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Multichannel molecular high-order harmonic generation from HeH^{2+} with the combination of a chirped laser and a unipolar pulse

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ABSTRACT

The high-order harmonics from the asymmetric molecule HeH^{2+} with the combination of a chirped laser and a unipolar pulse is investigated by solving non-Born–Oppenheimer approximation time-dependent Schrödinger equation. We illustrate the multichannel molecular high-order harmonic generation by using a chirped laser pulse with carrier envelope phase (CEP) $\varphi = 0$ plus a unipolar pulse, and a chirped laser pulse with CEP $\varphi = \pi$ minus a unipolar pulse, respectively. Due to the presence of a permanent dipole moment, the three-step model is generalized to a four-step model in molecular high-order harmonic generation (MHOHG). The results show that an ultrabroad supercontinuum spectrum with a spectral width of about 65 orders from 163 orders to 228 orders and a two-plateau structure can be observed. Furthermore, the time–frequency analysis shows that the short quantum trajectory is left and the long one is suppressed with the combination of a chirped laser and a unipolar pulse.

1. Introduction

High-order harmonic generation (HHG) of atoms and molecules in the intense laser fields has been widely investigated because of its applications for generating attosecond (as) pulse, which is a tool for observation and control of the electronic dynamics [1,2]. A HHG spectrum shows a decrease of the amplitude for the first few harmonics followed by a broad plateau where all the harmonics have the same strength and finally a sharp cutoff at harmonic energy $I_p + 3.17U_p$, where I_p is the ionization potential and U_p is the ponderomotive energy of the free electron in the laser field ($U_p = I^2/4\omega^2$, I and ω are the intensity and the frequency of the laser pulse, respectively). The HHG spectra can be well understood by the semi-classical three-step model [3–5]: First the electron tunnels through the Coulomb potential barrier formed by the laser field, then the ionized electron travels freely and obtains additional kinetic energy; finally, it recombines with the parent ion and releases a harmonic photon. Compared with atoms, the molecular investigation is more complicated due to the resonance [6], nuclear motion [7], and so on.

There are always some investigations on HHG [8,9], and many efforts have been made to extend the cut-off energy and produce an isolated attosecond pulse, such as the chirped-pulse scheme [10], Terahertz (THz) field control [11], vibrational states control [12], and so on. Miao and Zhang [7] investigated the harmonic emission from HeH^{2+} based on Born–Oppenheimer and non-Born–Oppenheimer approximation, and found that the nuclear motion has significant influence on the harmonic efficiency. Li et al. [13] demonstrated a method to obtain an isolated sub-30-as pulse from He^+ by adding a 27th harmonics pulse to an intense few-cycle chirped laser pulse. Orlando et al. [14] produced a 170 as pulse by using a weak 820 nm pulse with a controlling short unipolar pulse. Pan et al. [15] theoretically presented that an ultrabroad XUV supercontinuum spectrum with a spectral width of 930-eV can be generated from the oriented CO driven by using the combination of an intense few-cycle chirped laser and a unipolar pulse, and an isolated 38-as pulse can be obtained. The unipolar pulses with peak field up to several hundreds of kV cm^{-1} and duration time down to 400 fs are currently generated [16]. The calculation illustrated that half-cycle pulses with a full width at half maximum as short as 0.5 fs can be generated [17]. Yuan and Bandrauk [18] used a half-cycle THz pulse which is similar to a static field to control quantum trajectory and a single circularly polarized attosecond was produced. Both the THz pulse and the unipolar pulse have only one peak and can achieve the quantum trajectory control. In this sense, the half-cycle Terahertz pulse and the unipolar pulse make similar contributions to the HHG.

In this paper, we investigate the molecular high-order harmonic generation (MHOHG) of HeH^{2+} molecule via solving non-Born–Oppenheimer time-dependent Schrödinger equation (TDSE). For the asymmetric system HeH^{2+}, the electrons populated on the $2p\sigma$ state generalize the three-step model to a four-step model in the process of MHOHG. The combination of a chirped laser and a

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unipolar pulse is adopted to broaden the laser peak, such that the ionized electron experiences much longer time acceleration. We aim at investigating the difference processes of MHOHG in the three-step model and the four-step model case, and achieving different multichannel in harmonic emission. Furthermore, we also present the time–frequency analysis to illustrate the underlying physical mechanism of MHOHG.

