High-order harmonic generation in sub-one-cycle regime*

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(Received 27 October 2011; revised manuscript received 27 December 2011)

We theoretically investigate the high-order harmonic generation from the hydrogen atom driven by the laser pulses with the durations less than the optical cycle. It is found that the switching term of the laser field may have an obvious influence on the cutoff, intensity or plateau structure of the high-order harmonic spectrum. Generally speaking, the switching term can shorten the cutoff of the high-order harmonic spectrum for a relatively longer pulse and extend the cutoff for a relatively shorter pulse.

Keywords: high-order harmonic generation, sub-one-cycle pulse, switching term

PACS: 42.65.Ky **DOI:** 10.1088/1674-1056/21/6/064216

1. Introduction

As is well known, when atoms or molecules are exposed to an intense laser field, one can observe the high-order harmonic generation (HHG) in the emitted radiation. The generic shape of HHG spectrum is described below:[1] at first, there is a sharp decline in conversion efficiency at low harmonic orders, then a plateau appears in which the intensity of the spectrum varies slowly with harmonic order and at last an abrupt cutoff occurs at energy about $I_{\rm p} + 3.17U_{\rm p}$,[2] where $I_{\rm p}$ is the ionization potential of the atom and $U_{\rm p}$ is the ponderomotive energy of the electron in the laser field. Usually, the physical origin of the HHG process can be described by the well-known quasi-classical three-step-model (TSM) proposed by Corkum:[3] Firstly, the electron tunnels through the barrier formed by the Coulomb potential and the laser field, then oscillates in the laser field and may finally return back and recombine with the parent ion. During the recombination, a photon is emitted with energy equal to the return kinetic energy (RKE) of the electron plus $I_{\rm p}$.

The HHG has attracted great interest around the world since its initial observations by McPherson *et al.*[4] and Ferray *et al.*[5] While in those days, the duration of the driver laser pulse contained many optical cycles of the laser field and only the discrete odd harmonics of the driver field could be generated.[6,7] with the development of laser technology, a pulse with a duration of only a few optical cycles can be obtained in experiment.[8] Therefore, a continuum spectrum appears near the cutoff of the HHG spectrum, which can be used to generate a single attosecond pulse.[9] Due to its application in attosecond pulse generation, the HHG has become a very hot topic in recent years and most of these works are concerned with single attosecond generation by different controlled ways.[10−15] However, to our knowledge, there are no detailed studies on the HHG in the sub-one-cycle regime. As proposed in Ref. [16], a sub-one-cycle pulse can be generated in dense media of two-level systems due to pulse splitting and reshaping of pulses of a few optical-cycles time duration and Chen *et al.* obtained sub-one-cycle pulse trains experimentally using 7 Raman sidebands generated by molecular modulation in H_2.[17] As a matter of fact, many physical processes in the sub-one-cycle regime have been studied till now. For example, Barth *et al.* employed sub-one-cycle pulses in quantum simulations of periodic electron circulation in a model Mg porphyrin,[18] Doslic explored the effect that the time derivative of the pulse envelope has on the Rabi dynamics[19] and

*Project supported by the National Natural Science Foundation of China (Grant No. 10734080, 11074263, 60978013, 60921004, and 10874194) and the Doctor Fund of Henan Polytechnic University (Grant No. B2011-076).

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Niu *et al.* found that 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.[20] In this paper, we will investigate the HHG in the sub-one-cycle regime.

2. Model and method

The interaction between hydrogen atom and laser pulse can be described by the following three-dimensional (3D) time-dependent Schrödinger equation with dipolar approximation [the atomic units (a.u.) are used in all equations in this paper, unless otherwise mentioned],

$$\mathrm{i}\frac{\partial\psi(\boldsymbol{r},t)}{\partial t}=\left[-\frac{1}{2}\bigtriangledown^2-\frac{1}{|\boldsymbol{r}|}-\boldsymbol{r}\cdot\boldsymbol{E}(t)\right]\psi(\boldsymbol{r},t), \quad (1)$$

where $\boldsymbol{r}$ is the position vector and $\boldsymbol{E}(t)=E(t)\boldsymbol{z}$ is the electric field of the laser pulse with polarization direction along z axis. Equation (1) can be solved by a partial-wave decomposition of the wave function together with a Peaceman–Rachford scheme, as described in Ref. [21]. When the wave function $\psi(\boldsymbol{r},t)$ is obtained, the mean acceleration can be calculated by means of the Ehrenfest's theorem[22]

$$d_{\mathrm{A}}(t)\equiv\langle\ddot{z}(t)\rangle=-\langle\psi(\boldsymbol{r},t)|\frac{z}{r^3}-E(t)|\psi(\boldsymbol{r},t)\rangle \quad (2)$$

with $r=|\boldsymbol{r}|$. The HHG power spectrum $P(\omega)$ can be obtained by taking the Fourier transform of $d_A(t)$

$$P(\omega)=\left|\int d_{\mathrm{A}}(t)\,\mathrm{e}^{\mathrm{j}\omega_0 t}\,\mathrm{d}t\right|^2. \quad (3)$$

