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Comparing classical and quantum dynamics of strong-field double ionization

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Abstract: We compare quantum mechanical and fully classical treatments of electron dynamics accompanying strong field double ionization. The major features seen in quantum mechanical simulations, including the double-ionization jets, are reproduced when using a classical ensemble of two-particle trajectories.

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

References and links

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In a semi-classical description, a photoionized electron can be envisioned as oscillating in the electric field of the photoionizing laser. In some cases, the electron can recollide with the atomic core [1, 2], possibly exciting or ionizing a second electron. Recent results of Ivanov [3] suggest that a semi-classical description can give good accounting of the total double ionization yield in multi-electron systems. Here we present a parallel examination of exact wave functions and classical trajectories for a two-electron quantum system for the frequency and intensity regime associated with the well-known correlation "knee." These comparisons have the ability to support or refute interpretations

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that are made in classical particle terms and shed light on the parameter range where classical interpretations of double photoionization are applicable.

As we have done before [4], we use a one-dimensional model to investigate double ionization. This model allows a full solution of the time-dependent Schrödinger Equation on a numerical grid [5] without needing to make ab initio assumptions regarding what will be important. A straightforward numerical solution is possible without any specific theoretical interpretation, classical picture, or perturbative orientation. The solutions provide evidence that can be compared with analytical calculations [6] as well as with 3-D numerical simulations [7], which for the time being are mostly restricted to much higher energy photons than are employed here.

For these reasons, the one-dimensional model is a flexible platform for developing and testing new ideas. In the present manuscript, the capabilities of this journal are utilized to display the time evolution of a two-electron wave function in slow-motion movie format as double ionization occurs. In atomic units (a.u.), our model atom has the Hamiltonian:

$$H(x,y) = \frac{p_x^2}{2} + \frac{p_y^2}{2} - \frac{2}{\sqrt{x^2+1}} - \frac{2}{\sqrt{y^2+1}} + \frac{1}{\sqrt{(x-y)^2+1}} + (x+y)E(t)\,, \qquad (1)$$

where x and y denote position variables for the electrons, and $E(t) = E_0 f(t) \sin \omega t$ is the electric field of the laser. E_0 is the peak field strength, ω is the laser (angular) frequency, and $f(t)$ is a pulse-shape function which we take to be trapezoidal.

Our quantum mechanical simulations start with the atom in the ground state and the Schrödinger Equation is propagated in time using a spectral method. Representative results are shown in Fig. 1 and its associated movie. The laser frequency is 0.0584 a.u., corresponding to 13-photon single ionization and 39-photon double ionization in our model. An equivalent ionization process would require 2.04 eV photons in the context of physical helium. The maximum laser intensity is $6.5 \times 10^{14}\,\mathrm{W/cm^2}$, which gives appreciable single and double ionization [8]. The time development of electron probability $|\Psi(x,y)|^2$ is plotted on a logarithmic scale during a four-cycle trapezoidal pulse (one cycle linear ramp on, two cycle plateau and one cycle linear ramp off). Population along the axes in Fig. 1 indicates one electron is near the origin and the other is far away and is a signature of single-electron excitation or ionization. Population moving out into the various quadrants indicates double ionization.

There are two double-ionization processes apparent in Fig. 1. Some population can be seen emerging in bursts directly from the axes [4]. These bursts have the interesting characteristics of being limited in their distance from the origin and of having electrons emerge in opposite directions. These characteristics suggest that these should be thought of as anti-correlated population bursts. One can also see double-ionization jets in which population emerges directly from the origin into the third quadrant (during the second quarter of each cycle) or into the first quadrant (during the fourth quarter of each cycle), corresponding to having both electrons emerge nearly simultaneously and on the same side of the nucleus. These jets emerge just after the bursts, and in the same direction as the electric force of the laser ($F(t) = -E(t)$).

