

**Carrier-envelope-phase effect in a long laser pulse with tens of optical cycles**Xi Zhao,<sup>1,2</sup> Jing Chen,<sup>3,4</sup> Panming Fu,<sup>2</sup> Xueshen Liu,<sup>1</sup> Zong-Chao Yan,<sup>5</sup> and Bingbing Wang<sup>2,\*</sup><sup>1</sup>*Institute of Atomic and Molecular Physics, Jilin University, Changchun 130012, China*<sup>2</sup>*Laboratory of Optical Physics, Beijing National Laboratory for Condensed Matter Physics, Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China*<sup>3</sup>*Key Laboratory of High Energy Density Physics Simulation, CAPT, Peking University, Beijing 100084, China*<sup>4</sup>*Institute of Applied Physics and Computational Mathematics, P.O. Box 8009, Beijing 100088, China*<sup>5</sup>*State Key Laboratory of Magnetic Resonance and Atomic and Molecular Physics, Wuhan Institute of Physics and Mathematics, Chinese Academy of Sciences, Wuhan 430071, China,**Center for Cold Atom Physics, Chinese Academy of Sciences, Wuhan 430071, China,**and Department of Physics, University of New Brunswick, Fredericton, New Brunswick E3B 5A3, Canada*

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The carrier-envelope phase (CEP) is a crucial parameter of a laser pulse, which can influence all dynamic processes in matter-laser interaction. In this work, we find a clear CEP effect in laser-driven atomic bound-bound transition for a long laser pulse. Especially, the CEP effect is attributed to the interference between the sum-frequency and the difference-frequency components within a two-photon transition, where the degree of this interference depends only on the atomic energy-level structure and is independent of the laser pulse duration. Therefore, the CEP effect can stay robust for a long laser pulse containing tens of laser cycles. This result provides a method to measure the CEP value of a long laser pulse and may thus shed light on the study of characteristics of high-frequency XUV and x-ray laser pulses.

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**I. INTRODUCTION**

The carrier-envelope phase (CEP) is a crucial parameter in describing the characteristics of a laser pulse, because it can influence dynamic processes when an atom or molecule interacts with a laser pulse [1–8]. Currently, many dynamical processes have been precisely investigated on the attosecond time scale, as few-cycle laser pulses are available for experiments [9–11]. Hence stable and precise control of a laser's CEP is essential in such experiments. For example, Bergues *et al.* [11] investigated experimentally the attosecond tracing of the correlated electron emission in nonsequential double ionization of an Ar atom in a near-single-cycle laser, where the CEP was well controlled. Moreover, Xie *et al.* [12] demonstrated experimentally that the fragmentation reactions of complex molecules can be controlled by the CEP of a few-cycle laser pulse on subfemtosecond time scales.

Even though the influence of CEP on a laser-driven dynamic process will decrease as the laser pulse duration increases, this influence is still an important factor if one wants to study precisely the dynamic process of matter-laser interaction [13,14]. Sansone *et al.* [13] demonstrated that the CEP effect can be observed clearly in a high harmonic spectrum with a multi-optical-cycle laser pulse. By using a two-color radio-frequency pulse, Jha *et al.* [14] found experimentally clear evidence of the CEP difference between the two fields on multiphoton transitions between Zeeman sublevels, as the pulses are many cycles long. However, the problem of how to measure and control the CEP of a relatively long laser pulse, with tens of optical cycles, is still a great challenge. This is because most CEP effects are attributed to the dependence of the space-time asymmetry of the laser's electric field on

CEP values [6,7,15,16]; hence these CEP effects will decrease rapidly as the pulse duration increases, such as the CEP effect on high harmonic generation [1,17] and the ionization yield [5,6,8,18,19]. In general, this kind of CEP effect can hardly be detected, as the laser pulse duration is longer by about eight optical cycles [15,16].

In this work, we provide a path to investigate the CEP effect in a long laser pulse, with tens of laser cycles, in atomic bound-bound transitions. In particular, we find that an obvious CEP effect on a two-photon three-level transition can be observed for a long laser pulse when the laser frequency is resonant with the first step transition frequency. Because the CEP effect originates from the interference between the sum-frequency and the difference-frequency components within a two-photon transition, where this interference is independent of the pulse duration, it can stay robust even for a long laser pulse, as the laser contains several tens of optical cycles. This result provides a pathway to measurement of the absolute value of the CEP of a long laser pulse.