3. Results and analyses

In our numerical simulations, we define the vector potential as

$$\begin{aligned}A(t)&=A_0f(t)\cos(\omega t+\phi)\\&=A_0\,\mathrm{e}^{-2\ln 2t^2/\tau^2}\cos(\omega t+\phi),\end{aligned} \quad (4)$$

where A_0, ω_0, and ϕ is the peak value, frequency, and the carrier-envelope phase of the vector potential respectively, $f(t)$ is the field envelope and τ is the full width at half maximum (FWHM). Then the electric field $E(t)$ can be obtained by

$$\begin{aligned}E(t)&=-\frac{\partial A(t)}{\partial t}=Ff(t)\sin(\omega_0 t+\phi)\\&\quad-\frac{F}{\omega_0}\frac{\partial f(t)}{\partial t}\cos(\omega_0 t+\phi),\end{aligned} \quad (5)$$

where $F=A_0\omega_0$ is the amplitude of the electric field of the laser pulse. In all of our calculations, F is 0.12 a.u. (corresponding to the intensity of 5×10^{14} W/cm^2) and ω_0 is 0.057 a.u. (corresponding to the wavelength of 800 nm). The first item on the right-hand side of Eq. (5) corresponds to a pulse with a sine-oscillating carrier field and the second item is the so-called switching term (ST), which includes the time derivative of the vector potential envelope. For many-cycle pulses, the ST can be negligible, while it plays an important role in physical processes for sub-one-cycle pulses. In the following, we will investigate the HHG spectrum in the sub-one-cycle regime both for the pulse with and without ST and find out the influence of the ST on the HHG process.

3.1. HHG spectra at $\phi=0$

We show in Fig. 1 the HHG spectra driven by the laser pulse with different durations $\tau=0.8T$, $0.6T$, and $0.4T$, where T is the optical period of the laser field. From Fig. 1, we can see that with the decrease of the laser pulse duration, the cutoff of the HHG spectrum shortens, both for the driver pulse with and without ST. Though the intensities of the HHG spectra driven by the two pulses do not have much difference, their cutoffs are quite different. For $\tau=0.8T$ and $0.6T$, the cutoff of the HHG spectrum with ST is a little lower than that without ST, as shown in Figs. 1(a) and 1(b) respectively; while as can be seen from Fig. 1(c), it turns out just the opposite, i.e., the cutoff of HHG spectrum with ST is higher than that without ST, for $\tau=0.4T$. This means that the ST has different effects on the HHG spectrum cutoff for different pulse durations.

In order to find out how the ST influences the cutoff of the HHG spectrum, we show the RKE of the electron versus the tunneling ionization time in Fig. 2, as well as the electric fields of the laser pulses. The RKE of the electron is obtained by solving the Newton equation $\mathrm{d}v(t)/\mathrm{d}t=-E(t)$, with v being the velocity of the electron. According to the ADK theory,[23] the tunneling ionization rate is highly nonlinear depending on the electric field intensity and only the electrons which are born at the time corresponding to a reasonably high electric field have significant effects. From Fig. 2, we can see that the RKE of the electron is dramatically modified due to the influence of ST. In Fig. 2(a), the maximum RKE (MRKE) for the pulse without ST is at peak A with value of $2.68U_{\mathrm{p}}$ (corresponding to the photon energy $I_{\mathrm{p}}+2.68U_{\mathrm{p}}$ which is about 60 harmonics) and we note peak A as A (2.68, 60) for conciseness. At the time the electrons with the

