Carrier-envelope phase dependence in single-cycle laser pulse propagation with the inclusion of counter-rotating terms

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We focus on the propagation properties of a single-cycle laser pulse through a two-level medium by numerically solving the full-wave Maxwell-Bloch equations. The counter-rotating terms in the spontaneous emission damping are included such that the equations of motion are slightly different from the conventional Bloch equations. The counter-rotating terms can considerably suppress the broadening of the pulse envelope and the decrease of the group velocity rooted from dispersion. Furthermore, for incident single-cycle pulses with envelope area 4π , the time-delay of the generated soliton pulse from the main pulse depends crucially on the carrier-envelope phase of the incident pulse. This can be utilized to determine the carrier-envelope phase of the single-cycle laser pulse.

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I. INTRODUCTION

The modern technological progress in ultrafast optics makes it possible to produce few-cycle laser pulses [1-3]. Recently, a single-cycle pulse with a duration of 4.3 femtoseconds has been generated experimentally [4]. Furthermore, great effort for the generation of extremely short pulses via few-cycle laser pulses has been made [5, 6], particularly, single-cycle gap solitons [7] and unipolar half-cycle optical pulses [8], respectively, generated in the dense media with a subwavelength structure. If the pulse duration approaches the optical cycle, the strongfield-matter interaction enters into the extreme nonlinear optics [9], and the standard approximations of the slowly varying envelope approximation (SVEA), and the rotating-wave approximation (RWA) are invalid [10, 11]. When the Rabi frequency of the few-cycle laser pulse becomes comparable to the light frequency, the electric field time-derivative effects will lead to carrier-wave Rabi flopping (CWRF) [12], which was observed experimentally in the semiconductor GaAs sample [13]. In this extreme pumping regime, the simple two-level system can still serve as a reference point [9, 14, 15].

For the few-cycle laser pulses, the absolute carrierenvelope phase (CEP) strongly affects the temporal variation of the electric field. These effects give rise to many CEP dependent dynamics, such as high-harmonic generation [16–18], optical field ionization [19, 20], atomic coherence and population transfer [21, 22], etc. The CEP dependent strong interactions also provide routines to determine the CEP of few-cycle ultrashort laser pulses. In particular, the strong-field photoionization provides very efficient tools to measure the CEP of powerful few-cycle femtosecond laser pulses for the first time [23]. Another promising approach to determine the CEP is introduced on the detection of the THz emission by down-conversion from the few-cycle strong laser pulse [24]. Recently, the angular distribution of the photons emitted by an ultrarelativistic accelerated electron also provides a direct way of determining the carrier-envelope phase of the driving laser field [25]. However, all these measurements of CEP are based on light amplification in strong-field regime.

Therefore, it is very meaningful to explore routines for determining the CEP of few-cycle laser pulse at relative lower intensities without light amplification. The nonperturbative resonant extreme nonlinear optics effects would be good candidates for measuring the CEP of few-cycle laser pulses with moderate intensities [14, 15]. However, the period of these CEP-dependent effects is π due to the inversion symmetry of light-matter interaction in twolevel systems. Thus, the sign of the few-cycle laser pulse still cannot be determined. In order to remove the π shift phase ambiguity, the violation of inversion symmetry should be considered [26]. In the presence of an electrical bias, the phase-dependent signal of ultrafast optical rectification in a direct-gap semiconductor film implies a possible technique to extract the CEP [27]. Moreover, the inversion-asymmetry media, such as polar molecules [28] and the asymmetric quantum well [29], could also be utilized to determine the CEP of few-cycle laser pulses.

In this paper, we introduce the counter-rotating terms



FIG. 1: (color online) Schematic of the light-matter interaction scheme. The green curve illustrates the incident few-cycle laser pulse. The blue curve illustrates the transmitted laser field. Symbol " \rightarrow " denotes the propagation direction axis.

