Influence of laser pulse on the autocorrelation function of H in a strong electric field

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The autocorrelation function of electronic wave packet of hydrogen atom in a strong electric field below the zero-field ionization threshold is investigated in the formalism of semiclassical theory. It is found that the autocorrelation depends on the applied laser pulse significantly. In the case of narrow laser pulse, the reviving peaks in the autocorrelation can be attributed to the closed orbits of electrons, which are related to the classical dynamics of the system. But this correspondence is wiped out with increasing the laser width because of the interference among the adjacent reviving peaks.

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During the past few decades, short-pulse tunable laser technology has been developed constantly and one of its physical applications is to excite electronic wave packets in atomic systems. The information of the evolving Rydberg wave packet is related to the correspondence between classical and quantum dynamics. There have been considerable theoretical and experimental works for Rydberg wave packet dynamics with or without the presence of external fields. For instance, the dynamics of the electronic wave function of hydrogen was investigated experimentally by Noordam *et al.*[1] and rubidium system in an electric field by Broers *et al.*[2,3]. And more recent results can be found in Refs. [4,5]. In these experiments, the wave packet dynamics in an electric field above the classical field ionization were investigated with a double-pulse technique and the absolute values of the autocorrelation functions $\langle\psi(0)|\psi(t)\rangle$ were measured, which reflect the underlying dynamics of the wave packet.

To investigate the autocorrelation function theoretically, we can in principle propagate the wave packet till time t and calculate the resemblance between the wave packet at time t and the initial wave packet. The wave packet can be represented by superimposing the eigenstates of the system[6−8]. Unfortunately, the description of wave packet by superimposing the eigenstates is a difficult task in most cases since it involves so many eigen functions. Du *et al.* related the autocorrelation function with the oscillator strength density which can be approximated by closed orbit theory[9,10]. Furthermore, they derived a general formula of autocorrelation function based on the closed orbits in a static electric field[11]. Yu *et al.* obtained the autocorrelation function of hydrogen atoms in magnetic fields[12].

In this letter, we investigate the autocorrelation of electronic wave packet of hydrogen atoms in an electric field in the framework of semiclassical theory which has an intuitive physical picture and provides some useful information of the correspondence between classical and quantum dynamics. The problem of an atom in an electrical field has been studied extensively in past years. Especially, Gao *et al.* recently studied closed orbits and recurrence for single-electron atoms in electric fields[13]. They argued that at high energy only one orbit exists, and new orbits are bifurcated with the energy below the zero-field ionization threshold. As we have known, hydrogen atom in electric field is an important model to establish a relation between quantum and classical dynamics picture. So its wave packet dynamics, when a pulsed laser is applied to H atomic system, has important theoretical meaning. By using the semiclassical closed orbits theory, we calculate the autocorrelation function of the first several closed orbits at a fixed scaled energy and analyze the influences of the laser pulse width and the orbit bifurcation on the autocorrelation function. Atomic units are used throughout the letter unless otherwise noted.

To investigate the autocorrelation function of H in an electric field in semiclassical framework, it is necessary to find the closed orbits of the active electron which starts and ends at the nucleus. The dynamics of a highly excited hydrogen atom in the presence of an external static electric field aligned along the z axis is described by a single-particle, nonrelativistic Hamiltonian. In cylindrical coordinates (ρ, ϕ, z), it can be written as

$$H = \frac{1}{2}\left(p_z^2 + p_\rho^2 + l_z^2/\rho^2\right) - 1/\left(\rho^2 + z^2\right)^{1/2} + Fz, \quad (1)$$

where F is the electric field strength.

By using a scaled-variable method (varying the electron energy and the electric field simultaneously to keep the scaled energy fixed), we can find that the closed orbits have simple patterns, and the associated recurrences emerge naturally. Transforming to scaled semiparabolic coordinates (u, v) and considering the case of $l_z = 0$, we have the transformed Hamiltonian

$$\tilde{H} = \frac{1}{2}\left(p_u^2 + p_v^2\right) + \frac{1}{2}\left(u^4 - v^4\right) - \varepsilon\left(u^2 + v^2\right) - 2, \quad (2)$$

where $p_u = \mathrm{d}u/\mathrm{d}\tau$, $p_v = \mathrm{d}v/\mathrm{d}\tau$, $\mathrm{d}\tau/\mathrm{d}t = 1/(u^2 + v^2)$, and $\varepsilon = E/F^{1/2}$ is the scaled energy.

