The dependence of high-order harmonic generation on the laser frequency and inter-nuclear distance from multi-atom molecular ions

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High-order harmonic generation from one-dimensional (1D) multi-atom molecular ions in an ultra-short laser field is theoretically investigated. The dynamics of the electron in a linearly polarized intense laser field is analyzed in terms of 1D Schrödinger equation with the Crank-Nicolson algorithm. The dependence of high-order harmonics on the laser frequency and the inter-nuclear distance is discussed. It is found that the optimum range of inter-nuclear distance should be changed to get extended harmonic generation for different laser frequency, and the lower frequency laser pulse is favorable to higher order harmonic generation as the inter-nuclear distance increases.

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High-order harmonic generation (HHG), as a potential tunable coherent light source in the extreme ultraviolet region[1−3] and a promising way for generating attosecond pulses[4] of high frequency radiation, has become one of the attractive research frontiers in recent years. The interaction of intense laser pulses with atoms, molecules, and clusters can give rise to high-order harmonics. HHG from atoms exposed to an intense ultra-short laser pulses has been investigated theoretically and experimentally in detail[5]. Generally, HHG from atoms in an intense laser field may well be understood[6]. When an atom is exposed to an intense laser field, the outside electron is firstly ionized by tunneling through the potential barrier, and then oscillates and accelerates as a free electron in the laser field, and finally the ionized electrons come back to their parent ions and transit back to the bound states, radiating high energy harmonic photons, i.e. generating high-order harmonic.

HHG from molecules and multi-atom molecular ions in an intense ultra-short laser pulse have many evident advantages over atoms for their higher degrees of freedom, in which laser-induced collisions of electrons with neighboring ions are expected to produce longer harmonic plateaus than that in single atomic system. Because of the potential advantages, the researches on the interaction of an intense laser field with more complex system of molecules[7−9] or clusters[10] have received great attention. They have not only revealed new mechanisms such as above-threshold dissociation[11], coulomb explosion[12], and enhanced ionization[13], but also given rise to the possibility of higher order harmonics[14,15]. Recently, Numico and his co-workers have shown that the interaction of a linear chain of molecular ions with an intense polarized laser field can generate a high-order harmonic spectrum with the maximum photon energy of even larger than $I_{\rm p} + 8U_{\rm p}$ with $I_{\rm p}$ and $U_{\rm p}$ representing the ionization potential and ponderomotive energy, respectively[16]. The cutoff of high-order harmonics can also be extended to $I_{\rm p} + 12U_{\rm p}$ for two electron systems of molecules and molecular ions due to the effect of electron-electron interactions[17]. In this paper, HHG from molecules or molecular ions with different inter-nuclear distance R is investigated, and the dependence of HHG on the inter-nuclear distance and the laser carrier frequency is discussed. The inter-nuclear distance and the laser carrier frequency have effects on the maximal kinetic energy of the ionized electron and thus on the generated harmonics. It is found that the lower frequency laser pulse is favorable to extended HHG as the inter-nuclear distance R increases.

The interaction of free electrons driven by the laser field $E_0 \cos(\omega t)$ can be classically described by the following equations[18] (au, i.e., atomic unit used below),

$$\ddot{Z}(t) = -E_0 \cos(\omega t + \varphi), \tag{1}$$

$$\dot{Z}(t) = -(E_0/\omega)\left[\sin(\omega t + \varphi) - \sin\varphi\right], \tag{2}$$

$$Z(t) = \alpha_0\left[\cos(\omega t + \varphi) + \omega t \sin\varphi - \cos\varphi\right], \tag{3}$$

where Z represents the distance from the ionized free electron to the parent ion or the neighboring ion, $\dot{Z}$ and $\ddot{Z}$ are the velocity and acceleration of the electron, respectively, E_0 is the maximum amplitude of the laser field, ω is the laser frequency, φ is the initial phase of laser field, $\alpha_0 = E_0/\omega^2$ is the quiver radius. The maximal kinetic energy is $E_{\rm k} = \dot{Z}^2_{\rm max}/2$.

To simulate the dynamics of the electron in a linearly polarized intense laser field, we solve the following time-dependent Schrödinger equation by the Crank-Nicolson method,

$$i\frac{\partial\psi(x,t)}{\partial t}=\left[-\frac{1}{2}\frac{\partial^2}{\partial x^2}+V(R,x)-xE(t)\right]\psi(x,t). \quad (4)$$

For small molecules or ionized clusters or metals, the multi-well potential[16] is a sum of softened Coulomb potential,

$$V(R,x)=\sum_{n=-(N-1)/2}^{(N+1)/2}-\frac{b}{\sqrt{a+(x+nR)^2}}, \quad (5)$$

where N is the number of atoms, here we take $N=7$, R is the distance between the neighboring atoms, and a and b are the parameters related to the softened potential, and can be used to adjust the energy of the ground state. Here, $a=b=1$ is used.

