Classical explanation for electrons above energy $2U_p$ in strong-field double ionization at 390 nm

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A classical ensemble is employed to investigate double ionization of helium for laser wavelength 390 nm and intensity 1.1 PW/cm^2, below the threshold for recollision impact ionization. Significant numbers of trajectories that lead to electron energy above $2U_p$ are shown to be present and to feature a time delay between recollision and final ionization. Processes through which nuclear and laser forces combine to give an electron final energy above $2U_p$ are identified.

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A classical particle that starts from rest in an oscillating laser field will not only quiver but also drift from its starting point, with maximum drift speed (in atomic units) of $\sqrt{4U_p}$, where U_p denotes the ponderomotive energy. If the laser turn off does not affect the drift velocity [1], the maximum final energy of the particle will be $2U_p$. This energy matches an observed cutoff for electrons produced in single ionization of atoms by intense long-wavelength laser pulses, consistent with having electrons start with low speed after tunneling ionization [2]. However, for double ionization (DI) of atoms, logarithmic plots of energy spectra show [3,4] electrons with energies to at least $4U_p$, suggesting a richer ionization dynamic.

It is widely accepted that double ionization of atoms by long-wavelength strong-field lasers [2–5] proceeds primarily through recollision [6], a process in which one electron is pushed outward by the linearly polarized laser field, then back, to transfer energy to the other electron. Because of a phase lag between laser force and velocity of the returning electron, the most energetic recollisions occur at about the time of a laser zero. Thus, impact ionization is likely to result in two slowly moving electrons traveling against a growing laser force. Such electrons are most likely to be pushed back in the opposite longitudinal direction [7] and finish with drift speed less than $\sqrt{4U_p}$. Thus, impact ionization would not be expected to produce the higher energy electrons present in double ionization. One accepted explanation [4,8] is backscattering of one of the electrons at recollision. The backscattering changes the phase of the electron's quiver relative to the laser oscillation. In single ionization, post-ionization backscattering is known to occur in a small fraction of trajectories. In this case, the returning electron does not lose any energy to the other electron, and the phase change from post-ionization backscattering can increase electron final energy to about $10U_p$ [9].

Double ionization can occur even if the recolliding electron returns with insufficient energy for impact ionization, since there remains the possibility of collisional excitation, with subsequent ionization at the next field maximum or during a subsequent half cycle [10]. It has been shown experimentally that there is no sharp intensity threshold [11,12] for the onset of DI. A recent combined experimental and theoretical effort [13] at laser wavelength 390 nm and intensity below the threshold for impact DI revealed the production of significant numbers of DI electrons with energy above $2U_p$. The effort included a three-dimensional quantum calculation that showed one electron of a DI pair could achieve energy above $2U_p$. To explain these electrons, the authors introduced a model which treated the recollision as a series of weak electric-field impulses. They inferred that the struck electron—which could ionize only due to combined effects of the recollision and the laser field—would have energy less than $2U_p$, while the recolliding and rescattering electron could achieve greater energy.

In the present paper we employ three-dimensional (3D) fully classical ensembles [7] to examine the dynamics of the recollision process for laser wavelength 390 nm and intensity 1.1 PW/cm^2, consistent with Ref. [13] and below the threshold for impact ionization [14]. Ensemble size is 400 000. We use trapezoidally shaped pulses of five and ten cycles, with respective one- and two-cycle turn on and turn off. As we show in Fig. 1, the ensemble model also shows significant numbers of double-ionization electrons that have energy above $2U_p$. In this model, the struck electron is more likely to achieve high energy than the recolliding electron.

One candidate mechanism for the production of the high energy electrons is post-DI rescattering, in which one of the ionized electrons returns to the core and rescatters. However, a quick sorting of the trajectories reveals that this mechanism is not responsible. This suggests that the explanation for the higher-energy electrons lies in the recollision dynamics.

For each DI trajectory we have determined the recollision time (defined as time of closest approach of the electrons after the first departure of one electron from the core) and the final ionization time [15]. In Fig. 2 we consider laser phase at recollision and at final ionization for all DI trajectories. The trajectories that lead to high-energy electrons are shown in green as the lower bands. For both pulse lengths, recollisions occur over a wide range of phases for both the low- and high-energy trajectories, albeit with a significant dip about 0.15 cycles after a field zero. Ionization times are more clustered, especially for the high-energy trajectories, suggesting final ionization occurs as the confining nuclear barrier becomes sufficiently suppressed.

To investigate the time period between recollision and final ionization, we have sorted the high-energy trajectories based on whether the electrons that finish with high or low energy were bound or free, starting at the time of recollision.

