

Phase-of-the-phase spectroscopy in the few-cycle regime

D. Habibović¹ and D. B. Milošević^{1,2}

¹*Faculty of Science, University of Sarajevo, Zmaja od Bosne 35, 71000 Sarajevo, Bosnia and Herzegovina*

²*Academy of Sciences and Arts of Bosnia and Herzegovina, Bistrik 7, 71000 Sarajevo, Bosnia and Herzegovina*



(Received 2 January 2026; accepted 3 March 2026; published 20 March 2026)

We present a theoretical study of the phase-of-the-phase spectroscopy in the few-cycle regime and analyze its explicit dependence on the carrier-envelope phase (CEP) and the pulse length. The process of above-threshold detachment of negative ions driven by ultrashort two-color laser pulses is investigated within the strong-field approximation formulated for few-cycle pulses with two carrier frequencies. While the CEP effects are known to be difficult to extract from the photoelectron yield of direct electrons, we demonstrate that the CEP leaves a clear and robust imprint on the phase of the phase, even when only direct electrons are considered. We analyze two-color configurations with parallel and orthogonal polarizations of the driving and perturbation pulses and identify characteristic CEP-dependent structures in both momentum-resolved and energy-averaged phase-of-the-phase spectra. The role of symmetry and its reduction in orthogonally polarized configurations is discussed in detail. Importantly, we show that the CEP dependence of the phase of the phase survives focal averaging and becomes particularly smooth under realistic experimental conditions. In addition, we demonstrate that the phase of the phase exhibits a pronounced sensitivity to the pulse length in the few-cycle regime, while converging for longer pulses, indicating its potential for characterizing ultrashort pulse durations. Our results establish phase-of-the-phase spectroscopy as a sensitive and experimentally viable tool for extracting information about strong-field processes driven by few-cycle laser pulses.

DOI: [10.1103/jdsz-p838](https://doi.org/10.1103/jdsz-p838)

I. INTRODUCTION

When an intense laser pulse interacts with matter a plethora of nonlinear processes can be induced [1–7]. Among these, particularly interesting is above-threshold detachment (ATD), a process in which negative atomic or molecular ions are exposed to a strong laser pulse which liberates the outermost electron, leaving the neutral atom or molecule behind. The freed electrons may go directly to the detector, in which case they are referred to as direct electrons. The liberated electrons which oscillate in the laser pulse may be driven back to the parent system and elastically rescatter off it. This scenario is referred to as high-order ATD (HATD) and the corresponding detachment probability is a few orders of magnitude lower than that of the direct electrons. The photoelectron spectrum is characterized by the peaks caused by the interference of the contributions of electrons from subsequent laser cycles (intercycle interference). The interference of the contributions corresponding to the electrons liberated within the same optical cycle is called intracycle interference [8,9] and it leads to various oscillatory structures in the photoelectron spectrum. The intercycle and intracycle interferences were first analyzed in [10] for the linearly polarized pulse, while the names were introduced in [11]. Strong-field detachment of negative ions, including the observation of above-threshold

detachment peaks and high-energy rescattering plateaus, has been confirmed experimentally in several studies (see, for example, Refs. [12–18]).

The photoelectron spectrum depends on the laser-pulse parameters and the characteristics of the target. Examples of laser-pulse parameters include the carrier frequency, the carrier-envelope phase (CEP), the envelope of the laser pulse, the ellipticity, and the intensity of the laser pulse. Examples of target characteristics include the type of ground state and the electron affinity. The influence of the CEP and the envelope of the pulse on the photoelectron spectrum is prominent only for ultrashort pulses (the so-called few-cycle pulses) for which the pulse consists of a small number of optical cycles. While the CEP is the most widely exploited control parameter in this regime, the finite pulse duration itself represents an equally important characteristic of few-cycle pulses, whose influence on phase-sensitive observables has so far remained largely unexplored in the context of phase-of-the-phase spectroscopy. One of the key features of the photoelectron momentum distribution induced by an ultrashort, linearly polarized laser pulse is its pronounced forward-backward asymmetry. In simple terms, electrons are not emitted equally in the direction of polarization and the opposite direction. The role of the CEP in shaping this asymmetry was first observed experimentally in Ref. [19] and later clarified theoretically in Ref. [20] for few-cycle pulses without CEP stabilization. Subsequent theoretical studies of stereo photoelectron emission, driven by few-cycle pulses with a stabilized CEP, were carried out in Ref. [21], and the first experimental demonstration followed in Ref. [22].

Particularly interesting are the laser pulses with two carrier frequencies. When producing such laser pulses one typically

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starts with a single carrier frequency and generates a weaker, second harmonic by passing the pulse through a nonlinear beta-barium borate crystal. By varying the optical path that the fundamental and the second harmonic travel, experimenters introduce subcycle delays that change the relative phase between the two components. This can be done using, for example, finely translatable fused-silica wedges, thin glass plates, or birefringent wedges or by adjusting an interferometer arm [23–26]. The relative phase between the laser-pulse components is an additional parameter which can be used to extract the information about the process. The phase-of-the-phase spectroscopy is a method which considers the change of the photoelectron spectrum as a function of the relative phase between the two laser-pulse components [27]. It utilizes laser pulses with various polarizations. For example, laser pulses with components that are linearly polarized in the same direction were employed in [27–36], while laser pulses with components that are linearly polarized in the mutually orthogonal directions were employed in [37]. Furthermore, laser pulses with circularly polarized components were used in [38,39], while pulses with elliptically polarized components were utilized in [40–42]. Usually, the first component of the laser pulse drives the process, while the second component has a much lower intensity and acts as a perturbation. The observable quantity is the phase of the change of the photoelectron yield with respect to the relative phase between the laser-pulse components. The phase-of-the-phase spectroscopy is sensitive to both the laser-pulse parameters and the characteristics of the target [38]. All of the studies mentioned above employed laser pulses which include a large number of optical cycles (more than ten) and thus can be treated as long pulses with a flat envelope. In this case, the CEP-related phenomena and the phenomena depending on the pulse length are absent.

Strong-laser-field and attosecond-scale processes are governed by the instantaneous electric field of few-cycle laser pulses, making precise knowledge of both the CEP and the pulse duration essential. The CEP determines the phase between the carrier wave and the pulse envelope and thus directly controls subcycle electron dynamics, while the pulse duration defines the temporal confinement of the interaction and the number of contributing optical cycles. Together, these parameters uniquely characterize the laser waveform for a given wavelength and intensity and are therefore crucial for understanding, controlling, and reproducing CEP-dependent phenomena. Reliable real-time measurement of both quantities is particularly important for experiments employing ultrashort pulses, where even small fluctuations can significantly alter the strong-field response (see [43] and references therein).

In the present paper we are interested in the few-cycle pulses for which the CEP represents an important experimental parameter. Regarding the photoelectron spectra, the CEP-dependent phenomena are pronounced only in the energy range where HATD electrons dominate the signal [44]. This can be explained by recalling that, in order to produce a strong yield of electrons, the laser field has to be intense both at the initial detachment moment and during the time after the electron rescattering. For few-cycle pulses, these two events are typically separated by nearly one optical cycle, and it is difficult to ensure that the field is strong during both events.

However, by selecting an appropriate CEP, one can align the field maxima with both the detachment and after-rescattering times. Consequently, the yield of HATD electrons is far more sensitive to the CEP than the yield associated with direct electrons even though it is usually a few orders of magnitude lower than the ATD yield.

