Correlations between the final momenta of electrons and their initial phase-space distribution during photoionization

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Abstract
We present both full quantum mechanical and semiclassical calculations for the photoionization of a hydrogen atom induced by a few-cycle linearly polarized infrared laser pulse. As a quantum treatment, we applied the direct integration of the time-dependent Schrödinger equation. In the three-dimensional semiclassical trajectory Monte Carlo approximation, it is assumed that the wavepacket propagation in the post-tunneling process can be well described within the classical framework. By comparing the results of two methods, we found good agreement in the final momentum distributions of the emitted electrons for longer exciting pulses. This allowed us to use the semiclassical approximation and clearly identify and separate the initial conditions that correspond to various regions in the final momentum distributions of the emitted electrons. We also illustrate the corresponding regions with typical electron trajectories.

Keywords: photoionization, semiclassical approximation, few-cycle pulses

(Some figures may appear in colour only in the online journal)

1. Introduction

Understanding the ionization process during atomic collisions is fundamental, both from the experimental and theoretical points of view. Especially, it is a challenging theoretical task to describe the ionization cross sections near the threshold region, when, for example, a strong laser field interacts with atoms or molecules. In this case, we have to treat a highly nonlinear phenomenon. For the theoretical description, one can apply the direct numerical solution of the time-dependent Schrödinger equation (TDSE) (see, e.g., [1–3]), or use the semiclassical approximations (SCA) [4–11]. In the semiclassical model, at first an electron tunnels out of atoms or molecules, and it moves towards a detector either in the laser field or in the combined field generated by the laser field and the remaining target ion. The SCA approach is similar to the classical trajectory Monte Carlo (CTMC) method, and it is based on the inclusion of the classical phase information of the motion. Over the past years, SCA has been widely used for investigation of laser-atom collisions, partly because it is much simpler than any other quantum treatment of the problem, and it provides also the possibility of the visualization of the electron trajectories. This fact is true even when a large number of electron trajectories have to be determined, typically 100 million, for the accurate description of the tunnel ionization.

The SCA method already demonstrated effectiveness in many applications. Among them, it was shown to be able to...
explain the cutoffs in high-order harmonic generation (HHG) [12, 13], the high-order above-threshold ionization (ATI) spectra [14], the maximum angles in the photoelectron angular distributions [15], and also the characteristic momenta of recoil ions in the nonsequential double ionization (NSDI) [16–20]. In the original formulation, the SCA approach neglects the effect of the Coulomb potential of the parent ion on the electron motion after ionization. Applying a more precise description, by taking into account the interaction with the remaining target ion, the so-called Coulomb focusing effect was observed [21]. Nowadays, the focus of the research turns toward the description of the low-energy electron structures in strong infrared pulse induced ionization of atomic targets, see [22–33].

Recently, in a combined experimental and theoretical work, the strong-field ionization of lithium was studied [34], and although a qualitative agreement between experiment and theory was found, there were significant deviations in the photoelectron angular distribution and in the ATI patterns. Arbo and coworkers [35] analyzed the angular and momentum distribution of electrons ionized by few-cycle laser pulses in the transition regime from multiphoton absorption to tunneling by solving the time-dependent Schrödinger equation and by a classical trajectory Monte Carlo simulation with tunneling (CTMC-T). This theoretical work encouraged the experimentalists to measure the momentum-space images of low-energy electrons generated by the interaction of short intense laser pulses with argon atoms at high intensities. CTMC-T has already been used in the study of nonadiabatic effects in tunneling ionization of atoms in elliptically polarized laser fields [36]. When the interference of the quantum mechanical states cannot be ignored, it can be taken into account by using the lowest order approximation of the appropriate Feynman path integral [37]. A similar approach, but disregarding the Coulomb potential, was used to investigate the holographic interference patterns in strong-field ionization of H [38] and of N₂, O₂, and CO₂ [39]. Very recently, the SCA method has been applied to the identification of resonance structures in the low-energy part of the photoelectron spectra [40] and to the study of the nonadiabatic subcycle electron dynamics in orthogonally polarized two-color laser fields [41]. As an important improvement of SCA, Shvetsov-Shilovski and coworkers [42] developed a semiclassical two-step approach (SCTS) for strong-field ionization that describes quantum interference and accounts for the Coulomb potential beyond the semiclassical perturbation theory.

