Carrier-envelope phase dependence of the spectra of reflected few-cycle laser pulses in the presence of a static electric field

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The carrier-envelope phase (CEP)–dependent spectral effect of the reflected few-cycle ultrashort pulse from a dense medium is investigated. It is shown that when adding static electric field, a pronounced CEP dependence of the reflected spectrum can be obtained. Moreover, the period of the CEP-dependent signal becomes 2π because the inversion symmetry for laser-matter interaction is broken, which allows for extraction of the CEP information in both sign and amplitude.

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It is well established that when pulse duration is down to a few cycles, the phase of the carrier wave with respect to the pulse envelope, i.e., the carrier-envelope phase (CEP), will become an important parameter [1,2]. Many strong-field processes, such as high-harmonic generation [3] and optical field ionization [4], depend sensitively on the CEP of the few-cycle laser pulse. Hence the abilities to stabilize and measure the CEP have been key points. Though the stability of the CEP has been greatly improved [5,6], only a few investigations on measuring the CEP are realized [7–9]. By detecting the difference in photoionization rates for opposite directions, the determination of the CEP of powerful few-cycle laser pulse has been demonstrated in experiment for the first time [8]. A simplified device for measurement and control of the CEP of few-cycle pulses with an accuracy of better than $\pi/10$ was proposed recently [9].

Resonant extreme nonlinear optics has attracted considerable interest because it potentially allows one to gain access to the CEP at relatively lower pulse intensities [10–15]. In extreme nonlinear optics, the electric field itself governs the behavior and, equivalently, the spectral width of the envelope can approach the carrier frequency or even exceed it [13]. As a result, new spectral signal around the two-level transition frequency occurs due to the broadband harmonic spectrum and resonant enhancement effect [13,15]. Another example is the carrier-wave Rabi flopping [10–12], which indicates the breakdown of the area theorem [16]. Moreover, it has been found that these resonant extreme nonlinear effects depend on the CEP of the initial incident few-cycle pulses [12–15]. Although these phenomena are predicted for a two-level system, they have been successfully demonstrated experimentally in semiconductors [11,13,14]. However, because of the inversion symmetry of the interaction, the period of the CEP-dependent spectral signal is π, which means that the sign of the few-cycle electric field would remain undetermined.

In order to remove this π-shift phase ambiguity, breaking the inversion symmetry for laser-matter interaction is crucial [17]. By adding a surface second-order process, the CEP-dependent optical rectification in the regime of resonant nonlinear optics implies that full extraction of the CEP should be possible [18]. Moreover, in our previous works, we have shown that the inversion-asymmetry medium, such as the polar molecule [19] and asymmetry quantum well [20], can be used to extract information about the CEP. In this paper, we propose an idea that considers the reflection of a few-cycle pulse from a dense medium in the presence of a static electric field. Our results show that when a static electric field is added, the inversion symmetry ($\phi \to \phi+\pi$) for the laser-matter interaction can be broken. This results in the strong variation in the spectral signal as a function of the CEP, and the period of the CEP-dependent signal is 2π.

When the pulse propagates in a dense medium, a reflected pulse can be generated. Since it was first predicted by Roso-Franco [21], there is a great deal of works devoted to studying the novel nonlinear optical effects [22–25]. For example, the Doppler shift of the self-reflected pulses was reported by Forysiak *et al.* [24], and it was proved later in an experiment by using a semiconductor medium [25]. For few-cycle laser pulses propagating in a resonant two-level dense medium, it has been found that the reflected pulse can also be formed, and a large redshift in the reflected pulse is predicted [26].

The pulse reflection can be successfully described by the Maxwell-Bloch equations [24,26]. As done in Refs. [24,26], we consider the propagation of the few-cycle ultrashort pulse in a two-level medium. To study the CEP effect of the few-cycle ultrashort pulse, the slowly-varying-envelope approximation and the rotating-wave approximation cannot be adopted. In our work, we solve numerically the full-wave Maxwell-Bloch equations without any standard approximations. The pulse initially propagates in the free-space region, then it partially enters the dense medium at $z=24\ \mu$m, and partially reflects backward. The backward reflected pulse is detected at $z=5\ \mu$m. Taking the initial laser field polarized along x direction, and propagating along z direction, the full-wave Maxwell equations may be written in the following form:

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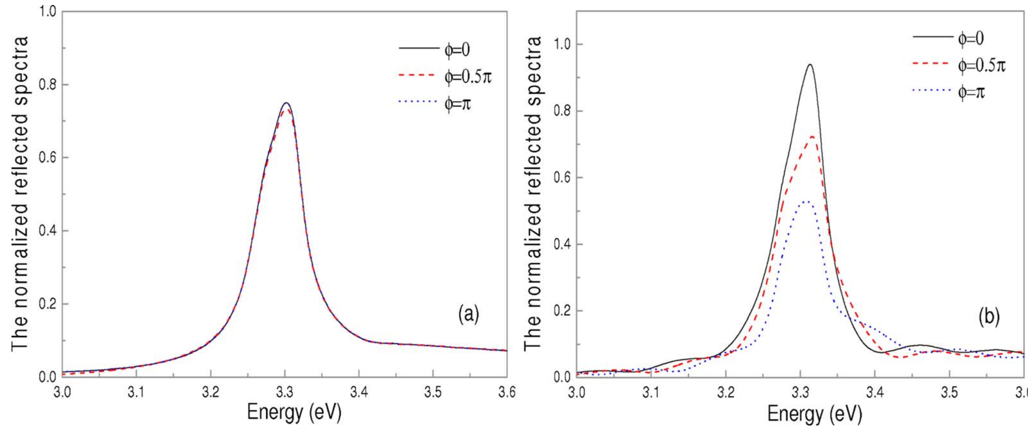

FIG. 1. (Color online) (a) The reflected spectra of the few-cycle laser pulse in the absence of a static electric field. (b) Same as (a) but for the presence of a static electric field.

$$\frac{\partial H_y}{\partial t} = -\frac{1}{\mu_0}\frac{\partial E_x}{\partial z}, \quad \frac{\partial E_x}{\partial t} = -\frac{1}{\varepsilon_0\varepsilon_s}\frac{\partial H_y}{\partial z} - \frac{1}{\varepsilon_0\varepsilon_s}\frac{\partial P_x}{\partial t}, \quad (1)$$

where μ_0 and ε_0 are the magnetic permeability and the electric permittivity in the vacuum, respectively. $P_x = 2Nd\,\mathrm{Re}[\rho_{12}]$ is the macroscopic nonlinear polarization which connects with the off-diagonal element of the density matrix in the medium, and ε_s is the relevant dielectric constant of the medium, while within the vacuum, $P_x=0$ and $\varepsilon_s=0$. d is the dipole matrix element. The Bloch equations take the forms

$$\frac{\partial \rho_{12}}{\partial t} = -i\omega_{12}\rho_{12} + i\frac{d}{\hbar}(E_x+F)n - \frac{1}{\tau_2}\rho_{12}, \quad (2)$$

$$\frac{\partial n}{\partial t} = i\frac{2d}{\hbar}(E_x+F)(\rho_{12}-\rho_{12}^*) - \frac{1}{\tau_1}n, \quad (3)$$

where ρ_{12} is the off-diagonal element of the density matrix, $n=\rho_{22}-\rho_{11}$ is the population difference between the excited and ground states, and ω_{12} is the transition frequency of the medium. E_x and F are the initial laser field and the static electric field, respectively. τ_1 is the excited-state lifetime; τ_2 is the dephasing time. The full-wave Maxwell-Bloch equations are solved by employing the finite-difference time-domain (FDTD) discretization scheme of Yee [27,28]. Mur absorbing boundary conditions [29] are incorporated with FDTD discretization in order to avoid the influence of the finite-space computational domain. The initial laser field propagating along z direction is

$$E_x(t=0,z) = E_0\,\mathrm{sech}[1.76(z-z_0)/c\tau_0]\cos[\omega_0(z-z_0)/c+\phi], \quad (4)$$

where E_0 is the peak field strength of the envelope. ϕ is the initial CEP, and τ_0 is the full width at half maximum (FWHM) of the pulse intensity envelope. The choice of z_0 ensures that the pulse penetrates negligibly into the medium at $t=0$.

