Journal of Electron Spectroscopy and Related Phenomena 173 (2009) 40–43

Contents lists available at ScienceDirect

Journal of Electron Spectroscopy and Related Phenomena

journal homepage: www.elsevier.com/locate/elspec

Time-resolved photodetached spectroscopy of H^- in electric field near an elastic surface

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ARTICLE INFO

Article history:
Received 9 January 2009
Received in revised form 10 April 2009
Accepted 10 April 2009
Available online 21 April 2009

Keywords:
Pulse width
Closed orbit
Autocorrelation function

ABSTRACT

The advances in femtosecond laser techniques facilitate the investigation of ultrafast electron dynamics on surfaces directly in time domain. The time-resolved spectroscopy of negative hydrogen ion in electric field near an elastic surface is investigated in the framework of semiclassical theory. We analyze the autocorrelation function and its dependence on the laser pulse width and external field strength. When the applied pulse width is very narrow, the reviving peaks in the time-resolved spectrum can be attributed to the closed orbits of electrons. With the increase of pulse width, the adjacent peaks interference mutually and finally merge. Furthermore, the number and amplitude of peaks in the time-resolved spectrum increases obviously with the increasing field strength.

1. Introduction

The study of the dynamics of electrons at surfaces is important for the understanding of a large variety of process, ranging from electron scattering at surfaces to charge transport dynamics across interfaces, relevant to design electronic devices [1–3]. The advances in femtosecond laser techniques facilitate the investigation of ultrafast electron dynamics on surfaces directly in time domain. The wave packet dynamics in real systems is usually very complicated because wave packet spreads and collapses with time evolution. A quantity that reflects the underlying wave packet dynamics is autocorrelation function, which is the overlapping between the wave function at time t and its initial state, $\langle\psi(t)|\psi(0)\rangle$, which can be measured by a pump-probe experiment. Many researchers investigate characteristic features of revival and fractional revival phenomena via analytic autocorrelation function expression with weighting probabilities modeled by a Gaussian distribution in different models [4–7].

Recently, a lot of attention has been paid to the photodetachment of H^- near an interface with or without external electric field [8,9] by the closed orbit theory [10]. And Wang et al. studied the photodetachment of H^- near two parallel interfaces with or without a static electric field [11,12]. But in their work, they only discussed the photodetachment in energy domain. In Ref. [13], Du related the autocorrelation function with the oscillator strength density which can be written as a sum of modified Gaussian functions that correspond to the closed orbits of the system. The closed orbit theory developed by Du and Delos is one of the effective techniques to deal with the photo-absorption phenomena of atoms or ions in strong external fields due to its clear physical picture and wide availability. Very recently the theory has been successfully applied to Rydberg hydrogen atom near a metal surface [14–15]. But the electronic wave packet dynamics of negative ion in a static electric field near an elastic surface is still an open problem so far. In this paper, we study the autocorrelation function of H^- in electric field near an elastic surface by the closed orbit theory to fill the gap. The surface electronic state in the system is similar to image state on metal surface but different in the following points: the surface electron is photodetached from a negative hydrogen ion, and a static electric field is applied perpendicular to the surface while the imaginary potential is ignored. By semiclassical theory, we calculate the autocorrelation of electronic wave packet of hydrogen negative ion in different electric fields near an elastic surface. The results show, as the laser pulse is very narrow, the reviving peaks in the autocorrelation can be attributed to the closed orbits of electrons. But with the increase of the pulse width, due to the interference between the adjacent peaks, this correspondence is wiped out. In addition, we find out that the number and amplitude of peaks in time-resolved spectrum increase obviously with the increasing field strength.

This paper is organized as follows: In Section 2, we describe the classical motion of the valence electron of H^- and give all the closed orbits. Then we present theoretical results of the autocorrelation function based on the closed orbit theory. Some remarks and discussions are given in Section 3. The conclusion is summarized in Section 4. Atomic units are used throughout the paper unless otherwise noted.

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doi:10.1016/j.elspec.2009.04.007

2. The electronic dynamics of negative hydrogen ion interacting with an elastic surface in a perpendicular electric field

The system configuration can be described as the following picture: a hydrogen negative ion H^- sits above an elastic surface which perpendicular to the electric field and a z-polarized laser is applied for the photodetachment. The H^- can be regarded as a one-electron system, with the active electron loosely bound by a short-range, spherically symmetric potential $V_b(r)$, where r is the distance between the active electron and the origin where the nucleus is. Take the direction of the electric field as the z axis and use the cylindrical coordinates (ρ, z, ϕ), the Hamiltonian governing the electron motion is

$$H = \tfrac{1}{2}(P_\rho^2 + P_z^2) + V_b(r) + V(z) \tag{1}$$

where $V_b(r)$ is the polarized central field of the H atom and this short-range potential can be negligible in our consideration. The ϕ motion has been separated due to the cylindrical symmetry of the system. The z component of the angular momentum is a constant of motion because of the symmetry and has been set to zero. And $V(z)$ is defined by the potential energy

$$V(z) = \begin{cases} \infty & \text{for } z \le -z_0 \\ Fz & \text{for } z > -z_0 \end{cases} \tag{2}$$

where F is the strength of electric field. The detached electron is bound in a z-direction linear potential well with an impenetrable flat surface at $z = z_0$ and moves freely in the ρ direction.