In the present work we calculate the momentum distribution of the electrons that are emitted from the hydrogen atom in the tunneling regime. The calculations to be presented below use both full quantum mechanical and semiclassical methods. We applied the direct integration of TDSE [38, 43, 44] and the semiclassical trajectory Monte Carlo (SCTMC) method [37, 42, 45, 46]. Although the full quantum mechanical treatment is the ‘ultimate’ way of describing atomic processes, sometimes it does not provide an intuitive picture. In other words, all physically relevant (or meaningful) questions could, in principle, be answered in terms of quantum mechanics, but the solution of the TDSE (especially when it can only be done numerically) may not reveal the physical mechanisms responsible for the observable effects. This is the point, when an appropriate semiclassical method, that is proven to be able to deliver measurable results close to the predictions of quantum mechanics, can be very useful. In this case, using the semiclassical model, we can ask and answer questions that, in extreme cases, can even be incompatible with the laws of quantum mechanics, but still add an important contribution to our understanding of the investigated physical process. In this way, the semiclassical model helps developing a clear physical picture which, finally, should be compatible also with quantum theory. Along this line, in the first part of this paper, we show that the full quantum mechanical model and the semiclassical approach provide very similar results for the momentum distribution of the liberated electrons in the process of tunnel ionization. Based on this, later on we focus on the semiclassical model and investigate the correlations between the initial phase-space positions of the (bound) electrons and their final momentum distribution. This allows us to assign an intuitive picture to the ionization mechanism, by identifying the initial conditions that correspond to the well separated interference maxima in the final momentum distributions. We also present typical electron trajectories from various regions of the momentum distribution. Exploiting the fact that it is straightforward to switch on/off the interaction with the core during the calculations, we demonstrate the strong influence of the field of the remaining ion on the outgoing electron trajectories. Atomic units are used throughout the paper unless indicated otherwise.

2. Theory

We use a few-cycle linearly polarized infrared laser pulse in our simulations. The polarization vector of the field is assumed to point along the z axis. Correspondingly, the only non-vanishing component of the vector potential is A_{z}, which (in the Coulomb gauge) has the following form:

\[ A_\text{z}(t) = -(F_0/\omega) \sin^2(\pi t/\tau) \sin(\omega t + \varphi_{\text{CEP}}). \]  

(1)

The \( \sin^2 \) envelope function above is assumed to be zero when \( t < 0 \) or \( t > \tau \), see [47].

The time dependence of the electric field \( F \) corresponding to the few-cycle laser pulse can be expressed by the vector potential. Using equation (1), we obtain:

\[ F(t) = F_1(t) + F_2(t), \]

(2)

with

\[ F_1(t) = F_0 \sin^2(\pi t/\tau) \cos(\omega t + \varphi_{\text{CEP}}), \]

(3)

\[ F_2(t) = \frac{\pi F_0}{\omega \tau} \sin(2\pi t/\tau) \sin(\omega t + \varphi_{\text{CEP}}), \]

(4)

where \( \omega \) is the angular frequency corresponding to a central wavelength of 800 nm and \( \tau \) is the temporal length of the n-cycle pulse, which is given by \( \tau = 2\pi n/\omega \). Note that this definition of the cycle number \( n \) is different from the usual
one that is based on intensity full width at half maximum (FWHM). Additionally, unless otherwise, we use \( \tau = 21.4 \text{ fs} \) (885 a.u.) corresponding to 7.8 fs (322 a.u.) intensity FWHM \( (n = 8) \) and \( F_0 = 25 \text{ GV/m} \) (0.05 a.u.). These parameters are experimentally achievable using current femtosecond amplifiers [48]. We assume \( \varphi_{CEP} = 0 \).

Figure 1 shows a few examples of the electric field waveforms to be used in the following calculations.

2.1. Time-dependent Schrödinger equation

The time-dependent Schrödinger equation of the system can be written as:

\[
\frac{i}{\hbar} \frac{\partial}{\partial t} \psi(\mathbf{r}, t) = H(t) \psi(\mathbf{r}, t),
\]

where the Hamiltonian takes the form

\[
H(\mathbf{r}, t) = H_0(\mathbf{r}) + H_1(\mathbf{r}, t) = \left[ -\frac{\Delta}{2} - \frac{1}{r} \right] + F(t) z
\]

in the coordinate representation. The Hamiltonian in (6) was decomposed into a free atomic \( H_0 \) and an interaction term \( H_1 \). We note that the position vector \( \mathbf{r} = (x, y, z) \) corresponds to the relative (electron–nucleus) coordinates, i.e., we are working in the center of mass frame. Additionally, although we use length gauge here, the fact that \( F(t) \) originates from a vector potential \( (1) \) that is zero both before and after the pulse, the gauge dependent effects are avoided. The light-atom interaction described by the inter interaction term \( H_1 = F(t) z = F(t) r \cos \theta \), which is written by using the dipole approximation, that can be shown to be valid in the parameter range considered here [49, 50].