The laser field is given by

$$E(t)=E_0 f(t)\sin(\omega t+\varphi), \quad (6)$$

where $f(t)$ is the envelop function of the laser field. The ground state can be supposed as the initial state, and thus the HHG spectrum from the initial time to the final time can be calculated by

$$P_h(\omega)\propto|\mathrm{d}(\omega)|^2=|\int_{-\infty}^{+\infty}a(t)\exp(i\omega t)\mathrm{d}t|^2, \quad (7)$$

with the expectation value of the dipole acceleration

$$a(t)=\langle\psi(x,t)|-\frac{\partial V(x)}{\partial x}+E(t)|\psi(x,t)\rangle. \quad (8)$$

HHG spectra from the molecules or molecular ions with the inter-nuclear distance R of 10, 19, 25, and 32 au for different laser frequency are calculated respectively with the laser intensity $I=2.0\times10^{14}$ W·cm^{-2} ($E_0=0.0755$ au) and initial phase $\varphi=0$. For the laser wavelength $\lambda=800$ nm ($\omega=0.057$ au), it is clear that the harmonics are extended to higher order as R increases[15,19]. Furthermore, the results for the excitation laser wavelength of 600 nm indicate that harmonic generation displays a little difference from that of $\lambda=800$ nm. As shown in Fig. 1, high-order harmonic plateau is extended to longer as R increases from 10 to 25 au, however the harmonic order is decreased as R increases to more than 32 au. The maximal kinetic energy of the gained electron is reduced, as a result, the harmonic order is decreased for $R=32$ au. For the laser wavelength of 400 nm, it is found that the cutoff of high-order harmonics red-shifts at the inter-nuclear distance from 10 to 19 au, and it red-shifts much more for the inter-nuclear distance of $R=25$ and 32 au. Figure 2 shows the high-order harmonic spectra at the inter-nuclear distance of $R=7$, 10, 13, and 19 au, respectively. The cutoff of high-order harmonics is extended to higher order as the inter-nuclear distance increasing from 7 to 10 au, whereas it shifts to lower order as the inter-nuclear distance increasing more than 13 au. According to Eqs. (1)—(3), the highest harmonic order are 18, 19, 16, 15 for the inter-nuclear distance of $R=7$, 10, 13, and 19 au, respectively. Here, it can be seen that the extended harmonics can be achieved as the inter-nuclear distance decreasing at the laser wavelength of 400 nm.

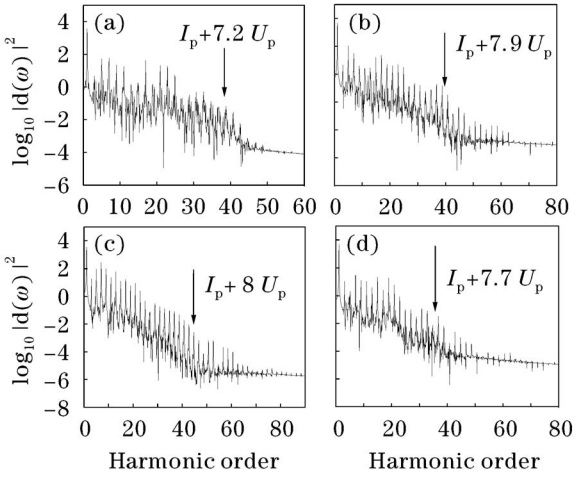

Fig. 1. HHG spectra from 7-atom molecular ions with the inter-nuclear distances of R =10 (a), 19 (b), 25 (c), and 32 au (d) in an ultra-short strong laser field at $I=2\times10^{14}$ W/cm^2 and $\lambda=600$ nm.

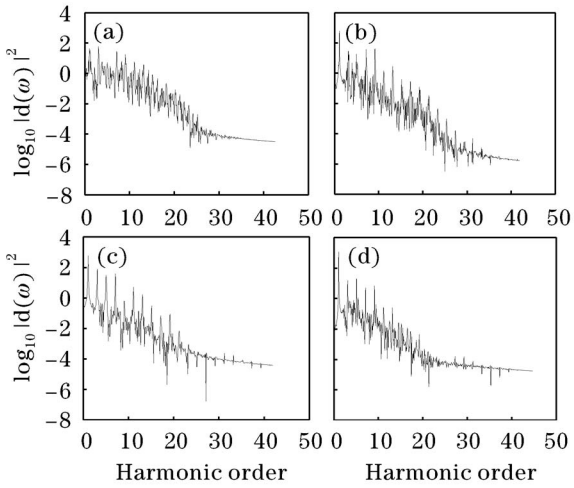

Fig. 2. HHG spectra from 7-atom molecular ions with the inter-nuclear distances of $R=7$ (a), 10 (b), 13 (c), and 19 au (d) in an ultra-short strong laser field at $I=2\times10^{14}$ W/cm^2 and $\lambda=400$ nm.

According to the classical theory, the resulting high-order harmonics depend on the maximal kinetic energy of the ionized electron, which is related to the interaction of the laser frequency ω and the inter-nuclear distance R, so different harmonic order can be obtained at different kinetic energy. The quiver radii are is 23, 13, and 5.8 au corresponding to the laser wavelengths of 800, 600, and 400 nm, respectively, so the optimal inter-nuclear distance for extended HHG is decreased. Consequently, extended harmonics can be only generated at optimum range of inter-nuclear distance for different laser frequency. With the increase in the inter-nuclear distance, high-order harmonics can be well extended to higher order at the laser wavelength of 800 nm, and the cutoff shifts to lower order from 800 to 600 and then to 400 nm, and so the lower frequency laser pulse is favorable to higher order harmonics.

In conclusion, HHG from one-dimensional (1D) multi-atom molecular ions in an ultra-short field is theoretically analyzed. The maximal kinetic energy depends on

the laser frequency and the inter-nuclear distance, and the quiver radius decreases at the laser wavelength from 800 to 600 and then to 400 nm. So, with the increase in the inter-nuclear distance, high-order harmonics can be well extended to higher order at the laser wavelength of 800 nm, but the harmonic order is decreased at the laser wavelengths of 600 and 400 nm, and thus the lower frequency laser pulse is favorable to higher order harmonics as the inter-nuclear distance increasing. Also, different range of inter-nuclear distance should be chosen to extended harmonics for different laser frequency.

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