The effect of electric field maximum on the Rabi flopping and generated higher frequency spectra

Yueping Niu\textsuperscript{1}, Ni Cui, Yang Xiang, Ruxin Li, Shangqing Gong and Zhizhan Xu

State Key Laboratory of High Field Laser Physics, Shanghai Institute of Optics and Fine Mechanics, Chinese Academy of Sciences, Shanghai 201800, People’s Republic of China
E-mail: niuyp@mail.siom.ac.cn

\textit{New Journal of Physics} 10 (2008) 103028 (10pp)
Received 19 July 2008
Published 31 October 2008
Online at \url{http://www.njp.org/}
doi:10.1088/1367-2630/10/10/103028

\textbf{Abstract.} We investigate the effect of the electric field maximum on the Rabi flopping and the generated higher frequency spectra properties by solving Maxwell–Bloch equations without invoking any standard approximations. It is found that the maximum of the electric field will lead to carrier-wave Rabi flopping (CWRF) through reversion dynamics which will be more evident when the applied field enters the sub-one-cycle regime. Therefore, under the interaction of sub-one-cycle pulses, the Rabi flopping follows the transient electric field tightly through the oscillation and reversion dynamics, which is in contrast to the conventional envelope Rabi flopping. Complete or incomplete population inversion can be realized through the control of the carrier-envelope phase (CEP). Furthermore, the generated higher frequency spectra will be changed from distinct to continuous or irregular with the variation of the CEP. Our results demonstrate that due to the evident maximum behavior of the electric field, pulses with different CEP give rise to different CWRFs, and then different degree of interferences lead to different higher frequency spectral features.

\textsuperscript{1} Author to whom any correspondence should be addressed.
1. Introduction

The impressive progress achieved recently in the generation of ultrashort laser pulses consisting of only a few optical cycles [1]–[3] has inspired great interest in researchers and has been widely used either to probe the properties of matter or to control the physical processes. When the laser pulse duration approaches the optical cycle, the interaction of strong-field–material enters into the extreme nonlinear optics [4]. Several new features arise due to the breakdown of standard approximations such as the slowly varying envelope approximation (SVEA) and rotating wave approximation (RWA). Ziolkowski et al [5] showed that cubic-polynomial features occur at the null-field points of the pulse because of the time-derivative behavior of the field. Hughes [6] predicted that these time-derivative effects will lead to CWRF and subsequently to the formation of higher spectral components on the propagation pulses. Such signatures of CWRF were presented experimentally by Mücke et al in GaAs [7]. Further works by Hughes [8] demonstrated that realistic optical pulses interacting with two-level atoms can lead to the generation of soft-x-ray transients, which recall some similarities with high-order harmonics in high-field atomic physics. This alterative prediction is another direct manifestation of CWRF. When the pulse duration enters the single-cycle and even sub-one-cycle regime, unexpected ionization efficiency is predicted when single-cycle pulses are applied [9]. Tarasishin et al [10] failed to find soliton behavior for a sub-one-cycle $2\pi$ pulse since it displayed noticeable distortions and lengthening in the process of propagation. Došlić [11] found shortening of the Rabi inversion period and incompleteness of inversion under resonant sub-one-cycle conditions.

When the few optical-cycle ultrashort pulse is considered, the strong dependence of the electric field on the carrier-envelope phase (CEP) gives rise to CEP sensitive dynamics [12]–[23]. Most of these studies concentrate on angular distributions of photoelectrons emitted from atomic targets [12]–[14], high-harmonic generation (HHG) [15, 16], photoemission yields from metal surfaces [17, 18], control of transition probabilities in dipolar molecules [19, 20], etc. In the context of HHG, for example, it has been shown that the attosecond temporal structure of coherent soft x-ray emission directly depends on the CEP of the driving pulse [15]. Moreover, CEP control of ultrafast optical rectification has also been investigated in resonantly excited GaAs [22]. At the same time, the above CEP-dependent phenomena also provide routes to determine the CEP itself. Recently, isolated single-cycle attosecond pulses with stable CEP are generated which should lead to new applications in ultrashort physics [1].

