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Probing quantum states of Rydberg electrons by half-cycle pulses

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Recently performed experiments[1] have demonstrated that half-cycle pulses (HCPs), i.e. unimodular electromagnetic pulses, are a useful new spectroscopic tool which is particularly well suited for investigating the dynamics of weakly bound Rydberg electrons. Typically their pulse durations range from the subpicosecond to the nanosecond regime and these pulses have already been produced with electric field strengths up to 10^5V/cm. So far work in this context has concentrated mainly on studies of total ionization or survival probabilities and on energy-resolved ionization spectra of Rydberg electrons [1, 2, 3]. Thus it has been shown with the help of a classical picture of the ionization process that energy-resolved ionization spectra yield direct information about the initial momentum distribution of a Rydberg electron. However, this way any phase information about the initial quantum state is lost.

In the following we address the question whether this phase information can be obtained from energy- and angle-resolved ionization probabilities. For this purpose a (multidimensional) semiclassical description of the ionization process of Rydberg electrons by half-cycle pulses is presented. In this theoretical approach it is particularly apparent how phase information about the initial quantum state of the Rydberg electron manifests itself in the angle- and energy-resolved ionization spectra. Furthermore, this way a detailed understanding of the ionization dynamics is obtained which is based on the underlying classical dynamics. In order to emphasize the essential physical aspects our subsequent discussion focuses on the sudden-ionization approximation [2] in which the ionizing HCP can be approximated by a delta-function in time. However, it should be mentioned that besides numerical advantages as far as the treatment of the Coulomb problem is concerned the presented semiclassical approach is also well suited for describing all effects which might arise from finite pulse durations or from spatial variations of realistic HCPs.

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Before we present the main ideas of the general multidimensional semiclassical description let us first of all summarize the quantum mechanical description of ionization by an HCP in the sudden-ionization approximation [2]. In the subsequent treatment we shall use Hartree atomic units with $e = \hbar = m_e = 1$ (e and m_e denote the electronic charge and mass).

Let us consider an atom initially prepared in a highly excited energy eigenstate $\mid nl\rangle$. Thereby its principal quantum number n is assumed to be large and its angular momentum quantum number l is small. This Rydberg electron is ionized at time $t = 0$ by a HCP. In the sudden-ionization approximation the HCP changes the momentum of the Rydberg electron abruptly by the amount

$$\vec{p}_0 = -\int_{-\infty}^{\infty} dt \vec{E}(t). \tag{1}$$

Thereby $\vec{E}(t)$ denotes the electric field strength of the HCP. The sudden ionization approximation is valid provided the Kepler period $T_{orb} = 2\pi(n-\alpha)^3$ of the Rydberg electron is much larger than the pulse duration of the HCP (α denotes the quantum defect of the Rydberg electron [4]). The electronic state $\mid \psi_0\rangle$ immediately after the application of the HCP is given by

$$\mid \psi_0\rangle = e^{i\vec{p}_0\cdot\hat{\vec{x}}} \mid nl\rangle. \tag{2}$$

Its subsequent time evolution is determined by the atomic Hamiltonian (in the absence of the HCP). Thus the angle- and energy resolved ionization probability is given by

$$\frac{d^3 P_{ion}}{d\epsilon \wedge d\Omega} = \sqrt{2\epsilon} \mid \langle \vec{p}(\epsilon, \Omega) \mid \psi(t \to \infty)\rangle \mid^2 \tag{3}$$

with $\epsilon = \vec{p}^2/2$ and Ω denoting the final energy and solid angle of the emitted electron. In the case of a hydrogen atom, for example, which is prepared in an s-state initially and which is ionized by a linearly polarized HCP, i.e. $\vec{p}_0 = p_0 \vec{e}_z$, a partial wave analysis yields

$$\langle \vec{p}(\epsilon, \Theta, \phi) \mid \psi(t \to \infty)\rangle = \tag{4}$$
$$\frac{1}{\sqrt{2\pi}|\vec{p}|} \sum_{l'=0}^{\infty} e^{i\sigma_{l'}} P_{l'}(\cos\Theta) \int_0^{\infty} dr S_{n,l=0}(r) j_{l'}(|\vec{p}_0| r) F_{l'}(\epsilon; r).$$

Eq.(4) describes the probability amplitude of detecting an ionized electron with asymptotic momentum $\vec{p}$. The spherical angles of the emitted electron are denoted (Θ, ϕ) with $d\Omega = \sin\Theta d\Theta \wedge d\phi$. $P_{l'}(x)$ is the Legendre polynomial of order l', $S_{n,l=0}(r)$ is the radial wave function of the initial state $\mid nl = m = 0\rangle$ and $F_{l'}(\epsilon; r)$ is the regular Coulomb wave function with energy $\epsilon > 0$ and angular momentum quantum number l'. The Coulomb scattering phase is denoted $\sigma_{l'}$ and $j_{l'}(x)$ is a spherical Bessel function of order l'. Eqs.(3)and (4) determine the quantum mechanical angle- and energy resolved ionization probability in the sudden-ionization approximation.