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(CRT) in the spontaneous emission damping, and investigate the influence of CRT on the propagation dynamics of nonamplified single-cycle laser pulses in two-level media. The CRT should be considered for such ultrashort pulses interacting with the medium with strong relaxation processes, because the CRT can notably suppress the broadening of the pulse envelope and the decrease of the group velocity arising from dispersion. Furthermore, when the incident single-cycle pulse with envelope area $\Theta = 4\pi$ propagates through the two-level medium, it splits into two pulses. The stronger main pulse moves faster than the weaker generated soliton pulse, and the pulse time-delay between them shows a pronounced CEP dependence. Therefore, in the presence of a static electric field, we present a simpler approach for measuring the CEP of the few-cycle laser pulses, by detecting the time-delay of the generated soliton pulse.

II. APPROACH

A. Maxwell Equations

We consider the propagation of a few-cycle laser pulse in a resonant two-level medium along the z axis, as shown in Fig. 1. The pulse initially moves in the free-space region, then it penetrates the medium on an input interface at z = 0 and propagates through the medium, and finally, it exits again into the free space through the output interface at z = L. With the constitute relation for the electric displacement for the linear polarization along the x axis, $D_x = \epsilon_0 E_x + P_x$, the full-wave Maxwell equations for the medium take the form:

$$\frac{\partial H_y}{\partial t} = -\frac{1}{\mu_0} \frac{\partial E_x}{\partial z},\tag{1a}$$

$$\frac{\partial E_x}{\partial t} = -\frac{1}{\epsilon_0} \frac{\partial H_y}{\partial z} - \frac{1}{\epsilon_0} \frac{\partial P_x}{\partial t}, \qquad (1b)$$

where E_x and H_y are the electric and magnetic fields, respectively. μ_0 and ϵ_0 are the magnetic permeability and the electric permittivity in the vacuum, respectively. The macroscopic nonlinear polarization $P_x = -Nd_{12}u$ is connected with the off-diagonal density matrix element $\rho_{12} = \frac{1}{2}(u + iv)$ and the population inversion $w = \rho_{22} - \rho_{11}$, which are determined by the Bloch equations below.

B. Master Equation

The Hamiltonian of the two-level system we considered can be described by [30]:

$$H = \sum_{k} \hbar \omega_{k} a_{k}^{\dagger} a_{k} + \hbar \omega_{0} S_{z} + \hbar \Omega(t) (S^{+} + S^{-}) + i \sum_{k} (\vec{g}_{k} \cdot \vec{d}_{21}) \{ a_{k}^{\dagger} (S^{+} + S^{-}) - \text{H.c.} \}, \qquad (2)$$

where ω_0 is the transition frequency, and \vec{d}_{21} is the electric dipole moment of the transition between the upper state $|2\rangle$ and the lower state $|1\rangle$. $a_k^{\dagger}(a_k)$ is the creation (annihilation) operator for photons with momentum $\hbar k$ and energy $\hbar \omega_k$, while $\vec{g}_k = \sqrt{\frac{2\pi\hbar\omega_k}{V}}\vec{e}_{\lambda}$ describes the vacuum-atom coupling and \vec{e}_{λ} represents the unit polarization vector with $\lambda \in \{1,2\}$. $S^+ = |2\rangle\langle 1| (S^- = |1\rangle\langle 2|)$ is the dipole raising (lowering) operator of the two-level system, $S_z = (|2\rangle\langle 2| - |1\rangle\langle 1|)/2$ is the inversion operator. $\Omega(t) = d_{12}E_x/\hbar$ is the Rabi frequency of the incident laser field.

In the usual Born-Markov and mean-field approximation, but without the rotating-wave approximation, the master equation of the system is determined by

$$\dot{\rho}(t) + i \left[\omega_0 S_z + \Omega(t) \left(S^+ + S^- \right), \rho \right] = -\gamma \left[S^+, (S^+ + S^-) \rho \right] + \text{H.c.}, \quad (3)$$

where an overdot denotes differentiation with respect to time. Here, $[S^+, S^+\rho(t)]$ and its hermitian conjugate term represent the counter-rotating terms (CRT) for the spontaneous emission damping, which are neglected under the rotating-wave approximation when the duration of the laser field pulse τ_p is much larger than ω_0^{-1} . However, for the few-cycle pulses, even the single-cycle or sub-cycle pulse, the CRT become indispensable and cannot be neglected. In the following, we will investigate the effects of the CRT on the propagation dynamics of the single-cycle laser pulse in the two-level medium.