Since several numerical predictions based on two-level model have been demonstrated experimentally in semiconductors [11–14], here we consider the medium parameters based on the ZnO band gap [13], namely, $E_g=\hbar\omega_{12}=3.3$ eV, $d=0.19$ e nm, $\varepsilon_s=4.0$, $N=4\times10^{20}$ cm^{-3}, $\tau_1=\infty$, and $\tau_2=50$ fs. The incident pulse has a FWHM with the intensity of $\tau_0=5$ fs and a central energy $\hbar\omega_0=1.4$ eV. The amplitude of laser field we used is $E_0=2.44\times10^8$ V/cm, yielding pulse area of about 5π within the medium. [The pulse area is defined as $A=\frac{d}{\varepsilon_s\hbar}\int\tilde{E}(t)dt$, where $\tilde{E}(t)$ is the envelope of the pulse.]

We first investigate the CEP dependence of the reflected pulse in the absence of a static electric field ($F=0$). Figure 1(a) shows that a peak near 3.3 eV (i.e., the resonant frequency) can be generated in the reflected spectrum, which is consistent with the transmission case [13,15]. With the CEP increasing, the reflected spectrum is little changed.

Then we consider the influence of a static electric field on the reflected spectrum. As shown in Fig. 1(b), the addition of a static electric field having strength 3% of the laser electric field strength can dramatically affect the reflected pulse. For $\phi=0$, the reflected spectrum around the resonant frequency is enhanced, while for $\phi=\pi$, the spectrum is much weaker. One thing that should be highlighted is that, as in the transmission case [13,15], the spectral peak around the resonant frequency is still a special result for the few-cycle pulses.

What is the underlying physical mechanism for the large variation with the CEP under the assistance of the static electric field? By analysis, we found that this CEP-dependent phenomenon is due to the fact that the static electric field enlarges the CEP-dependent variation in the peak electric strength of the few-cycle pulse. Figure 2 shows the total electric fields $E=E_x(t)+F$ for $\phi=0$ and $\phi=\pi$, respectively. The dashed line represents the case of $F=3\%E_0$, while the solid line represents the case of $F=0$. It shows that when $\phi=0$, the maximum of the electric field points to the direction of the static electric field, which is equivalent to increasing the electric field amplitude [see Fig. 2(a), dashed line]. While when $\phi=\pi$, the maximum of the electric field points in opposite direction of the static electric field, which is equivalent to decreasing the electric field amplitude [see Fig. 2(b), dashed line].

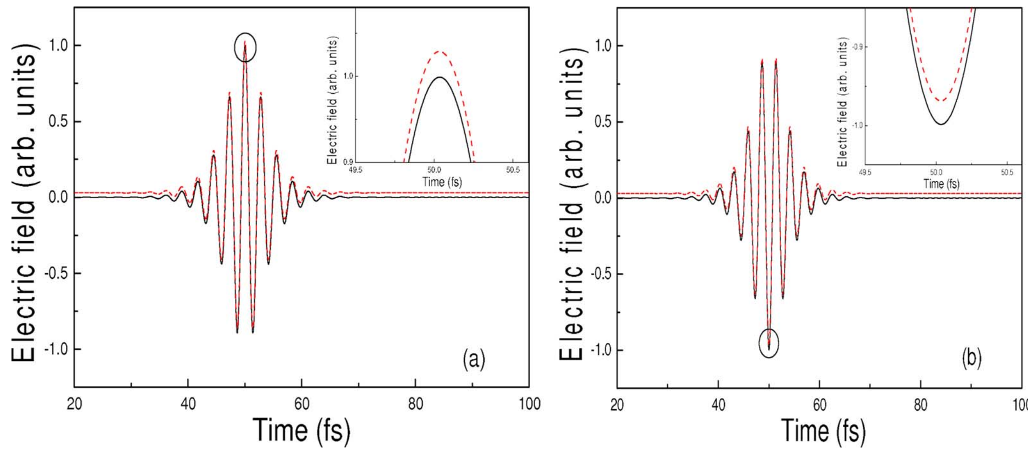

FIG. 2. (Color online) The normalized total electric fields $E = E_x(t) + F$ for (a) $\phi = 0$ and (b) $\phi = \pi$. The dashed line represents the case of $F = 3\% E_0$, while the solid line represents the case of $F = 0$. The insets show the amplified electric fields around the peak region.

In order to demonstrate the validity of our analysis, we further performed the reflected spectra for different electric fields with three peak field strengths under the conditions of $\phi = 0$ and $F = 0$. As shown in Fig. 3, surprisingly enough, the obtained reflected spectra for different peak field strengths without the static electric field are consistent with those for different initial CEPs ϕ in the presence of the static electric field [see Fig. 1(b)].