We demonstrate next the degree to which classical simulations reproduce quantum mechanical results for one-dimensional helium. We start with the movie and still image found in Fig. 2 which displays classical results for the same conditions as in Fig. 1. Each dot in the classical simulation represents the trajectory of a two-particle system. The degree of similarity between the classical and quantum mechanical simulations is remarkable, with the major dynamical features of double-ionization in the quantum mechanical case reproduced classically. Specifically, we see the same two double-ionization processes previously mentioned now appearing in a classical framework. We interpret

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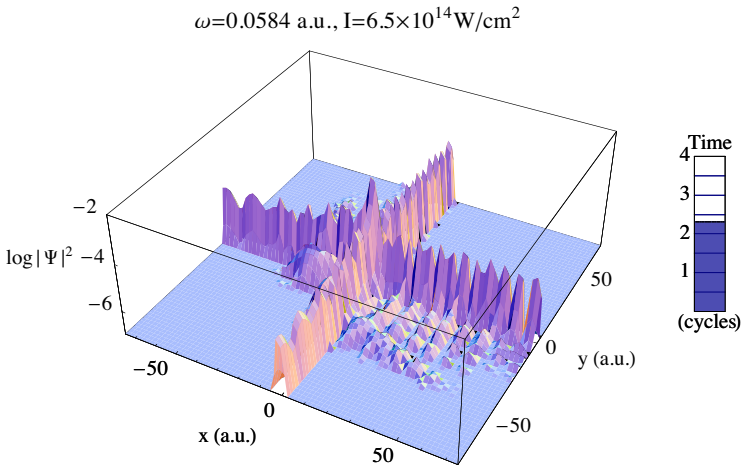

Fig. 1. $|\Psi(x,y)|^2$ during a four-cycle trapezoidal (1+2+1) pulse with laser frequency 0.0584 a.u. and intensity 6.5×10^{14} W/cm^2. The scale is logarithmic, beginning at 10^{-7}. The still image shows an ionization burst in the fourth quadrant and jets emerging into the third quadrant early in the third cycle. The animation (.7MB) shows the time development for the full pulse.

trajectories moving along an axis in position space as single ionization. Trajectories far from both axes are associated with double ionization.

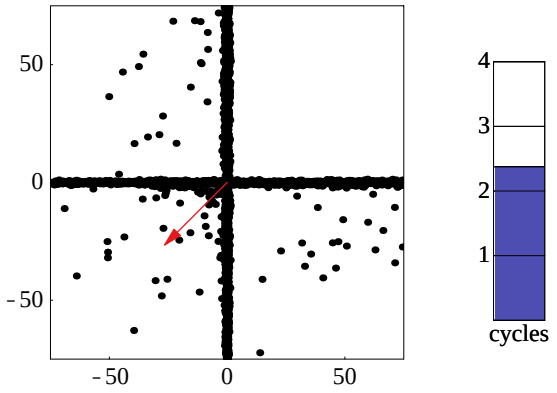

Fig. 2. (1.4 MB) Position space distribution for a particular numerical experiment using a classical ensemble and the same laser parameters as in Fig. 1. The red arrow indicates the direction and magnitude of the electric force.

In order to establish that the classical-quantum correspondence is not simply fortuitous, we allow for alternative pulse parameters. In Fig. 3 we show results of another sequence of simulations for a different pulse shape and frequency. There, we choose a laser frequency of $\omega = 0.1837$ a.u. corresponding to five-photon single ionization and 13-photon double ionization. The similarity between quantum and classical dynamics is clear. The spatial extent of population moving along each axis is nearly identical, as is the timing of the appearance of double ionization bursts and jets.

These similar dynamics were obtained by numerically solving two different types of equations whose equivalent behavior is not guaranteed from the outset. In the quantum mechanical situations, we solve Schrödinger's equation using the Hamiltonian in Eq. (1).

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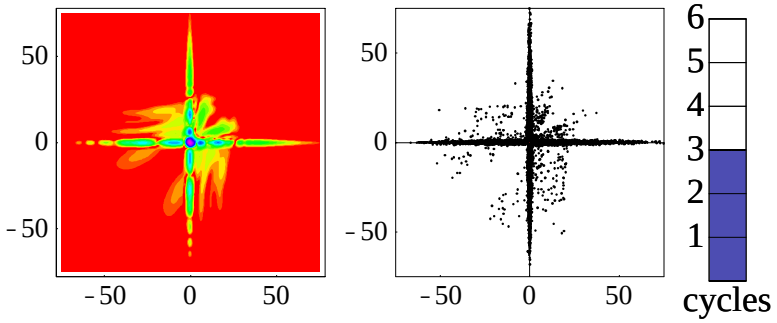

Fig. 3. (1.6 MB) Position space distribution for a particular numerical experiment using both classical and quantum simulations. The laser frequency is $\omega = 0.1837$ a.u. and the peak intensity is $6.5 \times 10^{14}\mathrm{W/cm}^2$. The pulse envelope is six cycles long with two cycle linear ramp on and off and a two cycle plateau. The time chosen for the still image is more than one-half cycle later than those of Figs. 1 and 2. As a result, the double-ionization jets appear on the opposite side of the nucleus.

