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Modulation of multiphoton resonant high-order harmonic spectra driven by two frequency-comb fields*

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The frequency-comb structure in the extreme ultraviolet (XUV) and vacuum ultraviolet (VUV) regions can be realized by the high-order harmonic generation (HHG) process driven by frequency-comb fields, providing an alternative approach for the measurement of an unknown frequency in XUV or VUV. We consider the case of two driving frequency-comb fields with the same repetition frequency and the carrier frequencies of fundamental- and third-harmonics, respectively. The many-mode Floquet theorem (MMFT) is employed to provide a nonperturbative and exact treatment of the interaction between a quantum system and the frequency-comb laser fields. Multiphoton transition paths involving both fundamental- and third-harmonic photons are opened due to the coupling of the third-harmonic frequency-comb field. The multiphoton transition paths are superpositioned when the carrier-envelope-phase shifts (CEPs) fulfill the matching condition. And the interference of the multiphoton transition paths can be controlled by tuning the relative envelope delay between the fields. We find that the quasienergy structure, as well as the multiphoton resonant high-order harmonic generation (HHG) spectra, driven by the two frequency-comb fields can be coherently controlled via the interference of multiphoton transition paths. It is also found that the spectral intensities of the generated harmonics can be modulated, and the modulation behavior is harmonic-sensitive.

Keywords: high-order harmonic generation, two frequency-comb laser fields, relative envelope delay

PACS: 32.80.Qk, 42.65.Ky, 42.50.Hz **DOI:** 10.1088/1674-1056/27/5/053202

1. Introduction

In the past two decades, high-order harmonic generation (HHG) from the interaction between matter and intense laser fields has been proposed as an alternative method to obtain coherent light sources in the extreme ultraviolet (XUV) and vacuum ultraviolet (VUV) regions.[1–4] Besides the fundamental physics of the laser–matter interaction, the manipulate of the features of the generated spectra has also drown researchers' attention due to potential applications.[5–10] When the matter–laser interaction is driven by a train of identical laser pulses with a fixed time interval, which corresponds to a frequency-comb structure in the frequency-domain, the frequency-comb structure is preserved in the harmonics in the HHG spectrum.[11–14] It provides an approach to bring the frequency-comb structure into XUV and VUV regions, which will allow the measurement of an unknown frequency in XUV or VUV owing to the precise and direct link between known and unknown frequencies. The generation of XUV and VUV frequency comb spectroscopy has led to the advancements in development of optical atomic clock,[15–20] high-precision optical frequency spectroscopy,[21–28] and test of quantum electrodynamics.[14,29–31]

It has been theoretically proved that HHG driven by a frequency-comb field can be dramatically enhanced via multiphoton resonance by tuning the carrier-envelope phase (CEP) shift of the driving field.[32,33] The employment of different color laser pulses has also been proved to be one of the schemes to enhance the intensities of the HHG spectra.[34–48] And the spectra can be modulated by tuning the relative envelope delay τ_d and/or the relative phase between pulses φ. For the case of two multiple-cycle pulses, the modulation features of the spectra by tuning τ_d are similar to those by tuning φ. When two trains of pulses are employed, the modulation features of the spectra by tuning τ_d may be different from those by tuning φ. Recently, a two-level system driven by intense fundamental- and weak second-harmonic frequency-comb fields was investigated in Ref. [49]. It was shown that the frequency-comb HHG spectra can be enhanced via constructively interference between the multiphoton transition paths.

In this paper, we theoretically investigate the coherent control of the quasienergy structure and multiphoton resonant high-order harmonic generation (HHG) driven by two frequency-comb fields. Additional multiphoton transition paths involving photons from both fields are opened, and the coherent control is achieved via interference of the multiphoton transition paths by tuning the relative envelope delay between the fields. We consider the case of two frequency-comb fields with the same repetition frequency and the carrier frequencies of fundamental- and third-harmonics, respectively. When the CEP shifts of the two fields are matched, different multiphoton transition paths can interfere with each other.