Semiclassical description of ionization by a HCP

For highly excited Rydberg states, i.e. $n \gg 1$, and for sufficiently weak field strengths of the HCP, i.e. $\mid \vec{p}_0 \mid^2 /2 \ll 1$, the ionization process can be described into the underlying classical dynamics of the ionization process. Furthermore, in this theoretical approach it becomes particularly apparent how phase information about the initial quantum state of the Rydberg electron manifests itself in the angle- and energy-resolved ionization spectra.

Let us first of all consider briefly the main theoretical ingredients of such a semiclassical description for the case of hydrogenic Rydberg states.

Semiclassically the energy eigenstate of a Rydberg electron with energy $\epsilon_n = -1/2n^2$ which is prepared in an s-state is approximately given by [5]

$$\begin{aligned}\langle \vec{x} \mid n, l = m = 0\rangle &= \frac{1}{\sqrt{8}\pi n^{3/2} r \sqrt{p(r)}}[\exp(i\int_0^r drp(r) - i\pi/2 - i\pi/4) + c.c.] \\ &= A_{cl}(\vec{x})\{\exp[iW_+(\vec{x})] + \exp[iW_-(\vec{x})]\}.\end{aligned} \tag{5}$$

Thereby $p(r) = \sqrt{2(\epsilon_n + 1/r)}$ and $r = \mid \vec{x} \mid$ denote the electronic radial momentum and the distance from the nucleus. Eq.(5) is valid for positions of the Rydberg electron which are located well inside the classically allowed region. The state immediately after the application of a HCP is given by Eq.(2). Within the framework of a semiclassical description [6] this quantum state corresponds to two Lagrangian manifolds $L_\pm = \{(\vec{x}, \vec{p}_\pm(\vec{x}))|\vec{p}_\pm(\vec{x}) = \nabla W_\pm(\vec{x}) + \vec{p}_0 \equiv \pm p(r)\vec{e}_r + \vec{p}_0\}$ in the classical phase space of the electron.

After the application of the HCP the time evolution of this semiclassical quantum state is characterized by the bi-valued family of classical trajectories $\vec{x}(t; \vec{x}^{(0)}, \vec{p}_\pm(\vec{x}))$ which start at all possible points $\vec{x}^{(0)}$ in the support of $A_{cl}(\vec{x})$ with initial momenta $\vec{p}_\pm(\vec{x})$ and which evolve according to the classical Coulomb Hamiltonian $H = \vec{p}^2/2 - 1/r$. The probability amplitude $\langle \vec{p} \mid \psi(t \to \infty)\rangle$ of detecting an ionized Rydberg electron with momentum $\vec{p}$ is determined by all those classical trajectories with assume the asymptotic, final momentum $\vec{p}$. With each of these trajectories one can associate [6]

- a classical action (eikonal)

$$S = \int \vec{x} \cdot d\vec{p} + \vec{x}^{(0)} \cdot \vec{p}^{(0)}, \tag{6}$$

- a classical probability density

$$P^{(cl)} = \left| \frac{dp_x^{(f)} \wedge dp_y^{(f)} \wedge dp_z^{(f)}}{dx^{(0)} \wedge dy^{(0)} \wedge dz^{(0)}} \right|^{-1}, \tag{7}$$

- and a Morse index μ which is equal to the number of sign changes of $\frac{dp_x^{(f)} \wedge dp_y^{(f)} \wedge dp_z^{(f)}}{dx^{(0)} \wedge dy^{(0)} \wedge dz^{(0)}}$ times their multiplicities.

Thereby $\vec{p}^{(f)}$ denotes the final momentum of the ionized Rydberg electron. In terms of these classical quantities the probability amplitude of detecting an ionized Rydberg electron with asymptotic momentum $\vec{p}$ is given by

$$\langle \vec{p} \mid \psi(t \to \infty)\rangle = \sum_j \sqrt{P_j^{(cl)}} e^{-i(S_j + \frac{\pi}{2}\mu_j)} A_{cl}(\vec{x}_j^{(0)}) e^{i[W_j(\vec{x}_j^{(0)}) + \vec{p}_0 \cdot \vec{x}_j^{(0)}]}. \tag{8}$$

Depending on whether the initial momentum of the classical trajectory j is positive or negative one has to choose for W_j either the outgoing or incoming component $W_\pm$.

