

Photoionization of H atoms in few-cycle laser pulses

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Received 21 July 2005 / Received in final form 30 September 2005

Published online 13 December 2005 – © EDP Sciences, Società Italiana di Fisica, Springer-Verlag 2005

Abstract. Photoionization of hydrogen atoms in few-cycle laser pulses is studied numerically. The total ionization probability, the instantaneous ionization probability, and the partial ionization probabilities in a pair of opposite directions are obtained. The partial ionization probabilities are not always equal to each other which is termed as inversion asymmetry. The variation of asymmetry degree with the CE phase, the pulse duration and the pulse intensity is studied. It is found that the pulse intensity affects the asymmetry degree in many aspects. Firstly, the asymmetry is more distinct at higher intensities than that at lower intensities when the pulse duration exceeds 4 cycles; secondly, the maximal asymmetry in lower intensities varies with the CE phase visibly while at higher intensities not; thirdly, the partial ionization probabilities equal to each other for some special CE phases. For lower pulse intensities, the corresponding value of CE phase is always 0.5π and 1.5π , while for higher pulse intensities, the corresponding value varies with the pulse intensity. Similar phenomena were observed in a recent experiment using few-cycle radio-frequency (RF) pulses.

PACS. 42.65.Ky Frequency conversion; harmonic generation, including higher-order harmonic generation – 42.65.Re Ultrafast processes; optical pulse generation and pulse compression – 32.80.Rm Multiphoton ionization and excitation to highly excited states (e.g., Rydberg states)

1 Introduction

During the past decades, there has been increasing interest in the generation and application of short laser pulses. High-power ultrashort pulses with durations as short as only few optical cycles are now available as research tools [1–3]. Few-cycle pulses are different from the multi-cycle pulses which are fully characterized by frequency, intensity and polarization, an additional parameter is required to specify a few-cycle pulse: the initial phase φ of the carrier wave with respect to the envelope of the pulse, i.e., the so-called carrier-envelope (CE) phase. The time variation of the electric field of the laser pulse depends on the CE phase, and so do the physical processes induced by the laser pulse.

The CE phase-dependent phenomena in nonlinear optical processes such as high-order harmonic generation (HHG) [4,5] and above-threshold ionization (ATI) [6–8] were extensively studied both theoretically and experimentally. Using a stereo-ATI spectrometer, Paulus et al. demonstrated the CE-phase effect in the photoionization of Kr atoms [9]. Recently, the stabilization of the CE phase for amplified pulses was achieved [10], so the ques-

tion of determining the value of the phase had to be addressed. While not all the parameters of phase-stabilized laser pulses can be continuously controlled easily and the laser intensity can not be increased to an arbitrary value, Gurtler et al. presented a novel idea to simulate the photoionization of few-cycle pulse [11]. By measuring of the ionization of Rydberg atoms subject to few-cycle radio-frequency (RF) pulses, they showed the asymmetry in ionization of few-cycle RF pulses and the variation of asymmetry with the pulse intensity and the CE phase. The RF experiment suggests a direction for further experiments in optical domain.

In this paper we perform a theoretical study to the photoionization of hydrogen atoms in few-cycle laser pulses. By numerically solving the 1-D time-dependent Schrodinger equation (TDSE), the photoionization probabilities in a pair of opposite directions are obtained, by which we study the CE phase-dependent phenomena. We focus on the asymmetry in photoionization and present a physical insight to intensity-dependent asymmetry. The intensity-dependent asymmetry was shown both experimentally [11] and theoretically [12], but an intuitive physical explanation is presented in this paper firstly.

About the numerical solution of TDSE, there are many works and we just cite those related to the present

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work. Chelkowski and Bandrauk obtained the CE phase-dependent distributions of photoelectron in few-cycle pulse [12]. A pronounced CE-phase dependence of the photocurrent was found in a metal surface [13] and the CE phase effects in the dissociation of the HD^+ molecular were studied in the framework of TDSE [14].