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Our calculation indicates that the quasienergy structure of the driven system can be modulated by tuning the relative envelope delay. Meanwhile, the spectral intensities of the generated harmonics can be modulated, and the modulation behavior is harmonic-dependent, and related to the multiphoton transition paths that contribute to the spectrum.

The paper is organized as follows. We present the many-mode Floquet theorem (MMFT) for the treatment of the interaction between quantum systems and two frequency-comb laser fields with different carrier frequencies and the same repetition frequency in Section 2. The coherent control of the quasienergy structure and HHG power spectra is investigated in Section 3. And the conclusion is presented in Section 4.

2. Theoretical method

For a quantum system driven by laser fields, the time-dependent Schrödinger equation can be written as

$$\left[\hat{H}(\boldsymbol{r},t)-\mathrm{i}\frac{\partial}{\partial t}\right]\Psi(\boldsymbol{r},t)=0, \tag{1}$$

with the Hamiltonian

$$\hat{H}(\boldsymbol{r},t)=\hat{H}_0(\boldsymbol{r})-\hat{\mu}(\boldsymbol{r})\cdot E_0(t), \tag{2}$$

where $\hat{H}_0(\boldsymbol{r})$ is the unperturbed Hamiltonian of the quantum system, and $\hat{\mu}(\boldsymbol{r})$ is the electric dipole moment operator. For the case of two driving frequency-comb fields, the laser field $E_0(t)=E(t)+E'(t)$, where[32]

$$E(t)=\sum_{k=-\infty}^{\infty}E_k\,\mathrm{e}^{\mathrm{i}\omega_k t}+\mathrm{c.c}, \tag{3}$$

$$E'(t)=\sum_{k=-\infty}^{\infty}E'_k\,\mathrm{e}^{\mathrm{i}\Omega_k(t-\tau_\mathrm{d})}\,\mathrm{e}^{\mathrm{i}\varphi}+\mathrm{c.c}, \tag{4}$$

with the relative carrier phase φ and the relative envelop delay τ_d between the two fields. The comb frequencies are given as

$$\omega_k=\omega_0+k\omega_\mathrm{r},\quad \Omega_k=\Omega_0+k\omega_\mathrm{r}, \tag{5}$$

with the main angular frequencies

$$\begin{aligned}\omega_0&=\left[\frac{\omega_\mathrm{c}-\omega_\delta}{\omega_\mathrm{r}}\right]\omega_\mathrm{r}+\omega_\delta,\\ \Omega_0&=\left[\frac{\Omega_\mathrm{c}-\omega'_\delta}{\omega_\mathrm{r}}\right]\omega_\mathrm{r}+\omega'_\delta,\end{aligned} \tag{6}$$

where [ ] is the round function, ω_c and Ω_c are the carrier frequencies, ω_r is the repetition frequency, $\omega_\delta(=\Delta\phi/\tau)$ and $\omega'_\delta(=\Delta\phi'/\tau)$ are the offset frequencies, and $\Delta\phi$ and $\Delta\phi'$ are the CEP shifts of the two fields, respectively. The field amplitudes can be expressed as

$$E_k=\frac{f_0\sigma\omega_\mathrm{r}}{\sqrt{2\pi}}\,\mathrm{e}^{-\sigma^2(\omega_0-\omega_\mathrm{c}+k\omega_\mathrm{r})/2}, \tag{7}$$

$$E'_k=\frac{f'_0\sigma\omega_\mathrm{r}}{\sqrt{2\pi}}\,\mathrm{e}^{-\sigma^2(\Omega_0-\Omega_\mathrm{c}+k\omega_\mathrm{r})/2}, \tag{8}$$

with the peak amplitudes f_0 and f'_0, and the standard deviation of a Gaussian function σ.