Various numerical methods can be used to solve the TDSE as a partial differential equation (see for example reference [51]). For our purposes, the most efficient approach was found to be based on combining spherical harmonics expansion and a finite difference (FD) method [1, 12, 52, 53]. With the use of the alternating direction implicit (ADI) method [54], the time evolution of the system takes the following form:

\[
\exp[-2\tau i\hat{H}] \approx [1 + i\tau \hat{H}_0]^{-1}[1 + i\tau \hat{H}_1]^{-1} \times [1 - i\tau \hat{H}_0][1 - i\tau \hat{H}_1],
\]

where \( \tau \) denotes the half step size in time. In our calculations, the wave function \( \psi(\theta, \phi, t) \) was expanded in terms of the spherical harmonics:

\[
\psi(\theta, \phi, t) = \sum_{\ell=0}^{\ell_{\text{max}}} \frac{\Phi_\ell(r, t)}{r} Y_\ell^0(\theta).
\]

Due to the cylindrical symmetry, we can restrict our calculations to \( m = 0 \) (which means that \( \psi \) has no \( \phi \) dependence). Note that equation (8) means a separation of the \( 1/r \) dependence of the wave function, resulting in the following equations for \( \Phi_\ell(r, t) \):

\[
\hat{H}_0 \Phi_\ell(r, t) = \left[ -\frac{\Delta}{2} \left( \frac{\partial^2}{\partial r^2} - \frac{\ell}{r^2} \right) - \frac{1}{r} \right] \Phi_\ell(r, t).
\]

Since \( \hat{H}_1 \) does not contain derivatives with respect to \( r \) (the interaction \( \hat{H}_1 = \hat{H}_1 \) depends only on the \( z \) coordinate) the nonzero matrix elements are well known, and can be expressed using

\[
\langle Y_\ell^0|\cos \theta|Y_{\ell'}^{0} \rangle = \frac{\ell + 1}{\sqrt{(2\ell + 1)(2\ell' + 3)}}.
\]

The expansion (8) together with the coefficients above results in a system of coupled differential equations representing the TDSE (5).
where the equation takes a separable form. It is convenient to introduce parabolic coordinates, where

\[
\eta = \frac{2l_p}{3F} + 2 \sqrt{\frac{p}{3}} \cos \left( -\sqrt{\frac{3}{p}} \arccos \left( \frac{3\eta}{2p} \right) \right),
\]

\[
p = \frac{4\beta_2}{F} - \frac{1}{3} \left( \frac{2l_p}{F} \right)^2, \quad q = \frac{1}{F} + \frac{8l_p\beta_2}{3F^2} - \frac{2}{27} \left( \frac{4\beta_2}{F} \right)^3.
\]

For the first step of our calculation we need to describe the tunneling mechanism. To this end a probability weight

\[
w(F, v_\parallel) = w_0(F) \frac{v_\parallel}{F^2} \exp \left( - \sqrt{\frac{2l_p}{F}} \frac{v_\parallel^2}{F} \right)
\]

is assigned to the trajectory, where

\[
w_0(F) = \frac{4}{F} \exp \left( - 2 \sqrt{\frac{(2l_p)^2}{3F}} \right),
\]

and \(v_\perp\) is the component of the initial velocity that is perpendicular to the polarization direction \(v_\parallel\) is the parallel one, which is assumed to be zero \([45]\). The equation \((19)\), can be derived by using the Landau-Dykhne adiabatic approximation \([58, 59]\).

For the second step of our model, we have to propagate the electrons ‘born’ in the first step according to the Newtonian equations of motion

\[
r = -\frac{\mathbf{r}}{r^3} - \mathbf{F}(t)
\]

and we also have to assign a phase to each trajectory according to the following equation \([41]\):

\[
\Phi(t_0, v_{\parallel 0}) = -\int_{t_0}^{\infty} \left[ \frac{v_{\parallel}^2(t)}{2} - \frac{2}{r(t)} + I_p \right] \, dt,
\]

that can be derived by investigating the lowest order contribution of the Feynman path integral \([37]\) (a similar calculation can be found in the appendix of \([45]\)). Finally, we have to calculate the asymptotic momenta of the electrons according to Kepler rules. Here, we denote the angular momentum by \(\mathbf{L}\) and the Laplace–Runge–Lenz vector by \(\mathbf{R}\) \([60]\). Then we can write:

\[
p = \frac{p^2}{2} - \frac{1}{2} \mathbf{L} \cdot \mathbf{x} - \frac{F}{1 + \frac{p^2L^2}{2}},
\]

\[
p^2 = \frac{p_{f}^2}{2} - \frac{1}{2}, \quad \mathbf{L} = \mathbf{r}_f \times \mathbf{p}_f, \quad \mathbf{R} = \mathbf{p}_f \times \mathbf{L} - \frac{\mathbf{r}_f}{r_f}.
\]

