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Low-order harmonic generation of hydrogen molecular ion in laser field studied by the two-state model*

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The low-order harmonic generation of hydrogen molecular ion interacting with a linearly polarized laser field has been investigated theoretically by using a simple two-state model. The validity of the two-state model is carefully examined by comparing the harmonic spectra of hydrogen molecular ion obtained from this model with those from the three-dimensional time-dependent Schrödinger equation. When combined with the Morlet transform of quantum time-frequency spectrum, the two-state model can be used to study the dynamical origin of the low-order harmonic generation of hydrogen molecular ion driven by low-frequency pulses. In addition, some interesting structures of the time profiles for low order harmonics are obtained.

Keywords: low-order harmonic generation, hydrogen molecule ion, two-state model

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1. Introduction

The high-order harmonic generation (HHG) that results from the interaction of intense laser field with atoms, ions, and molecules, is a non-linear process during which the medium will emit the high harmonics of the generation beam. During the past few years, most researchers have focused on the harmonics at the plateau and near the cutoff region[1–8] owing to their important applications (e.g., they render technologies for the extreme-ultraviolet and soft-x-ray sources on the attosecond time scale), which leads to the observation and control of their dynamic electronic behaviors.[9–12] As research moves on, near- and below-threshold harmonics have also aroused attention.[13–21] Some basic issues have been investigated, such as carrier envelope phase effects of laser pulse,[19] electron trajectory contribution,[20] inversion symmetry of localization of the electron density in molecular system.[21] For much lower harmonics,[22–24] their high repetition-rate and intensity are of importance. However, very little research has investigated this aspect.

Compared with the higher order harmonics, the low-order harmonic generation (LOHG) is rather complicated due to the influence of the atomic or molecular potential. Although LOHG process can be precisely simulated by solving a three-dimensional (3D) time-dependent Schrödinger equation (TDSE), it is necessary to adopt a suitable model to probe the mechanism and dynamical origin of the LOHG. In a recent study, we have investigated the generation of the 3rd, 5th, and 7th harmonics of hydrogen molecular ion (H_2^+) in a strong laser field.[25] Our 3D-TDSE simulations showed that under some conditions, spectra exhibited some fine sub-peak structures around the main harmonics. By adopting a two-state model, the origin for these subnormal structures was well-explained as interference in the cycles of the multiphoton-radiation emitted from the dipole transfer of the two lowest bound states. This simple model can also help to analyze the radiation processes. Following this work, we perform a more detailed simulation on the LOHG of H_2^+ in this paper. We investigate the laser wavelength and peak intensity dependence of the LOHG. By comparing the results with those obtained from solving 3D TDSE, the validity of the two-state model that we adopt to study the mechanism of LOGH is clarified.

The rest of this article is organized as follows. In Section 2, we introduce the theoretical framework. In Section 3, the results and discussion of the LOHG in H_2^+ are presented. Our conclusions are given in Section 4.

2. Theory

In the past, the two-state model has been used many times to describe the high-order harmonic radiation at the plateau and near the cutoff region in an intense low-frequency laser field.[26–29] For H_2^+, the ground states $|1\sigma_g\rangle$ and first excited states $|1\sigma_u\rangle$ (abbreviated as $|g\rangle$ and $|u\rangle$) are strongly coupled

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with each other. Therefore, the adiabatic two-state model is a good choice to simulate the LOHG of H_2^+, offering a very useful model to understand the dynamic origin of the lower-order harmonic.

