Role of excited states in the elliptically polarized harmonic generation*

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We theoretically investigate the contribution of the excited state to the ellipticity of the harmonics from H_2^+ at different orientation angles irradiated by a linearly polarized laser pulse. It is found that the first excited state has a significant influence to the ellipticity of the harmonics, and the contribution of higher excited states to the ellipticity can be neglected. Moreover, the conclusion is not dependent on the laser intensity.

Keywords: high-order harmonic generation, intense laser, excited state, ellipticity

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1. Introduction

High-order harmonic generation (HHG) is a main focus of intense laser–matter physics owing to its potential of producing ultrashort attosecond pulses,[1–7] and short-wavelength light sources.[8,9] The physical mechanism of the HHG process can be well understood by the three-step model.[10] Firstly, the electron tunnels through the barrier formed by the Coulomb potential and the laser field, then it oscillates quasi-freely under the drive of the laser field and gains kinetic energy, and finally it can recombine with the parent ion and emit a photon in the transition back to the ground state. According to this model, various schemes have been proposed to extend the harmonic spectrum or produce isolated attosecond pulses.[11–22] In addition, the polarization state is another important characteristic of the harmonic emission due to the potential applications of the elliptically polarized harmonics and the elliptically polarized attosecond pulses. HHG polarization has previously been studied in atomic gases where the laser ellipticity was varied.[23,24] In such case, the harmonic efficiency drops significantly with increasing ellipticity of the driving laser.[25] Recently, the elliptically polarized harmonics generated from the oriented molecules driven by a linearly polarized laser are also extensively regarded. For example, it has been demonstrated that elliptically polarized harmonics can be applied in HHG spectroscopy of oriented N_2 to track multichannel dynamics during strong-field ionization.[26] Moreover, because the information of the molecular system participating in the harmonic generation can be imprinted in the polarization characteristics of harmonics,[27–29] the elliptically polarized harmonics can provide a sensitive probe of the molecular system.

So far, the elliptically polarized harmonics have been experimentally produced from the molecular alignment with a good level driven by a linearly polarized laser pulse.[28,30] In theory, the harmonic spectrum is the Fourier transform of the dipole acceleration.[31] Thus, in order to accurately describe HHG, a fully time-dependent wave function is required. Very recently, the angle dependence of polarization has been explored,[32] where the effect of excited states has not been clarified yet. In this work, we mainly numerically study the role of the excited states in the elliptically polarized harmonic generation from H_2^+ irradiated by a linearly polarized laser pulse. It is found that the first excited state can significantly influence the ellipticity of the harmonics. Moreover, when the laser pulse is intense enough, the first excited state still plays an important role in determining the ellipticity of the harmonics, and the contribution of higher excited states to the ellipticity of the harmonics can be still neglected.

2. Model and method

In this work, we use the example of H_2^+ to investigate the role of the excited states in the elliptically polarized harmonic generation. Figure 1 presents a sketch of our coordinate system, which respectively defines the molecular axis and the vertical direction of the molecular axis as the x axis and the y axis, with the direction of the driving laser field lying in the x–y plane. The angle between the direction of the driving laser field and the molecular axis (x) is denoted as θ. In this system, the two-dimensional (2D) time-dependent Schrödinger equation (the atom units are used throughout this paper) can be given by

$$\mathrm{i}\frac{\partial\psi(x,y,t)}{\partial t} = \left[\frac{P_x^2+P_y^2}{2} + V(x,y) - xE(t)\cos(\theta)\right.$$

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$$-yE(t)\sin(\theta)\Big]\psi(x,y,t), \tag{1}$$

where $E(t)$ is the external electric field, and $V(x,y)$ is the Coulomb potential and can be written as

$$V(x,y) = \frac{-Z}{\sqrt{(x+R/2)^2+y^2+0.5}} + \frac{-Z}{\sqrt{(x-R/2)^2+y^2+0.5}}, \tag{2}$$

where Z is the effective charge, and R is the inter-nuclear separation (for $Z=1$ and $R=2$ a.u., the ground state energy for H_2^+ reproduced here is $I_p = 1.11$ a.u.).

