

# Asymmetry flip in photoelectron angular distribution of rare gas atoms in intense circularly-polarized few-cycle laser fields

Shinichi Fukahori<sup>1</sup>, Kaoru Yamanouchi<sup>1,\*</sup>, and Gerhard G. Paulus<sup>2</sup>

<sup>1</sup>Department of Chemistry, School of Science, The University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-0033, Japan

<sup>2</sup>Institut für Optik und Quantenelektronik, Friedrich-Schiller-Universität, Max-Wien-Platz 1, 07743 Jena, Germany

**Abstract.** An analytical formula representing the photoelectron kinetic energy at which the ejection direction of photoelectrons generated by an intense circularly-polarized few-cycle laser pulse flips was derived and was used for determining the laser pulse duration.

## 1 Introduction

The waveform of an ultrashort laser pulse with a few optical cycles, called a few-cycle laser pulse, is characterized by its carrier-envelope phase (CEP). It has been known that photoionization processes of atoms and molecules and photodissociation processes of molecules are affected sensitively by the CEP of intense few-cycle laser pulses [1]. For the investigation of the CEP dependence of the photo-induced dynamical processes, the CEP-tagging method has been widely used, that is, the CEP of each laser shot is recorded by an apparatus called a phasemeter in coincidence with photoelectrons and/or photoions produced by the target atoms or molecules in another vacuum chamber [1,2]. However, it is not an easy task to estimate the absolute CEP at the spatial point where atoms and molecules interact with the laser field because the CEP recorded by the tagging method is in most cases a relative value having a constant offset from the absolute CEP.

In our recent study [3], we showed that the absolute CEP can be determined in a straightforward manner as long as we photoionize Ar atoms by a near-infrared circularly-polarized few-cycle laser pulses ( $1 \times 10^{14} \sim 5 \times 10^{14}$  W/cm<sup>2</sup>) and record the energy-resolved photoelectron angular distribution by taking advantage of the fact that the emitting angle of the photoelectrons ejected from Ar takes a constant value in the high kinetic energy range above 30 eV [3]. In addition, we found that the ejection direction of the photoelectrons is flipped by  $\pi$  when the photoelectron kinetic energy goes across the kinetic energy of  $\sim 20$  eV to the higher kinetic energy range.

In the present study, we examine this phenomenon called the asymmetry flip on the basis of the tunnel ionization theory and derived an analytical formula representing the kinetic energy of photoelectrons,  $E_{\text{flip}}$ , at which the flip occurs. We also show that this

---

\* Corresponding author: [kaoru@chem.s.u-tokyo.ac.jp](mailto:kaoru@chem.s.u-tokyo.ac.jp)

analytical formula can be used for determining the temporal duration of the circularly-polarized few-cycle laser pulses.

## 2 Theory

When photoelectrons are generated by tunnelling at time  $t = t_0$  in the circularly-polarized laser electric field whose profile is given by

$$\mathbf{E}(t) = E_0 \exp \left[ -2(\ln 2) \left( \frac{\omega t}{2\pi N} \right)^2 \right] (\hat{\mathbf{x}} \cos \omega t - \hat{\mathbf{y}} \sin \omega t), \quad (1)$$

where  $E_0$  is the amplitude of the laser field,  $\omega$  is the laser frequency and  $N$  is the number of optical cycles in the laser field, the photoelectrons tunnelling at  $t_0 = 0$  are ejected towards the  $+y$  direction. On the other hand, the photoelectrons tunnelling at  $t_0 = -T/2$  and  $T/2$  are ejected towards  $-y$  direction, where  $T = 2\pi/\omega$  is the period of the optical cycle. Because the kinetic energy of the photoelectrons at  $t_0 = -T/2$  and  $T/2$  is smaller than that at  $t_0 = 0$ , it is considered that the photoelectron is ejected towards  $+y$  direction in the high energy range, while the photoelectron is ejected towards  $-y$  direction in the low energy range.

