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Phase jump in resonance harmonic emission driven by strong laser fields*

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We present a theoretical investigation of the multiphoton resonance dynamics in the high-order-harmonic generation (HHG) process driven by a strong driving continuous wave (CW) field along with a weak control harmonic field. The Floquet theorem is employed to provide a nonperturbative and exact treatment of the interaction between a quantum system and the combined laser field. Multiple multiphoton-transition paths for the harmonic emission are coherently summed. The phase information about paths can be extracted via the Fourier transform analysis of the harmonic signals which oscillate as a function of the relative phase between driving and control fields. Phase jumps are observed when sweeping across the resonance by varying the frequency or intensity of the driving field. The phase variation as a function of driving frequency at a fixed intensity and as a function of the intensity at a fixed driving frequency allows us to determine the intensity dependence of the transition energy of quantum systems.

Keywords: phase jump, high-order harmonic generation, Stark-shifted transition energy

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1. Introduction

The interaction between matter and strong laser field has been a subject of intensive studies for decades, owing to both fundamental and applied interest. High-order harmonic generation (HHG) driven by strong laser fields provides a source of coherent ultrashort pulses in the extreme ultraviolet (XUV).[1–4] It allows the investigation of atomic or molecular dynamics driven by external fields with femtosecond even attosecond resolution,[5–15] such as, the light-induced states of atoms driven by laser pulses,[8,9] Autler–Townes splitting effects,[10] polarization in the continuum,[11] and Auger decay and tunneling ionization processes.[12] The process of the high-order-harmonic generation from atoms or molecules can be understood by the semiclassical three-step model:[16] an electron is ionized, and accelerated in the laser field, then returns to its parent ion, giving rise to the emission of harmonic photons. The effect of resonance on the HHG process has been employed in several approaches for the enhancement of HHG spectral efficiency.[17–28] It is shown that, the bound-bound resonances can influence the ionization step in high-order-harmonic generation,[27,28] while the resonance in continuum can enhance the harmonic emission via autoionization states.[25,29] Beaulieu *et al.* investigate the roles of two different resonances in high-order-harmonic generation and demonstrate the temporal properties of these two XUV emission mechanisms.[30]

Besides the enhancement of the high-order-harmonic generation, resonances also can induce spectral phase jumps in the harmonic emission when sweeping across the resonance.[31,32] The significant distortions of the phase of near-resonant harmonic can be employed to extract structural and dynamic information about the generating system. Haessler *et al.* showed that the resonance considerably changes the relative phase of neighboring harmonics emitted from a tin plasma.[31] Ferré *et al.* demonstrated a method for the identification and isolation of the different channels in multi-channel high-order-harmonic generation in polyatomic molecules via the phase jump across the resonant harmonic emission.[32] The key to obtain the phase information of harmonic emission is to measure the response of the system to additional control parameters. The above two works both employ the "reconstruction of attosecond bursts by interference of two-photon transition" (RABBIT) technique,[33] in which a target gas is photonionized by the harmonic radiation with a weak controllable fundamental infrared laser field. The sideband in photonelectron spectrum oscillates as a function of delay between harmonic and fundamental fields, due to the interference between two quantum paths those corresponding to absorption of one harmonic and absorption or emission of an infrared photon. Thus the oscillation of sideband in photonionization spectrum driven by the combination of harmonics

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allows us to obtain the phase information of the harmonic radiation, unfolding the structural and dynamic information about the target. Besides the photonionization spectrum, HHG spectrum also carries the phase information, and it would be better if the phase information can be directly extracted from the HHG spectrum.

In this paper, we theoretically present an alternative phase measurement of resonant harmonic emission driven by the combination of strong driving fundamental and weak control harmonic fields. The HHG process is calculated by employing the Floquet theory[34] for the nonperturbative treatment of the interaction between intense laser fields and atomic systems. The control harmonic field opens multiple multiphoton-transition paths that connect the same initial and final states. And the relative phases between paths can be extracted via Fourier Transform analysis of the HHG spectra as a function of the relative phase between the driving and control fields. Our numerical calculation shows that, phase jumps occur when sweeping across the resonance by varying the frequency or intensity of the driving field. The phase variation as a function of the frequency and intensity of the driving field allows us to determine the intensity dependence of the transition energy.