The fact that the HATD yield is low represents an experimental challenge if one wants to analyze the CEP-dependent effects or to extract the value of the CEP from the photoelectron yield. In the present paper we demonstrate that the information about the CEP can be extracted using phase-of-the-phase spectroscopy by taking into account only the direct electrons. Furthermore, we show that the phase of the phase provides direct access to the few-cycle character of the driving pulse itself, exhibiting a strong dependence on the pulse length that vanishes in the long-pulse limit. To calculate the photoelectron yield, we use our theory based on the strong-field approximation (SFA) [4] (for more details on the SFA see [45]). This theory was recently generalized to the case of the few-cycle pulses with two carrier frequencies [46–48]. After the electron is liberated from the ion, the remaining system is a neutral atom, so the Coulomb interaction is absent. Consequently, the results obtained using our SFA theory (which by itself neglects the Coulomb interaction) are in excellent agreement with those obtained using more advanced theory based on the solutions of the time-dependent Schrödinger equation [49].

The paper is organized as follows. In Sec. II we present our SFA theory of the ATD process and introduce the basics of the phase-of-the-phase spectroscopy. Also, we define our laser pulse and elaborate the focal-averaging procedure. In Sec. III we present our numerical results for collinearly and orthogonally polarized two-color pulses and analyze the CEP dependence of the phase of the phase. We also investigate its dependence on the pulse length in the few-cycle regime. In Sec. IV we provide a summary and discuss our conclusions. Atomic units are used unless otherwise stated.

II. THEORY

A. Laser pulse

Our laser pulse is a sum of two components, i.e., $\mathbf{E}(t) = \mathbf{E}_1(t) + E_2(t)\hat{\mathbf{e}}_j$, with

$$\begin{aligned} \mathbf{E}_1(t) &= E_0 f(t) \cos(\omega t + \phi_{\text{CEP}}) \hat{\mathbf{e}}_x, \\ E_2(t) &= \kappa E_0 f(t) \cos[2(\omega t + \phi_{\text{CEP}}) + \varphi], \end{aligned} \quad (1)$$

where E_0 is the amplitude of the first laser-pulse component, ω is the fundamental carrier frequency, $f(t)$ is the envelope, ϕ_{CEP} is the CEP, and φ is the relative phase between the laser-pulse components. The ratio of the amplitudes of the two laser-pulse components is denoted by κ , while $\hat{\mathbf{e}}_j$, $j = x, y$, are the unit polarization vectors. For a small ratio κ , the component $\mathbf{E}_1(t)$ is much stronger than the component $E_2(t)$ and thus it is solely responsible for the detachment, while the component $E_2(t)$ behaves as a small perturbation. For this reason, we call $\mathbf{E}_1(t)$ a driving pulse and $E_2(t)\hat{\mathbf{e}}_j$ a perturbation pulse. For $\hat{\mathbf{e}}_j = \hat{\mathbf{e}}_x$ the polarization of the perturbation pulse is in the same direction as the polarization of the driving pulse,

while for $\hat{\mathbf{e}}_j = \hat{\mathbf{e}}_y$ the polarization of the perturbation pulse is orthogonal to the polarization of the driving pulse.

B. Strong-field-approximation theory and the phase of the phase

During the interaction of the negative ion with the ultrashort laser pulse, the outermost electron can be detached from the parent system. The probability of detection of such an electron with momentum $\mathbf{p}$ and energy $E_p = \mathbf{p}^2/2$ is given by $W_p = |M_p|^2 d^3\mathbf{p}/(d\Omega_p dE_p) = p|M_p|^2$, where $d\Omega_p$ is the solid-angle element, $p = |\mathbf{p}|$, and M_p is the probability amplitude. Within the framework of the SFA, which neglects the interaction of the detached electron with its parent system, the probability amplitude M_p can be written as [4]

$$M_p = -i \int_0^{\tau_p} dt \langle \tilde{\mathbf{p}} + \mathbf{A}(t) | \mathbf{r} \cdot \mathbf{E}(t) | \psi_0 \rangle e^{iS(\tilde{\mathbf{p}}, t)}. \quad (2)$$

Here $\mathbf{A}(t) = -\int^t \mathbf{E}(t') dt'$ is the vector potential of the field $\mathbf{E}(t)$, $\tilde{\mathbf{p}} = \mathbf{p} - \mathbf{A}(\tau_p)$, $\tau_p = n_p T$ is the total pulse duration expressed as an integer multiple of the laser field period T , and t is the detachment time. The modified action is [50]

$$S(\tilde{\mathbf{p}}, t) = I_p t + \int_0^t dt' [\tilde{\mathbf{p}} + \mathbf{A}(t')]^2/2, \quad (3)$$

where I_p is the electron affinity. The initial state is modeled using a linear combination of the Slater-type orbitals

$$\psi_0 = \psi_{lm}(\mathbf{r}) = \sum_a C_a \frac{(2\zeta_a)^{n_a+1/2}}{\sqrt{(2n_a)!}} r^{n_a-1} e^{-\zeta_a r} Y_{lm}(\Omega). \quad (4)$$

The coefficients C_a and ζ_a can be found in [52].

The detachment probability $W_p(\varphi)$ depends on the relative phase φ between the laser-pulse components. It can be expanded in the Fourier series

$$W_p(\varphi) = \frac{A_0(\mathbf{p})}{2} + \sum_{n=1}^{\infty} A_n(\mathbf{p}) \cos(n\varphi) + \sum_{n=1}^{\infty} B_n(\mathbf{p}) \sin(n\varphi), \quad (5)$$

where the coefficients are

$$\begin{aligned} A_0(\mathbf{p}) &= \int_0^{2\pi} W_p(\varphi) d\varphi / \pi, \\ A_n(\mathbf{p}) &= \int_0^{2\pi} W_p(\varphi) \cos(n\varphi) d\varphi / \pi, \\ B_n(\mathbf{p}) &= \int_0^{2\pi} W_p(\varphi) \sin(n\varphi) d\varphi / \pi. \end{aligned} \quad (6)$$

Assuming that the 2ω component of the pulse is much weaker than the ω component, we can neglect all the terms in the Fourier series except the first two, and the detachment probability $W_p(\varphi)$ becomes

$$W_p(\varphi) \approx A_0(\mathbf{p})/2 + \Delta W_p \cos[\varphi + \Phi(\mathbf{p})]. \quad (7)$$

Here $\Delta W_p = \sqrt{A_1^2(\mathbf{p}) + B_1^2(\mathbf{p})}$ is the relative phase contrast, while $\Phi(\mathbf{p}) = -\arctan[B_1(\mathbf{p})/A_1(\mathbf{p})]$ is the so-called phase of the phase. The phase of the phase (as well as the relative phase contrast) can be calculated for every point of the momentum plane, i.e., for every $\mathbf{p} = (p, \theta_p)$, where θ_p is the emission angle, i.e., the angle between the unit vector $\hat{\mathbf{e}}_x$ and the final

photoelectron momentum $\mathbf{p}$. Interesting information can also be extracted from the phase of the phase which is averaged over the photoelectron energy $E_p = p^2/2$ (or equivalently over the momentum intensity p) for the fixed value of the emission angle θ_p . We define the energy-averaged phase of the phase as $\tilde{\Phi}(\theta_p) = \int_{E_{p1}}^{E_{p2}} \Phi(\mathbf{p}) dE_p / \int_{E_{p1}}^{E_{p2}} dE_p$.

C. Focal averaging

The temporal distribution of an ultrashort pulse is taken into account via the time dependence of the envelope $f(t)$. However, the spatial distribution of the ultrashort laser pulse affects its interaction with an ion and it should also be taken into account for a detailed simulation of a potential experiment. In particular, the intensity of the laser pulse in the focal plane, where the interaction with ions usually takes place, is not constant and its distribution affects the process of detachment, i.e., the spectrum of the emitted photoelectrons. In order to calculate the focal-averaged spectrum, we use the method described in Refs. [53–55], according to which the detachment probability averaged over the laser-pulse intensity I in the focal plane can be written as

$$\langle W_p \rangle = \int_0^{I_0} W_p K(I, I_0) dI, \quad (8)$$

where the kernel is

$$K(I, I_0) \propto \frac{2I + I_0}{I^2} \sqrt{\frac{I_0 - I}{I}}, \quad (9)$$

with I_0 the peak intensity of the applied pulse. We denote the phase of the phase calculated using $\langle W_p \rangle$ by $\langle \Phi(\mathbf{p}) \rangle$.