RKE near peak A are born, the intensity of the electric field is very low which causes the low birth rate of the electrons. As a result, they have no apparent contributions to the HHG spectrum. Therefore, the cutoff of the HHG spectrum is determined by peak B (2.22, 52), just as we can see in Fig. 1(a) that an abrupt cutoff occurs near the 52nd order harmonic of the HHG spectrum without ST. For the pulse with ST, the MRKE is at peak C (2.28, 53). Though at the time the electric field intensity is a little higher than that at peak A, peak C still has no significant contributions to the HHG spectrum compared with peak D (1.99, 47). Therefore, the plateau of the HHG with ST ends at about the 47th order. As a result, the cutoff of the HHG spectrum with ST is lower than that without ST. In Fig. 2(b), the RKE with ST reaches its maximum at peak A (1.94, 46), which is larger than the peak value of B (1.72, 42), i.e., the MRKE without ST. However, the electric field at peak A is very low. For the reason mentioned above, peak A has no obvious contributions to the HHG spectrum and the cutoff of the HHG spectrum comes from peak C (1.57, 39) whose peak value is lower than that of peak B. Therefore, the HHG spectrum with ST has a lower cutoff than that without ST. When the pulse duration reduces to $0.4T$, the MRKE with ST becomes larger than that without ST, as shown in Fig. 2(c). So the HHG spectrum with ST has a higher cutoff than that without ST.

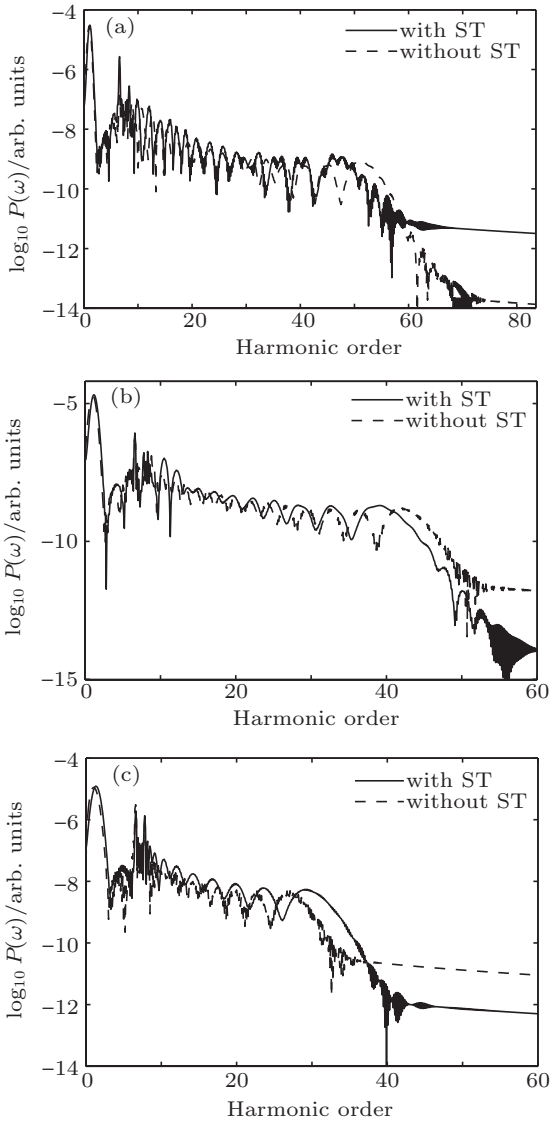


Fig. 1. The HHG spectra driven by the pulse with different durations at $\phi = 0$: (a) $\tau = 0.8T$, (b) $\tau = 0.6T$, and (c) $\tau = 0.4T$.