At first sight, the dramatic change in the reflected spectra seems a little hard to understand because the changes in the peak field strengths are very small. In fact, a 3% variation in the field amplitude corresponds to a 0.15π variation in the pulse areas in the medium. Since the resonant excitation depends sensitively on the pulse area, such variation in the pulse area can result in substantially different population distributions. That is the reason why the reflected pulses are so sensitive to the CEP and the intensity of the incident pulses. The population differences near the front face of the medium for three different cases are shown in Fig. 4. It can be seen that when the static electric field is absent, there is only a minute difference in the population distributions after the pulses for different CEPs. Moreover, the population distributions are the same for the CEPs $\phi = 0$ and $\phi = \pi$ [see Fig. 4(a)]. In contrast, when assisted by a 3% static electric field, the CEP-dependent difference becomes very obvious [see Fig. 4(b)]. Similar distributions can also be found for the case of variation in the field amplitude while keeping the CEP fixed [see Fig. 4(c)].

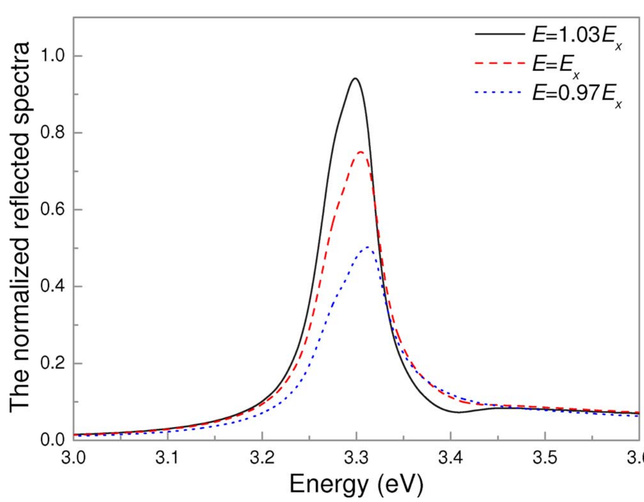

FIG. 3. (Color online) The reflected spectra for the initial incident electric fields with different peak field strengths under the conditions of $\phi = 0$ and $F = 0$.

The CEP-dependent population distributions can further affect the macroscopic coherent polarization $P_x(t)$ which acts as a source of the re-emitted field $E_{em}(t)$; i.e., $E_{em}(t) \propto -\frac{\partial P_x}{\partial t}$ [19,30] will directly induce the CEP dependence of the reflected spectra around the resonant frequency of the medium. The re-emitted spectra $[\hat{E}_{em}(\omega)]$ near the front face of the medium in the presence of a static electric field are shown in Fig. 5, which are consistent with our results on the reflected spectra.

Figure 6 shows the intensity of the reflected spectra near 3.3 eV as a function of the initial CEP of the incident few-cycle laser pulse for different static electric fields. The influence of the static field on the reflected spectrum is obvious. For $F = 0$, though the variation with the CEP is small, it can still be seen that the period of the CEP-dependent spectral signal is π due to the inversion symmetry, while when the static electric field is present, the period becomes 2π because the inversion symmetry is broken. Moreover, the variation with the CEP becomes much stronger. With increasing static electric field, the dependence of the reflected spectral signal on the CEP will be further enhanced. As a result, when assisted by a static electric field, the reflected spectrum suggests an alternative method for determination of the CEP of the initial incident few-cycle pulses in both sign and amplitude.

Since the reflected spectrum depends on both the CEP and the electric field amplitude, to extract the CEP information, one must calibrate the incident light intensity. This can be realized by recording the "normalized" contrast $(R = \frac{I - I'}{\bar{I}})$ of pairs of the reflected spectra I and I' at $\phi_1 = \phi$ and $\phi_2 = \phi + \pi$, where ϕ is varied in small steps, and $\bar{I}$ is equal to the