The rest of this paper is organized as follows. In Section 2, we present the Floquet Theory for the treatment of the quantum systems driven by strong driving fundamental and weak control harmonic fields. In Section 3, we study the phase jump of harmonic emission as varying the frequency and intensity of the driving field. This is followed by the conclusion in Section 4.

2. Theoretical method

We consider a quantum system driven by a strong cw laser field and its high-order harmonic fields generated from a train of ultrashort pulses in the time domain. The total electric field can be expressed as

$$E_{\rm t}(t) = E_{\rm d}\,{\rm e}^{{\rm i}\omega_{\rm d}t} + \sum_{k=-N}^{N} E_k\,{\rm e}^{{\rm i}\omega_k t}\,{\rm e}^{{\rm i}\phi} + {\rm c.c.}, \tag{1}$$

where $E_{\rm d}$ and $\omega_{\rm d}$ are the amplitude and frequency of the driving laser field, respectively, E_k are the amplitudes of harmonic fields with frequency components $\omega_k = \omega_0 + k\omega_{\rm r}$, and ϕ is the relative phase between the driving and harmonic fields. The repetition frequency $\omega_{\rm r}$ of the pulse train is $\omega_{\rm r} = 2\omega_{\rm d}$, and the offset frequency is $\omega_\delta = \omega_{\rm d}$. In the numerical calculation, N is chosen to ignore harmonics with amplitudes less than 1×10^{-10} a.u. (corresponding to an intensity of 3.51×10^{-4} W/cm^2). Then the Hamiltonian is given by

$$\begin{aligned}\hat{H}(\boldsymbol{r},t) &= \hat{H}_0(\boldsymbol{r}) - \mu(\boldsymbol{r})\cdot\boldsymbol{E}_{\rm d}\cos\omega_{\rm d}t - \sum_{k=-N}^{N}\mu(\boldsymbol{r})\cdot\boldsymbol{E}_k\cos\omega_k t \\ &= \hat{H}_0(\boldsymbol{r}) - \frac{1}{2}\mu_z E_{\rm d}[{\rm e}^{{\rm i}\omega_{\rm d}t} + {\rm e}^{-{\rm i}\omega_{\rm d}t}] - \sum_{k=-N}^{N}\frac{1}{2}\mu_z E_k[{\rm e}^{{\rm i}(\omega_0+k\omega_{\rm r})t}\,{\rm e}^{{\rm i}\phi} + {\rm e}^{-{\rm i}(\omega_0+k\omega_{\rm r})t}\,{\rm e}^{-{\rm i}\phi}],\end{aligned} \tag{2}$$

where $\hat{H}_0(\boldsymbol{r})$ is the unperturbed Hamiltonian of the atomic or molecular system, $\mu(\boldsymbol{r})$ is the electric dipole moment operator and μ_z is the component parallel to the polarization axis.

To investigate the interaction of an atomic system with the pulse train, the many-mode Floquet theory (MMFT)[34–36] and the generalized pseudospectral (GPS) method can be employed to solve the quasiperiodic time-dependent Schrödinger equation with the Hamiltonian (2), which is converted into an equivalent time-independent generalized Floquet matrix eigenvalue problem. In our case, there are frequency components $\omega_{\rm d}$, ω_δ, and $\omega_{\rm r}$, while there is only one independent frequency $\omega_{\rm d}$. As a result, a one-mode Floquet matrix eigenvalue equation can be constructed as

$$\sum_{\beta}\sum_{l'}\langle\alpha l|H_F|\beta l'\rangle\langle\beta l'|\lambda\rangle = \lambda\langle\alpha l|\lambda\rangle, \tag{3}$$

where the basis vectors $|\alpha l\rangle = |\alpha\rangle\otimes|l\rangle$ are employed, and λ is the quasienergy eigenvalues and $|\lambda\rangle$ is the corresponding eigenvetor. The one-mode Floquet matrix $H_{\rm F}$ can be constructed by