C. Bloch Equations

Based on the master equation (3), including the CRT in the spontaneous emission damping, the Bloch equations with CRT can be easily derived as follows:

i

$$=\omega_0 v,$$
 (4a)

$$\dot{v} = -\omega_0 u + 2\Omega(t)w - 2\gamma_2 v, \qquad (4b)$$

$$\dot{w} = -2\Omega(t)v - \gamma_1(w+1), \qquad (4c)$$

where γ_1 and γ_2 are the spontaneous decay rates of the population and polarization, respectively. The Bloch equations with CRT [Eqs. (4)] are slightly different from the conventional Bloch equations (see for instance Refs. [5, 11]):

$$\dot{u} = \omega_0 v - \gamma_2 u, \tag{5a}$$

$$\dot{v} = -\omega_0 u + 2\Omega(t)w - \gamma_2 v, \tag{5b}$$

$$\dot{w} = -2\Omega(t)v - \gamma_1(w+1), \qquad (5c)$$

in which the relaxation constants γ_1 and γ_2 are added phenomenologically.

D. Numerical Method

The propagation properties of the few-cycle laser pulse in the two-level medium can be modeled by the full-wave



FIG. 2: (color online) (a) The Rabi frequency of the incident single-cycle pulse with envelope area $\Theta = 2\pi$. (b) and (c) are the time-dependent electric fields and the corresponding population inversions at the distance $z = 90 \ \mu\text{m}$, respectively. The length of the two-level medium is choosen as $L = 110 \ \mu\text{m}$. The blue solid curves are for the case of Maxwell-Bloch equations with CRT, while the red dashed curves are for the case with the conventional Maxwell-Bloch equations. The black lines represent the pulse envelope.

Maxwell-Bloch equations beyond the SVEA and RWA, which can be solved by the iterative predictor-corrector finite-difference time-domain discretization scheme [11, 31]. For such an extremely short laser pulse, we define



FIG. 3: The corresponding spectra of the electric field in Fig. 2(b). The black dashed-dotted curve is the spectrum of the incident laser pulse. The blue curve depicts the case with CRT, and the red dashed curve the case without CRT.

the vector potential at z = 0 as in Refs. [32, 33]:

$$A_x(t) = A_0 \operatorname{sech}[1.76(t-t_0)/\tau_p] \sin [\omega_p(t-t_0) + \phi], \ (6)$$

where A_0 is the peak amplitude of the vector potential, ω_p is the photon energy, and ϕ being the CEP. τ_p is the full width at half maximum (FWHM) of the short pulse and t_0 is the delay. The electric field can be obtained from $E_x = -\partial A_x(t)/\partial t$. In what follows, we assume that the two-level medium is initialized in the ground state with u = v = 0 and w = -1. The material parameters are chosen as in Ref. [5]: $\omega_0 = 2.3 \text{ fs}^{-1}$ ($\lambda = 830 \text{ nm}$), $d_{12} = 2 \times 10^{-29} \text{ Asm}$, $\gamma_1^{-1} = 1 \text{ ps}$, $\gamma_2^{-1} = 0.5 \text{ ps}$, the density $N = 4.4 \times 10^{20} \text{ cm}^{-3}$. The incident pulse has a FWHM in single optical cycle $\tau_p = 2.8 \text{ fs}$ and the photon energy $\omega_p = \omega_0$. The Rabi frequency $\Omega_0 = -A_0 \omega_p d/\hbar = 1 \text{ fs}^{-1}$ corresponds to the electric field of $E_x = 5 \times 10^9 \text{ V/m or}$ an intensity of $I = 6.6 \times 10^{12} \text{ W/cm}^2$, and the incident pulse area is defined as $\Theta = \int_{-\infty}^{\infty} \Omega(t) dt$.

III. RESULTS AND DISCUSSION

Now, we focus on the effects of CRT on the propagation dynamics of single-cycle laser pulses in two-level medium by comparing the numerical results from the Maxwell-Bloch equations with CRT [Eqs. (1) and (4)] and without CRT [Eqs. (1) and (5)]. We use an incident single-cycle pulse with envelope area $\Theta = 2\pi$ for these simulations with the medium zone length: $z = 110 \ \mu$ m.