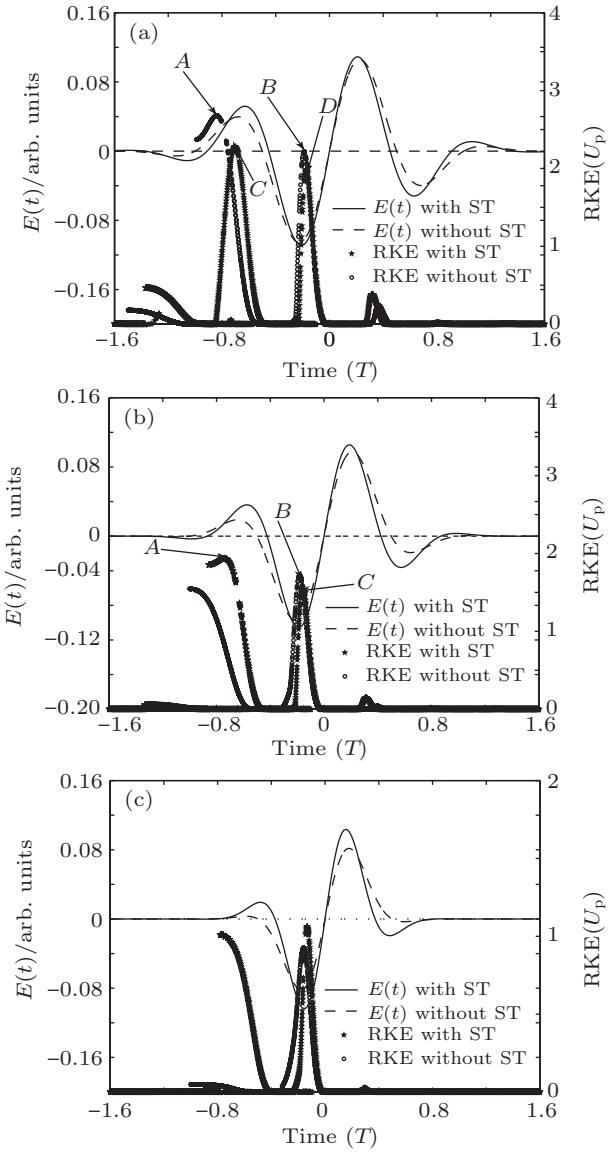


Fig. 2. The electric fields and the RKE for different pulse durations at $\phi = 0$: (a) $\tau = 0.8T$, (b) $\tau = 0.6T$, and (c)$\tau = 0.4T$.

3.2. HHG spectra at $\phi = \pi/4$

Figure 3 shows the HHG spectra for the pulses with different durations at $\phi = \pi/4$. In Fig. 3(a), the cutoff of the HHG spectrum with ST is at about the 54th order harmonic, which is about 9 harmonics lower than that without ST. In Fig. 3(b), the difference between them enlarges, which is about 12 harmonics. In Fig. 3(c), though the difference increases to about 14 harmonics, the intensity of the plateau of the HHG spectrum with ST is about two orders higher than that without ST. Apparently, the ST may influence not only the cutoff of the HHG spectrum but also its intensity. These phenomena can be interpreted from the RKE maps in Fig. 4. From Fig. 4(a), we can see that the MRKE with and without ST are at peaks A (2.36, 54) and B (2.81, 63), which agree well with the cutoffs of the HHG spectrum with and without ST in Fig. 3(a), respectively. In Fig. 4(b), the difference between the peak value of peaks A (1.98, 47) and B (2.58, 59) is larger than that in Fig. 4(a), so the difference between the two spectra in Fig. 3(b) enlarges in comparison with that in Fig. 3(a). In Fig. 4(c), the MRKE without ST is at peak B (2.11, 49), which is about 14 harmonics higher than that with ST, i.e., peak A (1.38, 35). Therefore the difference between the cutoffs of the two HHG spectra enlarges further. Note that the intensity of the electric field at peak A is comparable with that at peak B, both in Figs. 4(a) and 4(b). Accordingly, the intensities of the two HHG spectra have no significant difference, both in Figs. 3(a) and 3(b), while in Fig. 4(c), the electric field intensity at peak B is far lower than that at peak A. As a result, the HHG spectrum without ST has a plateau with intensity about two orders lower than that with ST and it can be expected that the cutoff with ST will be higher than that without ST for a shorter pulse duration, say $\tau = 0.3T$.

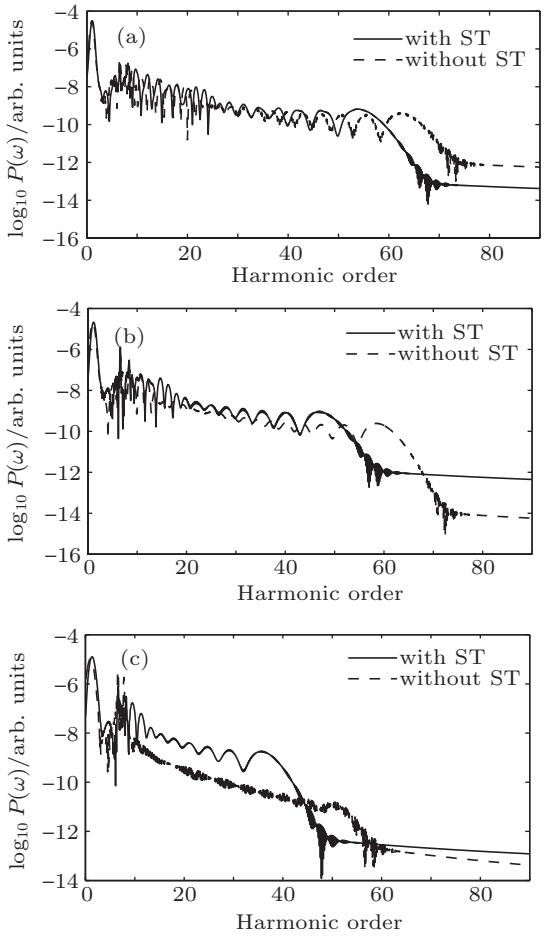


Fig. 3. The HHG spectra driven by the pulse with different durations at $\phi = \pi/4$: (a) $\tau = 0.8T$, (b) $\tau = 0.6T$, and (c) $\tau = 0.4T$.