The analogous classical problem involves solving a set of coupled differential equations for the forces on charged particles ($\vec{F}(x,y) = -E(t) - \nabla V(x,y)$). In atomic units, these equations are:

$$\ddot{x} = -E(t) - \frac{2x}{(x^2+1)^{3/2}} + \frac{(x-y)}{((x-y)^2+1)^{3/2}} \tag{2}$$

$$\ddot{y} = -E(t) - \frac{2y}{(y^2+1)^{3/2}} - \frac{(x-y)}{((x-y)^2+1)^{3/2}}\,. \tag{3}$$

The numerical solution method for coupled ordinary differential equations is well understood. Some care should be taken because the potentials are nonlinear functions of the dependent variables (x and y), but other than that one complication these equations can be solved using any of a number of well-known techniques.

Both classical and quantum simulations require us to provide initial values. In the quantum mechanical case the initial value we choose is the ground state wave function. This function is distributed in position and momentum space but has a definite energy. This serves as the starting point for creating a classical ensemble. We fix the energy of every two-particle trajectory in the ensemble to the ground state energy of the quantum system. The ensemble is populated by first placing a two-particle trajectory at the origin and randomly distributing the remaining energy between the two particles in the form of kinetic energy. This trajectory is allowed to move without field in the potential and the n^{th} element of the initial ensemble is generated by finding the position and momentum of the original trajectory at time $n\Delta t$. The time step Δt is chosen large enough to ensure that the entire region of space that is energetically allowed is filled. Fig. 4 shows an initial state of this ensemble using 10,000 trajectories.

We do not need to be overly concerned with just how we populate our initial state. As long as all the trajectories within the ensemble have a fixed energy, we can easily redistribute them into equilibrium using a relaxation method. As an example, we take an initial collection of particle pairs populated using a gaussian probability distribution in position space. Any particle pair in the classically forbidden region is discarded. The

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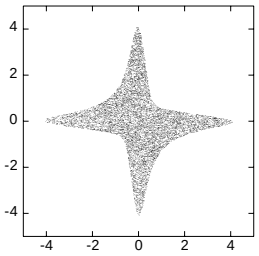

Fig. 4. Initial distribution of 10,000 two-particle trajectories in position space.

outcome is a distribution with some similarity to our original ensemble, but more sharply peaked at the origin. If we take this initial distribution and allow it to evolve freely, this non-equilibrium feature quickly is smoothed out. We show such a free evolution in Fig. 5 where population initially focused near the origin quickly disperses and by 10 a.u. of time is no longer visible. Further free evolution reveals nothing more than random fluctuations.

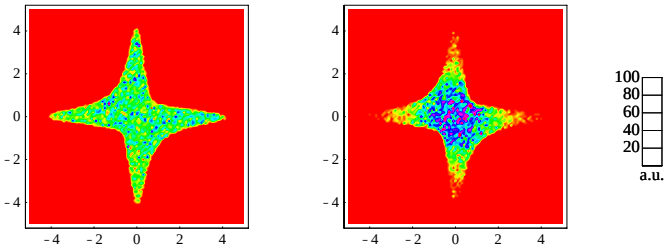

Fig. 5. The left hand still image shows an initial distribution of 10,000 two-particle trajectories populated by the method described in the text. The right hand still image shows a collection of 10,000 two-particle trajectories populated using a Gaussian probability distribution. The movie associated with this picture (2.5 MB) shows the free evolution of this collection. The data have been smoothed on a uniform grid of spacing $\delta x = 0.1$ a.u. to generate these contour plots. An atomic unit of time, in terms of fundamental constants is $t = 2\pi\hbar/2|E_g|$, where E_g is the ground state energy of the hydrogen atom. It is on the order of 10^{-17} s.

We have shown that a classical ensemble reproduces the basic dynamical features of a quantum mechanical two-electron system. When solving a one-dimensional two-electron problem, the computational time saving is an order of magnitude over the quantum calculation. Even more dramatic time savings are found as the complexity of the system increases. This makes a classical ensemble an ideal candidate for exploring multiple-ionization yields for more complex atoms or performing three dimensional calculations of helium on workstation-size machines.

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