**II. CEP EFFECT OF TWO-PHOTON TRANSITION**

We calculate the transition probabilities of excited states from the initial ground state of a hydrogen-like atom in a laser pulse by using the three-dimensional time-dependent Schrödinger equation [20] (atomic units are used throughout, unless otherwise stated):

$$i \frac{\partial \Psi(\mathbf{r}; t)}{\partial t} = [H_0(\mathbf{r}) + H_I(t)]\Psi(\mathbf{r}; t), \quad (1)$$

where the atomic and interaction parts of the Hamiltonian are, respectively,

$$H_0(\mathbf{r}) = -\frac{1}{2}\nabla^2 - \frac{Z_{\text{eff}}}{r}, \quad H_I(t) = \mathbf{r} \cdot \mathbf{e}(t), \quad (2)$$

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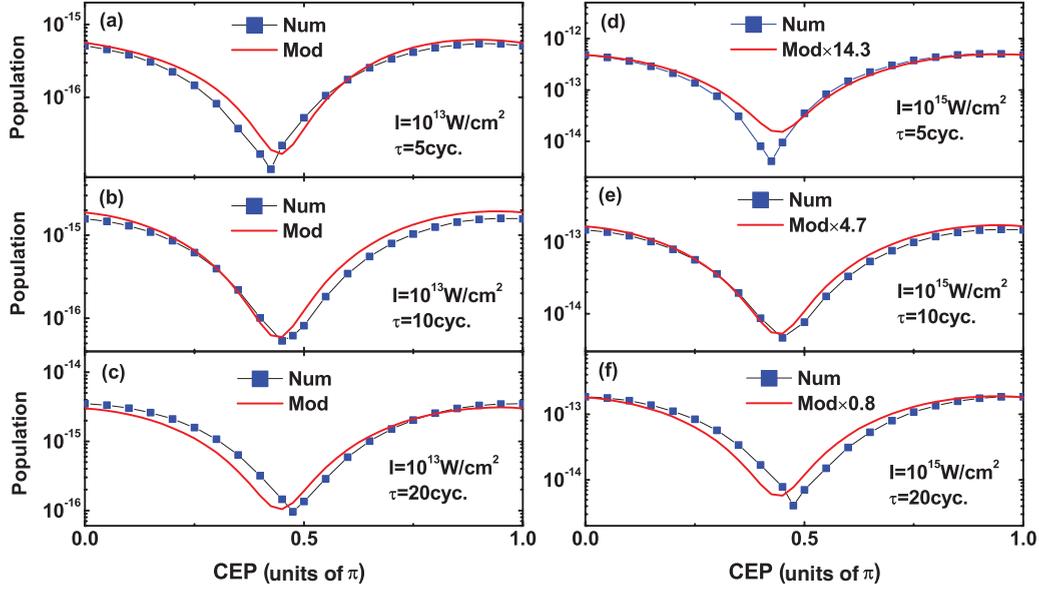


FIG. 1. (Color online) Population of  $3^2D$  as a function of the CEP, where the (blue) line with squares represents the numerical result by Eq. (1), and the (red) line is the model result with  $\tau = 5$  cycles (a, d), 10 cycles (b, e), and 20 cycles (c, f). The laser intensity  $I = 10^{13}$  W/cm $^2$  (a–c) and  $10^{15}$  W/cm $^2$  (d–f). The laser frequency  $\omega = 0.4345$ .

with  $Z_{\text{eff}}$  being the effective nuclear charge. Here we choose the value  $Z_{\text{eff}} = 1.08$  so that the ionization threshold is the same as that of Ar. Hence the energy difference between its ground state  $1^2S$  and  $2^2P$  is 0.4345 and that between  $2^2P$  and  $3^2D$  is 0.11. The laser's electric field can be written as

$$\mathbf{e}(t) = \hat{z}e_0 \exp(-\alpha^2 t^2) \cos(\omega t + \phi), \quad (3)$$

where  $e_0$  is the amplitude of the laser pulse,  $\omega = 0.4345$  is the laser frequency that equals the resonant transition frequency between the  $1^2S$  and the  $2^2P$  states,  $\alpha = \sqrt{2 \ln 2} / \tau$ , with  $\tau$  being the full width at half-maximum (FWHM) duration, and  $\phi$  is the CEP of the laser pulse.