$$\langle\alpha l|H_{\rm F}|\beta l'\rangle = H_{\alpha\beta}^{[l-l']} + l\omega_{\rm d}\delta_{\alpha,\beta}\delta_{l,l'}, \tag{4}$$

with

$$\begin{aligned}H_{\alpha,\beta}^{[l-l']} &= \varepsilon_\alpha\delta_{\alpha,\beta}\delta_{l,l'} + V_{\alpha,\beta}(\delta_{l+1,l'} + \delta_{l-1,l'}) \\ &\quad + \sum_{k=-N}^{N} U_{\alpha,\beta}^{(k)}(\delta_{l+k',l'}\,{\rm e}^{{\rm i}\phi} + \delta_{l-k',l'}\,{\rm e}^{-{\rm i}\phi}),\end{aligned} \tag{5}$$

where

$$\begin{aligned}&k' = [(\omega_{\rm c}-\omega_\delta)/\omega_{\rm r}] + 1, \qquad \varepsilon_\alpha = \langle\alpha|\hat{H}_0|\alpha\rangle, \\ &V_{\alpha,\beta} = -(1/2)E_{\rm d}\langle\alpha|\mu_z|\beta\rangle, \quad U_{\alpha,\beta}^{(k)} = -(1/2)E_k\langle\alpha|\mu_z|\beta\rangle,\end{aligned}$$

and ϕ is the relative carrier phase between the driving and harmonic fields. More details of the structure of $H_{\rm F}$ can be found in Refs. [37–39].

Solving the matrix eigenvalue problem (3) with the Floquet matrix (4), we can obtain a set of quasienergies $\lambda_{\gamma l}$

and the corresponding eigenvectors $|\lambda_{\gamma l}\rangle$ which satisfy the orthonormality condition. And the harmonic generation spectra can be expressed by

$$S(\omega_l) = \frac{4}{6\pi c^3 \omega_l^4}|\boldsymbol{d}_l|^2 = \frac{4}{6\pi c^3 \omega_l^4}|\sum_{\alpha,\beta}\sum_{l'}\langle\lambda_{\alpha,l'-l}|\mu_z|\lambda_{\beta,l'}\rangle|^2, \tag{6}$$

in which each harmonic element $\omega_l = l\omega_{\rm d}$.

3. Results and discussion

In this section, we investigate the phase jump of multiphoton resonant in atomic Hydrogen driven by a strong CW driving field and its harmonic control field corresponding to a train of ultrashort pulses. A case study focusing $1s\rightarrow 2p$ transition of atomic hydrogen is presented. The frequency of the driving field $\omega_{\rm d}$ is changed between 2.035 eV and 2.1 eV, while its intensity is changed from 0.1 $\mathrm{W\cdot cm^{-2}}$ to 7×10^{12} $\mathrm{W\cdot cm^{-2}}$. The control field is generated from a train of Gaussian pulses with carrier frequency $\omega_{\rm c} = 5\omega_{\rm d}$, repetition frequency $\omega_{\rm r} = 2\omega_{\rm d}$, peak intensity 1×10^{-9} $\mathrm{W\cdot cm^{-2}}$, and 1.5-fs full width at half maximum (FWHM). In our calculation, components with amplitudes less than 1×10^{-10} a.u. are ignored, thus only components with $\omega_k = 3\omega_{\rm d}$, $5\omega_{\rm d}$, and $7\omega_{\rm d}$ are included. The $1s\rightarrow 2p$ transition energy corresponds to the five-photon dominant resonance regime of the CW field ($5\omega_{\rm d}\approx\omega_{\rm res} = \lambda_{2p} - \lambda_{1s}$). According to the Floquet calculation, the quasienergy structure of the quasienergies can be represented by

$$\lambda_{\gamma m} = \lambda_\gamma + m\omega_{\rm d}, \tag{7}$$

where m is an integer and λ_γ are the energies of dressed states, which can be calculated by Eq. (3). The same quasienergy state can be populated via different dipole-transition paths, owing to different combinations of driving and control fields. And the relative phase ϕ between two fields allows us to coherently modulate the harmonic generation.[39]