In the two-state model, the time-dependent wave function is given by

$$\Psi(t) = C_1(t)\phi_1 + C_2(t)\phi_2, \tag{1}$$

where $C_n(t) = \langle\phi_1|\Psi\rangle$ denotes the overlap of the total wave function with the n-th field-dressed state. The field-dressed states of H_2^+ are given by[30–32]

$$\phi_1 = \cos(\theta)|g\rangle + \sin(\theta)|u\rangle, \tag{2}$$

$$\phi_2 = -\sin(\theta)|g\rangle + \cos(\theta)|u\rangle, \tag{3}$$

where

$$\theta = -\frac{1}{2}\arctan\left[\frac{2H_{gu}E(t)}{\Delta\varepsilon_{gu}}\right]. \tag{4}$$

Here, $H_{gu} = \langle g|z|u\rangle$ is the dipole matrix element and $\Delta\varepsilon_{gu} = \varepsilon_u - \varepsilon_g$ is the energy difference between the ground state $|g\rangle$ and the first excited state $|u\rangle$. The field-dressed eigenvalues can be obtained by

$$\varepsilon_{1,2}(t) = \frac{1}{2}\left[\varepsilon_u + \varepsilon_g \mp \sqrt{\Delta\varepsilon_{gu}^2 + 4|H_{gu}E(t)|^2}\,\right]. \tag{5}$$

In the two-state model, the dipole moment in length form and acceleration form can be respectively obtained as[32]

$$d(t) = H_{gu}[g(t)\cos(2\theta) + h(t)\sin(2\theta)], \tag{6}$$

$$a(t) = -\Delta\varepsilon_{gu}^2 d(t) + 2\Delta\varepsilon H_{gu}^2 E(t) \times [h(t)\cos(2\theta) - g(t)\sin(2\theta)], \tag{7}$$

with $g(t) = C_1^*(t)C_2(t) + C_2^*(t)C_1(t)$ and $h(t) = |C_1(t)|^2 - |C_2(t)|^2$.

Lastly, the spectra density of the emitted harmonic radiation can be calculated by the Fourier transforms of $d(t)$ or $a(t)$ as

$$S(\omega) = \frac{2\omega^4}{3\pi c^3}|d(\omega)|^2, \tag{8}$$

$$S(\omega) = \frac{2}{3\pi c^3}|a(\omega)|^2. \tag{9}$$

To verify the feasibility of the conclusion from the two-state model, we also display the LOHG calculated by solving 3D TDSE[25,33,34] as a comparison.

To probe the detailed spectra and temporal structures of the LOHG, we perform the time-frequency analysis on the induced dipole moment by means of the Morlet transform.[35–38] The function of the Morlet transform is given by

$$A_\omega(t_0,\omega) = \int a(t) w_{t_0,\omega}\,\mathrm{d}t = A_\omega(t_0), \tag{10}$$

where

$$w_{t_0,\omega} = \sqrt{\omega}W[\omega(t-t_0)], \tag{11}$$

$$W(x) = \frac{1}{\tau}\mathrm{e}^{\mathrm{i}x}\mathrm{e}^{-x^2/2\tau^2}. \tag{12}$$

We choose the window width parameter $\tau = 15$ in this paper.

3. Results and discussion

In our calculations, we use a linear polarized Gaussian-shape electric field

$$E(t) = E_0\,\mathrm{e}^{-2\ln 2(t^2/\tau^2)}\sin(\omega t). \tag{13}$$

The duration is 25 optical cycles. Figure 1 compares the LOHG of H_2^+ obtained from the two-state model and TDSE. The internuclear distance $R = 6$ a.u. and the laser peak intensity is 1×10^{14} W/cm^2, while the laser wavelength changes from 400 nm to 1400 nm. As can be seen, the results of the two-state model agree quite well with those from the TDSE in the long-wavelength region, while the 600-nm results are reliable only for the 1st and 3rd harmonics. Thus, we conclude that the electric dipole transition from the two field-dressed states in a long-wavelength laser pulse is very significant for the LOHG of H_2^+. However, for even shorter 400-nm wavelength, the two-state model is not suitable for studying low-order harmonics of H_2^+ with $R = 6$ a.u. We can see that the harmonic spectra in the low-order region present some fine sub-peak structures, which become increasingly obvious with the increase of the laser wavelength owing to the the interference in cycles of the multiphoton radiation during the dipole transition between the two lowest bound states. We have analyzed the origin of these fine sub-peak structures in detail in Ref. [25].