III. NUMERICAL RESULTS

We present our numerical results using the example of the F^- ion whose electron affinity is $I_p = 3.4$ eV. The intensity of the driving pulse is $I = E_0^2 = 1.5 \times 10^{13}$ W/cm², while the ratio of the electric-field amplitudes is $\kappa = 0.1$, so the intensity of the perturbation pulse is two orders of magnitude lower than the intensity of the driving pulse. The fundamental carrier frequency ω corresponds to a wavelength of 2000 nm and the pulse has a sine-squared envelope $f(t) = \sin^2[\omega t/(2n_p)]$.

A. CEP dependence of the phase of the phase for the perturbation pulse polarized along the polarization of the driving pulse

We first analyze the case in which the perturbation pulse is polarized in the same direction as the driving pulse. The laser pulse has four optical cycles ($n_p = 4$) so that the total pulse duration is $\tau_p = 26.7$ fs. In Fig. 1 we present the photoelectron momentum distribution for the F^- ion exposed to our laser pulse with the CEP $\phi_{\text{CEP}} = 30^\circ$ and the relative phase $\varphi = 0^\circ$. The distribution exhibits the reflection symmetry $p_y \rightarrow -p_y$ because both pulse components are polarized along the same direction $\hat{\mathbf{e}}_x$. In contrast, the symmetry $p_x \rightarrow -p_x$ is broken by the perturbation pulse and by the ultrashort nature of the driving pulse. However, since the perturbation is weak, the resulting distribution remains close to that obtained in the absence of the perturbation pulse. Furthermore, the momentum distribution changes only marginally with the

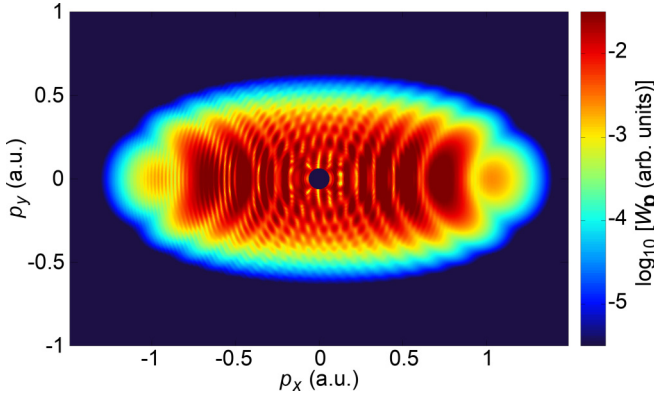


FIG. 1. Photoelectron momentum distribution of the F^- ion obtained using our laser pulse with $\phi_{\text{CEP}} = 30^\circ$ and the relative phase $\varphi = 0^\circ$. The polarizations of the driving and perturbation pulses are in the same direction.

CEP, making it difficult to determine the CEP directly from the photoelectron momentum distribution. In the following, we investigate how the phase of the phase depends on the value of the CEP.

In Fig. 2 we present the phase of the phase in the photoelectron momentum plane for ATD from the F^- ion driven by our laser pulse with the CEP $\phi_{\text{CEP}} = 0^\circ$ [Fig. 2(a)], $\phi_{\text{CEP}} = 90^\circ$ [Fig. 2(b)], and $\phi_{\text{CEP}} = 180^\circ$ [Fig. 2(c)]. The phase of the phase is predominantly positive for $p_x > 0$ and negative for $p_x < 0$, and the variety of oscillatory structures, present in the photoelectron momentum distribution, are clearly visible for all values of the CEP. These structures include the concentric rings originating from intercycle interference, as well as oscillations resulting from intracycle interference. More importantly, the overall value of the phase of the phase steadily increases with the increase of the CEP. Thus, the value of the CEP is clearly imprinted in the phase-of-the-phase spectra and this imprint is much stronger than the imprint of the CEP on the detachment probability. Finally, the fact that the phase of the phase approaches $\Phi \approx \pm\pi/2$ for $\phi_{\text{CEP}} = \pi/2$ [see Fig. 2(b)] implies that the ratio $B_1(\mathbf{p})/A_1(\mathbf{p})$ diverges for this value of the CEP. This behavior indicates that for $\phi_{\text{CEP}} = \pi/2$ the coefficient $A_1(\mathbf{p})$ becomes vanishingly small while $B_1(\mathbf{p})$ remains finite. This suppression does not imply negative contributions to the detachment probability itself, which is always positive, but instead results from the projection of the detachment probability onto the Fourier basis functions $\cos \varphi$ and $\sin \varphi$ that define the coefficients $A_1(\mathbf{p})$ and $B_1(\mathbf{p})$. For $\phi_{\text{CEP}} = \pi/2$, the dominant subcycle ionization events on either side of the pulse maximum generate a φ dependence of $W_p(\varphi)$ that leads to a near cancellation when weighted by $\cos \varphi$, while the corresponding projection onto $\sin \varphi$ remains finite. As a consequence, the first-order two-color modulation of the photoelectron yield is dominated by the sine term, which forces the phase of the phase to lock close to $\pm\pi/2$. The sign of the divergence depends on the emission direction: For $p_x > 0$ the coefficient $A_1(\mathbf{p})$ crosses zero from positive to negative values, yielding $B_1(\mathbf{p})/A_1(\mathbf{p}) \rightarrow -\infty$, while for $p_x < 0$ the opposite behavior occurs.

The phase of the phase for the values of the CEP $\phi_{\text{CEP}} > 180^\circ$ can be related to those calculated for

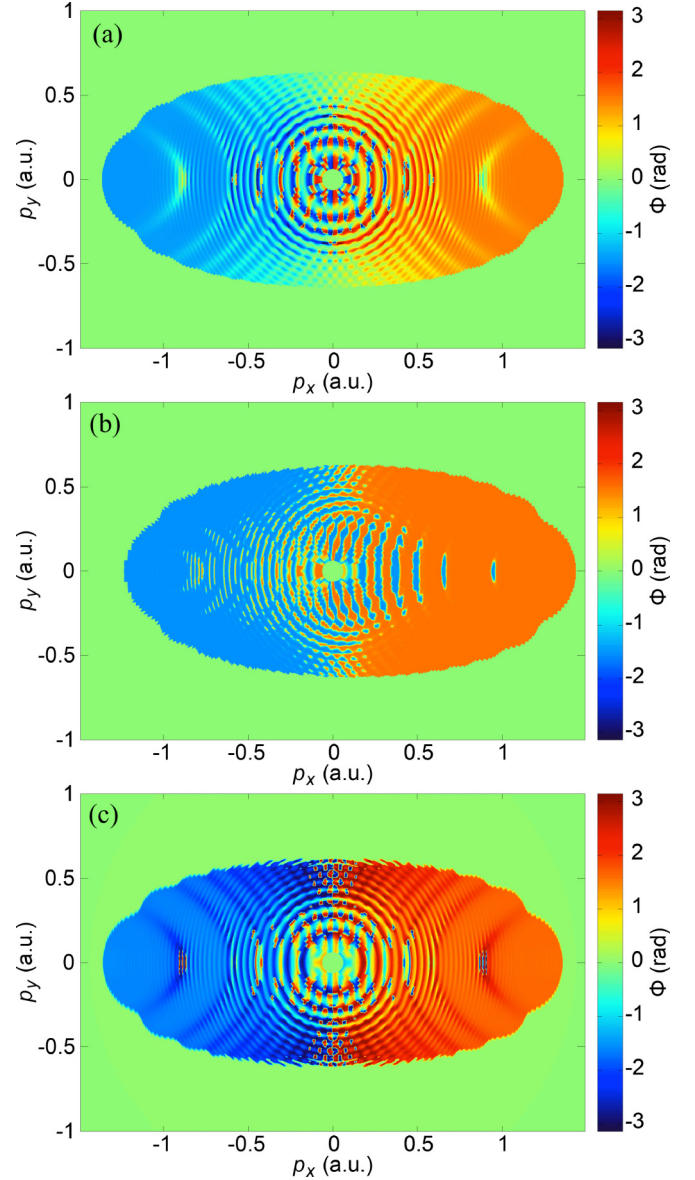


FIG. 2. Phase of the phase presented in the photoelectron momentum plane for ATD induced by exposing the F^- ion to our laser pulse with (a) $\phi_{\text{CEP}} = 0^\circ$, (b) $\phi_{\text{CEP}} = 90^\circ$, and (c) $\phi_{\text{CEP}} = 180^\circ$. The polarizations of the driving and perturbation pulses are in the same direction.