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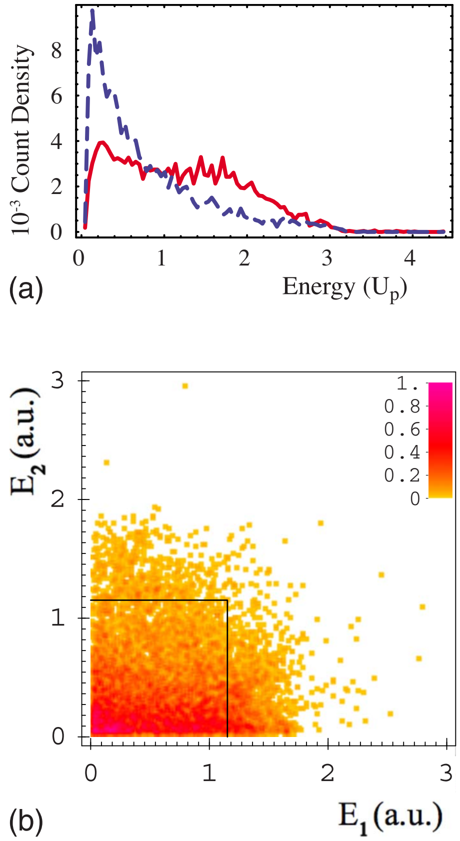

FIG. 1. (Color online) Final kinetic energies for all DI electrons from classical ensemble for laser wavelength 390 nm and intensity 1.1 PW/cm^2. On the top the count densities of the recolliding and struck electrons are plotted as the blue dashed curve and the red solid curve, respectively. On the bottom is a density plot showing the final energy E_2 of the recolliding electron vs final energy E_1 of the struck electron. Pulse length was five cycles. For these laser parameters U_p=0.58 au and the black box is drawn at $2U_p$.

Results are shown in Fig. 3. The rising magenta dotted curve indicates trajectories that have achieved double ionization. It clearly indicates the time lag between the recollision and the double ionization. The blue curve with square data points shows trajectories in which the electron that will achieve high energy is bound after the recollision, with the other electron free. The large-dashed green curve shows the converse, in which the electron that will achieve high energy is free and the other bound. The solid red curve includes trajectories in which both electrons are bound. Finally, a black curve along the bottom, visible only for the longer pulse, shows the few trajectories in which one electron is still bound and both electrons will achieve high energy.

The immediate question regards how an electron that is bound after recollision can achieve high energy. We answer this question with the example trajectory shown in the upper plot of Fig. 4 and in Fig. 5. Both plots begin just before the collision. In Fig. 4, the recolliding electron has come in from the right, and overshoots out the left. Meanwhile, struck electron loops around (or boomerangs) and exits to the right. In Fig. 5 we show a sequence of energy plots [16] for this trajectory. The first frame shows a time just before recollision. The recolliding electron, coded in blue, is traveling right to left and in the direction of the laser force. The collision is completed by the time of the second plot, with the electron that recollided overshooting the nucleus. That electron will experience a laser-induced direction change by the end of the sequence, and finish with energy less than $2U_p$. The struck electron is pushed in the $-z$ direction by the recollision, but remains bound, and the nucleus pulls it back toward the positive direction. The sign change of v_z occurs at about the time of the laser zero. The electron then traverses the nuclear well and emerges over the suppressed barrier. We refer to this process as the nuclear boomerang to distinguish from free-electron scattering and to emphasize the change in the direction of longitudinal motion.