This paper is arranged as follows: the calculation method is briefly described in Section 2; the total ionization probability and instantaneous ionization probability are investigated in Section 3; the variation of asymmetry with the CE phase, the pulse duration and the laser intensity is presented in Section 4; and Section 5 is for conclusions and discussions.

2 Schrödinger equation

We start from TDSE for a hydrogen atom interacting with an intense, linearly polarized few-cycle laser pulse. In the length gauge, the Schrödinger equation can be written as [15] (atomic units are used throughout this paper)

$$i\frac{\partial}{\partial t}\Psi(\mathbf{r}, t) = \left(-\frac{\nabla^2}{2} + V(\mathbf{r}) + \mathbf{r} \cdot \mathbf{E}(t)\right)\Psi(\mathbf{r}, t), \quad (1)$$

where $\Psi(\mathbf{r}, t)$ is the time-dependent wave function of the electron and \mathbf{r} denotes the position vector of electron with respect to its parent core. In the right side of the above equation, the first term in the large bracket is the kinetic energy term of electron, the second term is the Coulomb potential, and the last term is the dipole interaction in which $\mathbf{E}(t)$ is the electric field of the laser pulse. To avoid the time-consuming calculations, we adopt the 1-D model [16,17] to the considered problem. For linearly polarized laser pulses, the ionized electrons are mostly ejected out along the polarization vector [7, 17, 18]. In this treatment, the transverse spreading of wave packet was not taken into account and rescattering as well as recombination were neglected, since their contribution to the overall ionization is not dominant in linearly polarized pulses. Then, the Schrödinger equation reads:

$$i\frac{\partial}{\partial t}\Psi(x, t) = \left[-\frac{1}{2}\frac{\partial^2}{\partial x^2} + V(x) + xE(t)\right]\Psi(x, t), \quad (2)$$

and the soft-core model potential is used

$$V(x) = -\alpha/\sqrt{1+x^2}. \quad (3)$$

In the numerical simulation, chosen α as 0.775 a.u., the binding energy of the model atom has the binding energy of 13.6 eV. With the implicit Crank-Nicholson scheme and the 'trace-back' algorithm for solving the tridiagonal equations, we numerically calculate the evolution of electron wave function. The laser pulse used in the calculation is defined by the vector potential

$$A(t) = -(E_0 c/\omega)\text{sech}(t/\tau_0)\sin(\omega t + \varphi), \quad 0 \leq t \leq T_p = n_p T \quad (4)$$

from which the electric field can be determined $E(t) = -dA(t)/dt$. Here, φ denotes the CE phase, E_0 is the peak value of the electric field and τ_0 is related to the FWHM by $\tau_p = 1.76\tau_0$. In our calculations, we set $\omega = 0.056$, $E_0 = 0 \sim 0.6$. Correspondingly, the laser pulse is of central wavelength 800 nm and of peak intensity varying from 0 to about 1×10^{16} W/cm². The size of the x length is 512 bohr, i.e. $|x| < 256$ bohr. In the practical experiment, the distance between the atoms is about 200 bohr, so the calculation space is chosen in this scale. An absorbing mask function with an effective range of 32 bohr is applied at both ends of the x box. This choice ensures that the Coulomb force is negligible at such a large distance and the electron which reaches the distance will not return to the proton. The spatial step is chosen as 0.25 and the integration step in time is 0.03 a.u. Our results have been checked for convergence against the box size, the spatial step size, and the time step size.

The total ionization probability is defined as the absorption in the calculation boundaries and the absorption in one boundary is used to denote the partial ionization probability in the corresponding direction. For the long laser pulses, the ionization probabilities in a pair of opposite directions are always identical to each other [18]. While for few-cycle laser pulses, the spatial asymmetry of the electric field translates into different photoionization probabilities in the different directions [9]. With the definition of the asymmetry parameter a as [11]

$$a = \frac{P_+ - P_-}{P_+ + P_-}, \quad (5)$$

where P_+ and P_- are, respectively, the partial ionization probabilities in the positive and the negative x -direction, we study the asymmetry as a function of the pulse duration, the peak intensity, and the CE phase.