$\phi_{\text{CEP}} \leq 180^\circ$. In order to elaborate this, we use the energy-averaged phase of the phase $\tilde{\Phi}(\theta_p; \phi_{\text{CEP}})$, where we explicitly added the dependence on the CEP. In Fig. 3 we present the energy-averaged phase of the phase as a function of the emission angle θ_p and the CEP ϕ_{CEP} . For a fixed value of the CEP, the energy-averaged phase of the phase is the same for emission in the directions θ_p and $2\pi - \theta_p$, i.e., $\tilde{\Phi}(\theta_p; \phi_{\text{CEP}}) = \tilde{\Phi}(2\pi - \theta_p; \phi_{\text{CEP}})$. This happens because the laser pulse is linearly polarized so that the momentum distribution possesses the symmetry $p_y \rightarrow -p_y$ regardless of the value of the CEP and the value of the relative phase. In addition, the energy-averaged phase of the phase $\tilde{\Phi}(\theta_p; \phi_{\text{CEP}})$ is the same as the one for $\tilde{\Phi}(\pi - \theta_p; -\phi_{\text{CEP}})$ up to a sign. This can be explained by analyzing the symmetry properties

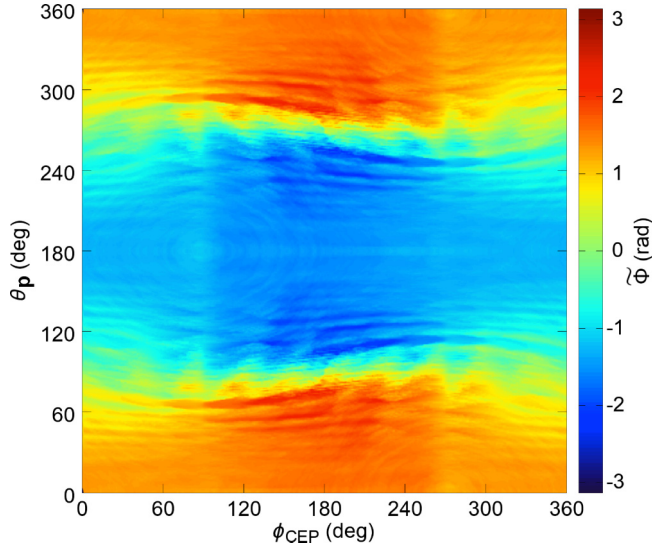


FIG. 3. Energy-averaged phase of the phase $\tilde{\Phi}(\theta_p; \phi_{\text{CEP}})$ as a function of the emission angle θ_p and the CEP ϕ_{CEP} . The polarizations of the driving and perturbation pulses are in the same direction.

of the detachment probability W_p and the coefficients (6) under the transformations $\phi_{\text{CEP}} \rightarrow -\phi_{\text{CEP}}$, $\theta_p \rightarrow \pi - \theta_p$ (which corresponds to $p_x \rightarrow -p_x$), and $\varphi \rightarrow -\varphi$. For the detachment probability we have $W_{-p_x, p_y}(-\phi_{\text{CEP}}, -\varphi) = W_{p_x, p_y}(\phi_{\text{CEP}}, \varphi)$, while for the coefficients (6) we can write $A_n(-p_x, p_y; -\phi_{\text{CEP}}) = -A_n(p_x, p_y; \phi_{\text{CEP}})$ and $B_n(-p_x, p_y; -\phi_{\text{CEP}}) = B_n(p_x, p_y; \phi_{\text{CEP}})$. Here we explicitly added the dependence on the CEP for the coefficients (6) and on the CEP and the relative phase φ for the detachment probability. Consequently, $\tilde{\Phi}(\pi - \theta_p; -\phi_{\text{CEP}}) = -\tilde{\Phi}(\theta_p; \phi_{\text{CEP}})$ and analogously for the phase of the phase itself. The special case of this symmetry happens for $\phi_{\text{CEP}} = 0^\circ$ and $\phi_{\text{CEP}} = 180^\circ$, in which case $\tilde{\Phi}(\pi - \theta_p) = -\tilde{\Phi}(\theta_p)$, i.e., the energy-averaged phase of the phase (as well as the phase of the phase itself) is invariant upon the transformation $p_x \rightarrow -p_x$ up to a sign [see Figs. 2(a) and 2(c)].

From an experimental perspective, one might worry that the oscillations present in the phase-of-the-phase spectra could hinder the extraction of the CEP. In practice, however, this should not pose a significant problem, because the oscillatory features of the photoelectron momentum distribution (which cause the oscillations in the phase-of-the-phase spectra) are expected to be largely washed out when the geometrical characteristics of the laser pulse are taken into account, i.e., after the focal averaging is performed. To illustrate this, in Fig. 4 we present results analogous to those shown in Fig. 2 but now including focal averaging. The oscillations present in the phase-of-the-phase spectra obtained without focal averaging are largely washed out. In addition, for all values of the photoelectron momentum, the phase of the phase depends on the value of the CEP. However, this dependence is more pronounced for the emission close to the direction which is orthogonal to the laser-pulse polarization direction. In this case, for the CEP equal to zero, the phase of the phase averaged over the focal plane is small ($\langle \Phi \rangle \approx 0$). As the CEP increases, $\langle \Phi \rangle$ also increases, and for $\phi_{\text{CEP}} = \pi/2$ it is $\langle \Phi \rangle \approx \pi/2$. Finally, for $\phi_{\text{CEP}} = \pi$ it