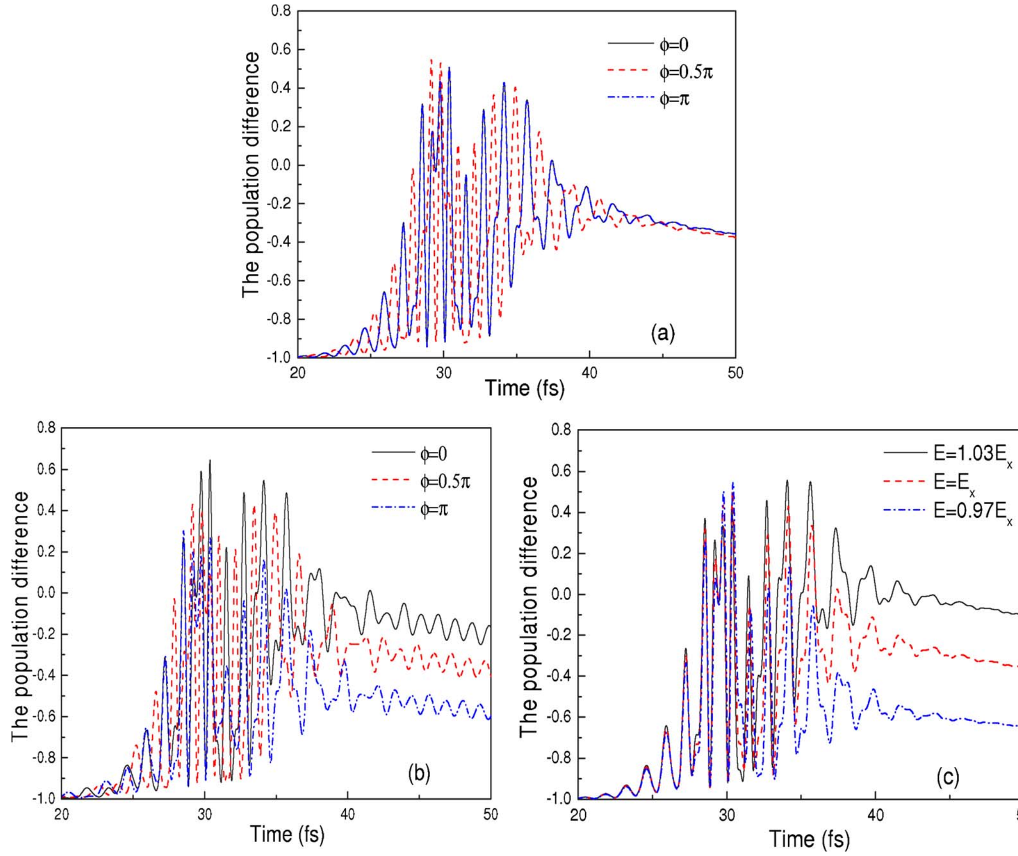

FIG. 4. (Color online) (a) The population difference n near the front face for the static electric field $F=0$. (b) Same as (a) but for the static electric field $F=3\%E_0$. (c) Same as (a) but for the CEP $\phi=0$ and a 3% variation in the laser field amplitude.

maximum value of I and I'. Division by $\bar{I}$ is equivalent to a process of normalization. For a certain ϕ, a single shot of the incident laser pulse is split into two beams with equal energy; one beam acting as reference is an additional π phase shift. In this method, the CEP $\phi=0$ can be identified from the largest positive value of R, while the largest negative value of R means $\phi=\pi$ [see Fig. 7(a)]. Comparing with the intensity of the reflected spectra [see Fig. 7(b)], recording the normalized contrast R can greatly reduce the influence of the intensity fluctuation of the incident pulse for various CEPs, which confirms the validity of our method. This method has been used in previous work in strong-field regime, in which

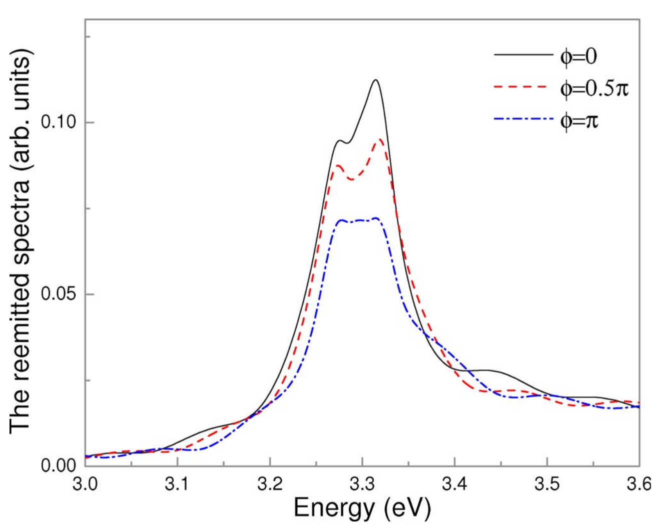

FIG. 5. (Color online) The re-emitted spectra $[\hat{E}_{em}(\omega)]$ near the front face of the medium in the presence of a static electric field.