In this paper, we will investigate the effect of the electric field maximum on Rabi flopping and the generated higher frequency spectra. It shows that this effect will cause evident reversion features and hence lead to CWRF in the sub-one-cycle pulse regime. Theory model and basic...
equations are presented in section 2. In section 3, the Rabi flopping dynamics of a two-level medium under the interaction of ultrashort pulses with different CEP is explored. With larger pulse area, the effect of electric field maximum on the generated higher frequency spectra is also studied in section 4. Section 5 is our conclusion.

2. Theory model and basic equations

The time-dependent Bloch equations describing the dynamics of a two-level medium with ground and excited states $|1\rangle$ and $|2\rangle$ interacting with an ultrashort pulse field $E_x$ reads

$$
\begin{align*}
\frac{\partial}{\partial t} u &= -\omega_{12} v - \gamma_2 u,
\frac{\partial}{\partial t} v &= \omega_{12} u + \frac{2dE_x}{\hbar} w - \gamma_2 v,
\frac{\partial}{\partial t} w &= -\frac{2dE_x}{\hbar} v - \gamma_1 (w - w_0).
\end{align*}
$$

Here, $\omega_{12}$ is the atomic transition frequency and $d$ the dipole moment. $u$ and $v$ are the dispersive and absorptive components of the off-diagonal density matrix element $\rho_{12}$. $w$ represents the population difference between the two levels and $w_0$ is the initial population difference. $\gamma_1$ and $\gamma_2$ means the population and polarization decay rate.

As pointed out by Ziolkowski et al [5], for ultrashort pulses in which the pulse duration (the full-width at half-maximum of the pulse intensity envelope) $\tau_p \ll 1/\gamma_1, 1/\gamma_2$, one has from equation (1) that

$$
\begin{align*}
\frac{\partial}{\partial t} u &\sim -\omega_{12}^2 u - \omega_{12} \left( \frac{2dE_x}{\hbar} \right) w,
\frac{\partial}{\partial t} v &\sim -\omega_{12}^2 v - \left( \frac{2dE_x}{\hbar} \right)^2 w + \frac{2d}{\hbar} w \left( \frac{\partial}{\partial t} E_x \right),
\frac{\partial}{\partial t} w &\sim -\omega_{12} \left( \frac{2dE_x}{\hbar} \right) u - \left( \frac{2dE_x}{\hbar} \right)^2 w - \frac{2d}{\hbar} v \left( \frac{\partial}{\partial t} E_x \right).
\end{align*}
$$

In [5], particular interest has been put on the impact of the time derivative of the pulse on the behavior of the two-level system. They found that when $E_x \sim 0$ and $\partial_t E_x \neq 0$ the time-derivative terms completely dominate the evolution of the nonlinear two-level system and the population difference $w$ acts like a cubic function near the points where $E_x \sim 0$. On the other hand, however, when $\partial_t E_x \sim 0$ and $E_x \neq 0$, the above equation becomes

$$
\begin{align*}
\frac{\partial}{\partial t} u &\sim -\omega_{12}^2 u - \omega_{12} \left( \frac{2dE_x}{\hbar} \right) w,
\frac{\partial}{\partial t} v &\sim -\omega_{12}^2 v - \left( \frac{2dE_x}{\hbar} \right)^2 v,
\frac{\partial}{\partial t} w &\sim -\omega_{12} \left( \frac{2dE_x}{\hbar} \right) u - \left( \frac{2dE_x}{\hbar} \right)^2 w.
\end{align*}
$$

Clearly $w$ now acts like a quartic function near the maximum of the transient electric field. Therefore, reversion features will occur in the population transition processes and this behavior will lead to CWRF.