Figure 2 shows the evolution of the field-dressed energies $\varepsilon_{1,2}(t)$ with emission time in laser fields with different laser wavelengths. In the two-state model, the highest order of the harmonic is decided by the maximum energy difference of the two adiabatic states. As see in the Figs. 2(a)–2(f), the maximum energy differences, corresponding to the highest order, are 2 (400-nm), 4 (600-nm), 5 (800-nm), 7 (1000-nm), 8 (1200-nm), and 9 (1400-nm), respectively. Hence, it is reasonable that the harmonic spectrum from the two-state model for the 600-nm laser pulse is consistent with the result of TDSE only for the 1st and 3rd harmonics. The two-state model is not suitable to study LOHG of H_2^+ generated by laser pulse with 400-nm and the shorter wavelength.

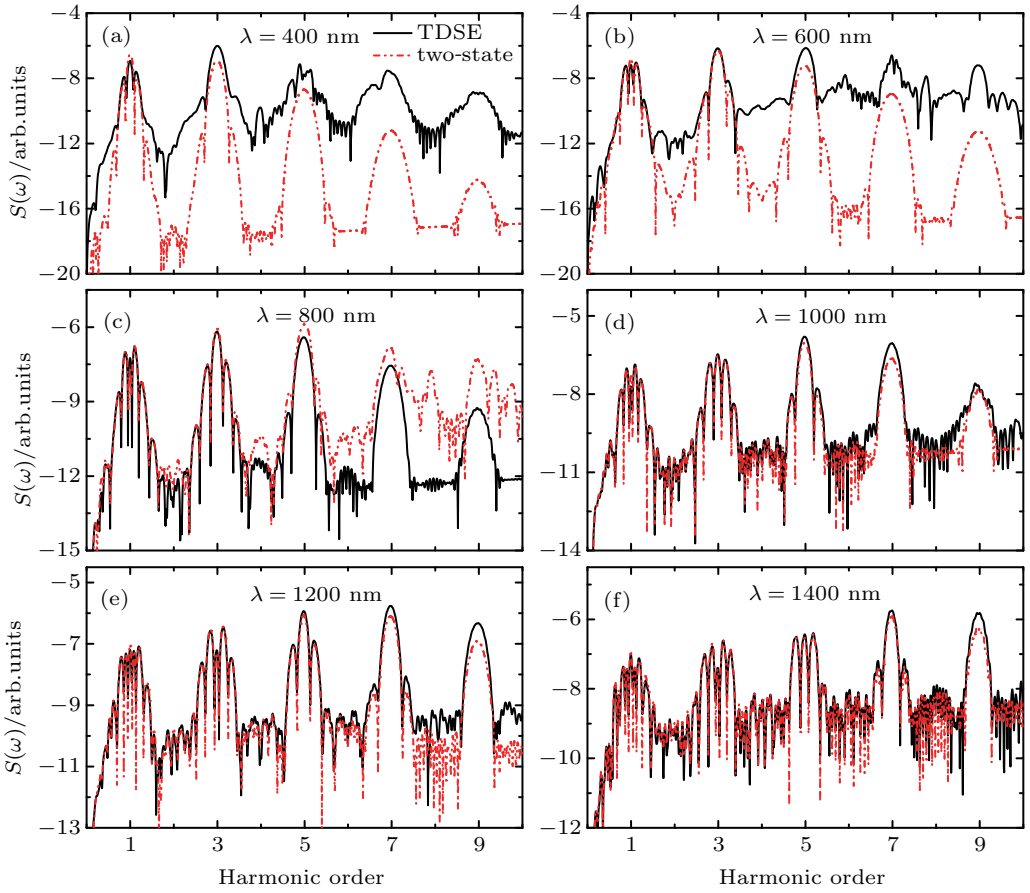


Fig. 1. (color online) Comparison of the power spectra of H_2^+ obtained from simulations of TDSE (black solid line) and the two-state model (red dash-dot-dot line) in laser fields with wavelengths of (a) 400 nm, (b) 600 nm, (c) 800 nm, (d) 1000 nm, (e) 1200 nm, and (f) 1400 nm. The laser intensity is 1×10^{14} W/cm^2 in all cases.