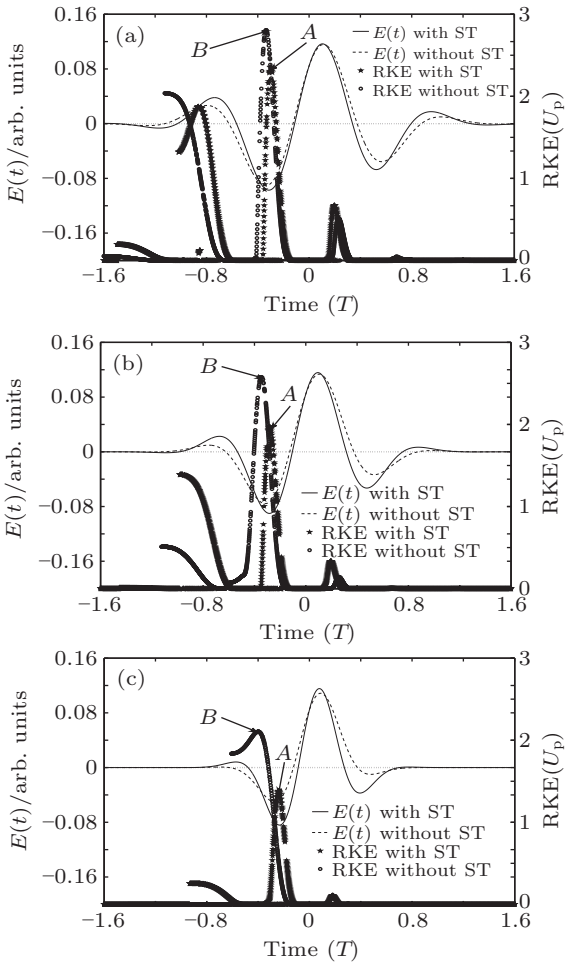


Fig. 4. The electric fields and the RKE for different pulse durations at $\phi = \pi/4$: (a) $\tau = 0.8T$, (b) $\tau = 0.6T$, and (c) $\tau = 0.4T$.

3.3. HHG spectra at $\phi = -\pi/4$

The HHG spectra generated by the pulse with different pulse durations are shown in Fig. 5. From

Fig. 5(a), we can see that the HHG spectrum has two plateaus, both for the pulse with and without ST. The cutoffs of the first plateau of the two HHG spectra are all at the 38th order harmonic, while for the second plateau, the HHG spectrum with ST has a lower cutoff. In Fig. 5(b), the HHG spectrum with ST still has two plateaus, while for the HHG spectrum without ST, only one plateau appears. For $\tau = 0.4T$, both of the two HHG spectra have only one plateau and the HHG spectrum with ST has a higher cutoff, as shown in Fig. 5(c). These phenomena mean that the HHG spectrum plateau structure is affected by the ST.

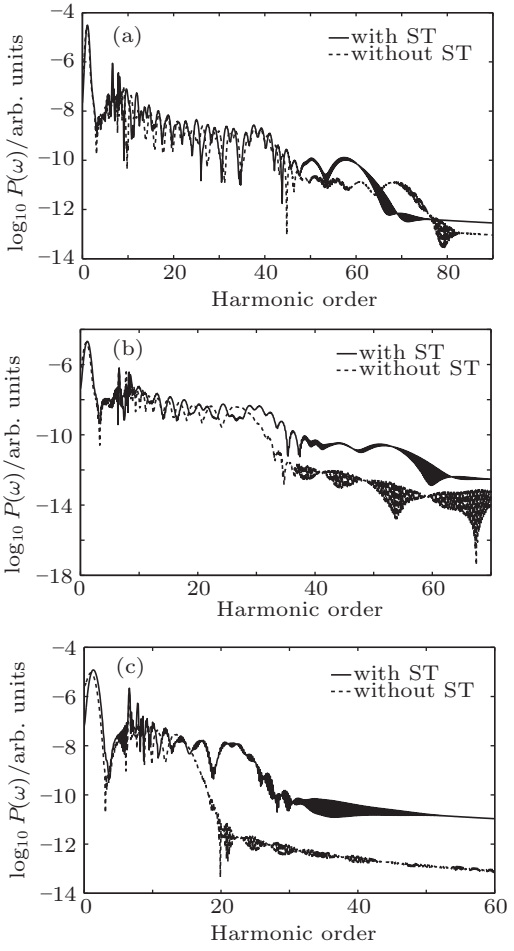


Fig. 5. The HHG spectra driven by the pulse with different durations at $\phi = -\pi/4$: (a) $\tau = 0.8T$, (b) $\tau = 0.6T$, and (c) $\tau = 0.4T$.