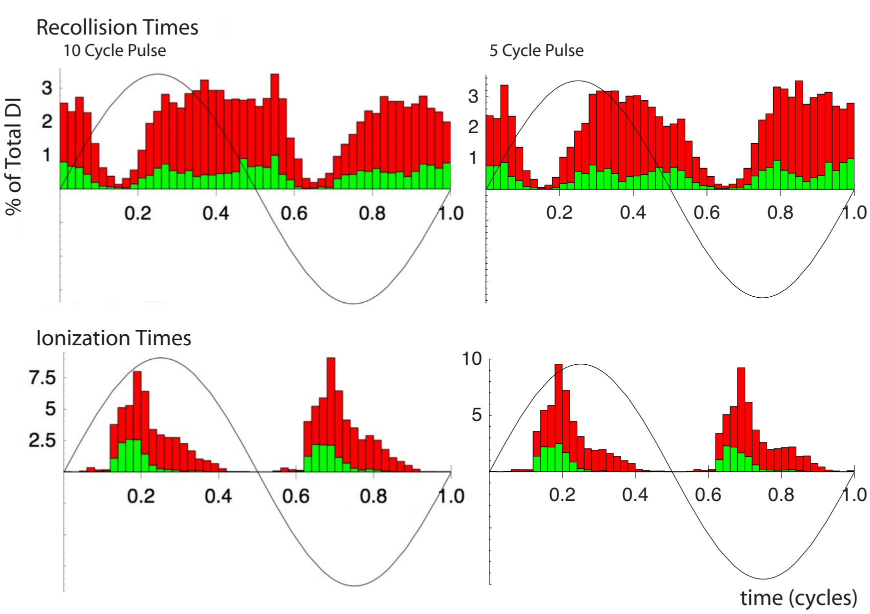

FIG. 2. (Color online) Percent of the trajectories vs laser phase for recollision (top plot) and ionization (bottom plot) times. In each plot, the lower green band indicates high-energy trajectories and the upper red band the remaining trajectories. Ionizations peak as the field grows in strength. Results are shown for both ten- and five-cycle pulses.

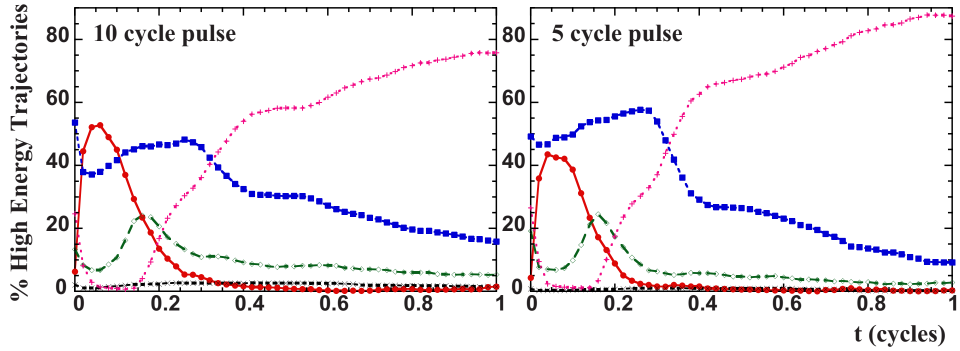

FIG. 3. (Color online) Percent of high-energy trajectories meeting specified conditions vs time since recollision for both ten- and five-cycle pulses. From top to bottom along the right: dotted magenta curve indicates both electrons are free; blue dashed curve indicates the high-energy electron is still bound but the other free; green curve with larger dashes has the low-energy electron bound but the other free. The solid red curve visible at shorter times indicates doubly excited atoms. The black dotted curve along the bottom denotes trajectories for which one electron is bound and both electrons finish with high energy.

In atomic units, an electron in one dimension exposed only to an oscillating electric force $(4U_p)^{1/2}\omega\sin(\omega t)$ has drift speed $v_d=|v_0\pm(4U_p)^{1/2}|$, where v_0 is the speed of the electron at the field zero, and with the + or − chosen based on whether the laser force after the field zero is in the same direction as v_0 or opposite. The case $v_0=0$ gives drift velocity $\sqrt{4U_p}$ and (for a linearly ramped turnoff) final energy $2U_p$. For an electron such as the recolliding one of Fig. 5 that is unbound after the collision and overshoots the core, there is partial cancellation between the two terms and final energy less than $2U_p$. The other electron, by contrast, experiences a change in direction by about the time of the field zero. This direction change has importance that goes beyond avoiding partial cancellation between v_0 and $(4U_p)^{1/2}$. The nucleus can increase the speed of the electron very early in the laser cycle and thereby influence how much work the laser does on the electron during that half cycle: $W=\int_{t_0}^{t_f}E_0\sin(\omega t)v_z dt$ depends on the instantaneous velocity, even though the impulse delivered by the laser during the half cycle is fixed. For our laser intensity and frequency, an electron that starts from rest at a field zero can achieve final energy up to $3.5U_p$ [17]. We will examine this point more fully elsewhere.