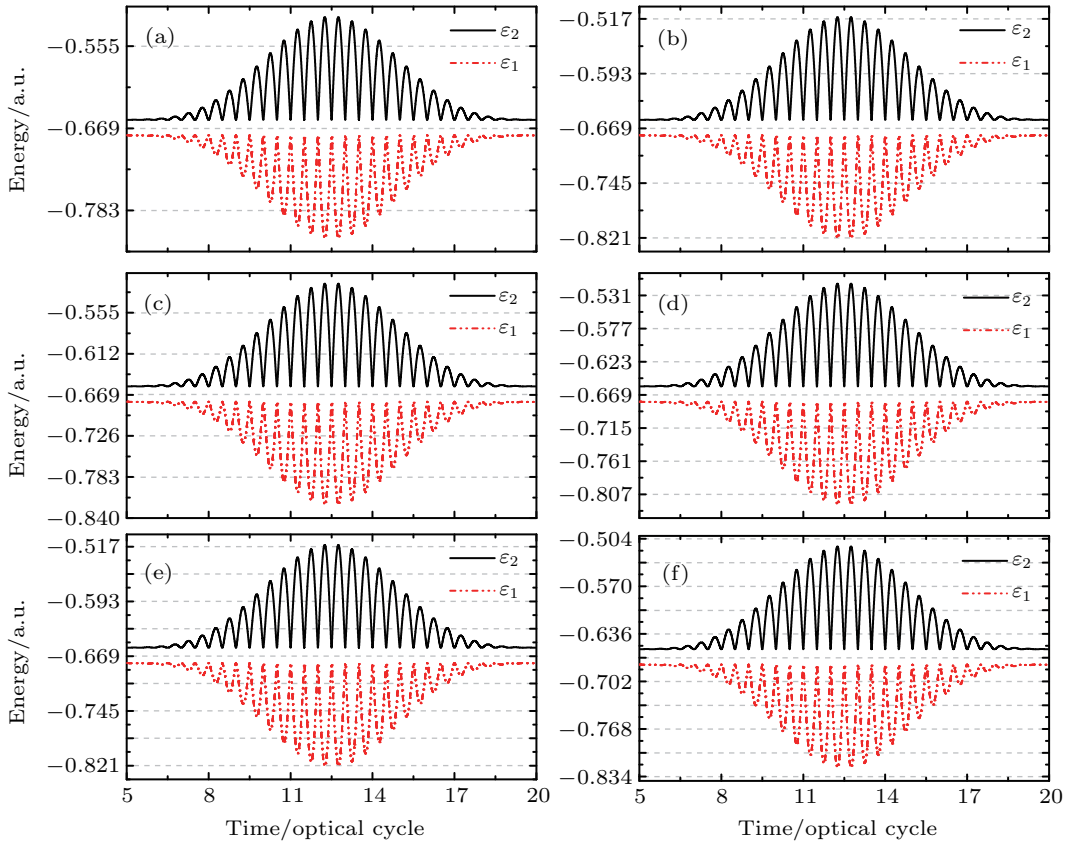


Fig. 2. (color online) The field-dressed energies $\varepsilon_1(t)$ and $\varepsilon_2(t)$ as a function of time. (a)–(f) The parameters used are the same as those in Fig. 1. The energy difference between the two horizontal neighbor gray-dash lines equals one photon-energy of the laser pulse.

Figure 3 presents the wavelet time-frequency spectra of H_2^+ with internuclear distance of 6 a.u. for different laser wavelengths. The top row is the spectra calculated from the TDSE, while the bottom row is the two-state model results. Clearly, with the same laser pulse, the emission time of harmonic in the two-state model is in good agreement with that in TDSE calculations. The higher the frequency, the narrower the emission time interval. The results further confirm that the two-state model can be used to investigate the dynamic origin of the LOHG of H_2^+ in the long-wavelength region. We also find that there are some minima along the emissions of the 3rd and 5th harmonics in the long-wavelength laser fields; that is, two for the 3rd harmonic in the 1000-nm, 1200-nm, and 1400-nm fields, one and two for the 5th harmonic in the 1200-nm and 1400-nm fields, respectively.