3 Total and partial ionization

In order to get a direct picture to the photoionization of atoms in few-cycle laser pulses and to disclose the corresponding relation between the electric-field strength and the instantaneous ionization probability, we first investigate the total ionization probability, the instantaneous ionization probability, and the influence of the CE phase on them. A representative correspondence relation is shown in Figure 1, in which the time-resolved total ionization probability and the instantaneous ionization probability of hydrogen atom subject to linearly polarized four-cycle pulses for $\varphi = 0$ and $\varphi = \pi/2$ are depicted. As shown in Figure 1a, a phase shift $\Delta\varphi$ of $\pi/2$ changes the peak value of the central oscillation near the envelope maximum considerably. The electric field shows one maximum for $\varphi = 0$, but has two maxima for $\varphi = \pi/2$ pointing to opposite directions, which we say the electric field is symmetric. Correspondingly, the ionization for $\varphi = 0$ is induced mainly from one half-cycle when the electric field reaches its maximum. While for $\varphi = \pi/2$, the ionization

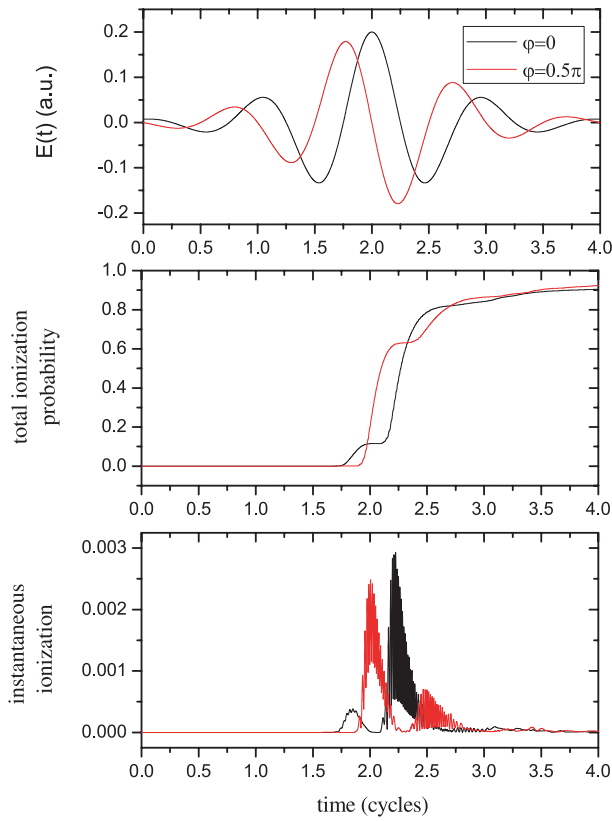


Fig. 1. (a) Time variation of the electric field of four-cycle pulses for two CE phases: $\varphi = 0$ and $\varphi = \pi/2$ with $E_0 = 0.2$ a.u. ($I = 1.4 \times 10^{15}$ W/cm²); (b) the corresponding time-resolved total ionization probabilities; and (c) the corresponding time-resolved instantaneous ionization probabilities. The black curves correspond to $\varphi = 0$; the red (dark gray) curves correspond to $\varphi = \pi/2$.

occurs mainly when the electric field reaches its two separated maxima, although the second maximum ionizes much less electrons than the first one, as shown in Figures 1b and 1c. Because the peak value of the pulse for $\varphi = 0$ is larger than that for $\varphi = \pi/2$, the maximal value of the instantaneous ionization probability for $\varphi = 0$ is obviously larger than that for $\varphi = \pi/2$, which means the difference in the electric-field strength is enlarged in the photoionization process. This feature of the nonlinear process such as HHG and ATI can be used to measure the value of CE phase [9,10]. Our calculation exhibits an intuitive corresponding relation between the CE phase and the instantaneous ionization. We note that whereas the total ionization profile is phase sensitive, the final fraction of ionization is approximately independent of φ . Similar results were also found with the help of numerical solutions of the TDSE [12,19] or calculations in the quasistatic approximation [1].