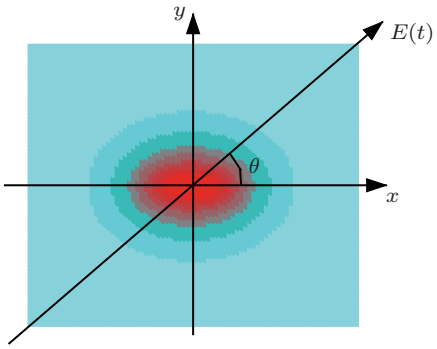


Fig. 1. (color online) A sketch of the laboratory coordinate system. (x,y) is the laboratory frame, the molecular axis is along the x axis, which forms the angle θ with the direction of the laser field.

The two-dimensional time-dependent Schrödinger equation can be solved accurately and efficiently by means of the Crank–Nicolson method. Once the time evolution of the wave function $\psi(x,y,t)$ is determined, the fully time-dependent induced dipole acceleration can be given by means of Ehrenfest's theorem

$$a_x(t) = -\langle\psi(x,y,t)|\frac{\partial V(x,y)}{\partial x}|\psi(x,y,t)\rangle,$$
$$a_y(t) = -\langle\psi(x,y,t)|\frac{\partial V(x,y)}{\partial y}|\psi(x,y,t)\rangle. \tag{3}$$

In order to investigate the role of the excited states in the elliptically polarized harmonics generation, the time-dependent induced dipole acceleration can be expressed as

$$a_x(t) = -\langle\varphi_i(x,y,t)|\frac{\partial V(x,y)}{\partial x}|\psi(x,y,t)\rangle + \text{c.c.},$$
$$a_y(t) = -\langle\varphi_i(x,y,t)|\frac{\partial V(x,y)}{\partial y}|\psi(x,y,t)\rangle + \text{c.c.} \tag{4}$$

Here, $\varphi_i = \sum_{n=0}^{i}\langle\phi_n|\psi\rangle\phi_n$ $(i=0,1,2)$, and ϕ_n denotes the wave function of n-th excited state of 2D H_2^+ in the free field. For example, for $\varphi_0 = \langle\phi_0|\psi\rangle\phi_0$, it indicates that only the transition between the time-dependent wavefunction and the ground state is considered in calculating the time-dependent induced dipole acceleration.

The harmonic spectrum is then obtained by Fourier transforming the time-dependent dipole acceleration. The two components can be obtained by $|a_x(q)|^2$ and $|a_y(q)|^2$. The $a_x(q)$ and $a_y(q)$ can be respectively given by

$$a_x(q) = \frac{1}{\sqrt{2\pi}}\int_0^T a_x(t)\,e^{-iq\omega t}\,dt,$$
$$a_y(q) = \frac{1}{\sqrt{2\pi}}\int_0^T a_y(t)\,e^{-iq\omega t}\,dt, \tag{5}$$

where T and q are the duration of the driving pulse and the harmonic order, respectively. The intensity (I) and phase (φ) of the harmonic components can be given by $I_j(q) = |a_j(q)|^2$ and $\varphi_j(q) = \arg[a_j(q)]$ $(j = x,y)$, respectively. The ellipticity ε is determined by the amplitude ratio and the phase difference of the two components[33]

$$\varepsilon = \sqrt{\frac{1+r^2-\sqrt{1+2r^2\cos(2\delta)+r^4}}{1+r^2+\sqrt{1+2r^2\cos(2\delta)+r^4}}}, \tag{6}$$

where $r = \sqrt{I_y/I_x}$ and $\delta = \varphi_y - \varphi_x$. The range of ellipticity is $0 \le \varepsilon \le 1$. The linear, elliptical, and circular polarization correspond to $\varepsilon = 0$, $0 < \varepsilon < 1$, and $\varepsilon = 1$, respectively.