The time-dependent Schrödinger equation with Hamiltonian (2) is polychromatic, and it can be solved by employing the MMFT.[50–54] For the case of fields with the same repetition frequency, there are three independent frequencies ω_0, Ω_0, and ω_r. And only two independent frequencies (ω_0 and ω_r) are left when the specific matching condition for the two CEP shifts is fulfilled.[49] Then the time-dependent Schrödinger equation can be converted into an equivalent two-mode Floquet matrix eigenvalue problem. And the two-mode Floquet matrix eigenvalue equation can be constructed as[49]

$$\sum_{\beta}\sum_{l'}\sum_{m'}\langle\alpha lm|H_\mathrm{F}|\beta l'm'\rangle\langle\beta l'm'|\lambda\rangle=\lambda\langle\alpha lm|\lambda\rangle, \tag{9}$$

where the basis vectors $|\alpha lm\rangle=|\alpha\rangle\otimes|l\rangle\otimes|m\rangle$ are employed. The two-mode Floquet matrix H_F can be constructed by

$$\begin{aligned}&\langle\alpha lm|H_\mathrm{F}|\beta l'm'\rangle\\ &=H_{\alpha\beta}^{[l-l',m-m']}+(l\omega_0+m\omega_\mathrm{r})\delta_{\alpha,\beta}\delta_{l,l'}\delta_{m,m'},\end{aligned} \tag{10}$$

with

$$\begin{aligned}&H_{\alpha,\beta}^{[l-l',m-m']}\\ &=\varepsilon_\alpha\delta_{\alpha,\beta}\delta_{l,l'}\delta_{m,m'}+\sum_{k=-N}^{N}V_{\alpha,\beta}^{(k)}(\delta_{l+1,l'}\delta_{m+k,m'}+\delta_{l-1,l'}\delta_{m-k,m'})\\ &+\sum_{k=-N'}^{N'}U_{\alpha,\beta}^{(k)}(\delta_{l+3,l'}\delta_{m+k',m'}\,\mathrm{e}^{-\mathrm{i}\Omega_k\tau_\mathrm{d}}\,\mathrm{e}^{\mathrm{i}\varphi}\\ &+\delta_{l-3,l'}\delta_{m-k',m'}\,\mathrm{e}^{\mathrm{i}\Omega_k\tau_\mathrm{d}}\,\mathrm{e}^{-\mathrm{i}\varphi}),\end{aligned} \tag{11}$$

where $k'=k+m_\delta$, $\varepsilon_\alpha=\langle\alpha|\hat{H}_0|\alpha\rangle$, $V_{\alpha,\beta}^{(k)}=-\frac{1}{2}E_k\langle\alpha|\mu_z|\beta\rangle$, and $U_{\alpha,\beta}^{(k)}=-\frac{1}{2}E'_k\langle\alpha|\mu_z|\beta\rangle$. More details of the structure of H_F can be found in Refs. [32] and [49].

Solving the matrix eigenvalue problem (9) with the Floquet matrix (10), we can obtain a set of quasienergies $\lambda_{\gamma lm}$ and the corresponding eigenvectors $|\lambda_{\gamma lm}\rangle$ which satisfy the orthonormality condition. And the harmonic generation spectra can be expressed as

$$\begin{aligned}S(\omega_{l,m})&=\frac{4}{6\pi c^3}|\boldsymbol{d}_{l,m}|^2\\ &=\frac{4}{6\pi c^3}\left|\sum_{\alpha,\beta}\sum_{l',m'}\langle\lambda_{\alpha,l'-l,m'-m}|\mu_z|\lambda_{\beta,l',m'}\rangle\right|^2,\end{aligned} \tag{12}$$

in which each harmonic element $\omega_{l,m}=l\omega_0+m\omega_\mathrm{r}$.

3. Results and discussion

In this section, we present a case study of the modulation of quasienergy structures and HHG power spectra generated from a two-level system driven by fundamental- and