Figures 1(a) and 1(b) present HHG spectra as a function of the relative phase between fields for two different driving frequencies $\omega_{\rm d} = 2.058$ eV and 2.064 eV, respectively. The intensity of the driving field is fixed at $I_{\rm d} = 1\times10^{12}$ $\mathrm{W\cdot cm^{-2}}$. Only the harmonics up to 11th-order, *i.e.* the so-called below-threshold harmonics, are presented. It is shown that, harmonic peaks strongly oscillate with the relative phase ϕ, and the oscillations of all harmonics depend on the driving frequency $\omega_{\rm d}$. The harmonic radiation can be understood as the dipole transition between quasienergy states, and the dependence of the harmonic spectra on the relative phase ϕ can be understood via the coherent superposition of multiphoton-transition paths.

The radiation signals can be expressed as

$$S(\omega_l) \propto |\sum_n \boldsymbol{d}_l^n|^2 = |\sum_n |\boldsymbol{d}_l^n| e^{i\phi_{\rm d}+i\phi_{\rm c}+i\varphi_l^n}|^2, \tag{8}$$

where $\boldsymbol{d}_l^n$ are the multiphoton transition probability amplitudes with n referring the photon number of the control field involved in the multiphoton-transition paths, and the phases of radiated harmonic fields consist of two parts: the phase term φ_l^n involved in the multiphoton-transition path, and the phases $\phi_{\rm d}$ and $\phi_{\rm c}$ of driving and control fields with relative phase $\phi = \phi_{\rm c} - \phi_{\rm d}$. In the following, the phase of the driving field $\phi_{\rm d}$ is set to be zero for convenient.

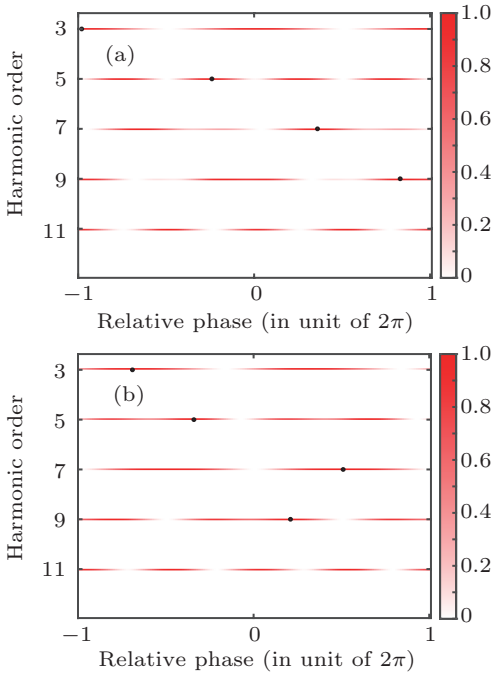


Fig. 1. HHG spectra as a function of the relative phase for $\omega_{\rm d} = 2.058$ eV (a) and $\omega_{\rm d} = 2.064$ eV (b). The positions of maximum intensities in 3rd, 5th, 7th, and 9th harmonics are labelled with red solid cycles.

In general, φ_l^n do not depend much on the photon energies, but change by π across the resonance if the multiphoton transition occurs via the resonant 2p state. A Fourier transform of the signals as a function of ϕ provides information on the multiphoton transition process, *e.g.* the relative phases between multiphoton transition paths which allows us to determine the AC Stark shift of the resonant transition energy between 1s and 2p states.

