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# Asymmetry of the photoionization rate of H atoms by a sequence of one-cycle laser pulses*

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Using an unperturbed scattering theory, the characteristics of H atom photoionization are studied respectively by a linearly- and by a circularly- polarized one-cycle laser pulse sequence. The asymmetry for photoelectrons in two directions opposite to each other is investigated. It is found that the asymmetry degree varies with the carrier–envelope (CE) phase, laser intensity, as well as the kinetic energy of photoelectrons. For the linear polarization, the maximal ionization rate varies with the CE phase, and the asymmetry degree varies with the CE phase in a sine-like pattern. For the circular polarization, the maximal ionization rate keeps constant for various CE phases, but the variation of asymmetry degree is still in a sine-like pattern.

**Keywords:** photoionization rate, ultrashort laser pulse, asymmetry
**PACC:** 3280, 3000, 4250, 4265

## 1. Introduction

With the rapid development in laser technology, it is now possible to produce intense laser pulses as short as only a few optical cycles.[1,2] For few-cycle laser pulses, the distribution of the electric field depends on the phase of the carrier wave with respect to the envelope, i.e. the carrier–envelope (CE) phase. More and more interest has been aroused in the CE phase-dependent phenomenon in few-cycle laser pulses, e.g. dynamics of strong-field photoionization of noble gas atoms [3−9] and generation of attosecond x-ray pulses.[10] Recently, Paulus *et al* experimentally investigated the influence of the CE phase on the photoionization of noble atoms,[4,5] and demonstrated the asymmetry for photoelectrons emitted into the two directions opposite to each other. Many theoretical studies confirmed the CE phase-dependent phenomenon in the photoionization by few-cycle laser pulses.[6−11]

The photoelectron rate for H atoms in a two-colour laser field depends on the relative phase between the two laser modes.[12,13] Recently, the asymmetry of the photoelectron rate of H atoms in a few-cycle laser pulse was studied.[14] In this paper, the asymmetry of the photoionization rate of H atoms respectively by a linearly- and by a circularly-polarized one-cycle laser pulse is studied in the frame of an unperturbed scattering theory.[15] We treat the Volkov state as an intermediate state and the final state of photoelectron as an electron–photon planar wave. Since the Volkov state describes a free electron moving in a laser planar wave, we neglect the Coulomb attraction of the parent ion to the ionized electron (strong-field approximation) and use a two-mode laser field to mimic the one-cycle laser pulse sequence.

## 2. Theory

The two laser modes used in this paper consist of a fundamental frequency $\omega$ and its second harmonic, and they are the same in both propagation direction and polarization. Different from the relevant theoretical study on two-colour photoionization,[16,17] the electric field strength of the fundamental mode is twice that of the harmonic mode. In such a modulation manner, the resultant electric field behaves as an infinite sequence of identical, one-cycle laser pulses, which

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can be expressed as follows:

$$\begin{aligned}E(t) =&E_0[2\sin(\omega t-\phi)-\sin(2\omega t-2\phi)]/4\\ =&E_0\sin^2(\omega t/2-\phi)\sin(\omega t-\phi),\end{aligned}$$

where $E_0$ is the peak value of the resultant electric field, and $\phi$ is the phase difference between the two modes. After a time displacement $t\to t+2\phi/\omega$ and introducing $T=2\pi/\omega$, the electric field can be rewritten as

$$E(t)=E_0\sin^2(\pi t/T)\sin(\omega t+\phi). \tag{1}$$

In such a field, the first term $E_0\sin^2(\pi t/T)$ on the right-hand side of the above expression can be regarded as the envelope of an infinite sequence of one-cycle pulses, while the second term $\sin(\omega t+\phi)$ acts as the carrier wave. Thus, the resultant field corresponds to an infinite sequence of one-cycle pulses, each shares a common phase $\phi$. Throughout this paper, we refer to the peak intensity of the resultant one-cycle pulses as the laser intensity and the phase difference $\phi$ as the CE phase.

