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Phase-dependent quantum interference between different pathways in bichromatic harmonic generation*

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This paper studies the harmonic generation of the hydrogen atom subjected to a collinear bichromatic laser field by numerically solving the time-dependent Schrödinger equation using the split-operator pseudo-spectral method. By adding a frequency variation to the additional field, the contributions of different pathways to particular order harmonic generation can be isolated. The quantum interference pattern between harmonic pathways, which influences the harmonic intensity, is found to be either constructive or destructive with respect to different relative phase of the two field components. Detailed description of up to the 35th-order harmonics and the harmonic pathways for a wide range of field parameters is presented.

Keywords: harmonic pathway, quantum interference, split-operator pseudo-spectral method
PACC: 3280K, 4265M, 3210A

1. Introduction

High-order harmonic generation (HHG) as a nonlinear laser-atom interaction process has been studied both theoretically and experimentally.[1] Efficient source of coherent light reaching the extreme ultraviolet frequency region which cannot be directly accessed by laser can be obtained through harmonic generation. Another intriguing application of HHG is its feasibility in attosecond pulse generation.[2,3] Semiclassical three-step model[4] gives out even quantitative explanation of the HHG processes and confirms the main characteristics of harmonic spectrum such as the broad plateau and the cut-off at the end of the plateau. In this model the electron is first ionized, and then excurses like a free electron under the influence of the laser field. When the ionized electron reaches the vicinity of the nucleus, it can recombine with the parent ion and emit a radiation. Lewenstein *et al*[5] proposed the fully quantum-mechanical version of three-step process using strong field approximation. However, there are harmonics generated at the lower end of the plateau which can not be explained by the three-step model because the harmonic energy can be lower than the ionization potential ($E_{\rm photon} < I_{\rm p}$). Comparing the contributions of bound-bound and bound-continuum transitions to the dipole acceleration, it has been revealed that the bound-bound transitions play the dominant role in lower energy harmonic generation.[6,7]

In our recent paper,[8] we calculated the harmonic generation when the hydrogen atom subjected to a collinearly polarized bichromatic field and provided the pathway analysis of the harmonic generation. We showed that the relative intensity of the two field components determines the harmonic pathway. The harmonic peak can be clearly observed in the spectrum when the leading pathway can be distinguished from other possible pathways, but when two harmonic pathways contribute equally, the destructive interference between different pathways decreases the harmonic intensity and can even lead to a "harmonic-collapse" phenomenon. In this paper, we extend our study of harmonic pathway by including the phase-dependent effect. We focus on the interference pattern between different pathways of the individual harmonic generation. The dependence of the harmonic intensity on the field parameters can be well understood through the pathway analysis.

2. Method

Direct numerical solutions to the time-dependent Schrödinger equation have been discussed in a lot of papers.[9−13] In this paper, we use the split-operator[9] pseudo-spectral method[10] to get the time-dependent

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wave function of a hydrogen atom which is subjected to the bichromatic field. The accuracy of this method has been demonstrated by the well agreement of the HHG power spectra in length and acceleration forms.[10] In our calculation, the harmonic spectrum is obtained from the Fourier transformation of the time-dependent dipole moments of the acceleration form

$$P(\omega) = \left| \frac{1}{t_f - t_i} \frac{1}{\omega^2} \int_{t_i}^{t_f} d_a(t) \mathrm{e}^{-\mathrm{i}\omega t} \right|^2 , \quad (1)$$

where the dipole moments are derived by

$$d_a(t) = \langle \psi(\boldsymbol{r}, t)| - \frac{z}{r^3} + E(t) \, |\psi(\boldsymbol{r}, t)\rangle \, . \quad (2)$$

The bichromatic laser field can be described as a time-dependent electric field

$$E(t) = E_0 \sin^2 \left(\frac{\pi}{\tau_p} t \right) [\sin(\omega_1 t) + \beta \sin(\omega_2 t + \phi)], \quad (3)$$

where τ_p is the pulse duration, the $\sin^2$ part describes the temporal envelope of the pulse, β is the amplitude ratio of the additional field to the fundamental field, and ϕ is the relative phase. The frequencies of the fundamental field and the additional field are $\omega_1 = 3.0\omega_0$ and $\omega_2 = 5.0\omega_0$, respectively, where $\omega_0 = 0.0143$ a.u. The pulse duration is $\tau_p = 20 \times \dfrac{2\pi}{\omega_0}$, which means 60 periods of the fundamental field.