The final momentum of the photoelectron ejected from an atom by the tunnel ionization at  $t = t_0$  is given by

$$\mathbf{p}_{\text{final}} \approx \mathbf{p}_0 + \frac{E_0}{\omega} \exp \left[ -2(\ln 2) \left( \frac{\omega t_0}{2\pi N} \right)^2 \right] (\hat{\mathbf{x}} \sin \omega t_0 + \hat{\mathbf{y}} \cos \omega t_0), \quad (2)$$

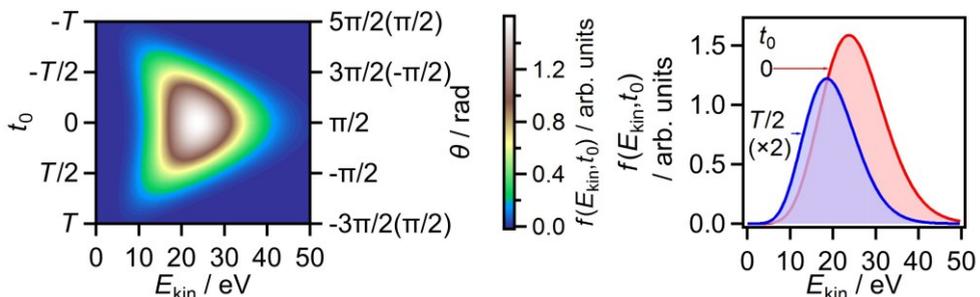
where  $\mathbf{p}_0$  is the initial photoelectron momentum at  $t = t_0$ , showing that the emitting angle  $\theta$  in the  $x$ - $y$  coordinate system is expressed as  $\theta = \pi/2 - \omega t_0$ . The photoelectron kinetic energy is calculated using  $\mathbf{p}_{\text{final}}$  as  $E_{\text{kin}} = |\mathbf{p}_{\text{final}}|^2/2$ .

By using the tunnel ionization rate  $W(t_0)$  and the distribution  $w(p_{\perp})$  of the initial momentum  $p_{\perp} = |\mathbf{p}_0|$  of the photoelectron perpendicular to the laser electric field  $\mathbf{E}(t_0)$ , both of which are derived by the theory of the tunnel ionization, the kinetic energy distribution of the photoelectrons ejected by the tunnel ionization at  $t = t_0$  is obtained as

$$f(E_{\text{kin}}, t_0) = W(t_0) w(p_{\perp}) / \sqrt{2E_{\text{kin}}}, \quad (3)$$

where the initial momentum  $p_{\perp}$  is given by

$$p_{\perp} = -\frac{E_0}{\omega} \exp \left[ -2(\ln 2) \left( \frac{\omega t_0}{2\pi N} \right)^2 \right] + \sqrt{2E_{\text{kin}}}. \quad (4)$$



**Fig. 1.** The kinetic energy distributions  $f(E_{\text{kin}}, t_0)$  when  $E_0 = 4.38 \times 10^{10}$  V/m,  $N = 1.7$ ,  $I_p = 15.76$  eV and  $\lambda = 722$  nm are adopted. (a) The two-dimensional map. The corresponding  $\theta$  values are also shown. (b) The red and blue lines show  $f(E_{\text{kin}}, 0)$  and  $f(E_{\text{kin}}, T/2)$ , respectively. The distribution  $f(E_{\text{kin}}, T/2)$  is multiplied by a factor of 2.

The kinetic energy distribution  $f(E_{\text{kin}}, t_0)$  of the photoelectrons ejected by the tunnel ionization of Ar is shown in Fig. 1(a).