*New Journal of Physics* 10 (2008) 103028 (http://www.njp.org/)
In the following numerical simulations, we use the vector potential $A_x(t) = F_0 \text{sech}[1.76(t - t_0)/\tau_p] \cos[\omega_p(t - t_0) + \phi]$, with $F_0$ being the peak amplitude of the vector potential, $\omega_p$ the photon frequency, $\tau_p$ the pulse duration and $\phi$ the CEP. Then, one can obtain the electric field $E_x(t)$ from $E_x(t) = -\partial A_x(t)/\partial t$. The two-level medium applied is assumed to have the parameters of Hughes [6]: $d = 4.24 \times 10^{-29} \, \text{C m}$, $\gamma_1 = \gamma_2 = 1 \, \text{ns}^{-1}$, $w_0 = -1$ and $\omega_{12} = 0.3 \, \text{fs}^{-1}$. The results that will be demonstrated in the following can be scaled to various material and laser parameters.

3. The effect of electric field maximum on the Rabi flopping dynamics

For the few-cycle ultrashort pulses, to name the result recovered by Hughes [6], although the two-level medium can be completely inverted and returned to its initial state by the $2\pi$ pulse, slight oscillations occur at the null-field points due to the time-derivative behavior of the input field. In this section, we will investigate the effect of the transient field maximum on the Rabi flopping dynamics. We simulate this by using a $2\pi$ pulse with $\omega_p = \omega_{12}$. As figure 1 shows, cubic-polynomial features caused by the time-derivative effect and quartic-polynomial features caused by the maximum of the transient field present simultaneously. With the shortening of the pulse duration, these effects manifest more evidently. Let us take $\tau_p = 5.25 \, \text{fs}$ for example.
and analyze it in figure 2. When $\phi = \pi/3$, there are four maxima (points A, C, E and G) and three null-field points (B, D and F) inside the whole pulse. When the first maximum A tends to reverse the transition, its neighboring null-field point B tends to oscillate, and hence the transition after point B is still to excite the population to the upper state. At the second maximum C, the transition reverses and hence some population goes back to the ground state. The de-excitation process to the ground state terminates till the null-field point D makes the transition slow down and even reverse again. In a similar way, the excitation and de-excitation processes present alternately. From the whole population transition process, one can see that not only the time-derivative effect of the field but also its maximum give rise to evident CWRF in the sub-one-cycle pulse regime.

Since the transient electric field of the sub-one-cycle pulse changes significantly when the CEP is varied, the population inversion is very sensitive to the CEP. We still take $\tau_p = 5.25$ fs for example. The pulse with $\phi = 0$ cannot invert all the population, whereas the pulse with $\phi = \pi/3$ can almost drive all the population to the excited state. Detailed analysis shows that the shadowed pulse area between the null-field point and the maximum of the field is larger for the case of $\phi = \pi/3$ (figure 2(a)) than that of $\phi = 0$ (figure 2(b)) and hence the former can excite more population to the upper state. In contrast to the conventional envelope Rabi flopping which might be classified as resulting from an integrated effect over the entire pulse envelope, the present result shows us that the Rabi flopping follows the transient electric field tightly through the oscillation and reversion dynamics caused by the time-derivative and maximum.
Figure 3. The generated higher frequency spectra at $z = 155 \mu m$ with $\tau_p = 63$ fs. Insets are their phase profiles.

effect of the electric field. As a result, the Rabi flopping dynamics of the two-level medium could be completely controlled instantaneously by applying pulses with different CEP.

4. The effect of electric field maximum on the generated higher frequency spectra

Hughes [6] has predicted that the electric field time-derivative effects will lead to CWRF and subsequently to the formation of higher spectral components on the propagating pulse because of strong carrier reshaping. As discussed in the above section, the maximum of the transient electric field can also give rise to CWRF, which manifests itself more evidently with the shortening of the pulse duration. Therefore, in this section, we will explore the effect of the transient electric field maximum on the generated higher frequency spectra through different CEP pulses of $8\pi$.