3. Results and discussion

In Figs. 2(a) and 2(b), we compare the x and y components of harmonic spectra from 2D H_2^+ for the orientation angle of 30° using Eqs. (3) and (4) with $i=0$, respectively. In the calculation, we adopt a linearly polarized 800-nm laser pulse with 10 cycles, and the amplitude of the laser field is set as 0.12 a.u. The electric field is expressed by $E(t) = E_0 f(t)\sin(\omega t)$. Here E_0 and ω are the amplitude and frequency of the laser field, respectively. The $f(t)$ is the envelope of the driving pulse and is considered as a trapezoidal shape with three-cycle linear climbing ramp. It can be seen from Figs. 2(a) and 2(b) that the contribution of the excited states to the harmonic spectra is not small enough and can be neglected compared with that of the ground state. Therefore, the excited states can play an important role in the elliptically polarized harmonic generation.

In the following, we further compare the ellipticity of the harmonics, which is shown in Fig. 2(c). It can be clearly seen that the ellipticity of harmonics obtained by Eq. (4) with $i=0$ (the red curves with the unfilled circle symbols) is evidently different from that obtained by Eq. (3). We conclude that the difference may come from the contribution of the excited states. In order to confirm our conclusion, figure 3(a) presents the ellipticity of the harmonics obtained by Eqs. (3) and (4) with $i=1$. For comparison, the results obtained by Eq. (4) with $i=0$ (the red curve with the unfilled circle symbols) is also presented. The parameters are the same as that in Fig. 2. It can be found that the ellipticity of the harmonics obtained by considering the contribution of the first excited state (the blue

curve with the unfilled triangle symbols) is in good agreement with that obtained by using Eq. (3). Similarly, we also present the ellipticity of the harmonics for the orientation angles of 45° and 60° in Figs. 3(b) and 3(c), respectively. All of them have a common character as shown in Fig. 3(a). Namely, for all the orientation angles, the ellipticity of the harmonics obtained by considering the contribution of the first excited state (the blue curve with the unfilled triangle symbols) is more accurate than that obtained by only considering the contribution of the ground state.

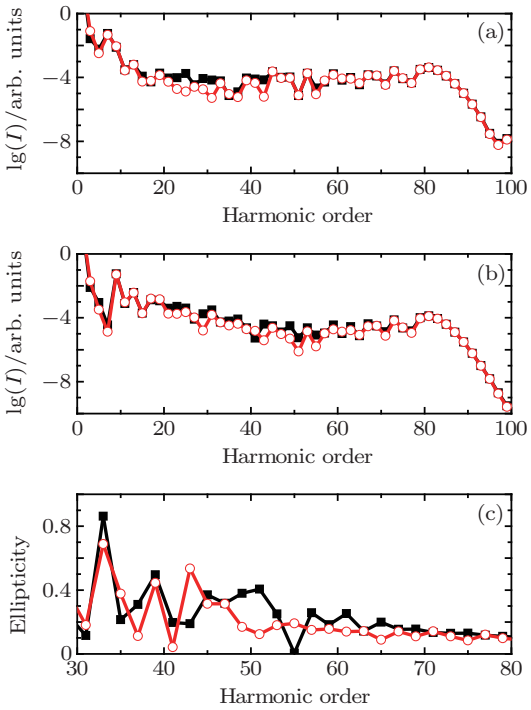


Fig. 2. (color online) (a) x and (b) y components of the harmonic spectra; (c) ellipticity of the harmonics from 2D H_2^+ for the orientation angle of 30°. The black curves with the filled square symbols denote the results obtained by Eq. (3), and the red curves with the unfilled circle symbols denote the results obtained by Eq. (4) with $i = 0$.