third-harmonic frequency-comb laser fields by varying the envelope delay. The parameters of the fundamental frequency-comb field are peak intensity 2.5×10^{15} W·cm^{-2}, carrier frequency 374.7 THz (corresponding to $\omega_c = 0.056954$ a.u. and wavelength 800 nm), and repetition frequency 10 THz (corresponding to $\omega_r = 1.51983\times10^{-3}$ a.u.) generated from a train of Gaussian pulses with 20 fs full width at half maximum (FWHM). The third-harmonic frequency-comb field has peak intensity 1×10^{14} W·cm^{-2}, which is 4% of the fundamental field, and its carrier frequency is 1124.1 THz (corresponding to $\Omega_c = 0.170862$ a.u.), i.e., the third harmonic of the fundamental field. The third-harmonic field is generated from a train of Gaussian pulses with 20 fs FWHM, which has repetition frequency 10 THz. The CEP shifts are set as $\Delta\phi/2\pi = \Delta\phi'/6\pi$, such that the matching condition is fulfilled.[49] For the two-level system, the energy separation between the levels is $\hbar\omega_{\alpha\beta} = \varepsilon_\beta - \varepsilon_\alpha = 0.28$ a.u., corresponding to the five-photon dominant resonance regime of the fundamental field ($5\omega_c \approx \omega_{\alpha\beta}$), and the transition dipole moment $\langle\alpha|\mu_z|\beta\rangle = 0.1$ a.u. is used.

Under the matching condition $\Delta\phi' = 3\Delta\phi$, we calculate the quasienergies and time-averaged transition probabilities as a function of $\Delta\phi$ when the delay is set as $\tau_d = 0$, which are presented in Figs. 1(a) and 1(b), respectively. The resonance can be achieved five times by varying the CEP shift, and these five peaks are separated exactly by $2\pi/5$. The resonance peaks of different multiphoton transition paths are coincident due to the superposition of the paths.[49] In Fig. 1(c), we also present the time-averaged transition probabilities as a function of $\Delta\phi$ for cases with different delays $\tau_d = 5T_c$, $15T_c$, $25T_c$ ($T_c = 2\pi/\Omega_c$). The resonance peak position changes as τ_d is varied, i.e., the quasienergy structure is modified. The modification of the quasienergy structure is due to the change of the overlap between pulses from the two laser fields, which mathematically is embodied in the phase of the off-diagonal elements with $\delta_{l+3,l'}\delta_{m+k',m'}$ and $\delta_{l-3,l'}\delta_{m-k',m'}$ in the Floquet matrix (10). Physically, when the pulses from the fundamental field overlap with those from the third-harmonic field, the total amplitude of the driving fields is modulated by varying τ_d. Thus, the Stark energy shifts of the levels, i.e., the quasienergy structure, are altered. It is reasonable to expect that the modification of the quasienergy structure is large when varying τ_d in the vicinity of 0, i.e., when the pulses from the two fields mostly overlap with each other. And the quasienergy structure is nearly unchanged with large values of τ_d ($> 30T_c$) where the pulses are hardly overlap with each other. We plot the CEP shifts of the resonance peaks $\Delta\phi_{res}$ as a function of τ_d in Fig. 2. It is shown that the resonance peak position oscillates with a period about T_c. And over multiple oscillation periods, the oscillation amplitude decreases with increasing τ_d.