In the following, the radiated 5th- and 7th-harmonics are taken as examples for discussion. The 5th-harmonic radiation signal can be expressed as

$$\begin{aligned} S(\omega_5) &\propto ||\boldsymbol{d}_5^0| e^{i\varphi_5^0} + |\boldsymbol{d}_5^1| e^{i\varphi_5^1+i\phi} + |\boldsymbol{d}_5^2| e^{-i\varphi_5^2+i2\phi}|^2 \\ &\propto |\boldsymbol{d}_5^0|^2 + |\boldsymbol{d}_5^1|^2 + |\boldsymbol{d}_5^2|^2 + 2|\boldsymbol{d}_5^0\boldsymbol{d}_5^1|\cos(\varphi_5^0 - \varphi_5^1 - \phi) \\ &\quad + 2|\boldsymbol{d}_5^1\boldsymbol{d}_5^2|\cos(\varphi_5^1 + \varphi_5^2 - \phi) \\ &\quad + 2|\boldsymbol{d}_5^0\boldsymbol{d}_5^2|\cos\left[2\left(\frac{\varphi_5^0+\varphi_5^2}{2} - \phi\right)\right], \end{aligned} \tag{9}$$

in which φ_5^n are the phase terms involved in the paths $\boldsymbol{d}_5^n$, respectively. $\boldsymbol{d}_5^0$ represents the dipole transition path in which five photons from the driving field absorbed and none from the control field absorbed, $\boldsymbol{d}_5^1$ represents the paths in which one 3rd-, 5th-harmonic or 7th-harmonic photon from the control field absorbed, and $\boldsymbol{d}_5^2$ represents the path in which two 3rd-harmonic photons from the control field are absorbed and one photon is emitted with frequency being the same as those from the driving field. The phase term φ_5^2 is with opposite symbol due to the emission of the driving photon. All paths $\boldsymbol{d}_5^n$ occur via the 2p state, so all φ_5^n change by π across the resonance. The last term in Eq. (9) corresponds to the half-cycle oscillation of 5th-harmonic signal in Fig. 1, and its phase, *i.e.*, $(\varphi_0+\varphi_2)/2$, changes across the resonance. After the Fourier-transition analysis of the 5th-harmonic signal, the phase of the second-order term is labelled as $\Phi_5^2=(\varphi_0+\varphi_2)/2$. Figure 2(a) shows Φ_5^2 as a function of ω_d. As expected, a phase jump occurs around $\omega_\mathrm{d}=2.064$ eV, corresponding to the AC Stark shifted transition energy between 1s and 2p states.

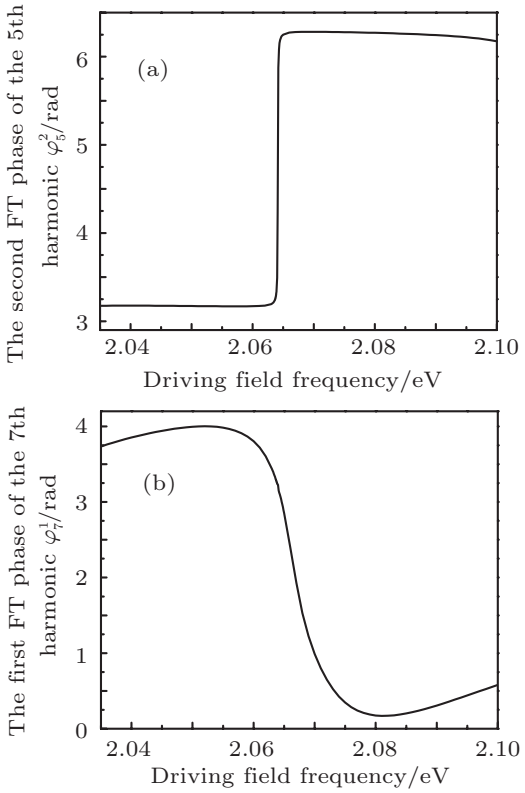


Fig. 2. (a) The phase of the 2nd-order term from Fourier-transition analysis of the 5th-harmonic as a function of ϕ, and (b) the phase of the 1st-order term from the Fourier-transition analysis of the 7th-harmonic as a function of ϕ, are plotted as varying the driving field frequency ω_d. The driving CW field intensity is $I_\mathrm{d}=1\times10^{12}$ W·cm^{-2}.