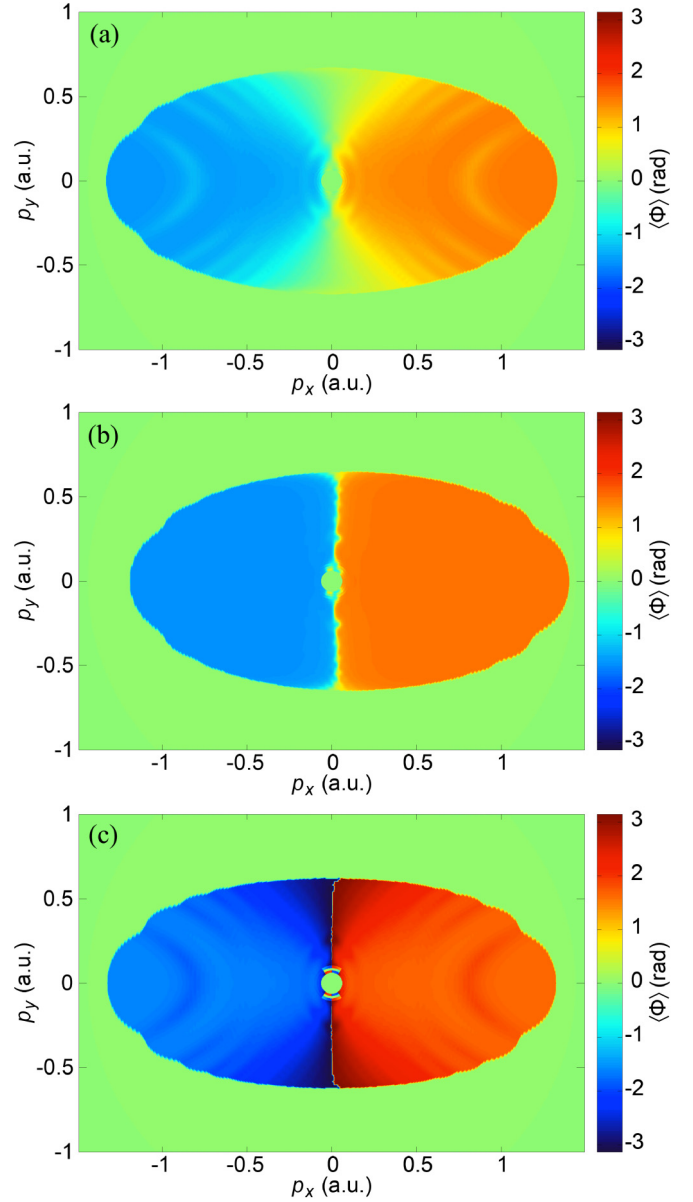


FIG. 4. Same as in Fig. 2 but with focal averaging.

approaches $\langle \Phi \rangle \approx \pi$. To provide clearer quantitative insight into the CEP dependence discussed above, in Fig. 5 we present the focal-averaged phase of the phase as a function of the photoelectron energy for selected emission angles and CEP values. While the momentum-resolved color maps reveal the overall structure, the representation in Fig. 5 allows one to directly read off the numerical values of the phase of the phase and to follow their systematic variations with both the CEP and the emission angle. It can be seen that the phase of the phase changes significantly with the CEP over a broad energy range, confirming the robustness of the CEP imprint.

A better way to quantify the obtained results is by analyzing the dependence of the phase of the phase on the CEP for fixed values of the photoelectron energy and the emission angle. In Fig. 6 we present the focal-averaged phase of the phase as a function of the CEP calculated for the photoelectrons with energy $E_p = 0.1$ a.u. which are emitted in the

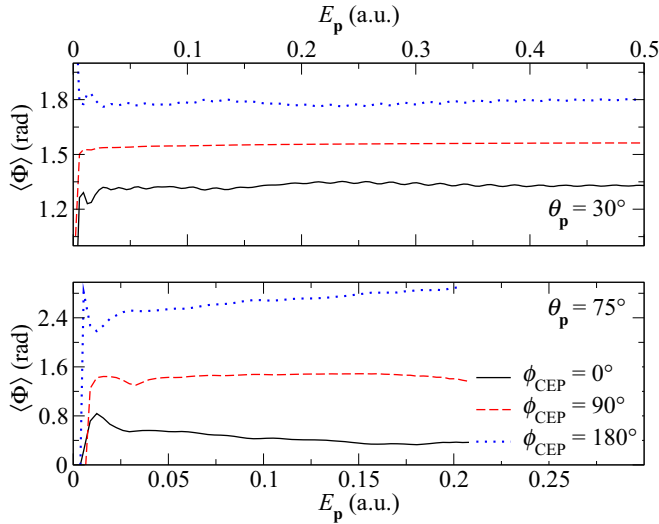


FIG. 5. Focal-averaged phase of the phase as a function of the photoelectron energy for the values of the CEP as indicated in the legend. The emission angle is indicated in each panel.

directions as indicated in the legend. For the chosen values of the emission angle, the phase of the phase is positive. It always depends on the CEP, but this dependence is more pronounced for the emission close to the direction orthogonal to the laser-pulse polarization. The dependence of the phase of the phase on the CEP is approximately symmetrical with respect to the transformation $\phi_{\text{CEP}} \rightarrow -\phi_{\text{CEP}}$. In addition, for $\phi_{\text{CEP}} = \pm\pi/2$ the focal-averaged phase of the phase is close to $\pi/2$. Similar conclusions hold for other values of the photoelectron energy. On the other hand, for the emission in the directions with $p_x < 0$ the conclusions are analogous to those for $p_x > 0$ but with the opposite sign of the phase of the phase. In conclusion, the imprint of the CEP on the photoelectron momentum distribution is minimal for the electrons which do not interact with the parent system after the detachment. However, this is not the case with the phase of the phase which significantly depends on the CEP even for the direct electrons.

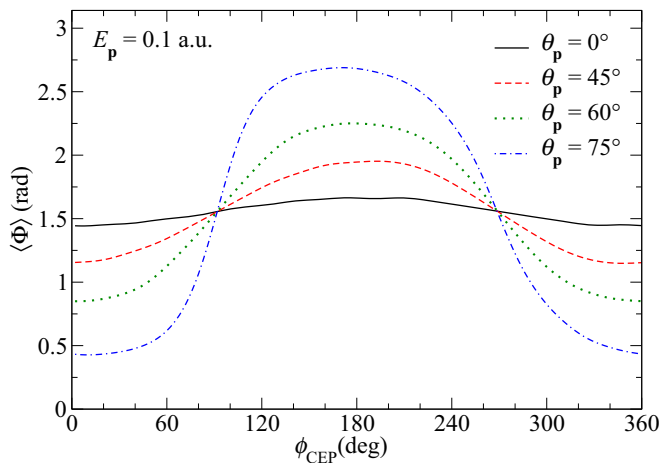


FIG. 6. Focal-averaged phase of the phase as a function of the CEP, calculated for the photoelectrons with energy $E_p = 0.1$ a.u., emitted in the directions as indicated in the legend.

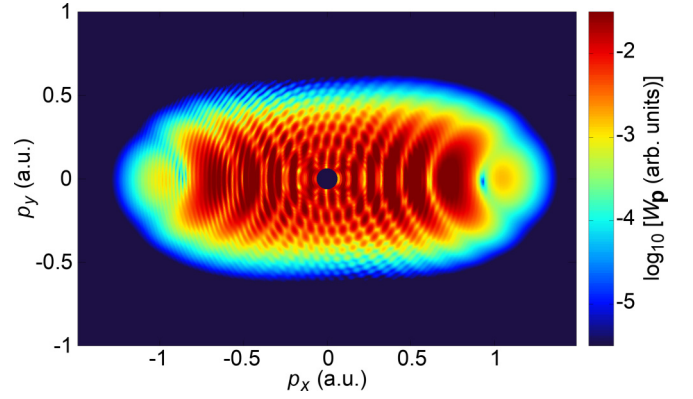


FIG. 7. Photoelectron momentum distribution of the F^- ion obtained using our laser pulse with $\phi_{\text{CEP}} = 30^\circ$ and the relative phase $\varphi = 0^\circ$. The polarization of the perturbation pulse is orthogonal to the polarization of the driving pulse.

When the focal averaging is performed to account for the real experimental conditions, the dependence of the phase of the phase on the CEP becomes particularly smooth and thus it can easily be used to extract the information about the value of the CEP from the experiment.

B. CEP dependence of the phase of the phase for the perturbation pulse with polarization orthogonal to the polarization of the driving pulse

Another interesting configuration assumes that the perturbation pulse is polarized in the direction orthogonal to the polarization direction of the driving pulse. In Fig. 7 we present the photoelectron momentum distribution for the F^- ion exposed to our laser pulse with the CEP $\phi_{\text{CEP}} = 30^\circ$ and the relative phase $\varphi = 0^\circ$. The polarization of the perturbation pulse is orthogonal to the polarization of the driving pulse. The photoelectron momentum distribution is similar to the one shown in Fig. 1. This is expected due to the fact that the driving pulse is the same in both cases. However, unlike to the distribution presented in Fig. 1, the reflection symmetry $p_y \rightarrow -p_y$ is broken because the polarizations of the driving pulse and the perturbation pulse are not along the same direction. The lack of the symmetry $p_y \rightarrow -p_y$ (as well as the symmetry $p_x \rightarrow -p_x$) is barely visible since the perturbation is weak and the influence of the ultrashort nature of the pulse on the direct-electron momentum distribution is not pronounced. Let us now analyze the CEP dependence of the phase of the phase for this pulse configuration.