To clearly observe these minima in the emission of the 3rd and 5th harmonics, Figure 4 depicts their time profiles in different laser fields. As can be see, the time profiles of the 3rd harmonic in the 400-nm and 600-nm, and the 5th harmonic in the 400-nm, 600-nm, and 800-nm laser fields present an usual variation trend, which are analogous to the laser field envelopes, while the profiles in other fields show the oscillatory radiation, leading to some fine structures of the time profiles. By choosing the appropriate incident laser pulse, the radiation of the 3rd or 5th harmonic of H_2^+ is able to become a means of changed-shape of the low-order harmonic.

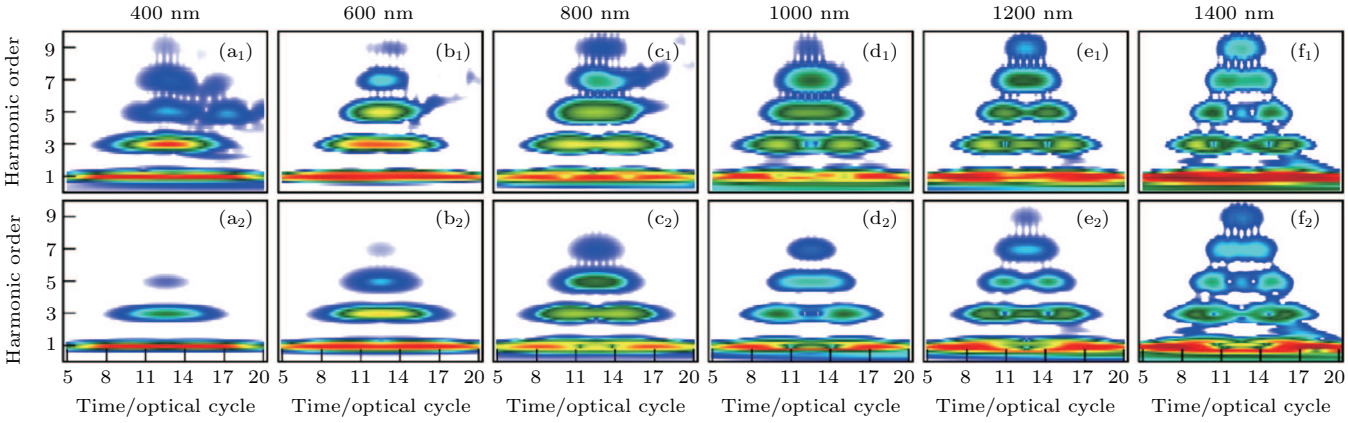


Fig. 3. (color online) The wavelet time-frequency spectra obtained from (a_1)–(f_1) TDSE and (a_2)–(f_2) two-state model. (a)–(f) The parameters used are the same as those in Fig. 1.

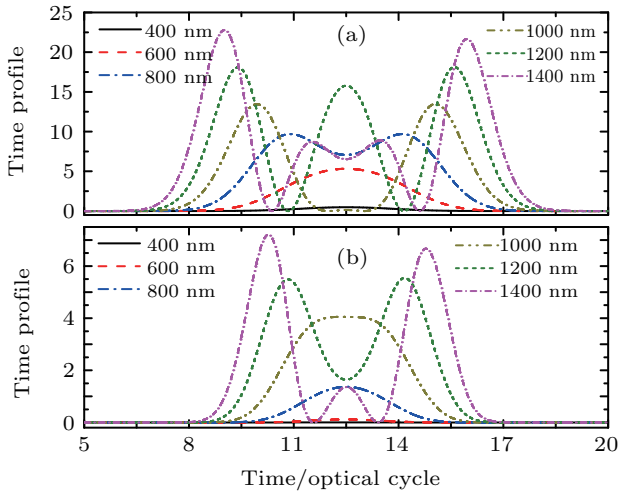


Fig. 4. (color online) The time profile of (a) the 3rd and (b) the 5th harmonics generated in laser fields with different wavelengths. The parameters used are the same as those in Fig. 1.