The state of an electron in one-cycle laser pulse sequence is described by a two-mode Volkov state defined as follows ($\hbar=1, c=1$):

$$\begin{aligned}|\psi_\mu\rangle =&V_{\rm e}^{-1/2}\sum_{j_1,j_2}\exp\{{\rm i}[P+(u_{p_1}-j_1)k_1\\ &+(u_{p_2}-j_2)k_2]\cdot r\}\\ &\times\chi_{-j_1,-j_2}(z)|n_1+j_1,n_2+j_2\rangle,\end{aligned} \tag{2}$$

where $V_{\rm e}$ denotes the electron's normalized volume, $\boldsymbol{P}$ is the electron's momentum in the electric field, the ponderomotive parameters $u_{p1}$ and $u_{p2}$ are defined as $u_{p1}=\dfrac{e^2\Lambda_1^2}{m_{\rm e}\omega_1}$ and $u_{p2}=\dfrac{e^2\Lambda_2^2}{m_{\rm e}\omega_2}$, where $2\Lambda_i$ is the classical amplitude of the $i$th mode, and $\Lambda_1=4\Lambda_2$, $\omega_1=\omega$, $\omega_2=2\omega$, $\omega_2=2\omega_1$, thus $u_{p2}=u_{p1}/32$. Vectors $\boldsymbol{k}_1$ and $\boldsymbol{k}_2$ are the wave vectors of two laser modes, respectively; $n_i$ is the background photon number in the $i$th ($i$=1, 2) mode, and $j_i$ denotes the number of transferred photons. Here $\chi_{-j_1,-j_2}(z)$ is the two-mode generalized phased Bessel (GPB) function:[17]

$$\begin{aligned}\chi_{-j_1,-j_2}(z)=&\sum_{q_1\cdots q_4}X_{j_1-2q_1-q_3-q_4}(\zeta_1)X_{j_2-2q_2-q_3+q_4}(\zeta_2)\\ &\times X_{q_1}(z_1)X_{q_2}(z_2)X_{q_3}(z_3)X_{q_4}(z_4),\end{aligned}$$

where the sum is performed over $-\infty\ \langle q_i(i=1,...,4)\langle\infty$, and $X_n(z)$ is the phased Bessel function related to the ordinary Bessel function by

$$X_n(z)=J_n(|z|{\rm e}^{{\rm i}n\arg(z)}.$$

The arguments of the GPB function are defined as

$$\begin{aligned}&\zeta_1=\frac{2|e|\Lambda_1}{m_{\rm e}\omega_1}\boldsymbol{P}\cdot\boldsymbol{\varepsilon}_1,\quad \zeta_2=\frac{2|e|\Lambda_2}{m_{\rm e}\omega_2}\boldsymbol{P}\cdot\boldsymbol{\varepsilon}_2,\\ &z_1=\frac{1}{2}u_{p_1}\boldsymbol{\varepsilon}_1\cdot\boldsymbol{\varepsilon}_1,\quad z_2=\frac{1}{2}u_{p_2}\boldsymbol{\varepsilon}_2\cdot\boldsymbol{\varepsilon}_2,\\ &z_3=\frac{2e^2\Lambda_1\Lambda_2\boldsymbol{\varepsilon}_1\cdot\boldsymbol{\varepsilon}_2}{m_{\rm e}(\omega_1+\omega_2)},\quad z_4=\frac{2e^2\Lambda_1\Lambda_2\boldsymbol{\varepsilon}_1\cdot\boldsymbol{\varepsilon}_2^*}{m_{\rm e}(\omega_1-\omega_2)}.\end{aligned} \tag{3}$$

In Eq.(3), $\boldsymbol{\varepsilon}_j$ ($j=1,2$) is the polarization vector of the $j$th mode, and defined as

$$\begin{aligned}\boldsymbol{\varepsilon}_j&=[\varepsilon_x\cos(\xi/2)+{\rm i}\varepsilon_y\sin(\xi/2)]{\rm e}^{{\rm i}\phi_j},\\ \boldsymbol{\varepsilon}_j^*&=[\varepsilon_x\cos(\xi/2)-{\rm i}\varepsilon_y\sin(\xi/2)]{\rm e}^{-{\rm i}\phi_j},\end{aligned} \tag{4}$$

where $\xi$ monitors the polarization degree, so that $\xi$=0 denotes linear polarization and $\xi=\pi/2$ circular polarization; $\phi_j$ is the phase angle of the $j$th ($j=1,2$) mode, which is related to the CE phase by

$$\phi_1=\pi,\quad \phi_2=-\phi,$$

where $\phi$ is the CE phase, and the polarization vectors satisfy the following relations:

$$\boldsymbol{\varepsilon}_j\cdot\boldsymbol{\varepsilon}_k=\cos(\xi){\rm e}^{{\rm i}(\phi_j+\phi_k)},\quad \boldsymbol{\varepsilon}_j\cdot\boldsymbol{\varepsilon}_k^*={\rm e}^{{\rm i}(\phi_j-\phi_k)}. \tag{5}$$

