is ionized at about t=2.25 cycles. It is pushed away from the atom, but returns for recapture at about t=3.25 c, and then by t=3.5 is poised for with-the-force recollision. We can test for this sequence by keeping only outer population (radius $> 6a.u.$) at times 2.50 and 3.00 c, and then applying a ring mask at t=3.5 c as in Fig.6. Results are shown in Fig. 7. No jets are visible in quadrant 1. We infer that reattachment does not contribute significantly to the formation of the with-the-force jets shown in the other figures.

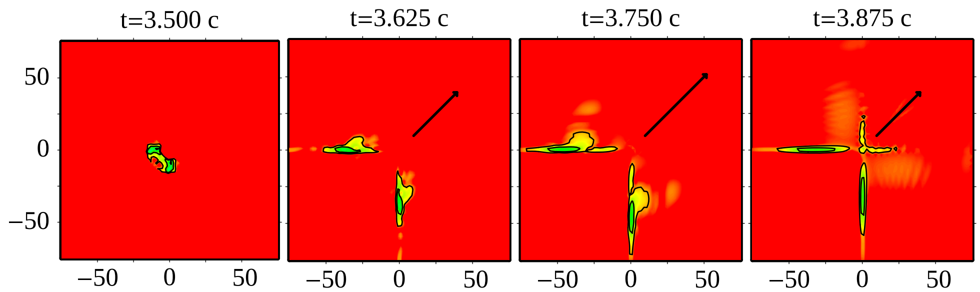

Fig. 7. Modulus squared of the spatial wavefunction vs. positions x_1 and x_2 after application of a ring mask at t=3.5c. Prior to times shown, all population within 6 $a.u.$ of the origin was masked at times 2.5 c and 3.0 c. Jets are not present.

Interestingly, some DI is visible in Fig. 7, emerging from along the negative real axes. Such emergent population is also visible in the first row of Fig. 6, but is not visible in Fig. 1, where the ring mask was applied only half a cycle after the laser reached full strength. We previously [15] have interpreted pulses of population that emerge from along the axes early in the half cycle as originating from population that has been excited through recollision, but where a time lag occurred between the recollision event and the escape of the inner electron. The time lag occurs because of the need for suppressing the potential-energy barrier, but no additional collision is needed. A similar interpretation is applicable here.

6. Summary

We have shown that in the aligned-electron model a small portion of the double ionization occurs via electron-electron collision in the direction of the laser force, for classical ensembles as well as quantum systems. By studying individual trajectories in the classical ensemble, we have learned that the speedup collisions are not high-energy recollision events but result to a large extent from excited bound states. These states may have remained bound from the start of the pulse and become excited by the laser, so that DI can occur without single ionization occurring first. Alternatively, the excited bound states may form after a previously ionized electron reattaches to the atomic core. The direct excitation mechanism is especially important during laser turn on, when it may account for one third of the total DI. Analysis of quantum systems using masking suggests that in the quantum case and for the laser parameters considered in the present work, only the laser excitation method contributes significantly. It contributes throughout the laser pulse.

The complicated dynamics of collisional processes leading to DI motivates careful study within simplified models. We have found that in our model one electron can become highly excited, and then be propelled across the inner atomic region where a collision with the other electron can lead to double ionization, without unambiguous single ionization having occurred. One can reasonably anticipate that similar behavior may be found in three-dimensional systems. In our model, the screening of the electron-electron interaction provides what is effectively a fixed impact parameter for collision. In a three-dimensional system one can expect variations in this impact parameter, but some of the three-dimensional trajectories can be expected to have small enough impact parameters for speed-up collisions to lead to double ionization. The three

Received 11 August 2004; revised 16 September 2004; accepted 20 September 2004

dimensional case will be considered more fully elsewhere.

Acknowledgments

This work has been supported by the National Science Foundation through grants PHY-009931 and PHY-0355035 to Calvin College. The authors acknowledge ongoing cooperation with J.H. Eberly's group at the University of Rochester, and especially acknowledge contributions of R. Panfili regarding computer programing for the classical model and P. Ho for a careful reading of the manuscript.

#5029 - $15.00 US Received 11 August 2004; revised 16 September 2004; accepted 20 September 2004