In order to isolate the contributions of different pathways,[8] we add a frequency variation to the additional field,

$$\omega_2' = \omega_2 + \Delta\omega, \quad (4)$$

where $\Delta\omega$ is much smaller than ω_0. In the power spectrum, the n-th-order harmonic peak may have a frequency shift of k times of $\Delta\omega$,

$$\Omega_n' = \Omega_n + k \cdot \Delta\omega, \quad (5)$$

where Ω_n' and Ω_n are the harmonic frequencies with and without the frequency variation of the additional field, respectively, while k is an integer number. Then one can deduce that the additional field contributes k photons to this order harmonic generation and the n-th-order harmonic pathway can be written as

$$H + m\gamma(3\hbar\omega_0) + k\gamma(5\hbar\omega_0) \rightarrow H + \gamma(n\hbar\omega_0), \quad (6)$$

where $3m + 5k = n$, and $m + k$ is an odd number.

3. Results and discussions

Phase-dependent effect in harmonic generation has been studied both theoretically[14–16] and experimentally.[17] Here we investigate the phase modulation of harmonic generation by concerning the quantum pathway of the harmonics. Figure 1 is the intensities of the 13th-, 15th-, and 29th-order harmonics for ϕ varying from 0 to 2π, while the field amplitude is $E_0 = 0.045$ a.u. The amplitude ratio is set to be $\beta = 0.35$ (solid symbols) and $\beta = 0.42$ (open symbols), respectively. A quasi-period of $\dfrac{2}{3}\pi$ of the harmonic intensity variation can be seen in Fig.1. This is caused by the symmetry of the field, for when ϕ is changed to $\phi + \dfrac{2\pi}{3}$, the two-colour field without the pulse envelope can be written as $\sin\left[3\omega_0\left(t - \dfrac{2\pi}{3\omega_0}\right)\right] + \beta \sin\left[5\omega_0\left(t - \dfrac{2\pi}{3\omega_0}\right) + \phi\right]$, it means a time translation of the field. In the case of $\beta = 0.35$, the maximum intensity of 15th harmonics (solid cycles) for different relative phases appears at $\phi = \dfrac{1}{3}\pi$ and it is two orders of magnitude higher than the minimum intensity which appears at $\phi = 0$. Similar profound phase modulation can be observed for the 29th harmonic (open triangles) when $\beta = 0.42$, and its maximum and minimum appear at $\phi = \dfrac{1}{2}\pi$ and $\phi = \dfrac{1}{6}\pi$, respectively. The 15th harmonic (open cycles) shows lower fluctuation when $\beta = 0.42$, and an offset phase of $\Delta\phi$ can be observed when the additional field intensity changes. However, the 13th harmonic intensity (solid squares) only shows a slight fluctuation with respect to different relative phase.

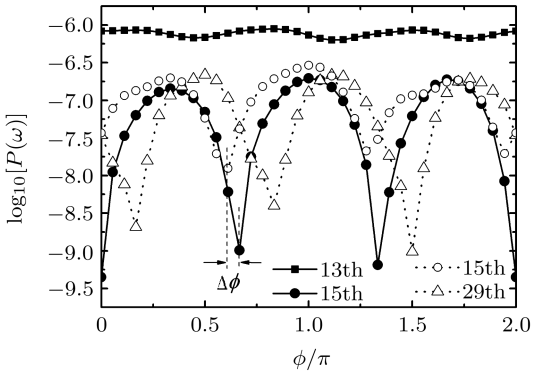


Fig.1. The harmonic intensity as functions of the relative phase. The squares, circles, and triangles correspond to the 13th-, 15th-, and 29th-order harmonic, respectively. The solid symbols connected with solid lines are for the relative amplitude ratio $\beta = 0.35$, while the open symbols connected with dotted lines are for the case $\beta = 0.42$. The laser field amplitude is set to be $E_0 = 0.045$ a.u.