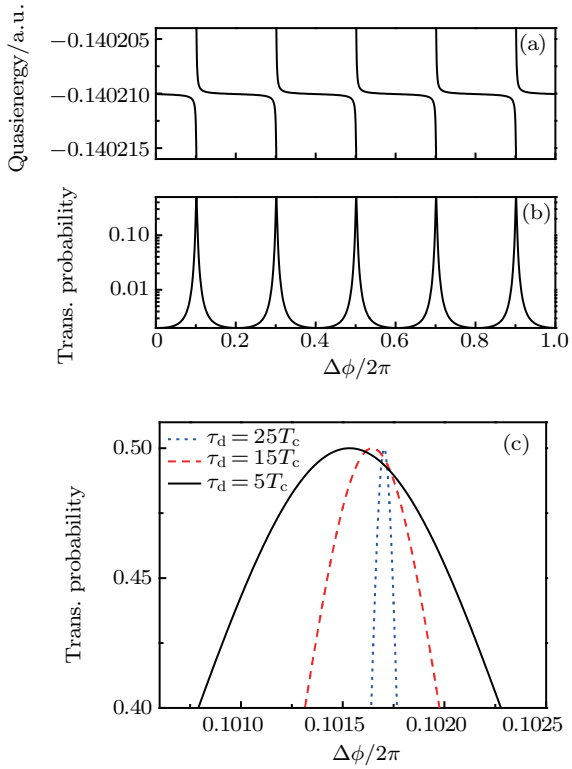


Fig. 1. (color online) (a) Plots of quasienergies and (b) time-averaged transition probabilities as a function of the CEP shift $\Delta\phi$ for the two-level system driven by fundamental and third-harmonic frequency-comb fields with their relative envelope delay $\tau_d = 0$. It has five resonance positions separated by $2\pi/5$ due to the five-photon dominant resonance. (c) Enlarged plots of the time-averaged transition probabilities as a function of the CEP shift, with envelope delay $\tau_d = 5T_c$ (black solid line), $15T_c$ (red dash line), and $25T_c$ (blue dotted line). The fundamental field has peak intensity 2.5×10^{15} W/cm^2, carrier frequency 374.7 THz, and repetition frequency 10 THz, and is generated from a train of Gaussian pulses with 20 fs FWHM. The third-harmonic field has peak intensity 1×10^{14} W/cm^2, carrier frequency 1124.1 THz, and repetition frequency 10 THz, and is generated from another train of Gaussian pulses with 20 fs FWHM. $\Delta\phi' = 3\Delta\phi$ is set to the fulfill the matching condition. The relative carrier phase is set as $\varphi = 0$.

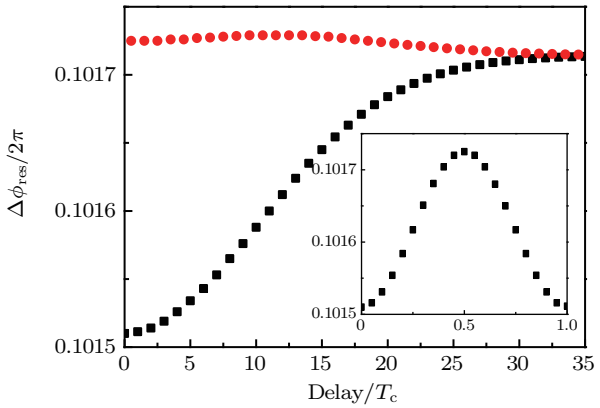


Fig. 2. (color online) The resonant CEP shifts as a function of the envelope delay τ_d from 0 to $35T_c$. $\Delta\phi_{res}/2\pi$ at $\tau_d = nT_c$ are represented with red circles and those at $\tau_d = (n+1/2)T_c$ are represented with black squares. The resonant CEP shifts as a function of τ_d from 0 to T_c are shown in the inset. The other parameters are the same as those in Fig. 1.