By means of the Volkov state in Eq.(2) the photoelectron rate is worked out to be

$$\begin{aligned}\frac{d^2W}{d\Omega_{P_{\rm f}}}=&\frac{(2m_{\rm e}^3\omega^5)^{1/2}}{(2\pi)^2}(q-\varepsilon_b)^{1/2}|\Phi_i(P_{\rm f})|^2\\ &\times\Bigg|\sum_{q_1,q_2}[(u_{p1}-q_1)+2(u_{p2}-q_2)]\\ &\times\sum_{j_1,j_2}\chi_{-j_1+q_1,-j_2+q_2}(z_{\rm f})\chi_{-j_1,-j_2}(z_{\rm f})^*\Bigg|^2,\end{aligned} \tag{6}$$

where $\Phi_i(\boldsymbol{P}_{\rm f})$ is the Fourier transformation of the initial wave function with binding energy $E_b$, $\varepsilon_b=E_b/\omega$, and $\boldsymbol{P}_{\rm f}$ is the final momentum of photoelectron. The argument $z_{\rm f}$ means that $\boldsymbol{P}$ in Eq.(3) is replaced by $\boldsymbol{P}_{\rm f}$ while the others are kept unchanged. $d\Omega_{P_{\rm f}}=\sin\theta_{\rm f}{\rm d}\theta_{\rm f}{\rm d}\phi_{\rm f}$ is the differential solid angle of the final photoelectron, where $\theta_{\rm f}$ and $\phi_{\rm f}$ are the scattering angle and the azimuthal angle, respectively; $q$ is related to the final kinetic energy of the photoelectron by $\dfrac{P_{\rm f}^2}{2m_{\rm e}}=q\omega-E_b$, where $q\omega=q_1\omega_1+q_2\omega_2$, $q_i=l_i-m_i$ ($i=1,2$) is the overall transferred photon number in the $i$th mode. The sum over $q_i$ is performed over all possible $q_1,q_2$ for a fixed $q$. Since most of the experimental observations of photoelectron rate are performed in the polarization plane defined by $\theta_{\rm f}=\pi/2$,

we fix $\theta_f = \pi/2$ in our calculations and vary $\phi_f$ from 0 to $2\pi$.

We identify one set of $q_i$ $(i = 1, 2)$ that satisfies $q\omega = q_1\omega_1 + q_2\omega_2$ for a fixed $q$ as a transition channel. The phase of each channel varies with the value of $q_i$, thus different channels correspond to different phases. When subjected to a space reflection, each transition channel produces an additional phase factor related to these $q_i$. Whether the transition rate changes or not is determined by the sum of $q_i$. When several channels are involved to form an ATI peak, the interference effect among channels will affect the total ionization rate. Thus, the photoelectron rates in a pair of opposite directions are not always equal to each other, which is termed inversion asymmetry. On the other hand, the inversion asymmetry reflects the asymmetric distribution of the electric field in the pulse envelope.[4] Because the electric field distribution varies with the laser polarization, the asymmetry degree also varies with the laser polarization.