In Fig.2, we plot the power spectra scale from $14\omega_0$ to $16\omega_0$ for $\phi = \dfrac{1}{3}\pi$ with respect to the ampli-

tude ratio (a) $\beta = 0.25$, (b) $\beta = 0.35$ and (c) $\beta = 0.42$. The additional field frequency is set to be $\omega_2 = 5.0\omega_0$ (solid lines) and $\omega_2 = 5.1\omega_0$ (dotted lines) for comparison to clearly show the 15th harmonic pathway. Same as that in the case $\phi = 0$,[8] two different pathways, i.e. $\{5\times3\omega_0\}$ and $\{3\times5\omega_0\}$, contribute equally to the 15th harmonic when $\beta = 0.35$, and the leading pathway of 15th harmonic changes with the increase of additional field intensity. But in the case $\phi = 0$ the "harmonic collapse" phenomenon takes place, while for $\phi = \frac{1}{3}\pi$ the 15th harmonic peak can still be observed when $\omega_2 = 5.0\omega_0$ and its intensity is larger than the two peaks in the spectrum of $\omega_2 = 5.1\omega_0$. It means that there are constructive interferences between different pathways for $\phi = \frac{1}{3}\pi$. The phase dependence of the harmonic intensity in Fig.1 should be attributed to different interference pattern for the variation of the relative phase of the two field components. Distinct phase modulation of the harmonic intensity can be observed when two quantum pathways contribute equally to a particular order harmonic; on the other hand, the harmonic intensity is less phase-sensitive when one pathway becomes dominant. For example, the pathway $\{3 \times 5\omega_0\}$ prevails against the pathway $\{5 \times 3\omega_0\}$ in the 15th harmonic generation when $\beta = 0.42$, as can be seen in Fig.2(c), so that in Fig.1, the intensity of 15th harmonic (open circles) shows lower fluctuation than that in the case $\beta = 0.35$. The slight fluctuation of 13th harmonic intensity can also be understood, for the secondary pathway is not comparable to the leading pathway in this order harmonic generation as has been shown in Ref.[8].

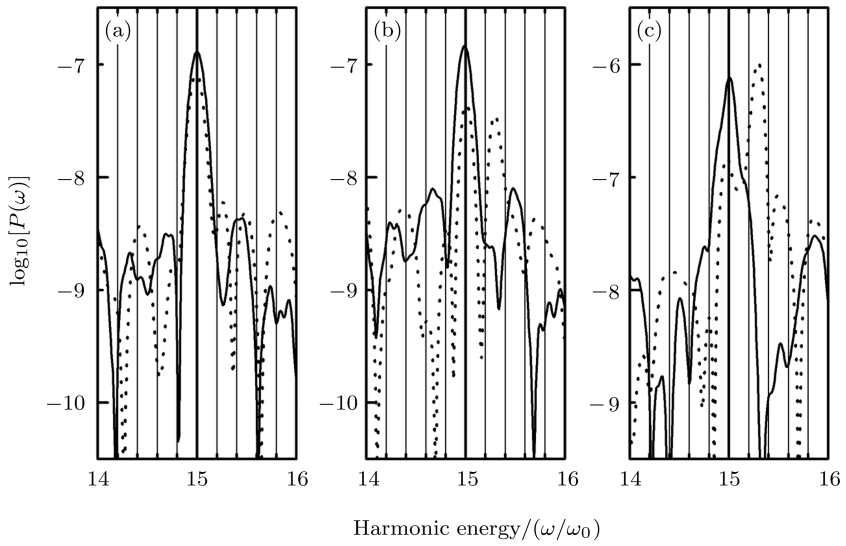


Fig.2. Harmonic power spectra for $\phi = \frac{1}{3}\pi$ with $\omega_2 = 5.0\omega_0$ (solid line) and $\omega_2 = 5.1\omega_0$ (dotted line), respectively. The relative amplitude ratio is set as (a) $\beta = 0.25$, (b) $\beta = 0.35$ and (c) $\beta = 0.42$. Other parameters are the same as those in Fig.1.