In the two-state model, the population transition of two adiabatic states is the main reason of HHG. Therefore, we present the evolution of population transition with emission time in different laser fields in Fig. 5. At the time the level crossings occur, the most population transition takes place (can be observed more clearly in Fig. 6). However, as the wavelength increases, the populations of the two adiabatic states differ greatly, which is responsible for the interesting structures of the time profiles in LOHG. Taking the case of 1000-nm as an example, we show in Fig. 6 again the time profiles of the 3rd and 5th harmonics, the field-dressed energies, and the population transition for the two states together. On the basis of the energy difference (the 1st harmonic not considered here) and relationship of population of the two field-dressed states, we divide the whole emission into three periods (shown by shadow regions in Fig. 6). In the 1st and 3rd periods, the population has the relation of $P_2 > P_1$, so only the 3rd harmonic has the strongest intensity. For the second period, in view of the energy, the 3rd and 5th harmonics would be emitted. Moreover, the total strength of these two harmonics should be smaller than that in the other two periods due to $P_2 < P_1$. Therefore, one minimum appears in the 3rd harmonic emission. However, the strong 5th harmonic still appears owing to a nonzero P_2. In the case of other wavelengths, these minima can be analyzed in the same way.

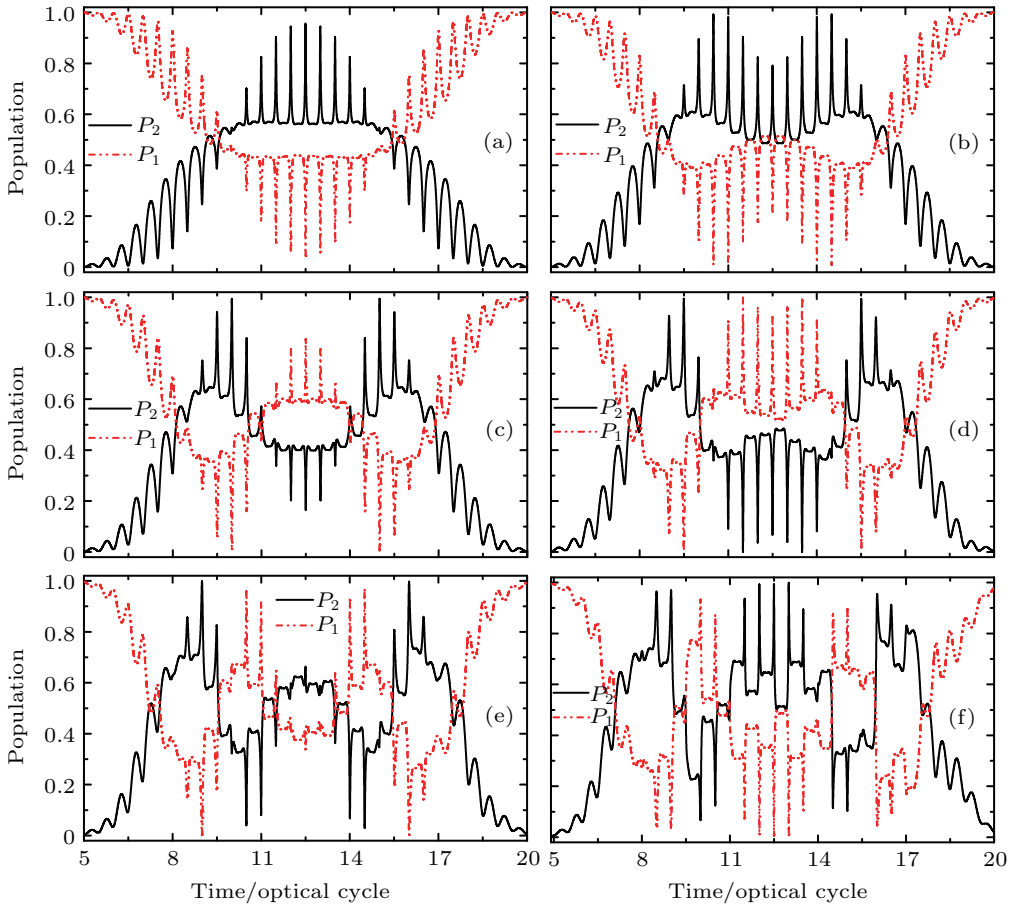


Fig. 5. (color online) The populations of states $|\phi_1\rangle$ (P_1, red dash-dot-dot line) and $|\phi_2\rangle$ (P_2, black solid line) as a function of emission time in different laser fields. (a)–(f) The parameters used are the same as those in Fig. 1.