Meanwhile, the 7th-harmonic radiation can be written as

$$\begin{aligned}S(\omega_7)&\propto||\boldsymbol{d}_7^0|\mathrm{e}^{\mathrm{i}\varphi_7^0}+|\boldsymbol{d}_7^1|\mathrm{e}^{\mathrm{i}\varphi_7^1+\mathrm{i}\phi}+|\boldsymbol{d}_7^2|\mathrm{e}^{\mathrm{i}\varphi_7^2+\mathrm{i}2\phi}|^2\\&\propto|\boldsymbol{d}_7^0|^2+|\boldsymbol{d}_7^1|^2+|\boldsymbol{d}_7^2|^2+2|\boldsymbol{d}_7^0\boldsymbol{d}_7^2|\cos(\varphi_7^0-\varphi_7^2-2\phi)\\&\quad+2|\boldsymbol{d}_7^0\boldsymbol{d}_7^1|\cos(\varphi_7^0-\varphi_7^1-\phi)\\&\quad+2|\boldsymbol{d}_7^1\boldsymbol{d}_7^2|\cos(\varphi_7^1-\varphi_7^2-\phi),\end{aligned}\tag{10}$$

where $\boldsymbol{d}_7^0$ represents the dipole transition path in which seven photons from the driving field absorbed, $\boldsymbol{d}_7^1$ represents the paths in which one 3rd-, 5th- or 7th-harmonic photon from the control field absorbed, and $\boldsymbol{d}_7^2$ represents the path in which two 3rd-harmonic photons from the control field and one photon from the driving field absorbed. φ_7^n are the phase terms involved in the paths $\boldsymbol{d}_7^n$, respectively. Since the multiphoton-transition path $\boldsymbol{d}_7^2$ occurs not via the 2p state, the phase φ_7^2 does not change as varying ω_d. On the other hand, paths $\boldsymbol{d}_7^0$ and $\boldsymbol{d}_7^1$ occur via the 2p state, and φ_7^0 and φ_7^1 change as varying ω_d. So the relative phases $\varphi_7^1-\varphi_7^2$ and $\varphi_7^0-\varphi_7^2$ change by π across the resonance, while $\varphi_7^0-\varphi_7^1$ does not. The one-cycle oscillation of 7th-harmonic signal in Fig. 1 corresponds to the last term in Eq. (10). Figure 2(b) shows the phase of first-order term after the Fourier-transition analysis of the 7th-harmonic signal, Φ_7^1, as a function of ω_d. Similar to Φ_5^2, a phase jump occurs around $\omega_\mathrm{d}=2.064$ eV, corresponding to the AC Stark shifted transition energy between 1s and 2p states. Note that, the one-cycle oscillation corresponds to not the last term in Eq. (10), but the sum of the last two terms. As a result, Φ_7^1 does not equal $\varphi_7^1-\varphi_7^2$, although it shows similar phase-jump behavior.

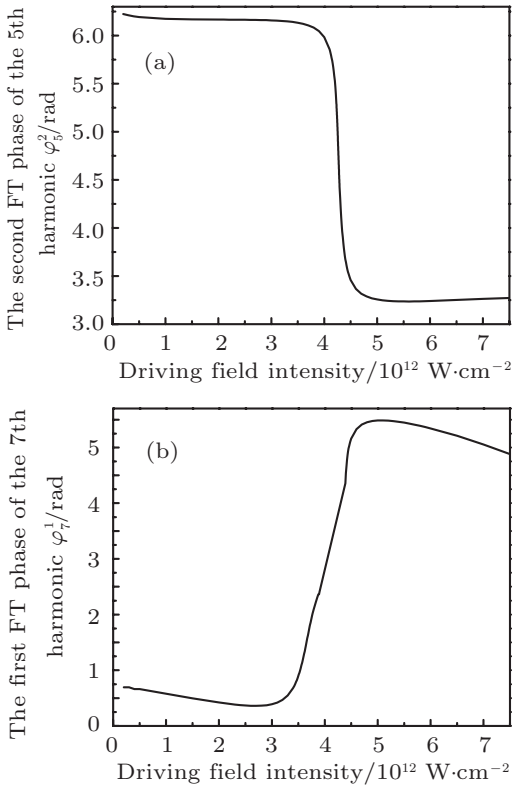


Fig. 3. (a) The phase of the 2nd-order term from Fourier-transition analysis of the 5th-harmonic as a function of ϕ, and (b) the phase of the 1st-order term from Fourier-transition analysis of the 7th-harmonic as a function of ϕ, are plotted as varying the driving CW field intensity I_d. The driving field frequency is fixed at 2.1 eV.