We focus on the population of the  $3^2D$  state, because this population strongly depends on the CEP of the laser pulse even though the pulse duration is tens of cycles long. Figure 1 presents the  $3^2D$  population as a function of the CEP when the laser intensity is  $I = 10^{13}$  W/cm $^2$  [Figs. 1(a)–1(c)] and  $10^{15}$  W/cm $^2$  [Figs. 1(d)–1(f)], and the FWHM pulse duration is 5 cycles [Figs. 1(a) and 1(d)], 10 cycles [Figs. 1(b) and 1(e)], and 20 cycles [Figs. 1(c) and 1(f)]. One may find that the CEP effect remains robust even when the laser pulse duration increases to 20 cycles. In the following, we try to answer three questions. First, What is the mechanism of this CEP effect? Second, Can the CEP effect be maintained for a long laser pulse? Third, How do we retrieve the absolute value of the CEP when the laser pulse is tens of laser cycles long?

In order to answer the first two questions, we now apply a three-level model to analyze the transition process of an atom in a laser pulse, where the three levels are denoted  $|0\rangle$ ,  $|1\rangle$ , and  $|2\rangle$ . The amplitudes of these three states in the laser field satisfy the following equations [21,22]:

$$\begin{aligned} \dot{a}_0(t) &= i\mu_{01}f_{01}(t)a_1(t), \\ \dot{a}_1(t) &= i\mu_{10}f_{10}(t)a_0(t) + i\mu_{12}f_{12}(t)a_2(t), \\ \dot{a}_2(t) &= i\mu_{21}f_{21}(t)a_1(t), \end{aligned} \quad (4)$$

where  $\mu_{jk} = \mu_{kj}$  is the transition dipole moment between two quantum states,  $|j\rangle$  and  $|k\rangle$ , and  $f_{jk}(t) = e(t) \exp(iE_{jk}t)$ , with  $E_{jk}$  being the energy difference between  $|j\rangle$  and  $|k\rangle$ .

As mentioned above, the frequency of the laser pulse equals the resonant transition frequency between state  $|0\rangle$  and state  $|1\rangle$ . Hence the populations of these two states will present a Rabi oscillation in the laser field. However, since the laser intensity  $I = 10^{13}$  W/cm $^2$  is weak, the ground state remains approximately one during the whole interaction process. Thus we may assume that  $a_0(t) \approx 1$  and  $|a_{1,2}| \ll 1$ . By using these equations, we have, from Eq. (4), that

$$a_1(t) \approx i\mu_{10} \int_{t_0}^t dt_1 f_{10}(t_1). \quad (5)$$

Consequently, we may express the population of  $|2\rangle$  as

$$a_2(t) \approx -\mu_{21}\mu_{10} \int_{t_0}^t f_{21}(t_1) \left( \int_{t_0}^{t_1} dt_2 f_{10}(t_2) \right) dt_1. \quad (6)$$

The population of state  $|2\rangle$  is a function of CEP, as shown by the solid (red) lines in Figs. 1(a)–1(c). One may find that the results from this three-level model agree well with the numerical calculation. Therefore, we further analyze Eq. (6) to determine the origin of this CEP effect. Using the expansion of function  $f_{ij}(t)$  for states  $|i\rangle$  and  $|j\rangle$  and the rotating-wave approximation, Eq. (6) can be rewritten as

$$a_2(t) = T_{++} + T_{+-}, \quad (7)$$

where

$$\begin{aligned} T_{++} &= -\exp(-2i\phi) \frac{e_0^2 \mu_{21} \mu_{10}}{4} \int_{t_0}^t e^{-\alpha^2 t_1^2} e^{i\Delta_{++}\omega t_1} dt_1 \\ &\times \int_{t_0}^{t_1} e^{-\alpha^2 t_2^2} e^{i(E_{10}-\omega)t_2} dt_2 \end{aligned} \quad (8)$$

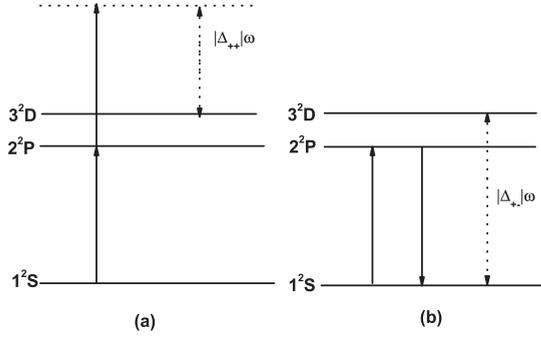


FIG. 2. Schema of (a) sum-frequency transition term and (b) difference-frequency transition term.