Figure 3 provides the individual harmonic intensity as functions of the relative amplitude ratio of the two field components. The intensity of the 15th harmonic (solid cycles) for $\phi = \frac{1}{3}\pi$ and the 29th harmonic (solid triangles) for $\phi = \frac{1}{2}\pi$ shows a monotonic increase for the increasing additional field intensity. This is coincident with Fig.1, for though a small offset phase of $\Delta\phi$ exists for the variation of the additional field intensity, the interference pattern of 15th and 29th pathways can remain to be constructive for $\phi = \frac{1}{3}\pi$ and $\phi = \frac{1}{2}\pi$. However, the phase-dependent line in Fig.1 is much steeper when it reaches the minimum, hence the destructive interference is strongly affected by the relative phase. This can be seen from the difference of 29th harmonic generation for the case of $\phi = \frac{2}{9}\pi$ (solid line with open triangles), and $\phi = \frac{1}{6}\pi$

(dotted line with open triangles) in Fig.3. The "harmonic collapse" phenomenon takes place in the 29th harmonic generation when $\beta = 0.46$, $\phi = \frac{2}{9}\pi$, while for $\phi = \frac{1}{6}\pi$, the minimum intensity of the 29th harmonic which appears at $\beta = 0.41$ is an order of magnitude larger than the former.

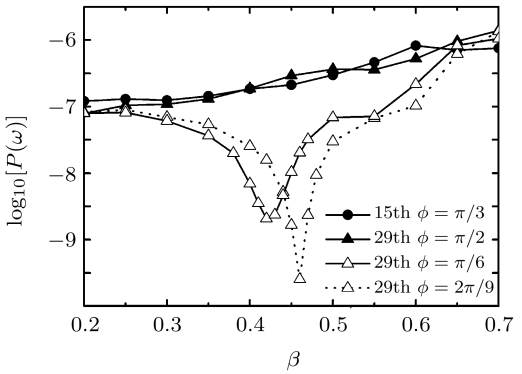


Fig.3. The harmonic intensity as functions of the relative amplitude ratio of the two field components. The circles and triangles are the 15th harmonic and the 29th harmonic, and the relative phase is set to be $\phi = \frac{1}{3}\pi$, $\phi = \frac{1}{2}\pi$, $\phi = \frac{1}{6}\pi$, and $\phi = \frac{2}{9}\pi$, respectively. Other parameters are the same as those in Fig.1.

It is convenient to use pathway analysis to study the harmonic intensity for the variation of the field parameters. In Table 1, we show the pathways of up to the 35th-order harmonics generation with the fundamental field kept fixed and the relative amplitude ratio varies from 0 to 0.7. Pathway I and Pathway II are the possible pathways of harmonic generation. The fourth column in Table 1 is the amplitude ratio where the two harmonic pathways have equal contributions and the fifth column is the relative phase where the interference pattern is destructive and the "harmonic collapse" phenomenon can be observed in the power spectrum. The harmonic pathway is mostly determined by the relative intensity between the two field sources. It is equivalent for the electron to absorb 5 photons of $3\omega_0$ from the fundamental field or 3 photons of $5\omega_0$ from the additional field to obtain a harmonic energy of $15\omega_0$, while the angular momentum conservation can still be fulfilled. As the additional field becomes stronger, the Pathway II which contains more photons of $5\omega_0$ becomes dominant. For example, two difference frequency mixing pathways contribute equally to the 7th order harmonic when $\beta = 0.14$, and the destructive interference takes place when $\phi = \frac{11}{18}\pi$. The leading pathways of the 9th-, 11th-, and 17th-order harmonics remain to be unchanged when the amplitude ratio varies from 0 to 0.7. To these order harmonics, the Pathway II in Table 1 requires the additional field to be much stronger than the fundamental one. But the stronger additional field intensity ($\beta > 0.7$) will allow the electrons easier tunnelling, ionization and the depletion of the bound states will decrease the lower-order harmonic intensity.