After this transformation, the Coulomb singularities have been vanished. Therefore, we can numerically integrate the motion equations for (u, v, p_u, p_v) of Eq. (2),

Table 1. Initial Angle, Action, and Scaled Period in the ρ-z Plane for the First Six Closed Orbits of Hydrogen in an Electric Field with $\varepsilon = -0.266$ at $F = 400$ kV/cm

Orbit	Initial Angle (deg.)	Action S (a.u.)	Period T (a.u.)
1	0.0	4.50906	1777.36601
2	0.0	9.05811	3554.73225
3	0.0	13.56717	5332.09833
4	13.0	13.56544	5744.74236
5	0.0	18.07623	7109.46456
6	15.0	18.04487	8151.91699

namely,

$$\dot{u} = \frac{\partial \tilde{H}}{\partial p_u}, \quad \dot{v} = \frac{\partial \tilde{H}}{\partial p_v}, \quad \dot{p}_u = -\frac{\partial \tilde{H}}{\partial u}, \quad \dot{p}_v = -\frac{\partial \tilde{H}}{\partial v}. \quad (3)$$

Using a standard fifth-order Runge-Kutta method to integrate the motion equations, the closed orbits can be found in a straightforward manner. The first six closed orbits are listed in the Table 1. For each closed orbit, we calculate its classical action S_k and scaled period T_k, where k runs over all of the closed orbits.

The time evolution of the wave packet is characterized by the autocorrelation function defined by

$$\psi^{\mathrm{AC}}(t) = \langle \psi(0) | \psi(t) \rangle, \quad (4)$$

where $|\psi(t)\rangle$ is the wave function at time t. This expresses the notion that we directly measure the overlap between the wave packet at time t and the initial wave packet $|\psi(0)\rangle$. The autocorrelation function can be calculated based on the closed orbit theory. We give the theoretical formula of the autocorrelation function by briefly following Ref. [11] and apply it to the system for H in the electric field. Let the initial state of the atomic system be $\psi_{\mathrm{i}}(r)$ and the wave packet generated by a short-pulse laser is $\psi(t)$. Assuming the short-pulse laser is in the form

$$f(t) = f_m \exp\left(-t^2/2\tau^2\right) \cos(\omega t + \phi), \quad (5)$$

where ω, f_m and τ are the frequency, peak amplitude and pulse width, respectively. The final state excited by the pulse is $\psi_{\mathrm{f}}(r)$, with energy E_{f} centered at $E_{\mathrm{f}}^{\mathrm{c}} = E_{\mathrm{i}} + \omega$ and a few $1/\tau$ width.

According to the time dependent perturbation theory, the autocorrelation function can be written as

$$\langle \psi(0) | \psi(t) \rangle = \int \mathrm{d}E \exp(-\mathrm{i}Et) \left| g(E - E_{\mathrm{i}}) \right|^2 \times \left[\frac{Df(E)}{2(E - E_{\mathrm{i}})} \right], \quad (6)$$

where

$$g(E - E_{\mathrm{i}}) = \int \mathrm{d}t f(t) \exp(-(E - E_{\mathrm{i}})t) \quad (7)$$

is the Fourier transformation of the short-pulse laser, $Df(E)$ is the oscillator-strength density. Using the rotating wave approximation, we obtain

$$g(E - E_{\mathrm{i}}) = \tau f_m \left[\frac{\pi}{2}\right]^{1/2} \mathrm{e}^{-\frac{(E - E_{\mathrm{i}} - \omega)^2 \tau^2}{2}} \mathrm{e}^{-\mathrm{i}\phi}. \quad (8)$$

In virtue of $|g(E - E_{\mathrm{i}})|^2$ is a Gaussian shape with width of $1/\tau$ and attains the peak when the energy $E_{\mathrm{f}}^{\mathrm{c}} = E_{\mathrm{i}} + \omega$, and the effective part of Eq. (6) is limited to an interval centered at E_{f} and $1/\tau$ wide. Considering this small energy interval, the oscillator-strength density can be approximated by using the formula of the standard closed orbit theory as

$$Df(E_{\mathrm{f}}^{\mathrm{c}} + \delta E) = Df_0(E_{\mathrm{f}}^{\mathrm{c}}) + \sum_k C_k(E_{\mathrm{f}}^{\mathrm{c}}) \sin\Big[T_k(E_{\mathrm{f}}^{\mathrm{c}})\delta E + \frac{1}{2}T_k'(E_{\mathrm{f}}^{\mathrm{c}})\delta E^2 + \Delta_k(E_{\mathrm{f}}^{\mathrm{c}})\Big], \quad (9)$$

where $T_k' = \mathrm{d}T_k/\mathrm{d}E_{\mathrm{f}}^{\mathrm{c}}$, δE is the deviation of energy from $E_{\mathrm{f}}^{\mathrm{c}}$, and the sum is over all the closed orbits of the system. We can expand the phases of the oscillations to the second order in the energy difference δE and set the amplitudes of the oscillations to constants. We can see that each oscillation in Eq. (9) corresponds to a closed orbit in this system and the oscillation is related to the stability property of the corresponding closed orbit, the laser polarization, and the initial quantum state.