In order to explore the physical origin behind the above phenomena, we show the RKE of the electron as a function of the tunneling time in Fig. 6, together with the electric fields of the laser pulses. As shown in Fig. 6(a), there are two dominant peaks A (3.08, 68) and B (1.54, 38) in the RKE without ST, which contribute to the HHG spectrum. Note that the electric field intensity at peak B is stronger than that at peak A, which means the intensity of the HHG spectrum which comes from peak B is higher than that which comes from peak A, therefore two plateaus appear in the HHG spectrum with cutoffs at the 38th and 57th order harmonics respectively. For the pulse with ST, there are also two dominant peaks in the RKE map, which cause the two plateaus of the HHG spectrum with ST in Fig. 5(a). The first one is peak C (2.52, 57) and the second one overlaps with peak B. So in Fig. 5(a), we can see that the first cutoff of the HHG spectrum with ST is the same as that without ST and the second cutoff is lower than that without ST. In Fig. 6(b), we can see there are two dominant peaks A (2.19, 51) and B (1.09, 30) in the RKE map with ST, so there are two plateaus in the HHG spectrum. However, for the pulse without ST, only one plateau appears in the HHG spectrum, though there are also two peaks C (2.99, 66) and D (0.93, 27) in the RKE map. This is because the electric field intensity at peak C is very low. Therefore,

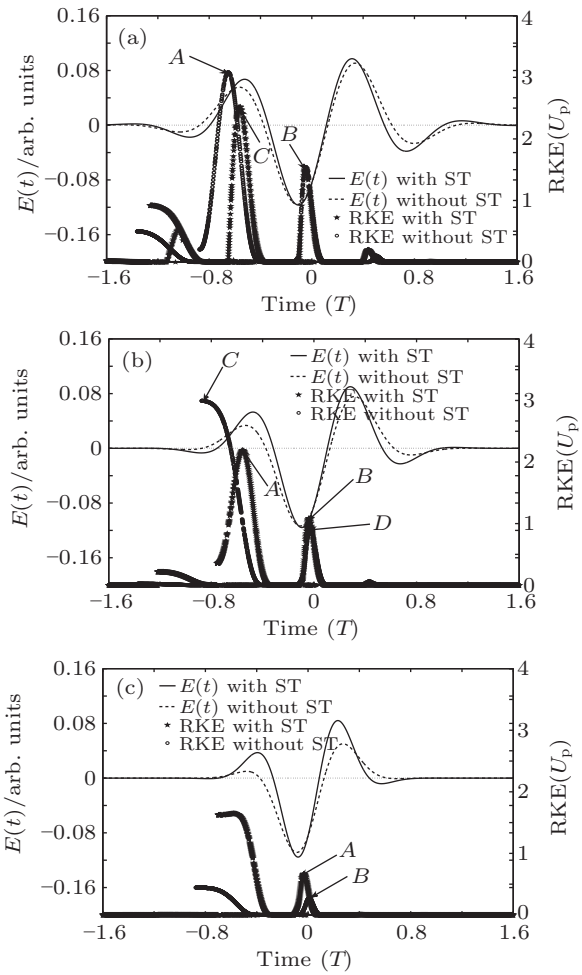


Fig. 6. The electric fields and the RKE for different pulse durations at $\phi = -\pi/4$: (a) $\tau = 0.8T$, (b) $\tau = 0.6T$, and (c) $\tau = 0.4T$.

peak C has no apparent contribution to the HHG spectrum so that there is no observable HHG spectrum above the 36th order harmonic. For the same reason, there is only one peak A (0.67, 22) that has an apparent contribution to the HHG spectrum with ST and peak B (0.27, 14) to that without ST, as shown in Fig. 6(c). Since the peak value of A is larger than that of peak B, the HHG spectrum with ST has a higher cutoff than that without ST.