and

$$T_{+-} = -\frac{e_0^2 \mu_{21} \mu_{10}}{4} \int_{t_0}^t e^{-\alpha^2 t_1^2} e^{i\Delta_{+-}\omega t_1} dt_1 \times \int_{t_0}^{t_1} e^{-\alpha^2 t_2^2} e^{i(E_{10}-\omega)t_2} dt_2, \quad (9)$$

where  $\Delta_{++} = (E_{21} - \omega)/\omega$  is the detuning parameter of  $T_{++}$ , and  $\Delta_{+-} = (E_{21} + \omega)/\omega$  is for  $T_{+-}$ . From Eqs. (8) and (9), one can see that the two terms of  $a_2(t)$  carry different CEP information, where  $T_{++}$  is proportional to  $\exp(-2i\phi)$ , while  $T_{+-}$  is independent of  $\phi$ . Therefore, if these two terms have a comparable contribution to  $a_2(t)$ , they may have an effective quantum interference in the population, causing a CEP effect on the population of  $|2\rangle$ .

Figure 2 presents schematically the two-photon transition process according to the two terms  $T_{++}$  and  $T_{+-}$ . As shown in Fig. 2(a),  $T_{++}$  indicates that the atom absorbs two photons, while  $T_{+-}$  indicates that the atom absorbs one photon and then emits another photon. Hence we define  $T_{++}$  as a sum-frequency transition and  $T_{+-}$  as a difference-frequency transition. It is known that, for a nonresonance transition, the population of the final state is determined by the detuning parameters of the state. To compare the contributions of  $T_{++}$  and  $T_{+-}$  to the  $|2\rangle$  state, we define the difference in the detuning parameters between the two terms as  $\delta \equiv |\Delta_{+-}| - |\Delta_{++}| = 2E_{21}/\omega$ , provided  $E_{21} \leq \omega$ . One can see that if the photon energy is much higher than the energy difference  $E_{21}$ , which is exactly the case in this work, the difference between the two detuning parameters is very small and the two terms may thus make comparable contributions to the population. Consequently, the two transition paths  $T_{++}$  and  $T_{+-}$  can interfere with each other effectively, causing the CEP effect on the population of state  $|2\rangle$ .

From the above analysis, one can expect that the maximum CEP effect may occur when the energy difference  $E_{21}$  is 0, which corresponds to the case of a two-photon transition process in a three-level system with two degenerate excited states, such as the three-level system of  $1^2S$ ,  $3^2P$ , and  $3^2D$ . Furthermore, one can also find that the CEP effect will decrease as the energy difference  $E_{21}$  increases, as shown in Fig. 3, where the CEP effect is characterized by the parameter  $M = \frac{2[P(\phi_{\max}) - P(\phi_{\min})]}{P(\phi_{\max}) + P(\phi_{\min})}$ , with  $P(\phi_{\max})$  being the maximum population when the CEP is equal to  $\phi_{\max}$  and  $P(\phi_{\min})$  being the minimum population when the CEP is equal to  $\phi_{\min}$  [21,23]. Figure 3

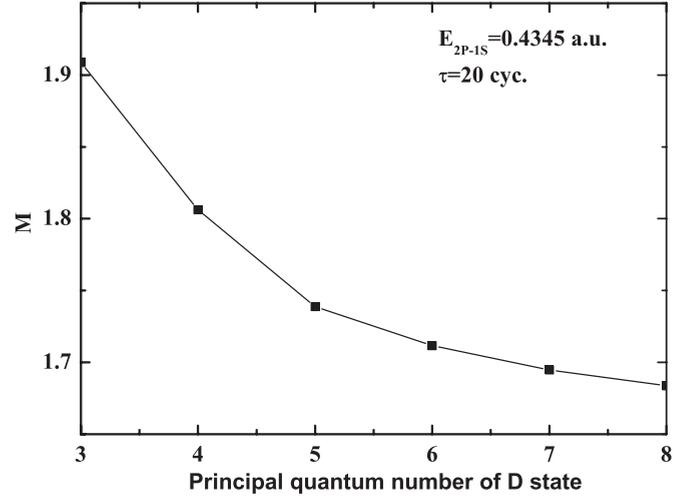


FIG. 3.  $M$  as a function of the principal quantum number  $n$  of the  $n^2D$  state of Ar, with  $I = 10^{13}$  W/cm<sup>2</sup>,  $\tau = 20$  cycles, and  $\omega = 0.4345$  a.u..

shows that the parameter  $M$  decreases with the principal quantum number  $n$  of the final state; this is because the energy difference between the intermediate state and the final state  $E_{21}$  increases with  $n$ .