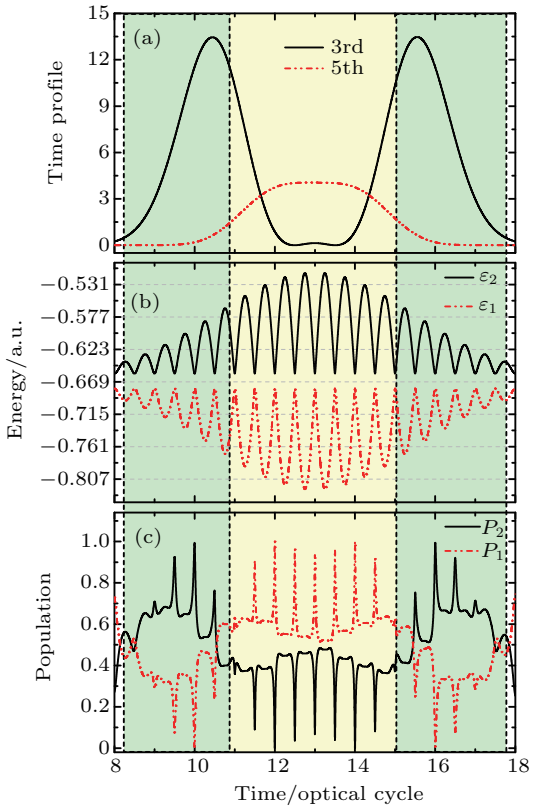


Fig. 6. (color online) (a) The time profiles of the 3rd and 5th harmonics. (b) The field-dressed energies $\varepsilon_1(t)$ and $\varepsilon_2(t)$. (c) The populations P_1 and P_2 of two adiabatic states. The laser wavelength is 1000 nm.

In Eq. (4), we can see that the laser peak has an important impact on the maximal field-dressed energy difference, which decides the highest harmonic generation. Figure 7 shows the LOHG of H_2^+ with $R = 6$ a.u. calculated from the two-state model for different laser peak intensities of $0.5I_0$, $1.5I_0$, $2.0I_0$, and $2.5I_0$ ($I_0 = 1 \times 10^{14}$ W/cm^2), the laser wavelength is fixed at 1000 nm. The harmonic spectra obtained from TDSE are also shown as a reference. Clearly, figures 7(a) and 7(b) show that the harmonic spectra obtained by the two-state model coincide exactly with those from TDSE calculations. However, there are some discrepancies in the more intense laser fields as shown in Figs. 7(c) and 7(d). In addition, with the increase of the laser intensity, the fine sub-peak structures in LOHG become less symmetrical and noticeable. That indicates that the two-state model can be used to investigate the LOHG of H_2^+ in a weak low-frequency laser field.

To explain the validity of the two-state model associated with the laser intensity, in Fig. 8 we show the time-dependent ionization probability for four different laser intensities. It is clearly seen that the more intense laser field, the higher ionization probability. The population of the higher excited states is also increased. Because the LOHG of H_2^+ with $R = 6$ a.u. is highly related to the transition between the ground state and