4 Variation of asymmetry with laser pulses

The asymmetry is one of the most attractive features in the photoionization of atoms in few-cycle pulses. The

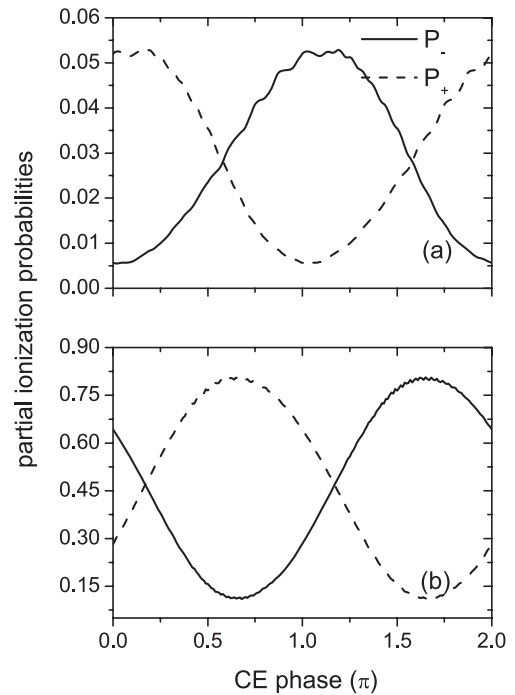


Fig. 2. P_+ and P_- as functions of the CE phase for laser intensity (a) $E_0 = 0.1$ a.u. ($I = 3.5 \times 10^{14}$ W/cm²) and (b) $E_0 = 0.2$ a.u. ($I = 1.4 \times 10^{15}$ W/cm²). The laser pulses are of four-cycle duration.

asymmetry degree changes dramatically with several pulse parameters, such as the CE phase, the pulse duration, and the laser intensity, so there exists an optimum condition under which the asymmetry is distinct.

4.1 Variation of asymmetry with CE phase

Since a larger value of the electric-field strength corresponds to a larger ionization probability, and the CE phase determines the value and the direction of the maximal electric field in the pulse envelope, the maximal ionization must be CE phase-dependent. We now pay our attention to the phase-dependent asymmetry in the partial ionization probabilities.

Figure 2 shows the variation of the partial ionization probabilities with the CE phase. Each of the partial ionization probabilities in the two opposite directions varies from its maximum to its minimum, then to its maximum with a periodicity of 2π , and equals to each other for some special CE phases. The oscillatory pattern is consistent with the recent experimental observations [20,21].

Since the ionization occurs mainly when the electric field reaches its maximum, it is expected that for some CE phases, such as 0.5π and 1.5π , for which the maxima of the electric field in the opposite directions are equal to each other, the partial ionization probabilities will be identical. This is the case for the laser pulses of lower intensity where the maximal ionization in a half-cycle is less than 0.5, but is not true for the laser pulses of higher intensities. The reason is obvious, if the pulse intensity is high enough that

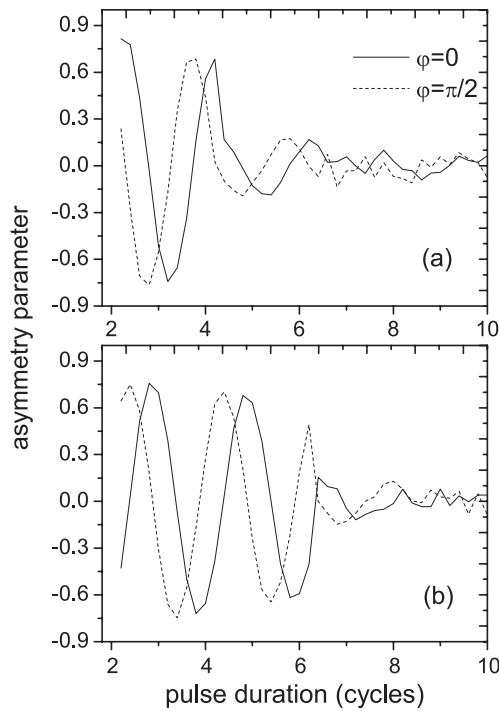


Fig. 3. The asymmetry parameter as a function of the pulse duration for laser intensities (a) $E_0 = 0.1$ a.u. and (b) $E_0 = 0.2$ a.u.