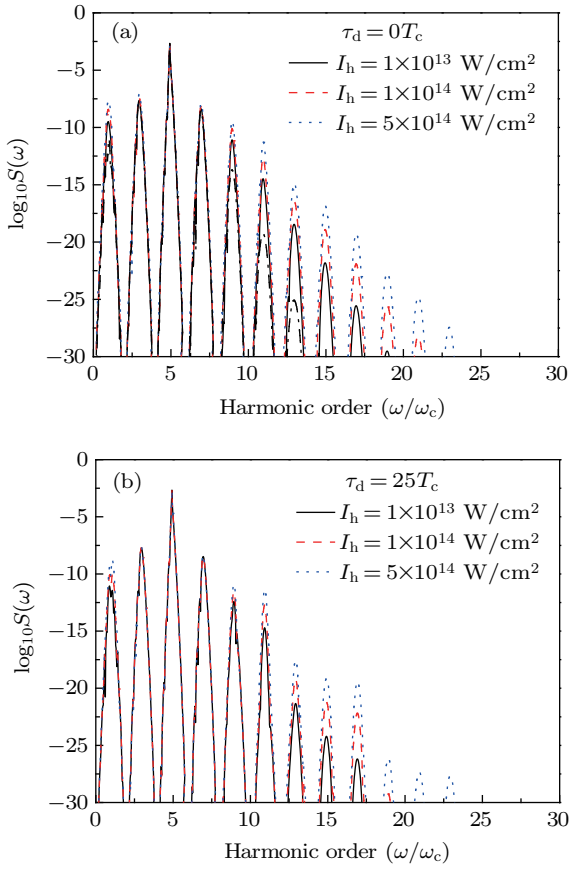


Fig. 3. (color online) HHG power spectra with (a) $\tau_d = 0$ and (b) $\tau_d = 25T_c$ by varying the peak intensity of the third-harmonic frequency-comb field. For the achievement of multiphoton resonance, (a) the CEP shift is tuned to $\Delta\phi/2\pi = 0.098788$ for $I_h = 1\times10^{13}$ W/cm^2 (black solid line), 0.101534 for $I_h = 1\times10^{14}$ W/cm^2 (red dash line), and 0.113857 for $I_h = 5\times10^{14}$ W/cm^2 (blue dotted line), and (b) the CEP shift is tuned to $\Delta\phi/2\pi = 0.098823$ for $I_h = 1\times10^{13}$ W/cm^2 (black solid line), 0.101703 for $I_h = 1\times10^{14}$ W/cm^2 (red dash line), and 0.114485 for $I_h = 5\times10^{14}$ W/cm^2 (blue dotted line). For comparison, the spectrum with $I_h = 0$ is presented wit the black dash-dotted line in (a). The other parameters are the same as those in Fig. 1.

Besides the quasienergy structure, the harmonic spectra can also be modulated by varying the envelope delay. Owing to the coupling of the third-harmonic driving field, additional multiphoton transition paths involving both fundamental- and third-harmonic photons are opened, and they contribute to the HHG power spectra, as shown in Fig. 3(a). Figures 3(a) and 3(b) present the HHG power spectra driven by the frequency-comb laser fields with varying peak intensity of the third-harmonic driving field at $\tau_d = 0$ and $25T_c$, respectively. Note that, for different values of I_h and τ_d, the CEP shifts are all tuned to achieve the multiphoton resonances. One can see that the spectral intensities of the harmonics are enhanced with increasing I_h. Generally, it is due to the strengthen of the multiphoton transition paths involving both fundamental- and third-harmonic photons. However, the enhancing feature of the case with $\tau_d = 0$ is quite different from that with $\tau_d = 25T_c$. For the case of $\tau_d = 25T_c$, the enhancement factors of specific harmonics 11th and 17th are larger than those of the others, and multiple plateaus are formed in the spectrum. It suggests that the overlap in the time domain between pulses from the two driving fields impacts the harmonic spectra in the frequency domain, and this should be related to the opened multiphoton-transition paths.