According to the standard area theorem, the pulse with area $\Theta = 2\pi$ can transparently propagate through the two-level medium without suffering significant lossness the so-called self-induced transparency (SIT) [34]. However, when the laser pulse envelope contains only few optical cycles, the standard area theorem breaks down because of the occurrence of CWRF [12]. From our numerical results, for the short propagation distance, the usual SIT regime is essentially recovered. However, at a further distance, the established conditions for SIT are destroyed due to the extreme nonlinear optical effects. Fig. 2(b) and Fig. 2(c) present the normalized electric-field pulses and the corresponding population inversions at the distance $z = 90 \ \mu m$ for different approaches, namely, the blue solid curves depict the case obtained from Maxwell-Bloch equations with CRT, while the red dashed curves are for the conventional approach without CRT. Compared with the incident single-cycle 2π pulse in Fig. 2(a), the electric-field pulses for both two cases become clearly broadened induced by the dispersion, and suffer the decrease in pulse amplitude. Accordingly, the population differences for both cases undergo an incomplete Rabi flopping with the CWRF [Fig. 2(c)].

However, there are notably different features between these two approaches. The electric-field pulse from the approach with CRT [blue solid curves in Fig. 2(b)] is evidently narrower than that in the case without CRT [red dashed curves in Fig. 2(b)]. This can be easily seen from the corresponding spectra shown in Fig. 3. The spectrum



FIG. 4: The transmitted electric field of the incident pulses with envelope area $\Theta = 4\pi$ for different initial CEPs: (a) 0 and (b) $\pi/2$. The length of two-level medium is $L = 80 \ \mu$ m.

of the case with CRT is obviously more broadened than that in the case without CRT, although both of them become narrower than the incident spectrum. The envelope peak from the approach with CRT is relatively larger than that in the case without CRT [Fig. 2(b)], hence, the former case lead to more population inversion at the leading edge of the electric-field pulse [Fig. 2(c)]. Moreover, there is a notably time delay of the electricfield pulses and the corresponding population inversions between the two approaches [Fig. 2(b) and (c)]. It means that the group velocity of the propagating pulse from the conventional approach without CRT is obviously smaller than that in the case with CRT. This difference in the group velocity is rooted from the different influence of dispersion effects for these two approaches. Comparing Eqs. (4) with Eqs. (5), there is no damping of the real part of polarization u in the Bloch equations with CRT, which indicates that the dispersion does not suffer lossness. That is to say, the presence of CRT evidently suppresses the strong dispersion effects, which lead to the broadening of pulse envelope and the decrease of the group velocity.

In addition, we also find that the influence of CRT on the propagation dynamics of the single-cycle laser pulses is significantly enhanced with the increase of the spontaneous decay rates. Therefore, the CRT is important and indispensable for the study of the propagation properties of few-cycle laser pulses in the medium with strong relaxation processes. In the following discussion, we will use our established full-wave Maxwell-Bloch equations with CRT [Eqs. (1) and (4)] to explore an approach for determining the CEP of the single-cycle laser pulse.

In what follows, we simulate the incident single-cycle pulses with larger envelope area, i.e. $\Theta = 4\pi$, propagating through the two-level medium with a length $L = 80 \ \mu \text{m}$. During the course of pulse propagation, the medium absorbs and emits photons and redistributes

energy in the pulse. The propagating pulses are altered in shape until it reaches a stable status after some propagation distance by splitting into two pulses, the strong main pulse and the SIT soliton pulse. However, the former moves faster than the latter, which is why the generated SIT soliton pulse breaks up from the main pulse. We show the transmitted pulses of the incident single-cycle pulse with pulse envelope area $\Theta = 4\pi$ for different CEP $\phi = 0$ and $\phi = \pi/2$ in Fig. 4. It can be seen that both of the transmitted pulses split into two pulses. There is a time delay between the main pulse and the soliton pulse defined as $t(\phi)$. The time-delay for the incident pulse with CEP $\phi = \pi/2 [t(\phi = \pi/2)]$ is evidently larger than that in the case with CEP $\phi = 0$ $[t(\phi = 0)]$. It demonstrates that the pulse time delay $t(\phi)$ is sensitive to the initial CEP of the incident pulse.