Based on the above analysis, we can easily understand why this kind of CEP effect can survive in a long laser pulse. The interference of the two transition paths  $T_{++}$  and  $T_{+-}$ , which determines the strength of the CEP effect, depends on the energy level structure (i.e.,  $\delta = 2E_{21}/E_{10}$ ), rather than the pulse duration, although the absolute value of the transition rate depends on the pulse duration. In the case of our present laser-atom system, we find that the CEP effect can be observed until the laser pulse duration is longer than 30 laser cycles.

We now consider the third question: How do we retrieve the absolute CEP value of a laser pulse using the present phenomenon? From Eqs. (7)–(9), we can see that if the relative phase between the two terms  $T_{++}$  and  $T_{+-}$  is 0, then the minimum value of the  $3^2D$ -state population corresponds to the CEP value of  $\pi/2$ . However, from Eqs. (8) and (9), we find that there is an additional relative phase between these two transition terms, which has to be considered. With  $\omega = E_{10}$ , the two terms can be rewritten in the form

$$T_{++} = (A_{++} + iB_{++})e^{-2i\phi}, \quad T_{+-} = A_{+-} + iB_{+-}, \quad (10)$$

where

$$A_{++} = -\frac{E_0^2 \mu_{21} \mu_{10}}{4} \int_{t_0}^t dt_1 e^{-\alpha^2 t_1^2} \cos(\omega \Delta_{++} t_1) \times \int_{-\infty}^{t_1} dt_2 \exp(-\alpha^2 t_2^2),$$

$$A_{+-} = -\frac{E_0^2 \mu_{21} \mu_{10}}{4} \int_{t_0}^t dt_1 e^{-\alpha^2 t_1^2} \cos(\omega \Delta_{+-} t_1) \times \int_{-\infty}^{t_1} dt_2 \exp(-\alpha^2 t_2^2) \quad (11)$$

and

$$B_{++} = -\frac{E_0^2 \mu_{21} \mu_{10}}{4} \int_{t_0}^t dt_1 e^{-\alpha^2 t_1^2} \sin(\omega \Delta_{++} t_1) \int_{-\infty}^{t_1} dt_2 \exp(-\alpha^2 t_2^2),$$

$$B_{+-} = -\frac{E_0^2 \mu_{21} \mu_{10}}{4} \int_{t_0}^t dt_1 e^{-\alpha^2 t_1^2} \sin(\omega \Delta_{+-} t_1) \int_{-\infty}^{t_1} dt_2 \exp(-\alpha^2 t_2^2).$$
(12)

Therefore, since the population of the third state is  $|a_2|^2 = |T_{++} + T_{+-}|^2$ , it can be written as

$$|a_2|^2 = A_{++}^2 + A_{+-}^2 + B_{++}^2 + B_{+-}^2 + 2[(A_{++}A_{+-} + B_{++}B_{+-})^2 + (A_{++}A_{+-} - B_{++}B_{+-})^2] \cos(2\phi - \varepsilon),$$
(13)

where the additional relative phase of the two terms is

$$\varepsilon = \arccos \left\{ \frac{A_{++}A_{+-} + B_{++}B_{+-}}{\sqrt{(A_{++}A_{+-} + B_{++}B_{+-})^2 + (A_{++}B_{+-} - A_{+-}B_{++})^2}} \right\}.$$
(14)

We calculated  $T_{++}$ ,  $T_{+-}$ ,  $\varepsilon/2$ ,  $\pi/2 - \varepsilon/2$ , and  $\phi_{\min}$  for pulse durations of 5, 10, 20, and 30 laser cycles, as reported in Table I. The value of  $\phi_{\min}$  was obtained by numerical integration of Eq. (1). In Table I, we can see that the minimum value of the state  $|2\rangle$  population for the three-level model is  $\pi/2 - \varepsilon/2 = 1.41, 1.40, 1.41$ , and  $1.41$  rad for a pulse duration of 5, 10, 20, and 30 cycles, respectively. These results agree well with the  $\phi_{\min}$  listed in Table I. Consequently, we may obtain the absolute value of the CEP of a laser pulse by calculating the additional relative phase of the two quantum paths.