The sequence of events in Fig. 5 is very similar to the excitation and escape sequence presented in Ref. [7] for

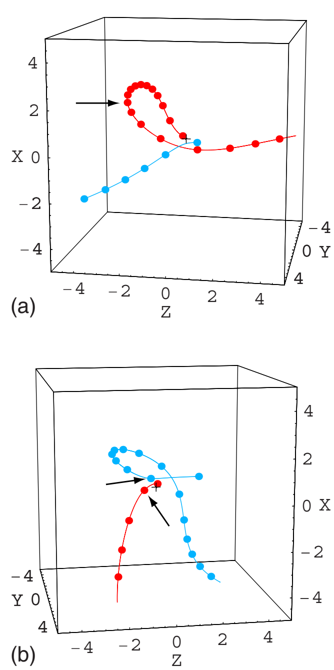

FIG. 4. (Color online) Trajectories of Figs. 5 (top) and 6. Curves begin same times as those plots, with successive dots indicating elapsed time of 0.02 cycles. Arrows show positions at zeros of the laser field. In each trajectory, the returning electron is incident from the right.

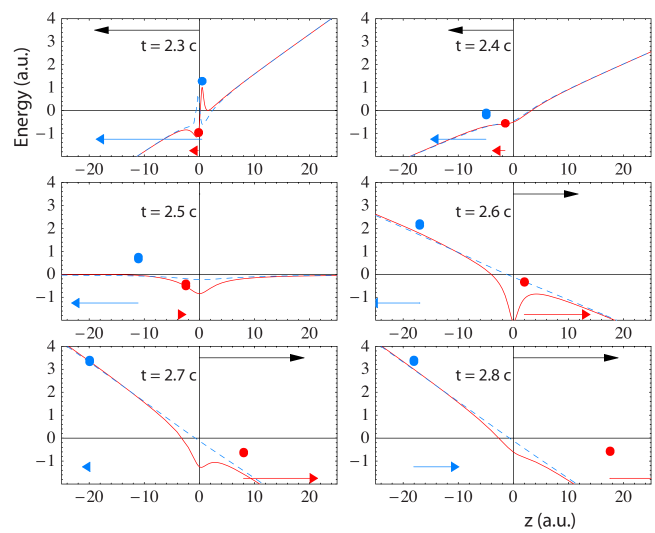

FIG. 5. (Color online) Sequence for upper trajectory of Fig. 4. Curves show effective potential energies for each electron as functions of z (the laser polarization axis). Upper arrows show laser force, and lower arrows show v_z. Vertical lengths of the "dumbbells" (more visible in Fig. 6) indicate kinetic energy for motion perpendicular to the z axis. At $t=2.5c$, the struck electron (color coded in red with solid curve) remains bound and has just started traveling back in the positive z direction. The laser force propels it over a suppressed barrier, and the electron finishes with energy greater than $2U_p$.

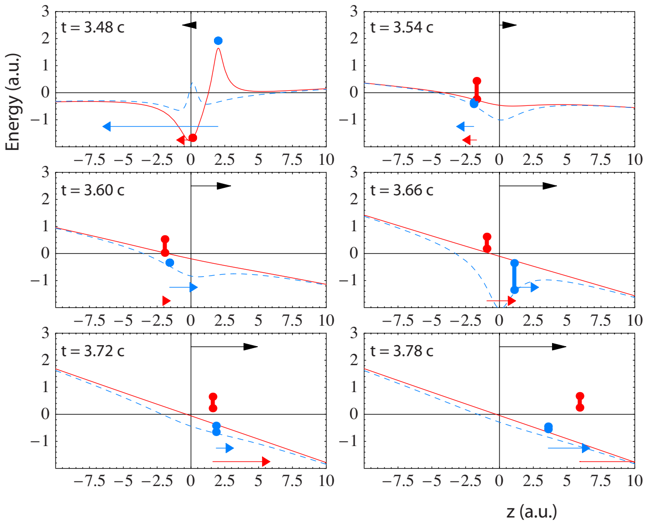

FIG. 6. (Color online) Effective-energy collage for lower trajectory of Fig. 4. The electron that is free after recollision achieves energy greater than $2U_p$. In this example, the recolliding electron is recaptured, but reionizes when the field is strong.

wavelength 780 nm. However, at the longer wavelength the electron speeds which lead to similar phase matching between electron motion and the oscillating laser field (so that a change in sign on v_z when the laser field is nearly zero is followed by over-the-barrier escape) do not lead to energies significantly above $2U_p$.

The nuclear boomerang also explains how the doubly excited complex can lead to a high-energy electron. In addition, we have found that if the two electrons ionize nearly simultaneously, then post-ionization electron-electron interaction can push one electron ahead so that it finishes with energy above $2U_p$.