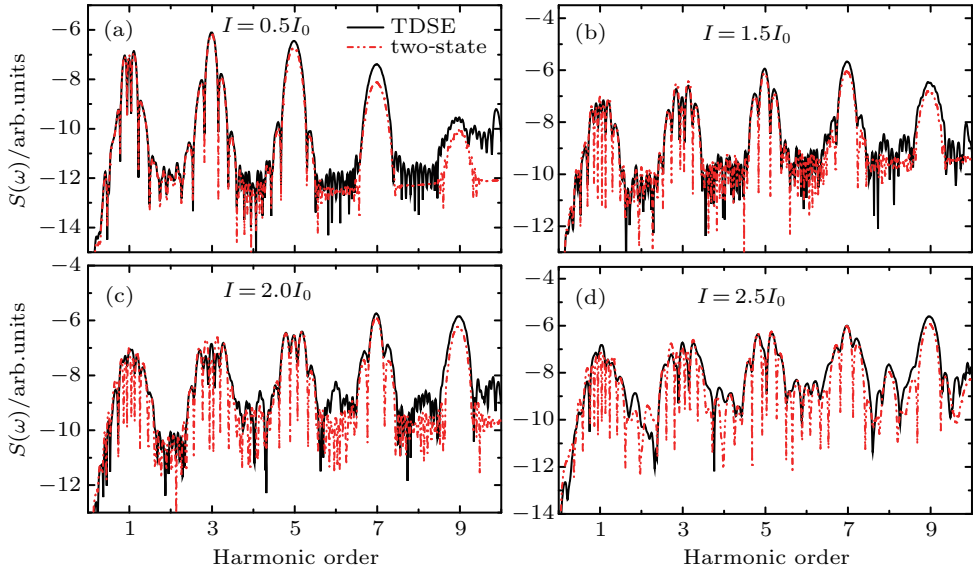


Fig. 7. (color online) Comparison of the power spectra of H_2^+ obtained from simulations of the two-state model (red dash-dot-dot line) and TDSE (black solid line) for laser peak intensities of (a) $0.5I_0$, (b) $1.5I_0$, (c) $2.0I_0$, (d) $2.5I_0$. Here, $I_0 = 1 \times 10^{14}$ W/cm^2. The laser wavelength is 1000 nm.

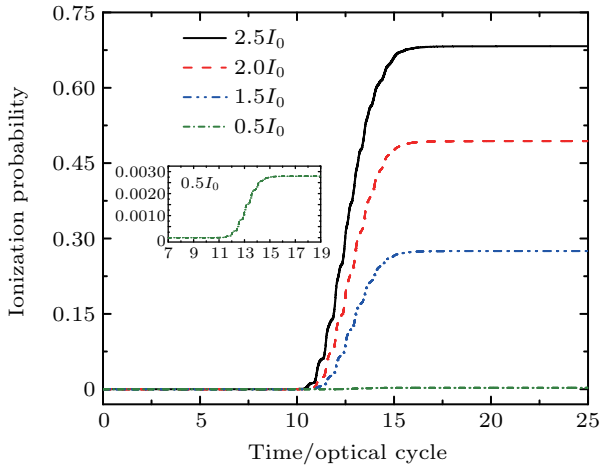


Fig. 8. (color online) Comparison of total ionization probability as a function of time calculated at laser intensities of $0.5I_0$, $1.5I_0$, $2.0I_0$, and $2.5I_0$. The other laser parameters used are the same as those in Fig. 7.

the first excited state, it is reasonable that the two-state model is no longer suitable to simulate the LOHG for laser intensities $I = 2.0I_0$ and $2.5I_0$ due to the high ionization probability.

4. Conclusion

In this paper, we employ the two-state model to study the LOHG of H_2^+ exposed to a linearly polarized laser field. The validity of the two-state model is carefully examined by comparing the harmonic and Molet time-frequency spectra obtained by this model with those of TDSE. Our results show that the two-state model can be used to study the dynamical origin of the LOHG of H_2^+ generated by a weak long-wavelength laser pulse. Once the validity of this model is established, it will be a simple and useful tool to study the LOHG of H_2^+. Some fine sub-peak structures in LOHG resulted from the interferences in cycles of the multiphoton-radiation emitted from the dipole transition between the lowest two bound states can also be observed. In addition, as the laser's wavelength increases, we find some interesting structures in the time profiles of LOHG of H_2^+, which can become a means of changed-shape of the low-order harmonic.

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