# 3. Results and analysis

The inversion asymmetry denotes the difference between the photoelectron rates in a pair of opposite directions. We use $P_{\rm left}$ to denote the photoelectron rate in a direction (i.e. $\phi_f = 0$), and $P_{\rm right}$ in its opposite direction (i.e. $\phi_f = \pi$). When the numbers of photoelectrons emitted into the left and the right directions are equal, the asymmetry degree is zero and $P_{\rm left} = P_{\rm right}$. Generally, the photoelectron rate in a direction is not always equal to that in its opposite direction, so the asymmetry degree is not always zero and $P_{\rm left} \neq P_{\rm right}$. In order to describe the asymmetry degree quantitatively, we introduce an asymmetry parameter as follows:[18]

$$a = \frac{P_{\rm left} - P_{\rm right}}{P_{\rm left} + P_{\rm right}}. \tag{7}$$

This parameter is expected to be an effective scale to characterize the photoelectron asymmetry:[18] if $a$ is zero, it denotes a symmetric ejection into two opposite directions, i.e. the number of photoelectrons emitted into the left direction is equal to that into the right direction; and if $a$ is $+1$ or $-1$, it denotes an asymmetric ejection, i.e. the ionized photoelectrons are all ejected into the left direction or the right direction.

In our calculations, the central wavelength is chosen as 800nm, and the wavefunction is chosen as the ground state wavefunction of H atoms with a binding energy of 13.6eV. According to Eq.(7), the asymmetry parameters of H atom in linearly- and circularly-polarized pulses are calculated respectively.

### 3.1. Asymmetry in linearly-polarized pulses

For linearly-polarized laser pulses, the CE phase determines the maximal electric-field strength in the pulse, and the simultaneous electric-field strength determines the rate of photoelectron ionized, so the maximal ionization is determined by the CE phase and the asymmetry varies with the CE phase.

Generally, the photoelectron rates are not identical in the left and right directions. We plot in Fig.1 the asymmetry parameter as a function of the CE phase with the peak laser intensity being $2 \times 10^{13}{\rm W/cm^2}$. From Fig.1 we find that the asymmetry parameter of H atoms ionized by one-cycle linearly-polarized pulse sequence depends on the CE phase significantly, the inversion asymmetry for photoelectrons emitted into a pair of opposite directions depends on the CE phase. For most CE phases, the asymmetry parameter $a$ is not equal to zero, i.e. the numbers of photoelectrons ejected into the left and right directions are not equal, thus the inversion asymmetry is demonstrated. And the asymmetry degree varies with the CE phase in a sine-like pattern. We choose Fig.1 (c) as an example to further show the details. When the CE phase is 0, the asymmetry parameter is close to –1.0, that is to say, most photoelectrons are ejected into the right direction, the minimal photoelectron distribution appears at $\phi_f = 0$, while the maximal photoelectron distribution occurs at $\phi_f = \pi$; when CE phase is $\pi$, the photoelectron distribution is in the opposite case: the asymmetry parameter is close to 1.0, i.e. the most ionized photoelectrons are ejected into the left direction, the minimal photoelectrons distribution appears at $\phi_f = \pi$, while the maximal photoelectrons distribution occurs at $\phi_f = 0$. A phase difference of $\pi$ means that the maximal electric field strengths of the two short pulses reside in two opposite directions. This phenomenon suggests that the electrons are ionized mostly when the electric field reaches its maximum. We can also find from Fig.1 that when the CE phase is $\pi/2$ or $3\pi/2$, the asymmetry parameter is 0, i.e. the number of photoelectrons emitted into the left direction is the same as that into the right direction.

Comparing Fig.1 (c) respectively with Figs.1 (a) and (b), we can see that for higher photoelectron energies, the asymmetry parameter curve is steeper, i.e. for the higher photoelectron energies, the variation curve of asymmetry parameter depending on the CE

phase changes more rapidly. Because the ionization rate of lower-energy electron is far larger than that of higher energy electron, the asymmetry degree of the overall ionization shows a sine-like pattern varying with the CE phase. This feature agrees with Paulus's observation.[4]