We also investigate the dependence of the multiphoton transition phase on the driving laser intensity. Figures 3(a) and 3(b) show the dependences of Φ_5^2 and Φ_7^1 on the driving laser intensity while keeping the driving frequency $\omega_d = 2.1$ eV, respectively. Phase jumps about π occurs around 4.2×10^{12} W$\cdot$cm^2, indicating the resonant multiphoton-transition between 1s and 2p states occurs.

Combining the above calculated phase Φ_5^2 (or Φ_7^1) as a function of the driving frequency ω_d for a fixed intensity and as a function of intensity for a fixed driving frequency allow us to determine the intensity dependence of the $1s \rightarrow 2p$ transition energy. The above procedure for the determination of phase is practicable in experiments. So we consider the numerical calculation as 'experimental' results. The 'experimental' result is shown in Fig. 4 with black solid line. On the other hand, the transition energy also can be theoretically calculated from Eq. (3), in which the transition energy is one of the quasienergy eigenvalues. The theoretical Floquet result of $1s \rightarrow 2p$ transition energy is shown in in Fig. 4 with red dash line. The 'experimental' and theoretical results agree with each other quite well. The discrepancy is due to the finite size of the Floquet matrix equation in the numerical calculation.

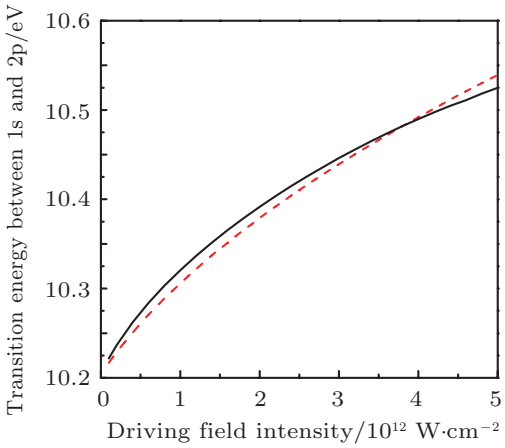


Fig. 4. The $1s \rightarrow 2p$ transition energy via phase measurement of the HHG spectra is plotted with black solid line. As a comparison, the result calculated within Floquet theory is presented with red dash line.

4. Conclusion

In the present study, we presented a theoretical investigation of the phase jump of the harmonic emission from a hydrogen atom driven by a strong CW laser field with a weak control harmonic field. The Floquet theory is used to accurately solve the interaction between the hydrogen atom and the combined laser field. The multiphoton-transition paths connecting the same initial and final states are coherently summed in the harmonic emission. The relative phases between paths are obtained via the Fourier-transform analysis of the harmonic signals as a function of the relative phase between the driving field and the control field. We observed the phase jumps when the resonance is crossed by varying the frequency and intensity of the driving field. Combining the phase variations as a function of the driving frequency for a fixed intensity and as a function of the intensity for a fixed driving frequency, the intensity dependence of the $1s \rightarrow 2p$ transition energy is determined. And the comparison with the results from the Floquet quasienergy calculation indicates that, the proposed method for the phase measurement of the harmonic emission can be employed to determine the intensity dependence of the transition energy of quantum systems. For experimental realizations, although the intensity 10^{12} W$\cdot$cm^2 at frequency 2 eV is hard to reach, the phase jumps in the vicinity of the resonance can also be observed by employing the driving laser field with long wavelength.

It is noted that, the HHG spectrum has been employed to probing the structure and electronic dynamics of molecules, in which the potential energy surface plays essential roles. Our approach can be applied to symmetric molecules or polar molecules to observe the potential energy surface of molecules driven by the laser fields.

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