During the evaluation, we coherently add the final probability amplitudes for each bin of the final momentum distribution and then take the norm. For the energy bin around \(p\), the following formula holds for the corresponding probability:

\[
P(p) = \sqrt{\sum_{j} w(t_{i0}, v_{\parallel j}) \exp \left[ i\Phi(t_{i0}, v_{\parallel j}) \right]}.
\]
3. Results and discussion

We determine the ionization probability densities of the H atom as a function of the deliberated electron’s momentum parallel \(p_z\) and perpendicular \(p_r\) to the polarization vector. Figure 2 shows the above-threshold ionization probability densities for different values of the number of cycles, \(n\), of the exciting infrared laser pulse. We found very strong forward-backward asymmetry in the momentum distribution for low values of \(n\). At the same time, the interference structure is considerably more rich for the TDSE approach than for SCTMC, but still we can identify the corresponding fan-like structure in SCTMC distributions also. We note that we found significantly more electrons in the backward direction for \((z < 0)\) \(n = 1\) than in the forward direction \((z > 0)\). On the contrary, for \(n = 2\), most electrons are emitted in the forward direction. According to figure 1, these observations are in agreement with the expectations. The ionization probability, as given by (20), is a strongly increasing function of the magnitude of the electric field. For \(n = 1\) the electron emission is dominated by negative field values, therefore the final momentum distribution corresponds to mostly backscattered electrons. Similarly, for \(n = 2\), the electron emission is dominated by positive field values, leading to final momentum distributions observably distorted towards the forward direction.

For larger values of \(n\), the strong forward-backward asymmetry becomes less pronounced or completely disappears. This observation is valid for both TDSE and SCTMC. The overall agreement between TDSE and SCTMC is improved also with increasing the pulse cycle number, since the applied semiclassical emission model is valid in the quasi-static regime [58, 59].

Semiclassical and CTMC simulations have many advantages. First, these methods can be applied for description of the photoemission from systems with nontrivial geometries, like nanotips [31, 62]. The classical or semiclassical calculations usually describe the general trend of the experimental data correctly, and the agreement between experiments and calculations are superior. Second, semiclassical simulations can help to identify the specific mechanism responsible for the relevant phenomena, and provide an illustrative picture of the mechanisms in terms of classical trajectories. Based on the qualitative agreement between TDSE and SCTMC in figures 2(g) and (h), in the following we focus on the case when the number of cycles is \(n = 8\) and analyze the momentum distributions of the ejected electrons as a function of the initial conditions. Figure 3 shows a magnification of figure 2(h) with boxes denoting the regions of our interest for further investigations. The numbers in the boxes will be our reference numbers later.

We sorted the events according to the boxes shown in figure 3 and visualized them as a function of tunnel exit points (figure 4), tunnel ionization time (figure 5), and initial perpendicular velocity measured from the polarization direction (figure 6). We found correlation between certain parts of the momentum distribution and the initial conditions. Although we used a relatively large box size, a clear separation is visible in the distributions. According to the deterministic behavior of the semiclassical treatment, we can predict the final momentum of the tunneled electrons from the initial conditions.

In figure 3, we selected the regions where \(v_p\) is close to zero. The boxes are symmetrically located almost in a line, except from box No. 3. The boxes with No. 3 and 4 are in the central ring in figure 3. The others are further and further away from the center, but the boxes No. 2 and 5 are in the same circular ring. For the case of the central ring (box No. 3 and 4) the tunnel exit points have positive \(z\) coordinates (see figure 4) and the birth times correspond to negative electric field (see figure 5). Boxes No. 2 and 5 are in the second ring, with box No. 2 being at the negative parallel momentum region, while box No. 5 is at the positive one. In the case of box No. 2, electrons born at the positive \(z\) values and the birth times correspond to negative electric field and the opposite holds for the case of No. 5. Generally, the periodicity between the electron’s birth time and distance can be identified in the entire momentum map. Figure 6 shows the initial perpendicular velocity distribution of electrons arriving to the given final momentum regions marked in figure 3. The velocity map is in coincidence with the peak structure of the birth time distributions. We note, that all these events correspond to final perpendicular momenta that are close to zero. For all selected cases the sign of the final \(p_z\) value is the opposite of \(z\) (the tunnel distance).