Assuming that the pulse propagates along the $z$-axis in the two-level medium, then the interaction property can be modeled by the above Bloch equations and the following Maxwell equations:

$$
\partial_t H_y = -\frac{1}{\mu_0} \partial_z E_x,
$$

$$
\partial_t E_x = -\frac{1}{\varepsilon_0} \partial_z H_y - \frac{1}{\varepsilon_0} \partial_t P_x.
$$

(4)
We apply a finite-difference time-domain (FDTD)-based algorithm to solve the coupled Maxwell–Bloch equations numerically. The pulse initially moves in free space, then is incident normally onto an input interface and finally exits again into free space through the output interface. The nonlinear polarization \( P_x = 2Ndu \), with \( N = 4 \times 10^{18} \text{ cm}^{-3} \) being the density of the two-level medium.

First, we show in figure 3 the frequency spectra at the propagation distance of \( z = 155 \mu \text{m} \) when \( \tau_p = 63 \text{ fs} \). Different CEP of \( \phi = 0, \pi/3 \) and \( \pi/2 \) are simulated. Obviously the generated higher frequency spectra and their phases (the insets of figure 3) are negligibly influenced by the variation of the CEP of the driving pulse. However, once the pulse duration is shortened to sub-one-cycle, the dependence of higher frequency spectra on CEP behavior is comparatively evident. Figure 4 depicts the generated higher frequency spectra and the phase profiles (the insets of figure 4) from pulses of \( \tau_p = 5.25 \text{ fs} \) with different CEP. When \( \phi = 0 \), distinct high-order harmonic peaks present, while when \( \phi = \pi/3 \), the higher frequency spectra exhibit

\[ (a) \quad \phi = 0 \]
\[ (b) \quad \phi = \pi/6 \]
\[ (c) \quad \phi = \pi/4 \]
\[ (d) \quad \phi = \pi/3 \]
\[ (e) \quad \phi = \pi/2 \]
\[ (f) \quad \phi = \pi \]

Figure 4. The generated higher frequency spectra at \( z = 155 \mu \text{m} \) with \( \tau_p = 5.25 \text{ fs} \). Insets are their phase profiles.
a continuum feature. For the case of \( \phi = \pi/2 \), however, irregular spectra come into being. Moreover, we can see clearly that the spectral phase is very sensitive to the CEP of the driving pulse.

As it is known that the generation of higher frequency spectra is mainly caused by the CWRF, hence the structure of the generated higher frequency spectra is predominantly determined by the characteristic of the CWRF. For the pulse duration of \( \tau_p = 63 \) fs, although the electric field has different CEP, the electric field maximum nearly has no effect on the CWRF and hence the CWRF exhibits similar characteristic (not shown here). Consequently, the generated higher frequency spectra from different CEP pulses exhibit similar features. However, when the pulse duration is shortened further, e.g. \( \tau_p = 5.25 \) fs, not only the slight oscillations but also the reversion features manifest remarkably in the CWRF. Since the electric field, and hence its maxima distribution, changes significantly with the CEP, these transient features will cause completely different carrier reshaping during the propagation process when pulses of different CEP are applied.

We plot the electric field with different CEP and the corresponding population dynamics at the propagation distance of \( z = 155 \) \( \mu \)m in figure 5. For \( \phi = 0 \), the CWRFs are mainly induced by the leading and back parts of the pulse (figure 5(a)). The two carriers have similar amplitude and their interference leads to the distinct high-order harmonic spectra. When

---

**Figure 5.** The population difference \( w \) (solid line) and the normalized electric field \( E_x \) (dashed line) at the propagation distance of \( z = 155 \) \( \mu \)m with \( \tau_p = 5.25 \) fs.
\( \phi = \pi/3 \), however, only the pulse tail causes CWRF (figure 5(b)). Hence, the produced higher frequency spectra are continuous. Note the case of \( \phi = \pi/2 \), both the front half-part and the tail of the pulse cause the CWRF (figure 5(c)), so the generated higher frequency spectra own a oscillation character but the distinct feature is not so definite. In a similar way, pulses with other CEP can lead to other different higher frequency spectral distributions. Thus, the CEP dependent higher frequency spectral distribution reveals the effect of the electric field maximum in the sub-one-cycle pulse regime.