In Fig. 8 we present the focal-averaged phase of the phase in the photoelectron momentum plane for the case in which the perturbation pulse is polarized orthogonally to the polarization of the driving pulse. The results are shown for the values of the CEP $\phi_{\text{CEP}} = 0^\circ$ [Fig. 8(a)], $\phi_{\text{CEP}} = 90^\circ$ [Fig. 8(b)], and $\phi_{\text{CEP}} = 180^\circ$ [Fig. 8(c)]. As in the collinearly polarized configuration, the phase of the phase exhibits a clear and pronounced dependence on the CEP over the entire momentum plane, demonstrating that the CEP imprint survives focal averaging also in the orthogonal two-color geometry. However, in contrast to the case of the perturbation which is collinear with the driving pulse, the momentum-resolved

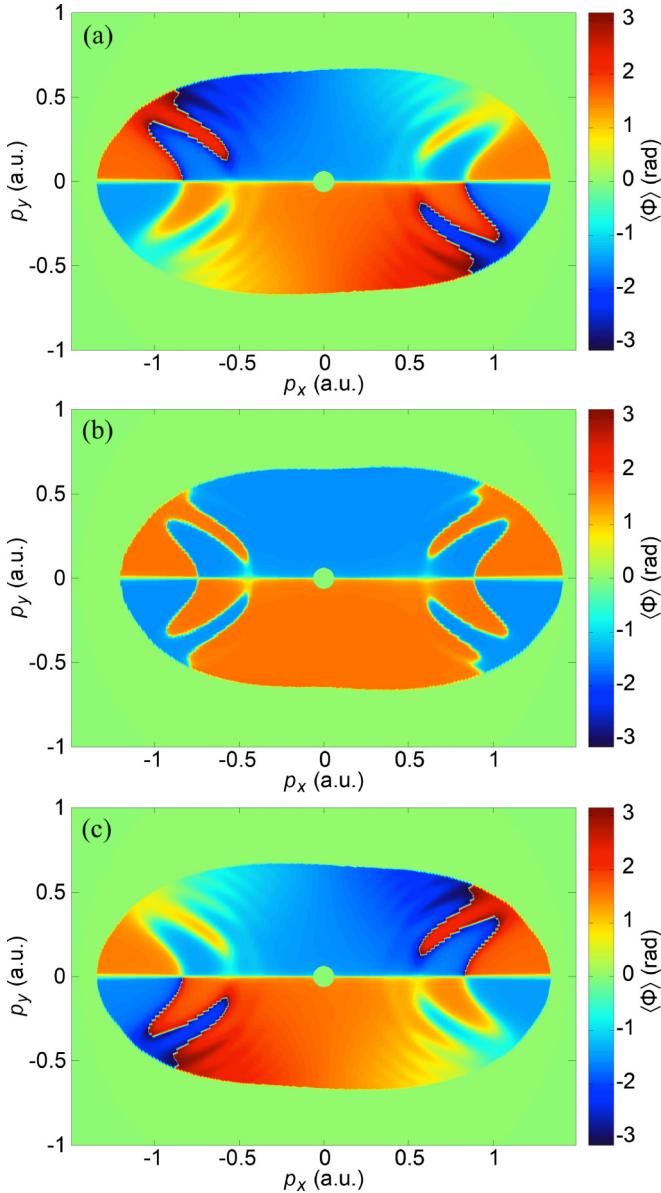


FIG. 8. Focal-averaged phase of the phase presented in the photoelectron momentum plane for ATD induced by exposing the F^- ion to our laser pulse with (a) $\phi_{\text{CEP}} = 0^\circ$, (b) $\phi_{\text{CEP}} = 90^\circ$, and (c) $\phi_{\text{CEP}} = 180^\circ$. The polarization of the perturbation pulse is orthogonal to the polarization of the driving pulse.

phase of the phase with orthogonal perturbation no longer exhibits reflection symmetry with respect to the transformation $p_y \rightarrow -p_y$. This asymmetry directly reflects the fact that the total electric field is no longer linearly polarized when the perturbation pulse is orthogonal to the driving pulse. As a consequence, the underlying symmetry of the photoelectron momentum distribution is reduced, and this reduction is clearly transferred to the phase-of-the-phase spectra. Despite this, the overall CEP dependence remains robust. For example, in the first quadrant of the momentum plane, the focal-averaged phase of the phase is relatively small for $\phi_{\text{CEP}} = 0^\circ$, while increasing the CEP leads to a continuous increase of its absolute value. In particular, for $\phi_{\text{CEP}} = 90^\circ$

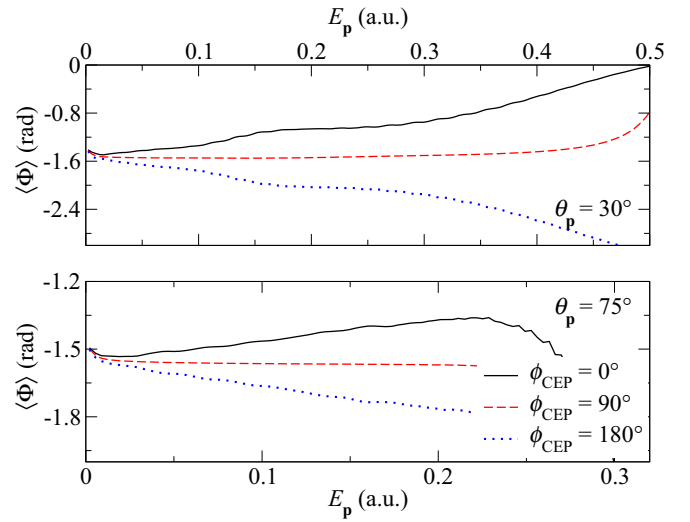


FIG. 9. Same as in Fig. 5 but for the case of the perturbation pulse orthogonal to the polarization of the driving pulse.

the phase of the phase approaches values close to $\pm\pi/2$ in extended regions of momentum space, indicating again a suppression of the cosine Fourier component relative to the sine component of the two-color modulation. For $\phi_{\text{CEP}} = 180^\circ$ the phase of the phase further increases towards values close to $\pm\pi$, consistent with the behavior observed for parallel polarization. This demonstrates that the phase of the phase remains a sensitive and reliable observable for extracting CEP information even when the perturbation field is polarized orthogonally to the driving pulse and when realistic experimental conditions are taken into account by focal averaging. At the same time, the orthogonal polarization configuration introduces additional asymmetries in the momentum plane, providing an extended degree of control and sensitivity compared to the collinear case.

Finally, to provide a clearer quantitative representation of the CEP dependence in the orthogonal polarization configuration, in Fig. 9 we present the focal-averaged phase of the phase as a function of the photoelectron energy for selected emission angles and CEP values. In contrast to the momentum-resolved color maps, this representation allows the exact numerical values of the phase of the phase to be directly identified and their variation with the CEP to be followed in detail. It can be seen that the phase of the phase remains strongly CEP dependent over a broad energy range, confirming that the CEP imprint persists also for the case of the perturbation which is orthogonal to the polarization of the driving pulse. In addition, the phase of the phase is negative for the chosen emission directions and its magnitude changes with the CEP, in agreement with the trends observed in the momentum-resolved spectra.