2. Theoretical model and numerical method

The MHOHG from HeH^{2+} can be investigated by solving the non-Born–Oppenheimer time-dependent Schrödinger equation (TDSE). The initial wave function can be constructed by the finite-element discrete variable representation method, which provides the block-diagonal sparse matrix representation of the kinetic operator and the diagonal matrix representation of the effective potential [19]. In one-dimensional (1D) model of HeH^{2+} molecule, we assume that the polarization direction of the laser field is parallel to the molecular axis. The time-dependent wave functions are obtained by using the second-order split-operator fast Fourier transform algorithm [20,21]. All the quantum wave packet calculations have been numerically performed using the attosecond resolution quantum dynamics program LZH-DICP [22–24]. In the dipole approximation and the length gauge, the time-dependent Schrödinger equation can be given by (in atomic units):

$$i\frac{\partial}{\partial t}\Psi(z,R,t) = [T_R + T_e + V_C(z,R) + V_l(z,t)]\Psi(z,R,t), \tag{1}$$

where $T_R = -\frac{1}{2\mu_\eta}\frac{\partial^2}{\partial R^2}$ and $T_e = -\frac{1}{2\mu_e}\frac{\partial^2}{\partial z^2}$ present the kinetic energy of the nuclei and the electron. $\mu_\eta = (M_{He}M_H)/(M_{He}+M_H)$ and $\mu_e = (M_{He}+M_H)/(M_{He}+M_H+1)$ are the reduced masses of nuclei and electron, M_{He} and M_H are the masses of the nuclei He and H. The $V_C(z,R)$ is the soft-core Coulomb potential for HeH^{2+} molecule:

$$V_C(z,R) = \frac{C_{He}C_H}{\sqrt{R^2+a}} - \frac{C_{He}}{\sqrt{(z-z_{He})^2+b}} - \frac{C_H}{\sqrt{(z-z_H)^2+b}}, \tag{2}$$

where z is the electronic coordinate (respect to the center of mass of the two nuclei) and R is the internuclear distance, $C_{He}=2$ and $C_H=1$ are the electric charges of nuclei, $z_{He} = \frac{-M_H R}{M_{He}+M_H}$ and $z_H = \frac{M_{He}R}{M_{He}+M_H}$ are the positions of He nucleus ($z_{He}<0$) and H nucleus ($z_H>0$). The soft-core parameters are adopted to $a=0.01$ and $b=0.3$, which corresponds to the initial state ($2p\sigma$ state) of HeH^{2+} molecule with ionization potential 1.03 a.u. and equilibrium distance 3.89 a.u. [25], which is close to the observation in experiment [26]. The light-molecule interaction term has the form [25,27]

$$V_l(z,t) = \left[\frac{C_{He}M_H - C_H M_{He}}{M_{He}+M_H}R + \left(1 + \frac{C_{He}+C_H-1}{M_{He}+M_H+1}\right)z\right]E(t). \tag{3}$$

We use a grid range from 0 a.u. to 30 a.u. with $\Delta R = 0.1$ a. u. on the R-axis and from -100 a.u. to 100 a.u. with $\Delta z = 0.2$ a. u. on the z-axis, respectively. To avoid reflections from boundaries, the absorbing positions of the mask function are 50 a.u. and 100 a.u. grid points away from the boundaries on the R-axis and z-axis, respectively [28]. The harmonic spectrum is proportional to the modulus squared of the Fourier transform of the dipole acceleration, which is given by [27]

$$a(t) = -\langle\Psi(z,R,t)|\frac{dV_C(z,R)}{dz} + \left(1 + \frac{C_{He}+C_H-1}{M_{He}+M_H+1}\right)E(t)|\Psi(z,R,t)\rangle. \tag{4}$$