Inserting Eqs. (8) and (9) into Eq. (6), replacing $(E - E_{\mathrm{i}})$ in the denominator of the integrand by ω, and carrying out the integral, we have

$$\psi^{\mathrm{AC}}(t) = \left[\frac{\tau f_m^2 \sqrt{\pi^3}(Df_0)}{4\omega}\right] \times \mathrm{e}^{-\mathrm{i}E_{\mathrm{f}}^{\mathrm{c}}t} \left\{ \mathrm{e}^{-t^2/4\tau^2} + \sum_k \left[G_k^-(t) + G_k^+(t)\right] \right\}, \quad (10)$$

$$G_k^{\pm} = \left[\frac{C_k}{2(Df_0)\alpha_k^{\pm}}\right] \times \mathrm{e}^{-\left[(t \pm T_k)^2/4\tau^2 \left(\alpha_k^{\pm}\right)^2\right] \mp \mathrm{i}(\Delta_k - \pi/2)}, \quad (11)$$

where $\alpha_k^{\pm} = \sqrt{1 \pm i\left[T_k'(E_{\mathrm{f}}^{\mathrm{c}})/2\tau^2\right]}$. Equation (10) is the autocorrelation function involving the laser-pulse parameters and the dynamical variables in the closed orbit theory. We can see that the autocorrelation function includes a sum of modified Gaussian terms. Each modified Gaussian term in the autocorrelation function corresponds to a parent oscillation term in the oscillator-strength density expressed in the closed orbit theory. The laser polarization, initial quantum state, and properties of the closed orbits of the system are known parameters. We can calculate the modified Gaussian terms in the autocorrelation function and the parent oscillation in the oscillator-strength density with the existing algorithms of closed orbit theory.

The theory can be applied directly to H atom in presence of a static electric field below the zero-field ionization threshold. The wave packet generated by a short-pulse laser of the form in Eq. (5) is $\psi(t)$ and the autocorrelation function can be written as

$$\langle \psi(0) | \psi(t) \rangle = C_0 M(t), \tag{12}$$

$$M(t) = \mathrm{e}^{-t^2/4\tau^2} + \frac{d}{\alpha_k^-} \mathrm{e}^{-\left[(t-T_k)^2/4\tau^2\left(\alpha_k^-\right)^2\right]+\mathrm{i}(\Delta_k-\pi/2)} + \frac{d}{\alpha_k^+} \mathrm{e}^{-\left[(t+T_k)^2/4\tau^2\left(\alpha_k^+\right)^2\right]-\mathrm{i}(\Delta_k-\pi/2)}, \tag{13}$$

where

$$C_0 = \frac{\tau f_m^2 \sqrt{\pi^3}(Df_0)}{4\omega} \mathrm{e}^{-\mathrm{i}E_\mathrm{f}^\mathrm{c} t},$$

$$d = \left[3F/8\sqrt{2}\left(E_\mathrm{f}^\mathrm{c}\right)^{3/2}\right] = [3F^{1/4}/8\sqrt{2}\varepsilon^{3/2}],$$

$$\alpha_k^\pm = \sqrt{1 \pm (\mathrm{i}/2\tau^2)\, T_k'}.$$

We can see from Eq. (13) that the autocorrelation function for H in a strong static electric field below threshold contains $(2k+1)$ peaks centered at $t=0$ and $\pm T_k$. The peak centered at $t=0$ in Eq. (13) comes from the nonoscillatory background term in the oscillator-strength density. The other peaks centered at $t=\pm T_k$ are related to the oscillatory terms in the oscillator-strength density in Eq. (9). For the system of H in a static electric field, there are several stable closed orbits with traveling time T_k. These closed orbits are corresponding to the oscillatory terms in the oscillatory-strength density in Eq. (9) and the peaks centered at $t=\pm T_k$ in the autocorrelation function.

By comparing Eq. (12) with Eq. (13), we find that $M(t)$ could be regarded as an effective autocorrelation function since the constant factor C_0 has no influence on the experimental measurement.

We compute the autocorrelation function of H in a static electric field below threshold and analyze the width of the laser pulse effect on the autocorrelation function. The time dependence of the absolute value of the autocorrelation function is entirely contained in the function $|M(t)|$, which is shown in Fig. 1 for positive time. The negative time part is symmetric: $|M(-t)| = |M(t)|$.