Now we investigate the symmetry properties of the CEP-dependent phase of the phase. Figure 10 shows the energy-averaged phase of the phase $\tilde{\Phi}(\theta_p; \phi_{\text{CEP}})$ as a function of the emission angle θ_p and the CEP ϕ_{CEP} for the case in which the perturbation pulse is polarized orthogonally to the driving pulse. In contrast to the results shown in Fig. 3, the symmetry $\tilde{\Phi}(\theta_p; \phi_{\text{CEP}}) = \tilde{\Phi}(2\pi - \theta_p; \phi_{\text{CEP}})$ for a fixed CEP is

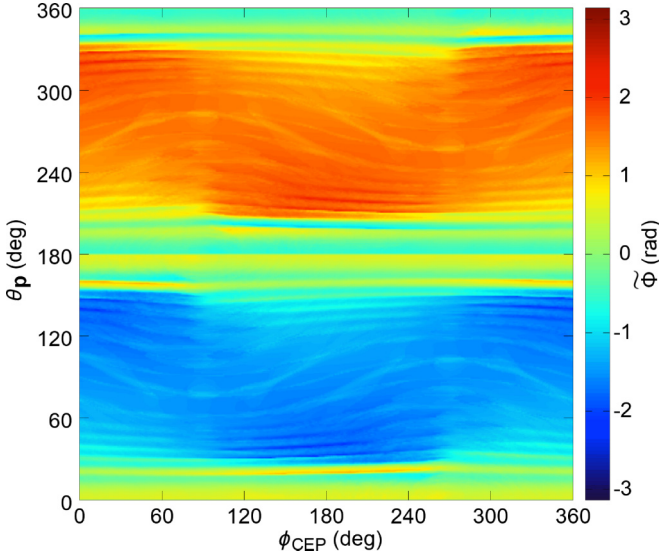


FIG. 10. Energy-averaged phase of the phase $\tilde{\Phi}(\theta_p; \phi_{CEP})$ as a function of the emission angle θ_p and the CEP ϕ_{CEP} . The polarization of the perturbation pulse is orthogonal to the polarization of the driving pulse.

no longer present, reflecting the fact that the total laser pulse is no longer linearly polarized and that the transformation $p_y \rightarrow -p_y$ is not a symmetry transformation of the photoelectron momentum distribution. Despite this, the energy-averaged phase of the phase exhibits a clear and smooth dependence on the CEP for all values of the emission angle.

The phase of the phase for $\phi_{CEP} > 180^\circ$ can be related to the results calculated for $\phi_{CEP} \leq 180^\circ$. In particular, the energy-averaged phase of the phase $\tilde{\Phi}(\theta_p; \phi_{CEP})$ is the same as $\tilde{\Phi}(\pi + \theta_p; -\phi_{CEP})$ up to a sign. This can be explained following the procedure elaborated for the case of a perturbation whose polarization is collinear to the polarization of driving pulse. The detachment probability W_p is invariant with respect to the simultaneous substitution $\phi_{CEP} \rightarrow -\phi_{CEP}$, $\theta_p \rightarrow \pi + \theta_p$ (which corresponds to $\mathbf{p} \rightarrow -\mathbf{p}$), and $\varphi \rightarrow -\varphi$, i.e., $W_{-\mathbf{p}}(-\phi_{CEP}, -\varphi) = W_{\mathbf{p}}(\phi_{CEP}, \varphi)$. For the coefficients (6) we can write $A_n(-\mathbf{p}; -\phi_{CEP}) = -A_n(\mathbf{p}; \phi_{CEP})$ and $B_n(-\mathbf{p}; -\phi_{CEP}) = B_n(\mathbf{p}; \phi_{CEP})$. Consequently, $\tilde{\Phi}(\pi + \theta_p; -\phi_{CEP}) = -\tilde{\Phi}(\theta_p; \phi_{CEP})$ and analogously for the phase of the phase itself. In conclusion, for the case of the perturbation parallel to the polarization of the driving pulse, the phase of the phase calculated for $-\phi_{CEP}$ and for the emission in the direction $\pi - \theta_p$ is the same as the one calculated for ϕ_{CEP} and for the emission in the direction θ_p . These two directions are related via reflection $p_x \rightarrow -p_x$. On the other hand, for the case of the perturbation orthogonal to the polarization of the driving pulse, the phase of the phase calculated for $-\phi_{CEP}$ and for the emission in the direction $\pi + \theta_p$ is the same as the one calculated for ϕ_{CEP} and for the emission in the direction θ_p . These two directions are related via rotation by the angle π ($\mathbf{p} \rightarrow -\mathbf{p}$). Finally, in Fig. 11 we present the focal-averaged phase of the phase as a function of the CEP calculated for the photoelectrons with energy $E_p = 0.1$ a.u. emitted in the directions as indicated in the legend. For the chosen values of the emission angle, the phase of the phase is

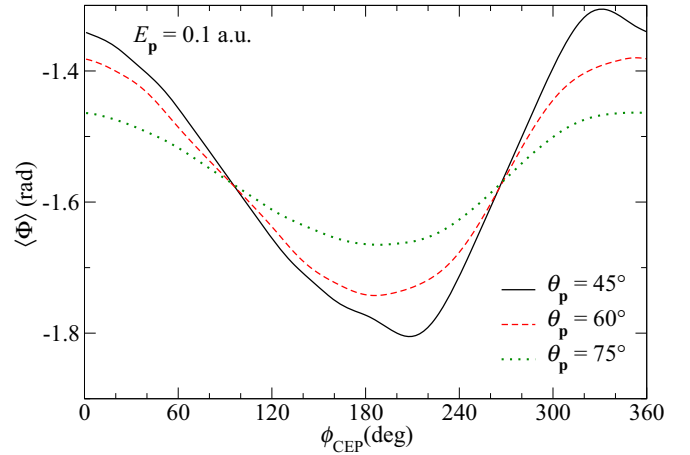


FIG. 11. Same as in Fig. 6 but for the perturbation pulse with polarization orthogonal to the polarization of the driving pulse.

negative. Contrary to the case of the perturbation pulse which is polarized along the polarization of the driving pulse, the CEP dependence is less pronounced for the emission close to the direction orthogonal to the driving-pulse polarization. Also, the dependence of the phase of the phase on the CEP is not as significant as in the case when the polarizations of the driving and the perturbation pulses are collinear. For $\phi_{CEP} = \pm\pi/2$ the focal-averaged phase of the phase is close to $-\pi/2$. Similar conclusions hold for other values of the photoelectron energy. On the other hand, for the emission in the direction $-\theta_p$ the conclusions are analogous to those for θ_p but with the opposite sign of the phase of the phase. In conclusion, the imprint of the CEP on the photoelectron momentum distribution is less pronounced in comparison with the collinear polarization case.

In summary, the orthogonally polarized two-color configuration preserves a strong and robust imprint of the CEP in the phase of the phase, even after focal averaging. While the reduced symmetry of the laser field leads to asymmetric angular structures in both the momentum-resolved and energy-averaged phase of the phase, the overall CEP dependence remains smooth and pronounced. This demonstrates that the phase of the phase provides a reliable observable for extracting CEP information also in the orthogonal polarization geometry while offering additional angular sensitivity compared to the collinearly polarized case.