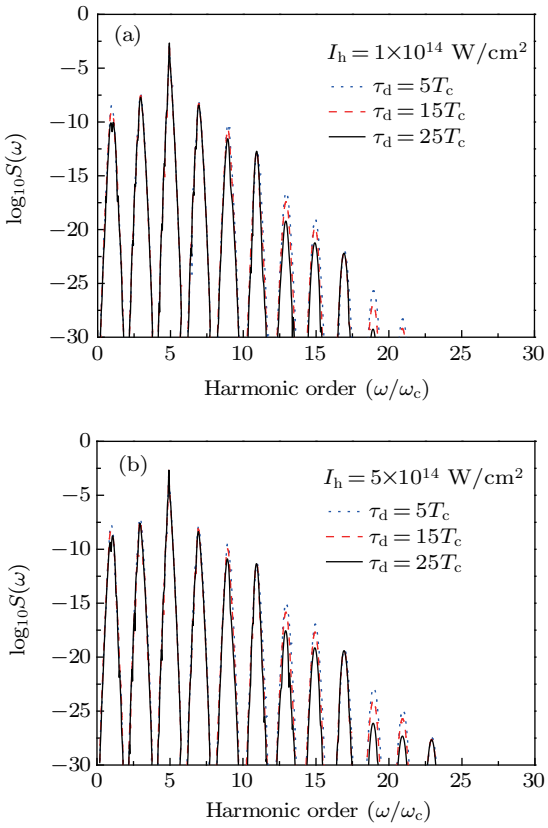


Fig. 4. (color online) HHG power spectra with (a) $I_h = 1\times10^{14}$ W/cm^2 and (b) $I_h = 5\times10^{14}$ W/cm^2 by varying the envelope delay τ_d. In each plot, the spectra with $\tau_d = 5T_c$, $15T_c$, and $25T_c$ are presented by black dash-dotted line, red dash line, and blue solid line, respectively. In panel (a), the CEP shift is tuned to $\Delta\phi/2\pi = 0.101534$ for $\tau_d = 5T_c$, 0.101645 for $\tau_d = 15T_c$, and 0.101703 for $\tau_d = 25T_c$ for the achievement of multiphoton resonance. In panel (b), the CEP shift is tuned to $\Delta\phi/2\pi = 0.113857$ for $\tau_d = 5T_c$, 0.114277 for $\tau_d = 15T_c$, and 0.114485 for $\tau_d = 25T_c$ for the achievement of multiphoton resonance. The other parameters are the same as those in Fig. 1.

For further investigating the dependence of the spectra on the envelope delay, the HHG power spectra by varying the envelope delays τ_d with $I_h = 1\times10^{14}$ W/cm^2 and 5×10^{14} W/cm^2 are calculated and presented in Figs. 4(a) and 4(b), respectively. For each case, the CEP shift is tuned to achieve multiphoton resonances. One can see that, as τ_d is varied from $5T_c$ to $25T_c$, the spectral intensities of the 11th and 17th harmonics are nearly unchanged, while those of the 9th, 13th, 15th, and 19th harmonics are suppressed. Considering the multiphoton transition paths under the multiphoton resonant condition, the electron is firstly promoted to the excited level via multiphoton resonant transition. After that, it absorbs additional multiple photons and transits back to the ground level, emitting a harmonic photon. The paths involving both fundamental- and third-harmonic photons are dependent on the envelope delay, since the multiphoton transition probabilities are determined by the field intensities in the overlap parts.

For instance, the 11th harmonic is mainly contributed by three multiphoton-transition paths taking the excited level as

the intermediate state, the first one via the absorption of 6 fundamental-harmonic photons, the second one via the absorption of 2 third-harmonic photons, and the third one via the absorption of 3 fundamental-harmonic photons and 1 third-harmonic photon. The first two paths are the main contribution and they are independent of the envelope delay, while the third path is dependent on the envelope delay. The 13th harmonic is also mainly contributed by three paths, the first one via absorption of 8 fundamental-harmonic photons, the second one via the absorption of 5 fundamental-harmonic photons and 1 third-harmonic photon, and the third one via the absorption of 2 fundamental-harmonic photons and 2 third-harmonic photons. However, only the first path is independent of the envelope delay. As a result, the spectral intensities of the 11th and 13th harmonics show different modulation behaviors as varying the envelope delay.

4. Conclusion

We present a theoretical investigation of the coherent control of the quasienergy structure and HHG power spectra driven by two frequency-comb fields via interference of multiphoton transition paths by tuning the relative envelope delay between the fields. We consider the case of two frequency-comb fields with the same repetition frequency and the carrier frequencies of fundamental- and third-harmonic frequencies. For accurate treatment of the interaction between a quantum system and the frequency-comb fields, the many-mode Floquet theory is employed. Multiphoton transition paths involving both fundamental- and third-harmonic photons are opened, and they can interfere with each other when the CEP shifts satisfy the matching condition. It is found that, not only the HHG power spectra, but also the quasienergy structure of the driven system can be coherently controlled. Our calculation results show that the quasienergies of the driven system can be tuned by varying the relative envelope delay when the pulses of the two fields overlap with each other. Meanwhile, owing to the dependence of the multiphoton transition paths on the relative envelope delay, the spectral intensities of the generated harmonics show different modulation behaviors when varying the relative envelope delay. Note that the effects of ionization and decay of levels, which are not included in this study of closed two-level system, also impact the quasienergy structure. And the study of realistic atomic systems including these effects is in process.

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