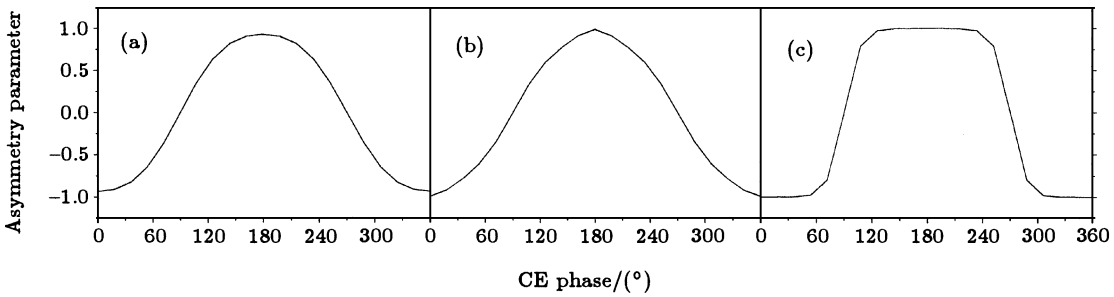



**Fig.1.** Calculated asymmetry parameter for the second (a), the fourth (b) and the seventh (c) ATI peaks depending on the CE phase for H atoms in a sequence of linearly-polarized one-cycle laser pulses. The laser peak intensity is chosen as $2\times10^{13}$W/cm$^2$ and the central wavelength is 800nm.

In Fig.2, we show the asymmetry parameter as a function of the CE phase at a higher laser peak intensity. Comparing Fig.2 with Fig.1, we find that for a higher intensity, the domains of the maximal and minimal photoelectron occupation are much wider. In Fig.2 (a) the plot shows a split, which is caused by the inherent property of photoionization.[19]

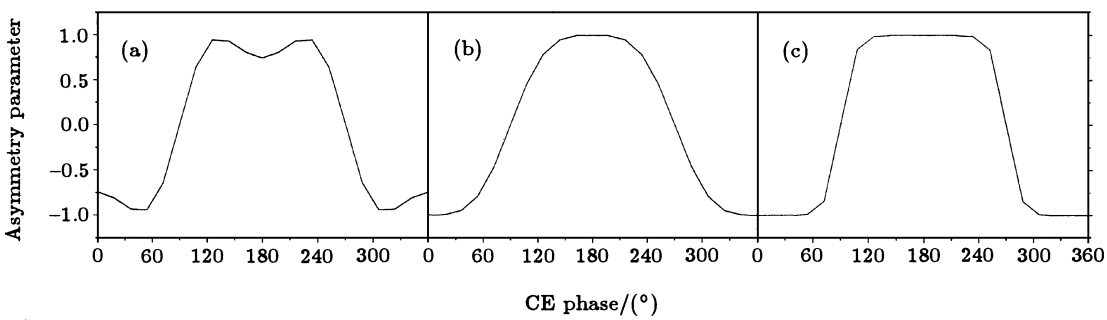



**Fig.2.** The same as those in Fig.1 but for laser peak intensity at $8\times10^{13}$W/cm$^2$.

### 3.2. Asymmetry in circularly-polarized pulses

We show in Fig.3 the asymmetry parameter as a function of the CE phase in circularly-polarized one-cycle laser pulses, the peak laser intensity is chosen as $8\times10^{13}$W/cm$^2$. Figures 3 (a)–(c) correspond to the second, the fourth and the seventh order ATI peaks respectively. It is evident from Fig.3 that the photoelectrons emitted into the left and right directions are inversion asymmetric for all ATI peaks with various CE phases, and the asymmetry degree also varies in a sine-like pattern. This is because, for ultrashort circularly-polarized laser pulses, the electric field of pulses changes with the CE phase. The CE phase determines the direction of the maximal field, and the intense-matter interaction depends on the electric field of the pulse. Consequently, the CE phase determines the direction of maximal photoelectron occupation. These results are consistent with the analysis of Paulus *et al.*[5] It can be also seen from Fig.3 that when the CE phase is $\pi/2$ or $3\pi/2$, the asymmetry parameter is 0, and the numbers of photoelectrons ejected into the left direction and right direction are equal.