the ionization probabilities is larger than 0.5 in the first strongest half-cycle, the ionization probabilities must be less than 0.5 in the following half-cycle, consequently, the partial ionization probabilities are not identical although the electric field is symmetric. However, this does not exclude the case that the partial ionization probabilities are equal to each other at higher laser intensities. For intense pulses, it is possible for a sub-strongest half-cycle leading to the ionization probabilities about 0.5 and in the following strongest half-cycle the atoms are depleted, the partial ionization probabilities will be equal to each other. The corresponding CE phase varies with the pulse intensity. The expected phenomena are verified by our numerical calculations, as shown in Figure 2. At lower pulse intensity, the symmetric partial ionization probabilities appear at about $\varphi = 0.5\pi$ and 1.5π , as shown in Figure 2a; for higher pulse intensity used in Figure 2b, the symmetric partial ionization appears at about $\varphi = 0.15\pi$ and 1.1π , and for other intensities, the symmetric ionization will appear for other value of CE phase.

4.2 Variation of asymmetry with pulse duration

In photoionization, the CE phase shows its influence via the asymmetric ionization probability. The role of CE phase varies with the pulse duration, thus the asymmetry varies with the pulse duration.

Figure 3 shows the variation of asymmetry parameter with the pulse duration for two different pulse intensities. The asymmetry parameter in Figure 3 vibrates periodically upon increasing pulse duration until the duration

exceeds 6 cycles. Comparing Figure 3a with Figure 3b, two points are distinct. One is that with a higher pulse intensity the asymmetry degree persists up to the pulses with longer durations. For low intensity as in Figure 3a, the asymmetry is inconspicuous once the duration exceeding 4 cycles, whereas for high intensity as in Figure 3b, the asymmetry is distinct even for 6-cycle pulses. The other is that for lower intensities as shown in Figure 3a, the maximum of the asymmetry degree reaches a larger value which is CE phase-dependent, while for higher intensities as shown in Figure 3b, the maximal asymmetry is relatively smaller and the variation of its value with the CE phase is subtle. For the same CE phase, the maximum in Figure 3a points to the minimum in Figure 3b. The reverse is attributed to an intensity effect and will be discussed in the following subsection.

Why the asymmetry is more distinct at higher intensity than that at lower intensity when the pulse duration exceeds 4 cycles? The answer lies in the photoelectron's kinetic-energy spectrum. An ultrashort laser pulse is comprised of many frequency components, and an bounded electron may absorb photon energy from different frequency components for its ionization. Thus, multichannel transition can be formed for the ionization in ultrashort pulses, here a transition channel means a possible combination of absorbed-photon numbers in the ionization process [22]. The interference effect among different transition channels leads to the inversion asymmetry in the photoionization. The number of transition channels varies with the pulse duration and the kinetic energy of photoelectrons. For photoelectrons of higher kinetic energy and for the laser pulses with shorter duration, the number of transition channels is larger. At lower pulses intensities, the ionization is contributed mainly from the photoelectrons with smaller kinetic energy [23], the multichannel ionization appears only for the very short pulses, so does the asymmetry. Although the asymmetry appears for the photoelectrons of higher kinetic energy, their contribution to the total ionization is small. At higher laser intensities, the ionization is contributed mainly by the photoelectrons of relatively higher kinetic energy, for them the multichannel transition comes easily into being for relatively longer pulses, thus, the asymmetry appears for relatively longer pulse duration at higher laser intensities, as shown in Figure 3b.