The lower part of Fig. 4 and Fig. 6 show an example trajectory in which the electron that is free immediately after the recollision achieves high energy. The process here is basically nuclear scattering, although a small impact parameter is not needed. In this particular trajectory the recolliding electron is recaptured in the collision, reionizes when the field is strong, and finishes with energy less than $2U_p$. The struck electron is ionized by the collision. The growing laser field and nuclear forces combine to change the sign of v_z. The result is final energy above $2U_p$. In this particular example the electron acquires significant transverse speed (final value 0.95 au vs final value 1.34 au for longitudinal speed), but other examples show scattering into the z direction so that the longitudinal velocity exceeds $\sqrt{4U_p}$ for the rescattered electron.

The classical ensemble model clearly reveals the important role that the nucleus can play in the recollision dynamic. In doing so, the model also reveals its own weakness. In setting up the ensemble, we prevented autoionization of the classical starting state by shielding the nuclear potential. Specifically, we replaced the Coulombic potential $-2/r$ with the shielded potential $-2/(r^2+a^2)^{1/2}$, where $a=0.825$ au. This shielding can be seen as an adaptation of the classical model to account for quantum mechanics in describing the ground state. Unfortunately, this adaptation may not be best for describing the recollision process, since a shielded nucleus will not give large-angle scattering.

To investigate the importance of the shielding parameter, we have adjusted the model by reducing the nuclear shielding to $a=0.01$ au once one electron achieves distance 5 au from the nucleus. To conserve total energy, we give each electron a compensating boost in radial kinetic energy. We find that the key ideas are not altered: there is still a time delay between recollision and final ionization (as would be expected from energy considerations) and the same basic processes occur. However, their relative importance changes, with nuclear scattering of the free electron becoming much more important [18]. This is not surprising, since a less shielded nucleus can scatter electrons into the longitudinal direction better. In addition, with reduced shielding, either electron—recolliding or struck—is almost equally likely to achieve high energy. A full analysis of the dependence on the shielding parameter will be presented elsewhere. It is not obvious *a priori* how much nuclear shielding classical models should employ at recollision. It can be argued that the uncertainty principle mandates that tightly bound electrons see a smoothed potential, while the equivalence of classical and quantum Coulomb scattering indicate free electrons see an unsmoothed potential. The recollision situation is intermediate between these.

We emphasize that there is considerable variety among trajectories, as indeed must be the case given the variety of recollision times shown in Fig. 2. For example, some trajectories feature multiple collisions, and some have a multiple-cycle time delay between recollision and final ionization. Nonetheless the trajectories we have shown illustrate key ideas and the basic processes involved in producing high-energy electrons.

In conclusion, we have found that classical ensemble model is consistent with Ref. [13] with regard to the production of significant numbers of DI electrons with energy above $2U_p$. In almost all trajectories that lead to high energy at least one electron remains bound for at least a portion of a laser cycle after recollision, and during this time the electron which will achieve high energy experiences a sign change in v_z (where z is the laser polarization axis). We have identified important processes that lead to high energy in the model. One involves the anticipated process of nuclear scattering of a free electron at recollision. The second process is an excitation-boomerang-escape sequence in which the electron that is bound after the recollision changes sign in v_z, escapes over a suppressed barrier, and achieves high energy. In both sequences there is a change of phase of electron oscillation relative to the laser phase. A less common mechanism is electron-electron interaction after nearly simultaneous ionization from a doubly excited state.

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[14] The maximum return energy can be estimated as $3.17U_p = 1.83$ au. For nuclear shielding parameter $a = 0.825$ (described in the text), our well depth after single ionization is -2.4 au, below the energy -2.0 au of the helium ion. For the electron-electron interaction we use shielding parameter 0.05 au.

[15] As in Ref. [7] we define an electron to be ionized if its energy (including the *e*-*e* interaction but excluding the laser) is greater than zero, has both $z^2 > 5$ au and z component of the net force away from the nucleus (thus being outside the well), or has $z^2 > 100$ au.

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[17] When the shielding parameter is reduced, there is a small starting region around $z = -5.6$ au that leads to final electron energy $3.5U_p$. If one allows initial perpendicular velocity $v_x = 0.1$, then there is a larger starting region near $z = -5.6$ au and $x = -1.65$ for which the electron scatters off the nucleus into the z direction, and finishes with energy $3.4U_p$.

[18] If we examine high-energy trajectories $0.10c$ before final ionization, we find that as the Coulomb shielding is reduced the percentage of high-energy trajectories in which the high-energy electron is bound and the low-energy free drops from 56 to 15 %, and the percentage in which the high-energy electron is free and the low-energy bound increases from 23 to 68 %. Also, the number of trajectories in which both electrons achieve high energy increases from 2 to 10 %. The remaining 7% of the high-energy trajectories are doubly excited at the chosen time.