3. Results and discussion

We investigate the MHOHG of HeH^{2+} with the combination of a chirped laser and a unipolar pulse. The combined laser fields are the chirped laser pulse plus a unipolar pulse, and the chirped laser pulse minus a unipolar pulse, which can be written as the form, respectively

$$E_1(t) = E(t) + E_{uni}(t), \tag{5}$$

$$E_2(t) = E(t) - E_{uni}(t). \tag{6}$$

The chirped laser is defined as

$$E(t) = E_0 f(t)\cos[\omega_0 t + \phi(t) + \varphi], \tag{7}$$

where $f(t) = \exp[-2\ln 2(t^2/\tau^2)]$ is the pulse envelope and E_0 is the peak amplitude of the electric field, ω_0 is the frequency, τ is the full width at half maximum (FWHM) which is chosen to be 7 fs. The laser intensity is $I = 5\times10^{14}$ W/cm^2 with the wavelength $\lambda = 800$ nm. The time-varying carrier envelop phase (CEP) of the fundamental pulse is given by $\phi(t) = \beta[(t-t_0)/\tau_0)]^2$ [13], here $\beta = 7.1$ is the chirp parameter of the pulse, $\tau_0 = 210$ is the adjustable parameter to control the steepness of electric field, and $t_0 = 216$ is used to adjust the sweep range of electric field [15]. The CEP φ is set to 0 and π which corresponds to the CEP of the $E_1(t)$ and $E_2(t)$, respectively. The unipolar pulse is defined as [14]:

$$E_{uni}(t) = k_0\theta(t-t_0')E_0\left[\frac{k_1(t-t_0')^3\exp[-8(t-t_0')/\tau'']}{\tau''^3} - \frac{k_2(t-t_0')^5\exp[-(t-t_0')/\tau'']}{\tau''^5}\right], \tag{8}$$

where $\theta(t)$ is the step function which makes the unipolar pulse starting at t_0', and τ'' is the pulse duration of the unipolar pulse, which are set as $t_0' = -0.06(2\pi/\omega)$ and $\tau'' = 7$ fs, respectively. k_0 is a real number to control the electric field strength of the unipolar pulse ($k_0 = 1$), the parameters k_1, k_2 are chosen to be $k_1 = 400$ and $k_2 = 10^{-15}k_1$.

Fig. 1(a1) shows the electric fields of $E_1(t)$, where the laser profiles of the chirp-free laser, the chirped laser, and the chirped laser with CEP $\varphi=0$ plus a unipolar pulse are depicted. Compared with the chirp-free laser, the electric field strength of the chirped laser near the positive peak B' is evidently broadened. When the unipolar pulse is added to the chirped laser, we can see that the electric field strength of the combined laser near the positive peak B'' is broadened and enhanced, which will influence the MHOHG processes remarkably.

Fig. 1(b1) shows the MHOHG spectra of HeH^{2+} molecule driven by the chirp-free laser, the chirped laser, and the chirped laser with CEP $\varphi = 0$ plus a unipolar pulse, which is expressed by $E_1(t)$. We can see that a harmonic spectrum driven by the chirp-free laser with modulations is generated, which comes from the contributions of several different electron trajectories. Compared with the chirp-free case, the MHOHG cutoff energy driven by the chirped laser is extended with a lower harmonic intensity. Meanwhile, when the unipolar pulse is added to the chirped laser, a continuum spectrum with a spectral width of about 65 orders from 163 orders to 228 orders is generated.

Fig. 1 (a2) shows the electric fields of $E_2(t)$, where the laser profiles of the chirp-free laser, the chirped laser, and the chirped laser with CEP $\varphi = \pi$ minus a unipolar pulse are depicted. Compared with the chirp-free laser, the electric field strength of the chirped laser near the negative peak B' is evidently broadened. When the minus unipolar pulse is added to the chirped laser, we can see that the electric field strength of the combined laser near negative peak B'' is broadened and enhanced, which is similar to the case as shown in Fig. 1(a1).

Fig. 1 (b2) shows the MHOHG spectra of HeH^{2+} molecule driven by the chirp-free laser, the chirped laser, and the chirped laser with CEP $\varphi = \pi$ minus a unipolar pulse, which is expressed by $E_2(t)$. Compared with the chirp-free case, we can see that the