At lower intensities, the value of the maximal asymmetry varies with the CE phase visibly. This phenomenon comes from the nonlinear dependence of ionization on the laser intensity. At lower intensities, a sub-strongest half-cycle ionizes less electrons. According to equation (5), a larger asymmetry can be reached. For the pulses with peak intensity just above the ionization threshold, the difference of the maximal electric field in a sub-strongest half-cycle to the ionization threshold varies with the CE phase, thus, the ratio of the electron yields in the sub-strongest and in the strongest half-cycles varies with the CE phase, then the possible maximum of the asymmetry varies with the CE phase. Because the electric field in shorter pulses varies more evidently with CE phase, this dependence

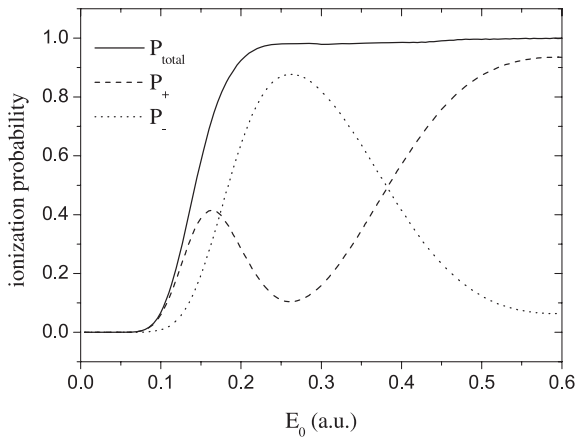


Fig. 4. The total ionization probability (solid line) and the partial ionization probabilities (dashed and dotted line respectively) as functions of the peak intensity of a four-cycle laser pulse with $\varphi = 0$.

of asymmetry on CE phase is more distinct for shorter pulses, as shown in Figure 3a.

Experimentally, Paulus et al. found that for pulse with duration longer than 6 cycles, the asymmetry is not significant [9]. In Grtler et al.'s experiment carried out for the ionization of Rydberg atoms in few-cycle RF pulses [11], the asymmetry exists for both sine and cosine pulses for strong pulses. Our calculations confirm those observations.

4.3 Variation of asymmetry with pulse intensity

Because the nonlinear dependence of photoionization on the laser intensity, the asymmetry is intensity-dependent. In this subsection we present a detailed investigation to the variation of asymmetry with the intensity. We will show that the trend of the asymmetric emission may change dramatically with pulse intensity and the pulse intensity determines the role of CE phase and pulse duration.

Figure 4 shows the total ionization probability and the partial ionization probabilities as functions of the peak intensity of laser pulse. The pulse is of four-cycle duration with the CE phase $\varphi = 0$. As shown in Figure 4, the ionization in very low intensity regime ($E < 0.05$ a.u.) is negligible. Upon increasing pulse intensity, the influence of the pulse intensity on the ionization asymmetry becomes obvious. Until the strength of the electric field reaches to 0.17 a.u., the emission to the positive x -direction is predominant: $P_+ > P_-$. For further higher laser intensities, the emission to the negative x -direction is predominant: $P_+ < P_-$, then the asymmetry is reversed. A second reversal occurs when the laser intensity is larger than 0.38 a.u. The same phenomenon is observed in Figure 2 which shows the partial ionization probabilities varying with CE phase at different laser intensities. When $\varphi = 0$, in the low intensity regime (Fig. 2a), more electrons emit to the positive x -direction whereas in the high intensity regime (Fig. 2b) more electrons emit to the negative x -direction.

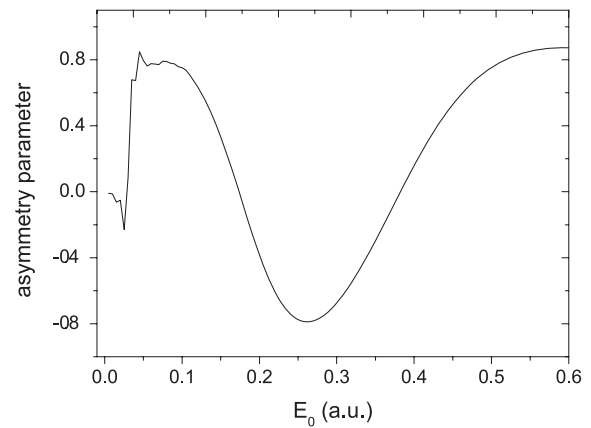


Fig. 5. Asymmetry parameter as a function of the peak intensity for a four-cycle laser pulse with $\varphi = 0$.