3.4. HHG spectra at $\phi = \pi/2$

Finally, we investigate the HHG spectra generated by the pulse with and without ST at $\phi = \pi/2$. The HHG spectra driven by the pulses with different durations are shown in Fig. 7. For $\tau = 0.8T$, the cutoff of the HHG spectrum with and without ST is at about the 58th order harmonic, which is about 11 harmonics lower than the cutoff of the HHG spectrum without ST, as shown in Fig. 7(a). When the pulse duration shortens to $0.6T$, the cutoff of the HHG spectrum with ST is at about the 52nd order harmonic, which is lower than that in Fig. 7(a), while for the HHG spectrum without ST, the cutoff is the same as that in Fig. 7(a) and two plateaus can be clearly seen in the HHG spectrum, as shown in Fig. 7(b). In Fig. 7(c), we can see the cutoff of the HHG spectrum with ST reduces to about the 42nd order harmonic, while no apparent HHG spectrum can be observed for the pulse without ST, for $\tau = 0.4T$. These phenomena can still be understood from the TSM. So once again, we show the RKE versus the tunneling time of the electron in Fig. 8, as well as the electric fields of the driver pulses. In Fig. 8(a), peaks A (2.55, 58) and B (3.13, 69) are the MRKE for the pulse with and without ST, respectively. Since the value of peak A is lower than that of peak B, in Fig. 7(a) the HHG spectrum with ST has a lower cutoff. In Fig. 8(b), the MRKE with ST is at peak A (2.18, 51) with peak value lower than that in Fig. 8(a) and the maximum of the RKE with ST is at peak B (3.13, 69) with peak

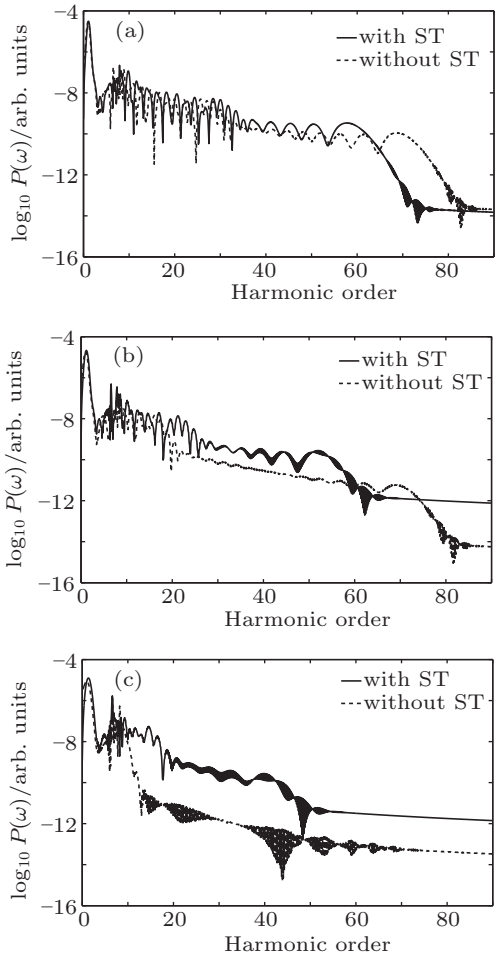


Fig. 7. The HHG spectra driven by the pulse with different durations at $\phi = \pi/2$: (a) $\tau = 0.8T$, (b) $\tau = 0.6T$, and (c) $\tau = 0.4T$.