To investigate the dynamics of photodetached wave packet of H^- in an electric field near an elastic surface in semiclassical framework, we have to find all the closed orbits of the active electron which starts and ends at the nucleus. We use the same scheme for the closed-orbits as in Ref. [8]. All closed-orbits are labeled by (j, n) where $j = 1, 2, 3, 4$ and $n = 0, 1, 2, \ldots$. Any closed-orbit (j, n) with nonzero n can be decomposed as one of the fundamental closed orbits and either or running n times. Particularly, Yang et al. already illustrated these closed orbits in detail [8]. They argue that it is only four fundamental closed orbits in this situation. The returning time T_j (where $j = 1, 2, 3, 4$) of the fundamental closed orbits are

$$\begin{aligned} T_1 &= \frac{2k}{F} \\ T_2 &= \frac{-2k + 2\sqrt{k^2 + 2Fz_0}}{F} \\ T_3 &= \frac{2\sqrt{k^2 + 2Fz_0}}{F} \\ T_4 &= \frac{2\sqrt{k^2 + 2Fz_0}}{F} \end{aligned} \tag{3}$$

where $k = \sqrt{2E/m}$ is the emanating momentum.

Clearly, we have $T_3 = T_4 = T$. The returning time of any closed orbit is $T_{jn} = T_j + nT$ ($j = 1, 2, 3, 4$ and $n = 0, 1, 2, \ldots$). And the periods of the four fundamental closed orbit change with respect to the energy $T_j' = [\mathrm{d}T_j/\mathrm{d}E]$ ($j = 1, 2, 3, 4$) can be expressed as

$$\begin{aligned} T_1' &= \frac{1}{F}\left(\frac{2}{E}\right)^{1/2} \\ T_2' &= \frac{-(2/E)^{1/2} + 2(2E + 2Fz_0)^{-1/2}}{F} \\ T_3{}' &= \frac{2(2E + 2Fz_0)^{-1/2}}{F} \\ T_4' &= \frac{2(2E + 2Fz_0)^{-1/2}}{F} \end{aligned} \tag{4}$$

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

$$\psi^{AC}(t) = \langle \psi(0) | \psi(t) \rangle \tag{5}$$

where $|\psi(t)\rangle$ is the wave function at time t. This expresses the notion that we directly measure the overlap of the wave function at later times with the initial state. The autocorrelation function can be related with the closed orbits. We give the theoretical formula of the autocorrelation function briefly following Ref. [13], and apply it to the system. Let the initial state of the atomic system be $\psi_i(r)$, and the wave packet generated by a short pulse laser is $\psi(t)$. Assuming the Gaussian short-pulse laser is in the following form

$$f(t) = f_m \exp(-t^2/2\tau^2) \cos(\omega t + \phi) \tag{6}$$

where ω, f_m and τ are the frequency, peak amplitude and pulse width, respectively.

In the formalism of the time dependent perturbation theory, the autocorrelation function can be written as

$$\langle \psi(0) | \psi(t) \rangle = \int \mathrm{d}E \, \exp(-iEt) |g(E - E_i)|^2 \left[\frac{Df(E)}{2(E - E_i)}\right] \tag{7}$$

where

$$g(E - E_i) = \int \mathrm{d}t f(t) \exp(-(E - E_i)t) \tag{8}$$

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

$$g(E - E_i) = \tau f_m \left[\frac{\pi}{2}\right]^{1/2} e^{-((E - E_i - \omega)^2 \tau^2/2)} e^{-i\phi} \tag{9}$$

In virtue of $|g(E - E_i)|^2$ is a Gaussian shape with width of $1/\tau$ and attains the peak when the energy $E_f^c = E_i + \omega$, the effective part of Eq. (7) 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 closed orbit theory as

$$\begin{aligned} Df(E_f^c + \delta E) = Df_0(E_f^c) + \sum_k C_k(E_f^c) \sin[T_k(E_f^c)\delta E + \tfrac{1}{2} T_k'(E_f^c)\delta E^2 \\ + \Delta_k(E_f^c)] \end{aligned} \tag{10}$$

where $T_k' = \mathrm{d}T_k(E_f^c)/\mathrm{d}E_f^c$, δE is the deviation of energy from E_f^c, and the sum is over all the closed orbits of the system. We can expand the phases of the oscillations to second order in the energy difference δE and set the amplitudes of the oscillations to constants. We can see that each oscillation in Eq. (10) 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. (10) and (9) into Eq. (7) and replacing $(E - E_i)$ in the denominator of the integrand by ω, and carrying out the integral, we have

$$\psi^{AC}(t) = \left[\frac{\tau f_m^2 \sqrt{\pi^3}(Df_0)}{4\omega}\right] \times e^{-iE_f^c t} \left\{ e^{-t^2/4\tau^2} + \sum_k [G_k^-(tt) + G_k^+(t)] \right\} \tag{11}$$

$$G_k^\pm = \left[\frac{C_k}{2(Df_0)\alpha_k^\pm}\right] \times e^{-[(t \pm T_k)^2/4\tau^2(\alpha_k^\pm)^2] \mp i(\Delta_k - \pi/2)} \tag{12}$$

where $\alpha_k^\pm = \sqrt{1 \pm i[T_k'(E_f^c)/2\tau^2]}$, Δ_k is mainly the phase accumulation as the wave propagates along the kth closed orbit and C_k is the amplitude determined to a large degree by the stability of the kth closed orbit. The parameters Df_0, C_k, and T_k in Eq. (11) have been omitted to simplify the notations. Eq. (11) is the autocorrelation