Focusing on the final momentum distribution of the emitted electrons, we also clearly identify and separate regions that correspond to qualitatively different initial conditions. The separation is especially noticeable for the distribution of the birth time of electrons arriving to different bumps of the momentum distribution. We note that the possible overlap between the peaks of the distributions is partly related to the relatively large box size and partly it can be attributed to the number of rescattering on the target nucleus. Recattering events wash out the clear separation.

Let us take one of the most important advantages of the classical treatment and check the typical trajectories. Figure 7 shows specific classical electron trajectories and their distance from the origin corresponding to the regions marked in figure 3. We found two completely different branches of trajectories for each region. In the first case, the electrons after tunnel ionization return to the target, following the shape of the oscillatory electric field and forming a regular quiver trajectory. However, in the second case, electrons approach the nucleus so closely that they can become bound again. The sharp direction changes in the radial distance correspond to...
Figure 2. Ionization probability densities for the H atom as a function of the electron momenta parallel and perpendicular to the polarization direction ($z$ axis). The electric field is defined by equation (3). Left column corresponds to the TDSE approach, while the right one shows results obtained using the SCTMC method. First row: 1 cycle pulse ($n = 1$), second row: 2 cycle pulse ($n = 2$), third row: 4 cycle pulse ($n = 4$), fourth row: 8 cycle pulse ($n = 8$).
rescattering events with some ‘swinging maneuver’. What can we deduce from this picture? If an electron, as a consequence of the interaction with the oscillatory electric field, returns to the nucleus in the proper phase it can gain energy. According to this expectation, we found that after each collision the electron indeed gains energy and after a few collisions the final electron energy is always much higher than its energy at the birth instants. This phenomenon is close to the Fermi-shuttle type ionization, that is well known either in cosmology [63] or in ion-atom collisions [64].

In all cases, the electron trajectories driven by the laser field show oscillations, i.e., they travel back and forth closer to and further away from the nucleus. This is partly due to the quiver motion of electrons interacting with the driving field, but also indicates the strong interaction with the target nucleus. As an illustration, figure 8 shows electron trajectories that correspond to the same initial conditions but with and without the interaction with the target nucleus being taken into account. We found drastic differences between the electron traces. If we only consider the interaction with the driving field, well-established regular quiver trajectories can be recognized. This is in contrast with the case when both the interaction with the driving field and the target nucleus were taken into account, when the electrons return twice to the nucleus (see the points A and B in figure 8(a), and also figure 8(b)).

Figure 3. Magnification of figure 1(h) (which represents the electron momentum distribution after an n = 8 cycle pulse). The numbered boxes mark the six regions that will be studied in detail.

Figure 4. The ‘birth distance’ (measured from the target nucleus) distribution of the photoelectrons arriving to the characteristic regions marked in figure 3.

4. Conclusions

We presented a full quantum mechanical calculation, together with a semiclassical one, to describe the process of above-threshold ionization. We focused on the tunneling regime for the hydrogen atom, and considered a few-cycle infrared exciting pulse. As a quantum treatment, we applied the direct integration of the time-dependent Schrödinger equation (TDSE). In the three-dimensional semiclassical trajectory Monte Carlo method, we assumed that the wavepacket propagation in the post-tunneling process can be well described within the classical framework. We found that our 3 dimensional semiclassical method describe reasonably well the momentum distribution of the photoelectrons. Furthermore, we found good agreement between our TDSE and SCTMS results in the final momentum distributions of the emitted electrons for relatively long exciting pulses. This fact allowed us to perform further analysis using the semiclassical approximation and clearly identify and separate the initial conditions that correspond to various regions in the final momentum distributions of the emitted electrons. We illustrated the corresponding regions with typical electron trajectories (see figures 7 and 8). Additionally, we have seen significant differences between the electron trajectories when only the interaction with the driving field was taken into account and when the interaction both with the driving field and the target nucleus played role. In the first case the electrons have regular oscillatory trajectories, while in the second one the electron can return to the nucleus several times.
Figure 7. In the first row, the time evolution of the distance from the origin is illustrated for some specific trajectories, (a) region 1, and (b) region 2, in figure 3. In the second row the corresponding planar motion is shown, (c) region 1, and (d) region 2. Different line colors help us to distinguish the trajectories with different initial conditions.

Figure 8. Part (a) Time dependence of the distance from the origin for identical initial conditions. The green line corresponds to the time evolution without taking into account the Coulomb interaction between the electron and the target nucleus, while the purple line corresponds to the case when both the interactions are included. (b) Same as panel (a) but showing the planar motion of the orbits.
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