Table 1. Pathway of the harmonics from the 1st order to the 35th order. The laser field parameters are chosen as: $E_0 = 0.045$ a.u., $\omega_1 = 3.0\omega_0$ ($\omega_0 = 0.0143$ a.u.), $\omega_2 = 5.0\omega_0$, and the pulse duration $\tau_p = 20 \times \frac{2\pi}{\omega_0}$.

harmonic frequency	pathway I	pathway II	β	ϕ
$7\omega_0$	$4\times3\omega_0 - 1\times5\omega_0$	$2\times5\omega_0 - 1\times3\omega_0$	0.14	$11\pi/18$
$9\omega_0$	$3\times3\omega_0$	$3\times5\omega_0 - 2\times3\omega_0$	-	-
$11\omega_0$	$2\times3\omega_0 + 1\times5\omega_0$	$4\times5\omega_0 - 3\times3\omega_0$	-	-
$13\omega_0$	$6\times3\omega_0 - 1\times5\omega_0$	$1\times3\omega_0 + 2\times5\omega_0$	0.06	$\pi/2$
$15\omega_0$	$5\times3\omega_0$	$3\times5\omega_0$	0.35	0
$17\omega_0$	$4\times3\omega_0 + 1\times5\omega_0$	$4\times5\omega_0 - 1\times3\omega_0$	-	-
$19\omega_0$	$8\times3\omega_0 - 1\times5\omega_0$	$3\times3\omega_0 + 2\times5\omega_0$	0.02	$5\pi/9$
$21\omega_0$	$7\times3\omega_0$	$2\times3\omega_0 + 3\times5\omega_0$	0.20	$\pi/9$
$23\omega_0$	$6\times3\omega_0 + 1\times5\omega_0$	$1\times3\omega_0 + 4\times5\omega_0$	0.47	$\pi/18$
$25\omega_0$	$10\times3\omega_0 - 1\times5\omega_0$	$5\times5\omega_0$	0.13	$5\pi/18$
$27\omega_0$	$9\times3\omega_0$	$4\times3\omega_0 + 3\times5\omega_0$	0.21	0
$29\omega_0$	$8\times3\omega_0 + 1\times5\omega_0$	$3\times3\omega_0 + 4\times5\omega_0$	0.42	$2\pi/9$
$31\omega_0$	$12\times3\omega_0 - 1\times5\omega_0$	$7\times3\omega_0 + 2\times5\omega_0$	0.05	$\pi/6$
$33\omega_0$	$11\times3\omega_0$	$6\times3\omega_0 + 3\times5\omega_0$	0.18	$11\pi/18$
$35\omega_0$	$10\times3\omega_0 + 1\times5\omega_0$	$5\times3\omega_0 + 4\times5\omega_0$	0.37	$5\pi/9$

4. Conclusion

In conclusion, we have presented theoretical study of the harmonic pathway in bichromatic field for different relative intensity and relative phase. The two collinearly polarized field components with frequency ratio 3:5 provide a wide range of harmonics which are generated by the bound-bound transitions. As has been analysed in our paper, the harmonic pathway plays an important role in the lower order harmonic generation. The harmonic pathway is mainly determined by the relative intensity, while the interference pattern between two different pathways has critical dependence on the relative phase. The harmonic intensity becomes strongly phase-sensitive when two pathways contribute equally to a particular order harmonic generation. For a fixed relative phase, the harmonic intensity increases monotonically for the increasing additional field intensity if the harmonic's leading pathway keeps unchanged or there is constructive interference between two different pathways, but when the interference pattern changes to be destructive, the harmonic intensity is decreased and can even lead to a "harmonic collapse" phenomenon. As can be seen in Table 1, the relative intensity where two different pathways have comparable contributions and the relative phase where these two pathways have destructive interference varies for different-order harmonic generation, so the intensity variation of each order harmonic for different field parameters (β, ϕ) should be analysed individually.

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