In order to exhibit characteristic properties of the ionization spectra and their relation to the underlying classical dynamics of the Rydberg electron let us consider a linearly polarized HCP with $\vec{p}_0 = p_0 \vec{e}_z$ and an initial hydrogenic Rydberg state $\mid nl = 0\rangle$.

Due to axial symmetry $d^3 P_{ion}/d\epsilon \wedge d\Omega$ is independent of ϕ. The initial positions $(r^{(0)}, \Theta^{(0)})$ of the classical trajectories with final asymptotic energy $\epsilon > 0$ are determined by energy conservation, i.e.

$$\frac{1}{2}[\vec{p}_{\pm}(r^{(0)}, \Theta^{(0)}) + \vec{p}_0]^2 - 1/r^{(0)} = \epsilon \tag{9}$$

or

$$1/r^{(0)} = -\epsilon_n + \frac{(\epsilon - \epsilon_n - \vec{p}_0^2/2)^2}{2\vec{p}_0^2 \cos^2(\Theta^{(0)})}. \tag{10}$$

Thus there is one value of $r^{(0)}$ for each value of $\Theta^{(0)}$. The initial positions of those classical trajectories which yield a particular final energy ϵ and emission angle Θ are a dynamical property of the Coulomb problem and can be determined in a straightforward way.

In particular, one can distinguish two characteristic dynamical regimes:

1. $0 < \epsilon < \epsilon_n + \vec{p}_0^2/2$
   In this case trajectories with $0 < \Theta^{(0)} < \pi/2$ $(\pi/2 < \Theta^{(0)} < \pi)$ start with negative (positive) radial momenta.

2. $\epsilon_n + \vec{p}_0^2/2 < \epsilon$
   In this regime of high asymptotic energies trajectories with $0 < \Theta^{(0)} < \pi/2$ $(\pi/2 < \Theta^{(0)} < \pi)$ start with positive (negative) radial momenta.

Characteristic examples for the dependences of the final observation angle Θ on the initial emission angle $\Theta^{(0)}$ are depicted in Figs. 1 and 2. The deflection function shown

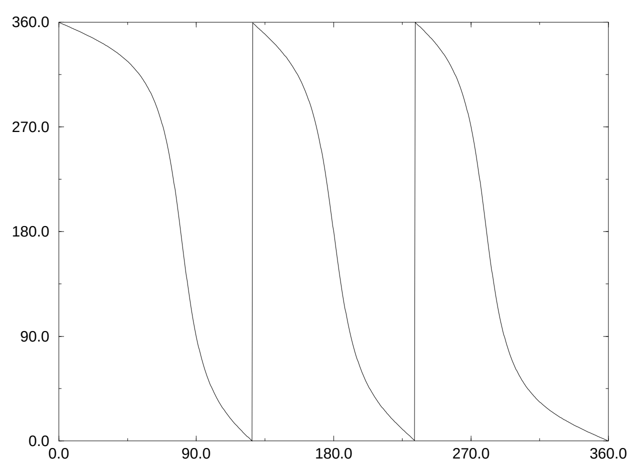

Figure 1. Deflection function, i.e. final angle Θ as a function of the initial emission angle $\Theta^{(0)}$ (in degrees); parameters are $n = 50$, $\mid \vec{p}_0 \mid = 0.05$a.u., $\epsilon = 10$meV.

in Fig. 1 corresponds to a case of small electron energies. It is apparent that there are three different initial emission angles $\Theta^{(0)}$ for each final angle Θ. Thus on the basis of Eq.(8) it is expected that the resulting angular- and energy resolved ionization probability displays a complex interference structure arising from quantum mechanical trajectories. Furthermore, there are classical trajectories with initial emission angles $\Theta^{(0)} \neq 0, \pi$ which are ionized along the direction of polarization of the HCP, i.e. with $\Theta = 0, \pi$. Due to the axial symmetry of the problem these trajectories give rise to the semiclassical glory phenomenon [7] which manifests itself in a divergence of the primitive semiclassical ionization amplitude of Eq.(8). However, this divergence can be removed in a straightforward way with the help of semiclassical uniformization methods [7].