For simplicity, we define the relative pulse time delay $\Delta t = t(\phi) - t(\phi = 0)$ to indicate the CEP dependence. We present the relative pulse time delay Δt as a function of the initial CEP of the incident pulse in Fig. 5 with blue circles. It is found that the relative pulse time delay Δt is related to the CEP of the incident pulse with a nearly cosinelike dependence. However, the time delay $t(\phi = \pi)$ is exactly the same as $t(\phi = 0)$, and hence, the period of the CEP-dependent pulse time delay is only π because of the inversion symmetry of light-matter interaction. This means that we cannot distinguish the incident pulse from the initial CEP $\phi \rightarrow \phi + \pi$.

In order to remove the π -shift phase ambiguity, we add a static electric field to break the inversion symmetry of the light-matter interaction [35]. As a result, the Rabi frequency terms in Bloch equations [Eqs. (4)] should change as $\Omega(t) \rightarrow \Omega(t) + f$, where f describes the strength of the static electric field. The presence of the



FIG. 5: The relative pulse delay between the transmitted soliton pulses as a function of the CEP of the incident pulses for different strengths of the static electric field. The solid lines are a guide for the eyes.

static electric field gives rise to the enhancement of the CEP-dependent variation in the peak electric strength of the single-cycle pulse, which will enhance the CEP dependence of the dynamics effects. The relative pulse time delay Δt of the transmitted soliton pulses as a function of the initial CEP of the incident single-cycle pulses for different static electric fields is presented in Fig. 5. Compared with the blue circles of f = 0, the influence of the static electric field on the relative pulse time delay is significant. Let us take $f = 2\% \Omega_0$, for example green squares in Fig. 5, then the relative time delay $\Delta t \neq 0$ at $\phi = \pi$, i.e., $t(\phi = \pi)$ is quite different from $t(\phi = 0)$ in the presence of the static electric field. The variation with the CEP of the incident pusle also becomes much stronger. The period of the CEP-dependent pulse time delay becomes 2π because the inversion symmetry is broken assisted by the static electric field. Moreover, with the increase of the static electric field, such as $f = 3\% \Omega_0$ and $f = 4\% \Omega_0$, the dependence of the relative pulse time delay on the initial CEP is further enhanced [red diamond and black circles in Fig. 5].

As a result, in the presence of the static electric field, if the relative time delay of the generated soliton pulses is calibrated, this effect suggests an approach for determining the CEP of the incident single-cycle laser pulses in both sign and amplitude. In addition, it should be pointed out that the pulse time delay might be much easier to detect compared with other features of the soliton pulse, such as the intensity and pulse duration [28]. Finally, in our discussion, the static electric filed strengths, which are a few percentages of the single cycle laser pulse strength, exceed a few MV/cm. In order to achieve this

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kind of strength of the static electric field in an experiment, we may proceed with a special case as suggested in Ref. [35] where an additional electric field with a much lower frequency (such as CO_2 laser field, terahertz pulses or a midinfrared optical parameter amplifier pulse) is used instead of the static electric field. The ultra-short dynamics can prevent the system from being destroyed or ionized.

IV. SUMMARY

In summary, we investigated the propagation properties of single-cycle laser pulses in a two-level medium including the counter-rotating terms in the spontaneous emission damping. We found that the counter-rotating term can efficiently suppress the broadening of the pulse envelope and the decrease of the group velocity. Thus, the counter-rotating term is important and indispensable for the study of the propagation dynamics of few-cycle laser pulses, even for single-cycle and sub-cycle pulses. Furthermore, we explored the CEP-dependence of the generated soliton pulse from the single-cycle laser pulse propagating through the two-level medium. The time delay of generated soliton pulses depends sensitively on the CEP of single-cycle incident laser pulse. Hence, the presence of the static electric field enhances the CEPdependence of the relative pulse time delay, which have an potential application in determining the CEP of the incident single-cycle laser pulse.

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