Next, we further study the contribution to the ellipticity of the higher excited states. Figure 4 shows the ellipticity of the harmonics obtained by Eqs. (3) and (4) with $i = 2$ at different orientation angles. For comparison, the results obtained by Eq. (4) with $i = 1$ (the red curve with the unfilled triangle symbols) are also shown. The parameters are the same as that in Fig. 3. From this figure, we do not see obvious difference between the results obtained by Eq. (4) with $i = 2$ and $i = 1$. Both of the two methods gain the exact ellipticity of the harmonics. In other words, for this case, the contribution of the second excited state to the ellipticity can be neglected. However, since the population of the excited state increases as the intensity of the driving laser pulse increases, does the second excited state have a significant influence for the ellipticity

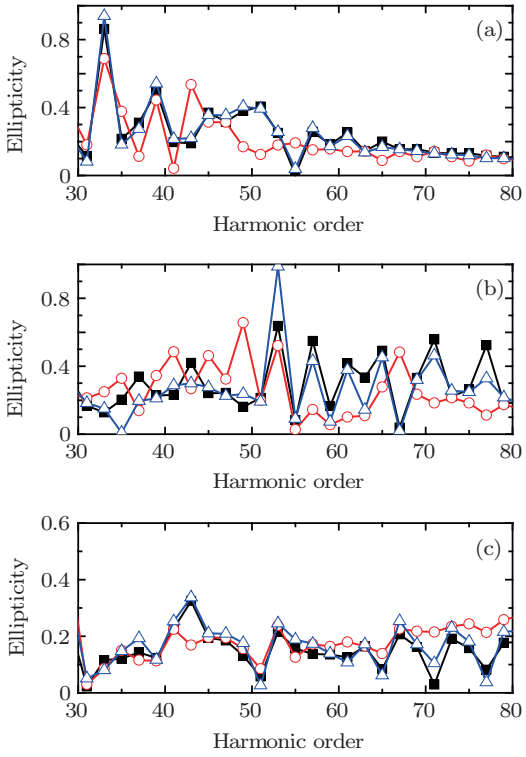


Fig. 3. (color online) Ellipticity of the harmonics from 2D H_2^+ for the orientation angles of (a) 30°, (b) 45°, and (c) 60°. The black curves with the filled square symbols denote the results obtained by Eq. (3), the red curves with the unfilled circle symbols and the blue curves with the unfilled triangle symbols denote the results obtained by Eq. (4) with $i = 0$ and $i = 1$, respectively.

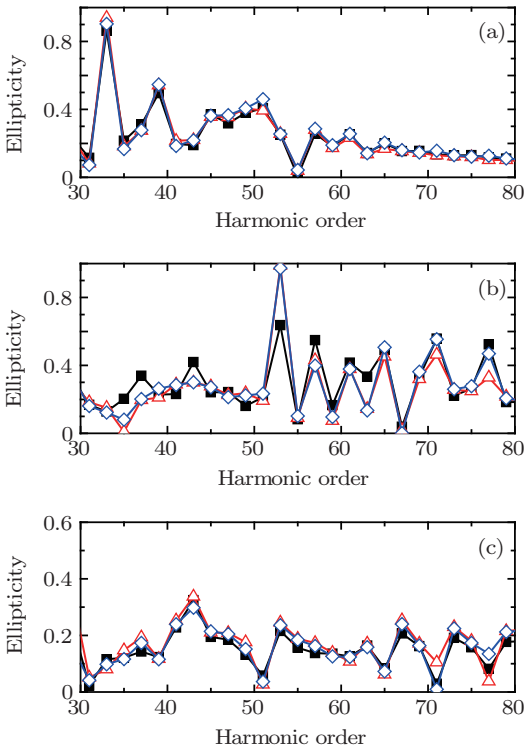


Fig. 4. (color online) Ellipticity of harmonic spectra from 2D H_2^+ for the orientation angles of (a) 30°, (b) 45°, and (c) 60°. The black curves with the filled square symbols denote the results obtained by Eq. (3), the red curves with the unfilled triangle symbols and the blue curves with the unfilled diamond symbols denote the results obtained by Eq. (4) with $i = 1$ and $i = 2$, respectively.