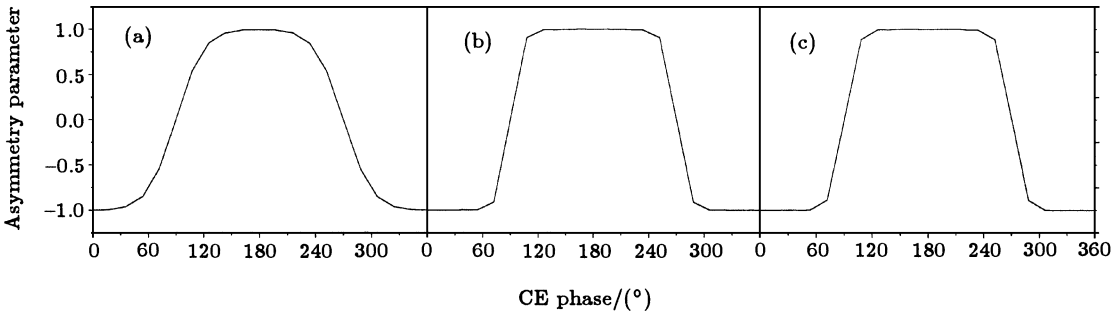



**Fig.3.** The same as those in Fig.2 but for circular polarization.

In order to show the difference in photoionization between by circularly-polarized pulses and by linearly-polarized pulses, we depict in Fig.4 photoelectron angular distribution for H atoms in circularly-polarized laser pulses in a polar coordinate system. The peak laser intensity is chosen as $8 \times 10^{13}\mathrm{W/cm^2}$ and the wavelength is 800nm. Figures 4 (a)–(c) correspond to different CE phases.

From Fig.4 we can see the following points. Firstly, the maximal ionization rate remains unchanged for various CE phases, but the position of the maximum varies with the CE phase. This phenomenon reflects the fact that the maximal field strength of the resultant circularly-polarized pulses keeps constant, but the position of the maximal electric-field strength varies with the CE phase. Secondly, due to the symmetry of the electric field with respect to its maximum, the photoelectrons emitted into two opposite directions are symmetric about an axis related to the CE phase. Finally, the asymmetry still exists when the CE phase is $\pi/2$ or $3\pi/2$ although the asymmetry parameter is 0. In this case, the maximal number of photoelectrons emitted appears at $\phi_{\mathrm{f}} = \pi/2$ and $3\pi/2$, respectively.

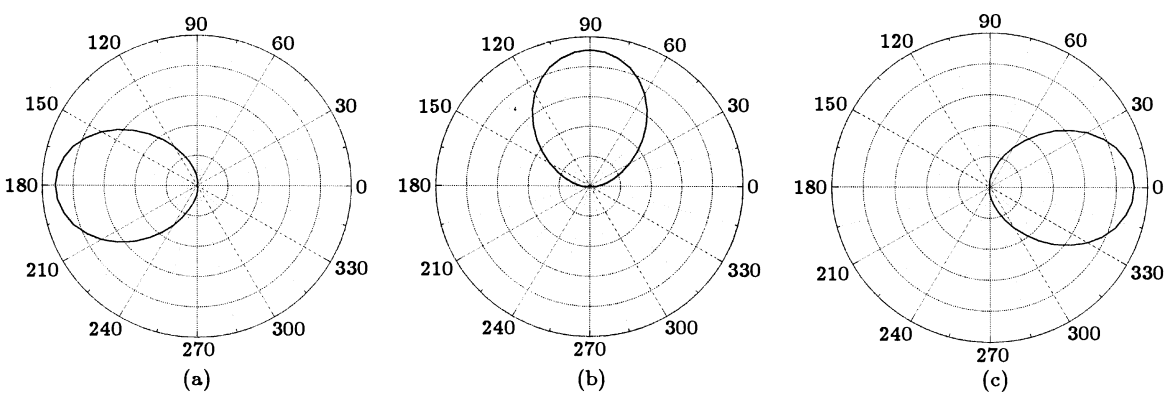



**Fig.4.** Calculated photoelectron angular distribution for the seventh ATI peak of H atom in a sequence of circularly polarized one-cycle pulses for the CE phase: (a) at 0, (b) at $\pi/2$ and (c) at $\pi$. The peak laser intensity we choose is $8\times10^{13}\mathrm{W/cm^2}$, and the central wavelength is 800nm.

# 4. Conclusions

We conclude as follows.

First, the photoelectron rates in a pair of opposite directions are not always equal to each other, and the asymmetry degree varies with the CE phase, the laser intensity, and the kinetic energy of the photoelectrons.

Second, in linearly-polarized one-cycle pulse sequence, the maximal ionization rate varies with the CE phase, and the asymmetry degree varies with the CE phase in a sine-like pattern.

Third, in circularly-polarized one-cycle pulse sequence, the maximal ionization rate keeps constant for various CE phases, but the variation of asymmetry degree is still shown to be in a sine-like pattern.

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