### 3 Results and discussion

When the emitting angle is  $\theta = \pi/2$ , that is, when the photoelectron is ejected towards  $+y$  direction, the corresponding  $t_0$  value becomes  $t_0 = 0$  because  $\theta$  is given by  $\theta = \pi/2 - \omega t_0$ . Therefore, the kinetic energy distribution at  $\theta = \pi/2$  can be expressed as  $f(E_{\text{kin}}, 0)$ . Similarly, when the emitting angle is  $\theta = -\pi/2$ , that is, when the photoelectron is ejected towards  $-y$  direction, two  $t_0$  values,  $t_0 = T/2$  and  $-T/2$ , contribute, and consequently, the kinetic energy distribution at  $\theta = \pi/2$  can be expressed as  $2f(E_{\text{kin}}, T/2) = f(E_{\text{kin}}, T/2) + f(E_{\text{kin}}, -T/2)$ .

As shown in Fig. 1(b), the distribution  $f(E_{\text{kin}}, 0)$  takes the larger value than  $2f(E_{\text{kin}}, T/2)$  in the range of  $E_{\text{kin}} > 19$  eV while it takes the smaller value than  $2f(E_{\text{kin}}, T/2)$  in the range of  $E_{\text{kin}} < 19$  eV, which means that the yield of the photoelectrons at  $\theta = \pi/2$  is larger than that at  $\theta = -\pi/2$  in the range of  $E_{\text{kin}} > 19$  eV while the yield at  $\theta = \pi/2$  is smaller than that at  $\theta = -\pi/2$  in the range of  $E_{\text{kin}} < 19$  eV. Because the flip of the ejection direction of the photoelectrons occurs when the the photoelectron yield at  $\theta = \pi/2$  and that at  $\theta = -\pi/2$  are balanced,  $f(E_{\text{flip}}, 0) = 2f(E_{\text{flip}}, T/2)$  holds at the asymmetry flip energy  $E_{\text{flip}}$ . From this requirement, we obtain the analytical formula,

$$\frac{E_{\text{flip}}}{U_p} = 2 \frac{1}{2N^2} + \frac{(\ln 2)h\omega}{2 \left( \frac{1}{2^{2N^2}} - 1 \right) I_p} \left[ 1 + \frac{1}{2N^2} \left( \sqrt{\frac{2}{I_p}} - \frac{1}{2} \right) \right] \gamma - \frac{2}{3} \gamma^2, \quad (5)$$

where  $U_p = E_0^2/(2\omega^2)$  is the ponderomotive energy,  $I_p$  is the ionization energy of a target atom and  $\gamma = \sqrt{I_p/U_p}$  is the Keldysh parameter. This formula shows that the asymmetry flip energy  $E_{\text{flip}}$  changes depending on the number of optical cycles  $N$ . Therefore, by substituting the  $E_{\text{flip}}$  value obtained experimentally and the laser-field intensity  $E_0$  into eq. (5), the temporal pulse duration of the few-cycle pulses can be estimated

In summary, on the basis of the semiclassical tunnel ionization theory, the analytical formula representing the asymmetry flip energy has been derived as a function of the number of optical cycles within the laser pulse. By recording the asymmetry in the photoelectron emission from atoms by circularly-polarized few-cycle intense laser pulses and deriving the asymmetry flip energy  $E_{\text{flip}}$ , the pulse duration of the few-cycle pulses can be estimated straightforwardly using the analytical formula (5) obtained above.

This work was supported by JSPS KAKENHI Grant No. JP15H05696 and by the German Research Foundation (DFG) Grant No. PA 730/7.

### References

1. S. Miura, T. Ando, K. Ootaka, A. Iwasaki, H. Xu, T. Okino, K. Yamanouchi, D. Hoff, T. Rathje, G. G. Paulus, M. Kitzler, A. Baltuška, G. Sansone, M. Nisoli, *Chem. Phys. Lett.* **595–596**, 61–66 (2014).
2. T. Wittmann, B. Horvath, W. Helml, M. G. Schätzel, X. Gu, A. L. Cavalieri, G. G. Paulus, R. Kienberger, *Nat. Phys.* **5**, 357–362 (2009).
3. S. Fukahori, T. Ando, S. Miura, R. Kanya, K. Yamanouchi, T. Rathje, G. G. Paulus, *Phys. Rev. A* **95**, 053410 (2017).