Figures 1(a)—(d) show the autocorrelations at four different widths of the pulse at a fixed electric field strength $F = 400$ kV/cm. In all cases, the peak centered at $t=0$ comes from the background term in the oscillator-strength density and is not related to any real orbit. The closed orbit theory gives the following physical picture for the wave packet reviving. The incoming laser pulse excites a wave packet that is localized in the region of the initial bound state of hydrogen atoms at the beginning. This wave then propagates away from the nucleus to large distance far from the nucleus and it can be approximated according to semiclassical approach which is correlated with classical trajectories. Eventually, these trajectories are turned back by the electric field, some of the orbits return to the vicinity of the nucleus, and the associated waves interfere with the outgoing waves to produce the observed oscillation in the time-reserved spectrum. One can see in Fig. 1(a) that the autocorrelation function produced by a short laser pulse shows six peaks (each closed orbit gives an oscillatory contribution to the autocorrelation function). It is found that when a new closed orbit is born, there appears a new oscillation in the time-resolved spectrum. This phenomenon can be explained in terms of wave packet theory. Since the oscillation in the autocorrelation function is generated by the interference of wave packets, when the wave packet returns to the nucleus along the orbit at T_k, the returning piece of the wave packet overlaps with $\psi(0)$ and produces the peak at T_k. Obviously, if the time duration becomes longer, there will be more peaks in the autocorrelation function. Figure 1(b) also shows six peaks centered at $T = T_1, T_2, T_3, T_4, T_5, T_6$ in the autocorrelation function. On the other hand, the width of the returning peak depends on the laser pulse width. The peaks go more widely with the increasing pulse width. The phenomena develop to some extent, and their mutual interference occurs. Figure 1(c) shows the interference and only five peaks appear in the autocorrelation function. Figure 1(d) shows the decreasing oscillation of the autocorrelation function further. We can see that when the laser pulse width becomes longer, the oscillations become smoother. The structure in Fig. 1 can be understood in the following way. The electronic wave packet created by a narrower laser pulse is more localized in space and it is easy to distinguish those belonging to different closed orbits. However, with the increasing laser pulse width, it is hard to distinguish them because of their extension in space and the interference between them. Because of this effect, the width of reviving peaks in the autocorrelation function grows and even two adjacent peaks merge into the recurrence spectrum. For example, when the pulse width $\tau = 200$ (a.u.) (Fig. 1(c)), the number of peaks reduces by one due to the merging. When τ is much smaller than the space between adjacent peaks, the autocorrelation function keeps its discrete peaks. Conversely, the mutual interfere effect of the discrete peaks is remarkable.

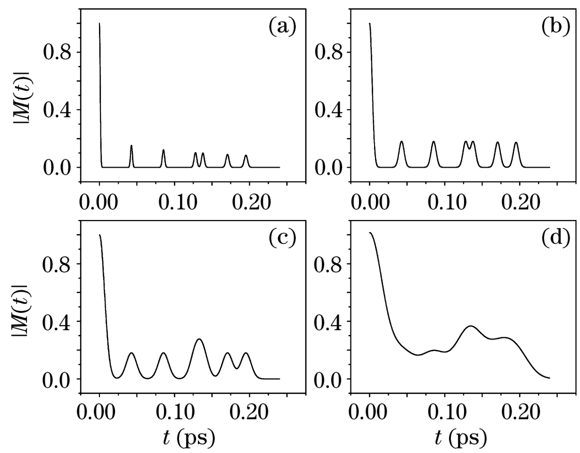

Fig. 1. Amplitudes of autocorrelation around the first six closed orbits for four different pulse widths. (a) $\tau = 30$ (a.u.), (b) $\tau = 100$ (a.u.), (c) $\tau = 200$ (a.u.), (d) $\tau = 500$ (a.u.). The strength of electric field is fixed at $F = 400$ kV/cm.

In summary, we examined the dynamics of the electronic wave packet of a hydrogen atom in an electric field when a short laser pulse was applied to the system.

Using a simple analytic formula from semiclassical closed orbit theory, we calculated the autocorrelation function of the wave packet below the zero-field ionization threshold for different pulse widths. The calculation results show that the autocorrelation function displays different amplitudes of oscillation for different pulses. The oscillation in the autocorrelation function can be attributed to the interference induced by the outgoing and returning electron waves travelling along the closed orbits. With the increasing laser pulse width, we have the growing width of reviving peaks in the autocorrelation function, and this eventually leads to their mutual interference. The interference is related to the duration of laser pulse. It may provide a visual demonstration for the understanding of the dynamical behavior of Rydberg atoms in strong electric fields. We are expecting that our results can be tested in a future experiment.

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