of the harmonics when the laser pulse is intense enough? In

order to answer this question, figure 5 shows the ellipticity of the harmonics for the orientation angle of 45° when the amplitude (E_0) of the laser field is 0.15 a.u. In Fig. 5(a), we present the ellipticity of harmonics obtained by Eqs. (3) and (4) with $i = 0$ and $i = 1$, respectively. It can be found that the ellipticity of the harmonics obtained by considering the contribution of the first excited state (the blue curve with the unfilled triangle symbols) is still in good agreement with that obtained by Eq. (3). In order to check the contribution of the higher excited state, we further present the ellipticity of harmonics obtained by Eqs. (3) and (4) with $i = 1$ and $i = 2$ in Fig. 5(b), respectively. From this figure, we do not see obvious difference between the results obtained by Eq. (4) with $i = 2$ and those obtained by Eq. (4) with $i = 1$. Both the results obtained by Eq. (4) with $i = 1$ and $i = 2$ are in good agreement with that obtained by Eq. (3). These results reveal that the ellipticity of the harmonics is mainly decided by the contribution of the ground state and the first excited state, and the contribution of higher excited states can be neglected even if the laser pulse is intense.

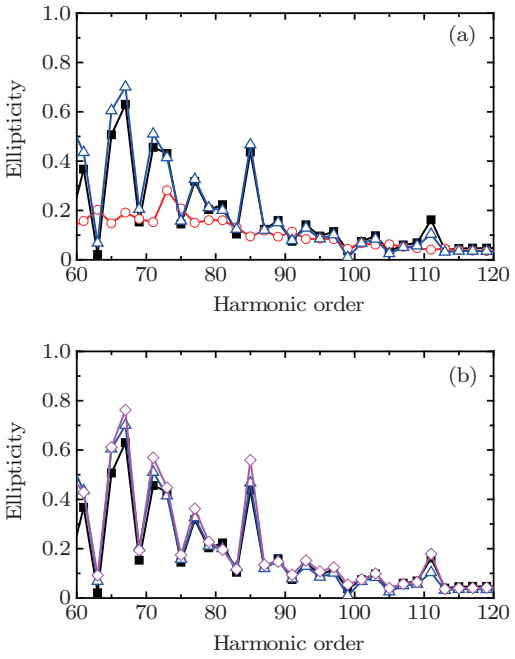


Fig. 5. (color online) Ellipticity of harmonics from 2D H_2^+ for the orientation angle of 45°. The black curves with the filled square symbols denote the results obtained by Eq. (3), the red curves with the unfilled circle symbols, the blue curves with the unfilled triangle symbols, and the magenta curves with the unfilled diamond symbols denote the results obtained by Eq. (4) with $i = 0$, $i = 1$, and $i = 2$, respectively.

Finally, we also investigate the contribution of the excited state to the ellipticity of the harmonics from H_2^+ with $R = 2.5$ a.u. for the orientation angle of 30°, which is shown in Fig. 6. The other parameters are the same as that in Fig. 2. The common conclusion can be obtained.

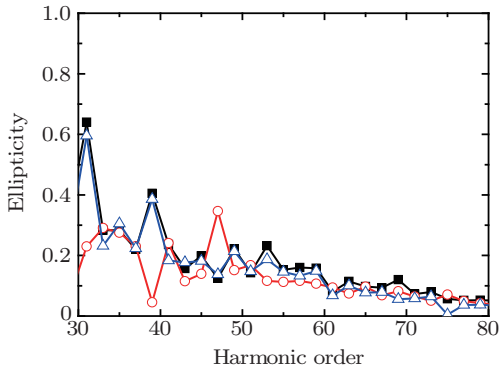


Fig. 6. (color online) Ellipticity of harmonics from 2D H_2^+ with $R = 2.5$ a.u. for the orientation angle of 30°. The black curves with the filled square symbols denote the results obtained by Eq. (3), the red curves with the unfilled circle symbols, and the blue curves with the unfilled triangle symbols denote the results obtained by Eq. (4) with $i = 0$ and $i = 1$, respectively.

4. Conclusion

In summary, we investigate the high-order harmonic generation from H_2^+ at different orientation angles irradiated by an intense linearly polarized laser pulse. It is found that the contribution of the excited states to the harmonic spectra is not small enough and can not be neglected compared with that of the ground state. Therefore, the excited states can play an important role in the elliptically polarized harmonic generation. Further investigations reveal that the first excited state has an important contribution to the ellipticity of the harmonics. Moreover, the conclusion is not dependent on the laser intensity.

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