Since the two-photon transition is a nonresonant transition with a large detuning parameter, one can find that the absolute value of the  $3^2D$  population is relatively small, as shown in Figs. 1(a)–1(c). In order to increase the absolute value of the  $3^2D$  population, we increase the laser intensity. Figures 1(d) and 1(e) present the population with the laser intensity being  $10^{15}$  W/cm<sup>2</sup>, where ionization of the atom is obvious. Apparently, the previous assumption for a weak laser condition  $a_0(t) \approx 1$  does not hold, such that the  $3^2D$ -state population cannot be obtained directly. Thus one may ask how to explain the CEP effect in Figs. 1(d)–1(f). To answer this question, we consider all the states which may contribute to the population of the  $3^2D$  state by one-photon transition (because the photon energy is much higher than the energy difference between the  $3^2D$  and the  $n^2P$  states with  $n = 2, 3, 4, \dots$  or the  $3^2D$  and  $m^2F$  states with  $m = 4, 5, \dots$ , we do not need to consider the multiphoton transition process). These states include  $n^2P$  states with  $n = 2, 3, 4, \dots$  and  $m^2F$  states with  $m = 4, 5, \dots$ . Analyzing the contributions of these states to the population of the  $3^2D$  state, we find that the contribution of each state can also be divided into two parts,  $T_+$  and  $T_-$ , where  $T_+$  corresponds to the  $n^2P$  ( $m^2F$ ) state absorbing one photon and

carrying the CEP with  $T_+ \propto \exp(i\phi)$ , while  $T_-$  corresponds to the  $n^2P$  ( $m^2F$ ) state emitting one photon and carrying the CEP with  $T_- \propto \exp(-i\phi)$ . To illustrate the absolute values of  $T_+$  and  $T_-$  for these states, Fig. 4 presents the contributions to the  $3^2D$  state from the  $2^2P$  state [Fig. 4(a)], the  $6^2P$  state [Fig. 4(b)], the  $4^2F$  state [Fig. 4(c)], and the  $5^2F$  state [Fig. 4(d)] and the corresponding values for the  $T_+$  and  $T_-$  parts. Here, the amplitudes of the  $n^2P$  ( $m^2F$ ) states are obtained by the time evolution of the wave function in the numerical calculation. One may find that the contributions of  $T_+$  and  $T_-$  remain comparable for all the transition processes, hence the interference between them is effective during the transition processes, which is very similar to the case of the  $T_{++}$  and  $T_{+-}$  terms in a weak laser field. This demonstrates once again that this kind of CEP effect is caused by the energy level structure of the atom, rather than the laser condition, where the energy difference between the  $3^2D$  and the  $n^2P$  ( $m^2F$ ) states is much smaller than the one-photon energy of the laser, resulting in the CEP effect's remaining for each transition of  $n^2P$ – $3^2D$  with  $n = 2, 3, \dots$  and  $m^2F$ – $3^2D$  with  $m = 4, 5, \dots$ . Therefore, the total population of the  $3^2D$  state shows the obvious CEP effect in such a strong laser field. Furthermore, one finds that the population of the  $3^2D$  state increases by more than two orders of magnitude as the laser intensity increases from  $10^{13}$  to  $10^{15}$  W/cm<sup>2</sup>, while the CEP effect on the bound-bound transition remains robust.

We should mention that the CEP effect for a long laser pulse in this work is quite different from the CEP effect for an ultrashort laser pulse in our previous paper [22], although in both cases the CEP effect on the  $1S$ – $3D$  transition comes from the interference between the sum-frequency and the

TABLE I.  $T_{++}$ ,  $T_{+-}$ ,  $\varepsilon/2$ ,  $\pi/2 - \varepsilon/2$ , and the CEP value at the minimum population  $\phi_{\min}$ .