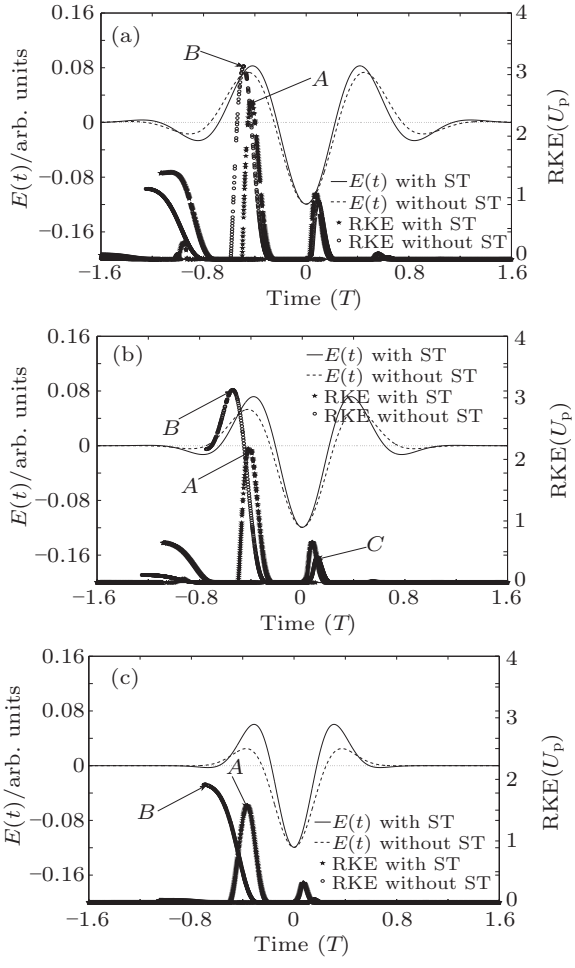


Fig. 8. The electric fields and the RKE for different pulse durations at $\phi = \pi/2$: (a) $\tau = 0.8T$, (b) $\tau = 0.6T$, and (c) $\tau = 0.4T$.

value equal to that in Fig. 8(a), so the cutoff of the HHG spectrum with ST shortens while that without ST does not change when the pulse duration varies from $0.8T$ to $0.6T$. As can be seen in Fig. 8(b), the electric field at peak B is much lower than that at peak C (0.41, 17), so apparently the HHG spectrum without ST has two plateaus with the first cutoff at about the 17th order harmonic. In Fig. 8(c), the MRKE with and without ST are at peaks A (1.57, 46) and B (1.91, 39), respectively. Though the peak value of B is higher than that of A, the electric field at peak B is far lower than that at peak A. As a result, in Fig. 7(c) one can see that for the pulse with ST the HHG spectrum is of the generic shape with cutoff at about the 39th order harmonic, while no obvious HHG spectrum can be observed for the pulse without ST.

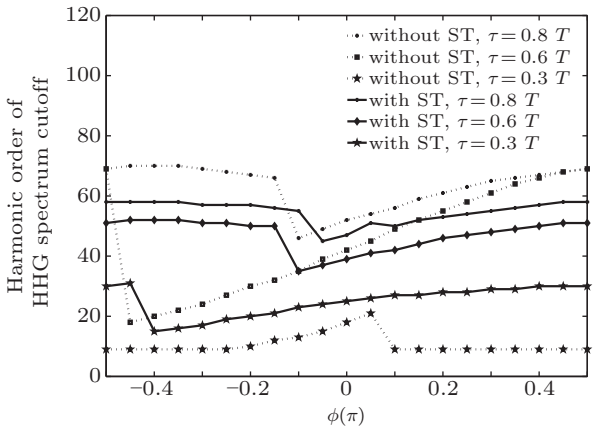


Fig. 9. The harmonic order of the HHG spectrum cutoff as a function of ϕ and τ.

From the above analyses, we can see that in the sub-one-cycle regime the ST has significant effects on the cutoff, intensity, and plateau structure of the HHG spectrum, due to the fact that the ST apparently modifies the electric field of the laser field, which not only changes the trajectories of the electrons, but also their tunneling rate. The former influences the RKE of the electrons which determines the harmonic orders of the photons. The latter influences the amount of electrons recombining with the core, which determines the intensity of the HHG spectrum. The effect of the ST on the HHG spectrum depends on the laser parameters ϕ and τ. In order to find out the effect of ST on the HHG spectrum cutoff, we show in Fig. 9 the harmonic order of the HHG spectrum cutoff as a function of ϕ and τ, both for the pulses with and without ST. As can be seen from this figure, the influence of ST on the cutoff varies with ϕ and τ. For $\tau = 0.8T$, the ST may lower the cutoff except at the phases near -0.1π. while for $\tau = 0.6T$, the ST may increase the cutoff in a larger phase range (about -0.45π–-0.1π). When the pulse duration τ reduces to $0.3T$, the ST may increase the cutoff in the whole phase range. This means that with the decrease of the pulse duration, the ST may increase the HHG spectrum cutoff in a large phase range. Generally speaking, for a relatively longer pulse, the ST may lower the cutoff and increase the cutoff for a relatively shorter pulse.

4. Conclusions

In this paper, we explored in details the HHG spectra driven by the pulses with duration less than the period of the laser field. We found that the ST has a dramatic influence which varies with the pulse duration and the CEP on the HHG spectrum. The characteristics of the HHG spectrum, such as the cutoff, intensity, and plateau structure, may be modified due to the influence of the ST. In general, for a relatively longer pulse, the HHG spectrum with ST has a lower cutoff than that without ST and has a higher cutoff than the latter for a relatively shorter pulse.

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