5. Conclusions

We have investigated the effect of the electric field maximum on the Rabi flopping and the generated higher frequency spectra. It is found that the effect of the electric field maximum acts like a quartic function and causes reversion features in the Rabi flopping. Therefore, not only the time-derivative but also the maximum of the electric field can lead to CWRF. Our results show that in the sub-one-cycle pulse regime, different CEP makes the transient field have different maximum distribution and therefore the reversion behavior leads to different CWRF characteristics. Hence, the Rabi flopping follows the transient electric field tightly and its dynamics could be controlled instantaneously through different CEP pulses. With larger pulse area, the higher frequency spectra generated from the sub-one-cycle pulses present distinct or continuum and even irregular features under different CEP conditions. Since the field maximum causes evident CWRF in the sub-one-cycle regime, the carrier reshaping after a long propagation distance is completely different when the CEP is varied, which leads to different higher frequency spectral features.

Acknowledgments

This work was supported by the National Natural Sciences Foundation of China (grant no. 60708008), the Project of Academic Leaders in Shanghai (grant no. 07XD14030), 973 program (grant no. 2006CB806000) and the Knowledge Innovation Program of the Chinese Academy of Sciences.

References

[1] Sansone G et al 2006 Science 314 443
[2] Cavalieri A L et al 2007 New J. Phys. 9 242
[3] Hauri C P et al 2007 Opt. Lett. 32 868
[4] Brabec T and Krausz F 2000 Rev. Mod. Phys. 72 545
[5] Ziolkowski R W, Arnold J M and Gogny D M 1995 Phys. Rev. A 52 3082
[6] Hughes S 1998 Phys. Rev. Lett. 81 3363
[7] Mücke O D, Tritschler T and Wegener M 2001 Phys. Rev. Lett. 87 057401
[8] Hughes S 2000 Phys. Rev. A 62 055401
[9] Liu C P and Nakajima T 2007 Phys. Rev. A 76 023416
[10] Tarasishin A V, Magnitskii S A, Shuvaev V A and Zheltikov A M 2001 Opt. Express 8 452
[11] Došlić N 2006 Phys. Rev. A 74 013402
[12] Dietrich P, Krausz F and Corkum P B 2000 Opt. Lett. 25 16

New Journal of Physics 10 (2008) 103028 (http://www.njp.org/)
[13] Paulus G G, Grasbon F, Walther H, Villoresi P, Nisoli M, Stagira S, Priori E and De Silvestri S 2001 Nature 414 182
[14] Zhang J, Feng X, Xu Z and Guo D S 2004 Phys. Rev. A 69 043409
[15] Baltuška A et al 2003 Nature 421 611
[16] Nisoli M, Sansone G, Stagira S, De Silvestri S, Vozzi C, Pascolini M, Poletto L, Villoresi P and Tondello G 2003 Phys. Rev. Lett. 91 213905
[17] Apolonski A et al 2004 Phys. Rev. Lett. 92 073902
[18] Lemell C, Tong X M, Krausz F and Burgdörfer J 2003 Phys. Rev. Lett. 90 076403
[19] Brown A and Meath W J 1998 J. Chem. Phys. 109 9351
[20] Cheng T and Brown A 2004 Phys. Rev. A 70 063411
[21] Mücke O D, Tritschler T, Wegener M, Morgen U and Kärtner F X 2002 Phys. Rev. Lett. 89 127401
[22] Van Vlack C and Hughes S 2007 Phys. Rev. Lett. 98 167404
[23] Zhang C, Song X, Yang W and Xu Z 2008 Opt. Express 16 1487