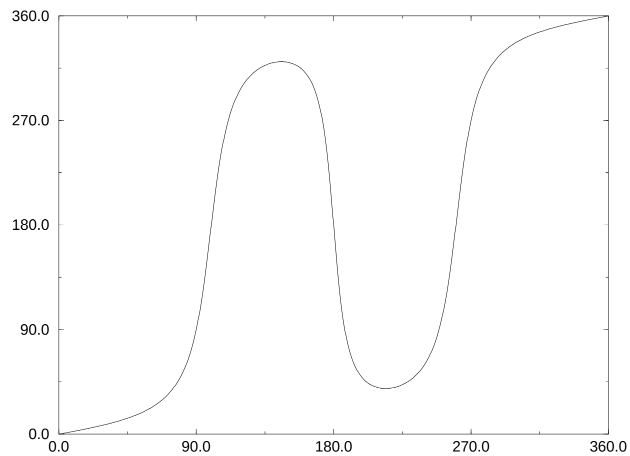

Figure 2. Deflection function, i.e. final angle Θ as a function of the initial emission angle $\Theta^{(0)}$ (in degrees); parameters are $n = 50$, $\mid \vec{p}_0 \mid = 0.05$a.u., $\epsilon = 40$meV.

The characteristic behaviour of the deflection function for high energies of the ionized electron, i.e. $\epsilon_n + \vec{p}_0^2/2 < \epsilon$, is shown in Fig. 2. At these energies electrons are emitted in the polarization direction of the HCP with initial angle $\Theta^{(0)} = 0$ only so that there is no longer a semiclassical glory phenomenon at $\Theta = 0$. However, in the opposite direction, i.e. for $\Theta = \pi$, a semiclassical glory phenomenon is still observable as there are classical trajectories leaving the atom in this direction with initial angles $\Theta^{(0)} \neq 0, \pi$. In addition, also an extremum of the deflection function is noticable around an emission angle of approximately $\Theta^{(0)} \approx 3\pi/4$ which gives rise to a semiclassical rainbow phenomenon [7]. At this emission angle the contributions of two classical trajectories coalesce and the primitive semiclassical ionization amplitude of Eq.(8) diverges. Again this divergence can be removed in a straightforward with the help of semiclassical uniformization techniques [7].

As an example, the resulting energy-resolved angular distribution is depicted in Fig. 3. The overall good agreement between the semiclassical result obtained on the basis of Eq.(8) and the exact quantum mechanical result of Eqs.(3) and (4) is apparent. The primitive semiclassical result of Eq.(8) deviates from the quantum mechanical one only in a small region around the rainbow and backward glory angles. According to Eq.(8) the oscillations of the ionization probability can be explained in a straightforward way in terms of quantum mechanical interferences between the probability amplitudes of the contributing classical trajectories. Thus the angle- and energy resolved ionization spectrum can be related directly to the underlying classical dynamics of the Rydberg electron under the influence of the ionizing HCP.

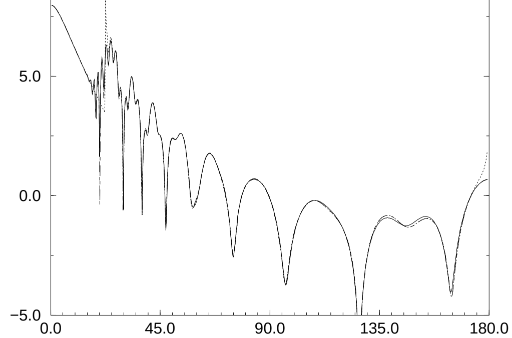

Figure 3. Energy-resolved angular distribution $\ln[d^3 P_{ion}/d\epsilon \wedge d\Omega]$ in the sudden-ionization approximation: exact quantum mechanical result of Eq.(4)(full), primitive semiclassical result of Eq.(8) (dotted) and uniform semiclassical result (dashed); parameters are $n = 50$, $| \vec{p}_0 | = 0.05$a.u., $\epsilon = 40$meV.

Outlook and conclusion

The presented multidimensional semiclassical approach offers an adequate description of ionization of Rydberg electrons by HCPs. Thereby the angle- and energy-resolved ionization spectra can be related directly to the underlying classical dynamics of the Rydberg electron. Within this semiclassical treatment the phase information contained in the quantum state is taken into account properly. Thus on the basis of this semiclassical treatment it is expected that also all phase properties of the initial quantum state of the Rydberg electron might be reconstructed from observed angle- and energy resolved ionization spectra. Work in this direction is currently in progress. This semiclassical approach offers also significant numerical advantages as it avoids the usual problems connected with the large extensions of Rydberg states and with the Coulomb singularity. It can be extended in a straightforward way to cases where the pulse durations are finite and where spatial variations of the ionizing HCP are no longer negligible.

Acknowledgments

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