The asymmetry reversal along the pulse intensity can be interpreted intuitively on the basis of the instantaneous ionization probability, as shown in Figure 1. The reversal is determined by the question that in which half-cycle the maximal ionization occurs. In low-intensity regime, the maximal ionization occurs mainly in the strongest half-cycle, and the adjacent sub-strongest half-cycles ionize much less electrons to the opposite directions, thus a large asymmetry appears. Along with the increasing laser intensity, although both partial ionization probabilities increase, the asymmetry decreases. When the laser intensity exceeds some critical value, the sub-strongest half-cycle in the leading edge predominates the ionization, thus the asymmetry reverses. The asymmetry degree increases with the increasing laser intensity, then decreases when reaches its second maximum. Similar analysis holds for further increasing laser intensity. We also notice that the maxima of the partial ionization probabilities are the same after the first reversal, which reflects the fact that the atom target is completely depleted during one half-cycle.

According to the above analysis, it is expected that the value of the second maximum is less than that of the first value. Our calculation confirms this expectation for some CE phases. This result is suggestive to the experimental observations. In practical experiment to measure the value of CE phase, a distinct asymmetric signal is expected. Figure 5 shows the influence of the pulse intensity on the asymmetry parameter. The peak intensity of the laser pulse changes continuously. Although the largest asymmetry appears when the laser intensity is very low, the asymmetry is hard to be observed experimentally, because the number of ionized electrons in both directions are too small to be detected. When the laser intensity varies from $E = 0.05$ a.u. to $E = 0.12$ a.u., a broad maximum for the asymmetry parameter appears, and the experimental observation on the phase-dependent ionization can be performed at this intensity regime. For further increased laser intensity, although the maximum of the asymmetry is kept unchanged, the intensity is hard to be achieved for the phase-stabilized few-cycle pulses used presently.

5 Conclusions and discussions

Photoionization of hydrogen atom in few-cycle laser pulses is studied by numerically solving the 1D TDSE. The total ionization probability, the instantaneous ionization probabilities, and the partial ionization probabilities in a pair of opposite directions are obtained. It is found that the partial ionization probabilities are not always equal to each other which is termed as inversion asymmetry. The variation of asymmetry degree with the CE phase, the pulse duration and the pulse intensity is studied. We have shown that the pulse intensity is the most important parameter and determines the role of the CE phase and pulse duration. Firstly, the partial ionization probabilities vary with the CE phase and show some sine-like manner; secondly, the partial ionization probabilities equal to each other for some special CE phases. If the pulse intensity is less than a critical value (for hydrogen and for 800 nm laser pulses, it is about 0.1 a.u.), the inversion symmetry appears when the CE phase is about 0.5π and 1.5π ; when the pulse intensity is larger than that critical value, the inversion symmetry still appears, but the corresponding value of CE phase varies with pulse intensity; thirdly, the asymmetry degree varies with the pulse intensity and the pulse duration. For the pulses with duration less than 4 optical cycles, the maximum of asymmetry degree is of a larger value for lower intense pulses than that for higher intense pulses, but for the pulses with duration larger than 4 optical cycles, the asymmetry is more obvious for the pulses with larger peak intensity; finally, the maximal asymmetry at lower intensities varies with the CE phase visibly while at higher intensities not.

When compared to the observation of Gürtler et al., where the few-cycle RF pulses are used to ionize Rydberg atoms [11], the similarity of our calculations to their observations is multifold. From the viewpoint of a scaling law, the similarity is apprehensible [24]. The scaling law states that the photoionization in strong field is determined by three dimensionless numbers: (1) $u_p = U_p/\hbar\omega$, the ponderomotive energy in units of the laser photon energy; (2) $\epsilon_b = E_b/\hbar\omega$, the atomic binding energy in units of the laser photon energy; and (3) q , the absorbed-photon number. Keep these three parameters unchanged, there is no difference for the ionization in RF pulses and that in laser pulses.

This work is supported by the Chinese National Natural Science Foundation under Grant No. 60408008, National Key Basic Research Foundation under Grant No. G1999075200 and Natural Science Key Foundation of Shanghai under Grant No. 04JC14036.

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