C. Dependence of the phase of the phase on the pulse length

After analyzing the dependence of the phase of the phase on the CEP, we now investigate its dependence on the pulse length. In Fig. 12 we present the focal-averaged phase of the phase for our laser pulse with the CEP $\phi_{CEP} = 0^\circ$ and for the emission in the direction $\theta_p = 45^\circ$. The polarizations of the driving and perturbation pulses are in the same direction and the number of optical cycles per pulse is indicated in the panel. The phase of the phase is highly sensitive to the pulse length, particularly for very short pulses. For example, for $n_p = 3$ the phase of the phase is around $\langle \Phi \rangle \approx 2.4$ rad, while for $n_p = 4$ it is around $\langle \Phi \rangle \approx 1.1$ rad (cf. the black solid and red dashed lines in Fig. 12). As the pulse length

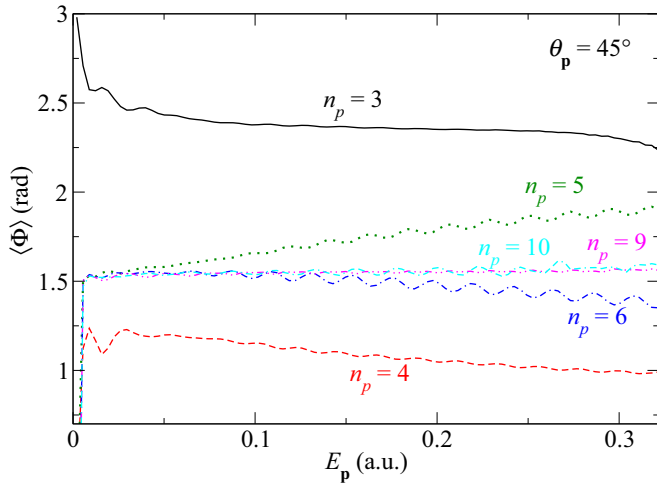


FIG. 12. Focal-averaged phase of the phase for ATD induced by exposing the F^- ion to our laser pulse with $\phi_{\text{CEP}} = 0^\circ$ and for the emission in the direction $\theta_p = 45^\circ$. The number of optical cycles per pulse is indicated in the panel. The polarizations of the driving and perturbation pulses are in the same direction.

increases, the difference between the values of the phase of the phase calculated with subsequent values of n_p decreases (cf. the green dotted and blue dash-dotted lines in Fig. 12 for $n_p = 5$ and 6, respectively) and it becomes negligible when the pulses become long (cf. the magenta dash-double-dotted and cyan double-dash-dotted lines in Fig. 12 for $n_p = 9$ and 10, respectively). This demonstrates a clear imprint of the pulse length on the value of the phase of the phase indicating that this observable can successfully be employed for measurement of the pulse duration.

A similar analysis can be performed for the case of the perturbation with the polarization orthogonal to the polarization of the driving pulse. In Fig. 13 we present the results analogous to those presented in Fig. 12 but for the case when the polarizations of the perturbation and the driving pulse are mutually orthogonal. The conclusions remain unchanged with the exception that the differences between the values of the

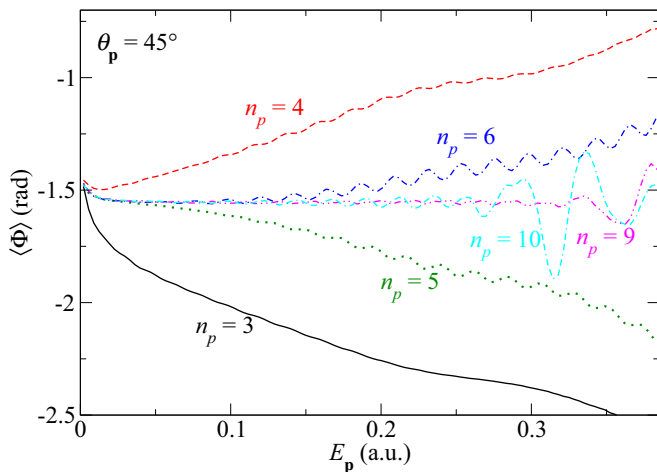


FIG. 13. Same as in Fig. 12 but for the perturbation pulse with polarization orthogonal to the polarization of the driving pulse.

phase of the phase calculated for subsequent values of n_p are less pronounced in comparison with the collinear case. Therefore, the phase of the phase does not only exhibit a strong dependence on the CEP, but it is also highly sensitive to the pulse length, which is another important parameter of few-cycle pulses.

In summary, the phase of the phase exhibits a strong and systematic dependence on the pulse length in the few-cycle regime, with pronounced variations for very short pulses and a rapid convergence towards a pulse-length-independent behavior as the number of optical cycles increases. This behavior reflects the intrinsically subcycle origin of the phase-of-the-phase signal and demonstrates that it is sensitive not only to the CEP of the field, but also to the finite temporal extent of the driving pulse. Consequently, the phase of the phase provides a direct and robust fingerprint of the few-cycle character of the laser pulse, complementing its sensitivity to the CEP discussed in the previous sections.

IV. CONCLUSION

In this paper we have investigated CEP-dependent effects in above-threshold detachment of negative ions driven by few-cycle two-color laser pulses within the framework of phase-of-the-phase spectroscopy. In addition, we analyzed the dependence of the phase of the phase on the pulse length. Using the strong-field approximation adapted to ultrashort pulses with two carrier frequencies, we demonstrated that the CEP and the pulse length leave a clear and robust imprint on the phase of the phase even when the analysis is restricted to direct electrons, whose yield is orders of magnitude higher than that of rescattered electrons and therefore experimentally more accessible.

For the case in which the perturbation pulse is polarized parallel to the driving pulse, we showed that the momentum-resolved phase of the phase exhibits a pronounced dependence on the CEP over the entire momentum plane. While the CEP dependence of the photoelectron yield itself is weak for the direct electrons, the phase of the phase provides a much more sensitive observable. The CEP information is encoded both in the overall offset and in the sign structure of the phase of the phase, with characteristic behavior near $\phi_{\text{CEP}} = \pi/2$, where the cosine component of the two-color modulation is suppressed. Importantly, after focal averaging the fine oscillatory structures are smoothed out, leading to a particularly clear and monotonic dependence of the phase of the phase on the CEP, especially for emission directions close to orthogonal with respect to the driving-pulse polarization.

We further extended our analysis to the case in which the perturbation pulse is polarized orthogonally to the driving pulse. In this configuration the reduced symmetry of the total laser field is directly reflected in the phase-of-the-phase spectra, which no longer exhibit reflection symmetry with respect to the polarization axis. Nevertheless, the CEP dependence remains strong, smooth, and robust across the momentum plane and after focal averaging. This demonstrates that phase-of-the-phase spectroscopy retains its capability for CEP retrieval also in orthogonally polarized two-color fields while offering at the same time an additional angular sensitivity due to the broken symmetry.

Beyond the dependence on the CEP, we showed that the phase of the phase is highly sensitive to the pulse length in the few-cycle regime. For very short pulses, changes of only a single optical cycle lead to substantial variations of the phase of the phase, while for longer pulses the observable rapidly converges to a pulse-length-independent behavior. This demonstrates that the phase of the phase not only encodes the information about CEP, but also provides a clear fingerprint of the finite temporal extent of ultrashort laser pulses. As such, it offers a complementary route for characterizing few-cycle pulse durations within strong-field measurements.

Overall, our results establish phase-of-the-phase spectroscopy as a powerful and experimentally feasible tool for accessing key properties of few-cycle strong-field processes, even when only direct electrons are considered. In addition to providing a clear and robust imprint of the CEP, we have shown that the phase of the phase exhibits a pronounced sensitivity to the pulse duration in the few-cycle regime, while converging to a pulse-length-independent behavior for longer pulses. This dual sensitivity demonstrates that the phase of

the phase encodes complementary information about both the CEP and the temporal structure of ultrashort laser pulses. The demonstrated robustness with respect to focal averaging and polarization geometry further highlights the potential of this method for realistic experimental implementations and for characterization of few-cycle laser pulses in strong-field and attosecond physics. Finally, our present analysis was performed within the strong-field approximation appropriate for negative ions. For atoms the inclusion of Coulomb corrections is expected to modify quantitative details of the spectra. However, the overall trends and the pronounced dependence of the phase of the phase on the CEP and the pulse duration are governed primarily by the laser-driven electron dynamics and are therefore expected to remain valid when the influence of the Coulomb potential is taken into account.

DATA AVAILABILITY

The data are available from the authors upon reasonable request.

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