| Pulse duration (cycles) | $T_{++}$                          | $T_{+-}$                          | $\varepsilon/2$ | $\pi/2 - \varepsilon/2$ | $\phi_{\min}$ |
|-------------------------|-----------------------------------|-----------------------------------|-----------------|-------------------------|---------------|
| 5                       | $(2.231 + i0.364) \times 10^{-8}$ | $(2.206 - i0.365) \times 10^{-8}$ | 0.159           | 1.41                    | 1.33          |
| 10                      | $(3.900 + i0.672) \times 10^{-8}$ | $(3.859 - i0.661) \times 10^{-8}$ | 0.170           | 1.40                    | 1.41          |
| 20                      | $(5.829 - i0.939) \times 10^{-8}$ | $(5.813 + i0.927) \times 10^{-8}$ | 0.160           | 1.41                    | 1.49          |
| 30                      | $(1.583 + i0.026) \times 10^{-7}$ | $(1.572 - i0.027) \times 10^{-7}$ | 0.165           | 1.41                    | 1.41          |

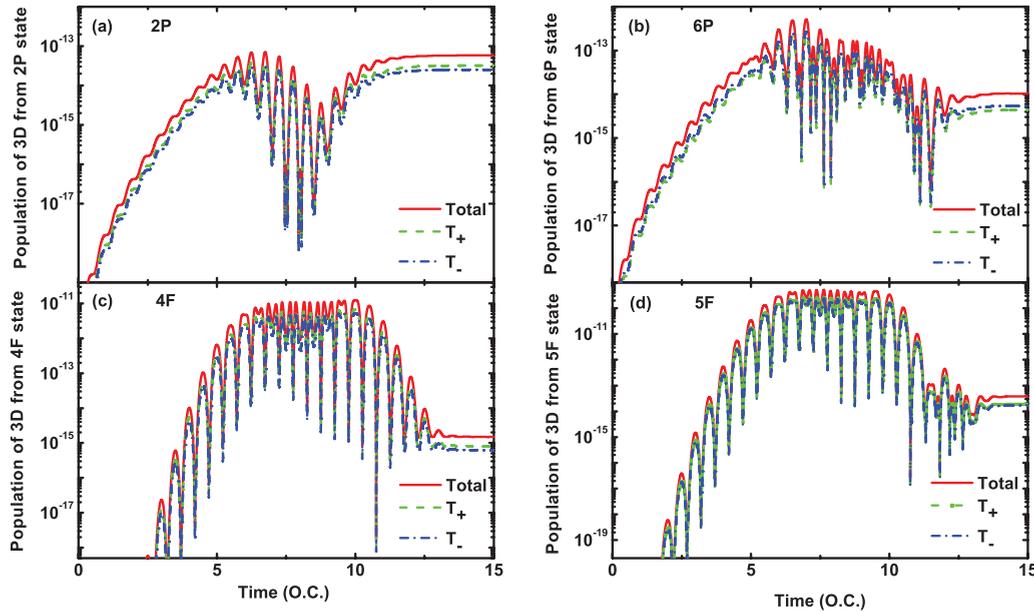


FIG. 4. (Color online) Population of the  $3^2D$  state contributed from the  $2^2P$  state (a), the  $6^2P$  state (b), the  $4^2F$  state (c), and the  $5^2F$  state (d).

difference-frequency transition subpaths. For an ultrashort laser pulse, it has a wide frequency bandwidth; hence the energies of the two photons in the two-photon transition may be different. Therefore, the CEP effect in an ultrashort laser pulse occurs when the laser frequency is approximately equal to the energy difference between  $1S$  and  $3D$ , i.e.,  $\omega = E_{3D-1S}$ . Especially, in the first step the atom absorbs one photon, and in the second step, it absorbs or emits another photon with the same frequency that is smaller than  $\omega$ , resulting in the sum-frequency and difference-frequency transitions possessing the same detuning and providing comparable contributions to the  $3^2D$  population [22].

### III. CONCLUSION

In conclusion, we have investigated the CEP effect on the two-photon bound-bound transition of an atom in a laser pulse with tens of optical cycles long. We have demonstrated that the CEP effect is caused by the interference between the sum-frequency and the difference-frequency subpaths in the two-photon transition. This interference comes from the natural

characteristics of the atom, where the energy differences between two neighboring levels satisfy  $|\Delta E_{2P-1S}| \gg |\Delta E_{3D-2P}|$ . Therefore, the degree of this interference is independent of the laser pulse duration, and the CEP effect can remain robust even when the pulse duration is tens of optical cycles long. We have also found that the CEP effect can survive in an intense laser pulse that can effectively ionize the atom.

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