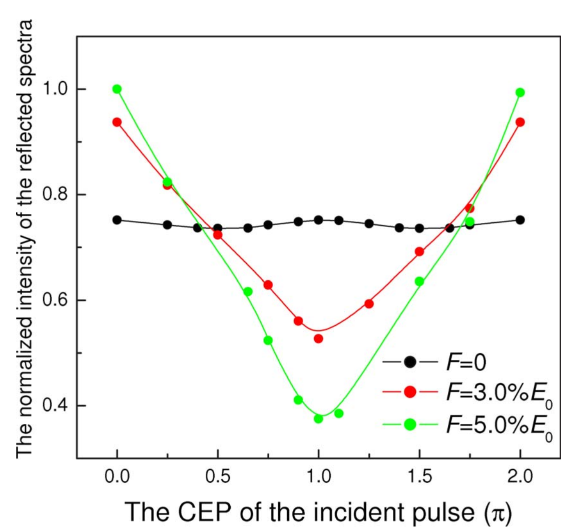

FIG. 6. (Color online) The normalized intensity of the reflected spectra around the resonant frequency as a function of the CEP for different static electric field strengths.

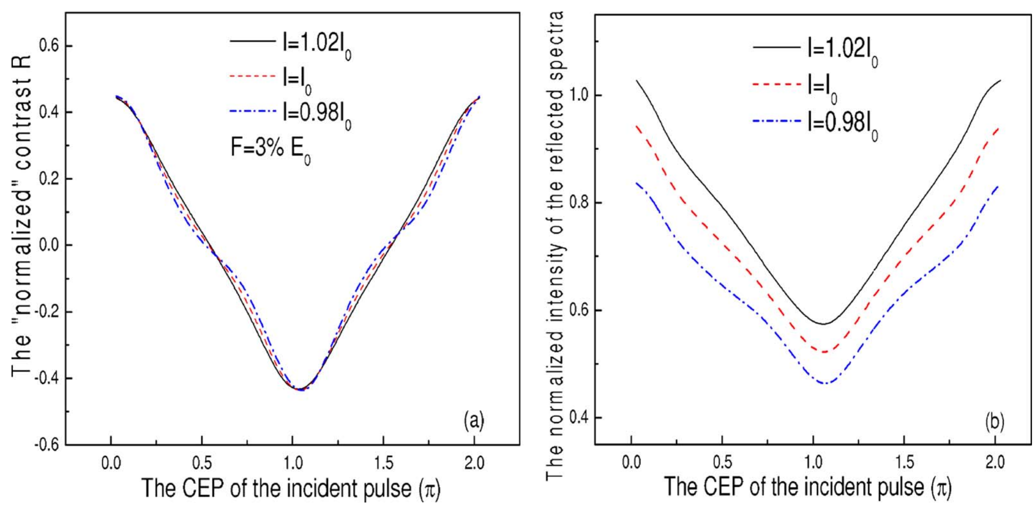

FIG. 7. (Color online) (a) The normalized contrast R and (b) the normalized intensity of the reflected spectra as functions of the CEP for different incident laser intensities.

the CEP can be evaluated with the highest possible accuracy by recording pairs of harmonic spectra [2]. The accuracy of the method depends on the laser-pulse stability, the precision of the detector, etc. [2,9].

The static electric field strength in our paper will be difficult to achieve directly in experiment nowadays. However, since the pulse duration we used is extremely short and the main role of the static electric field in our work is to enhance the CEP-dependent electric field variation, as suggested in Refs. [31,32], a laser field with a much lower frequency (such as CO_2 laser field and terahertz pulses) may be used instead of the static electric field. A midinfrared optical parametric amplifier pulses are also the possible candidates [33].

In summary, we have studied the reflected spectrum of the few-cycle ultrashort laser pulse from a dense medium in the presence of a static electric field. It has been found that a small modification of the electric field strength induced by the presence of the static electric field is sufficient to result in strong variation in the reflected spectra with the CEP. Moreover, the presence of the static electric field also breaks the dynamical symmetry that allows the determination of the CEP not only in its amplitude but also sign. Further analysis performed provides an explanation for this effect: The presence of the static electric field modifies the population distributions and the macroscopic coherent polarization, which leads to the CEP-dependent reflected spectra.

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