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Attosecond-resolution two-electron harmonic emission

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ABSTRACT

Two-electron harmonic emission from the He atom has been investigated by solving the two-electron time-dependent Schrödinger equation (TDSE), which exhibits an extended plateau and many new harmonic cutoffs beyond the classical single-electron harmonic cutoff. Theoretical analyses show that these extended new cutoffs are caused by the sequential double recombination and the nonsequential double recombination of the two electrons, which is a general characteristic for the two-electron harmonic emission and which is revealed through the investigation on laser parameter effects. Moreover, from analyzing the time-dependent wave functions, the motions of the two electrons and the single and double ionization time have been described.

1. Introduction

High-order harmonic generation (HHG) is an effective method to produce the ultrashort attosecond sources [1–7]. It can be used to efficiently explore the electron dynamics in atoms and molecules [8–10]. For single-electron model system, HHG process can be well explained by the 'three steps' model of ionization–acceleration–recombination [11]. Within this model, a maximum harmonic cutoff with $E_{\max} = I_p + 3.17U_p$ can be reached. Here, I_p is the ionization potential and U_p is the ponderomotive energy of the free electron in the laser field ($U_p = I/4\omega^2$). In single-electron model, only the outer electron is active while the others are frozen in their ground state. However, for a multi-electron system with electron correlation effect, the single-electron model is deficient and cannot explain the phenomena such as one photon/two photons sequential or nonsequential double ionization [12–15], double excited states harmonic generation [16,17], etc.

Recently, Koval et al. [18] theoretically investigated the two-electron harmonic generation in the intense pulse. And, a new mechanism named the nonsequential double recombination (NSDR) on the two-electron harmonic generation has been first discovered and illustrated [18]. However, there are still many unknown/unclear phenomena on the multi-electron dynamics, such as (i) the electron correlation effect on the multi-electron excitation, ionization or recombination; (ii) how do the multi-electrons move after they are excited and ionized; (iii) how do the classical single-electron phenomena change when the additional inner electrons are considered, etc. Therefore, multi-electron dynamics is necessary for better understanding the electron motions and harmonic emission. Thus, in this paper, by solving the two-electron time-dependent Schrödinger equation (TDSE), we further investigated two-electron harmonic emission and two-electron motion from the He atom irradiated by intense laser field. The theoretical results showed an extended plateau and many more new cutoffs beyond the classical single-electron and the two-electron nonsequential double recombination harmonic cutoffs. Further analyses revealed that these extended new cutoffs are caused by the sequential double recombination and the nonsequential double recombination of the two electrons. Finally, the time-dependent probability density shows the two-electron motion and the time for the single and double ionization.

2. Theoretical methods

We consider a two-electron model where each electron is allowed to move along the laser polarization direction. Then, the TDSE with the dipole approximation in the length gauge can be written as (atomic units are used throughout this paper unless stated otherwise),

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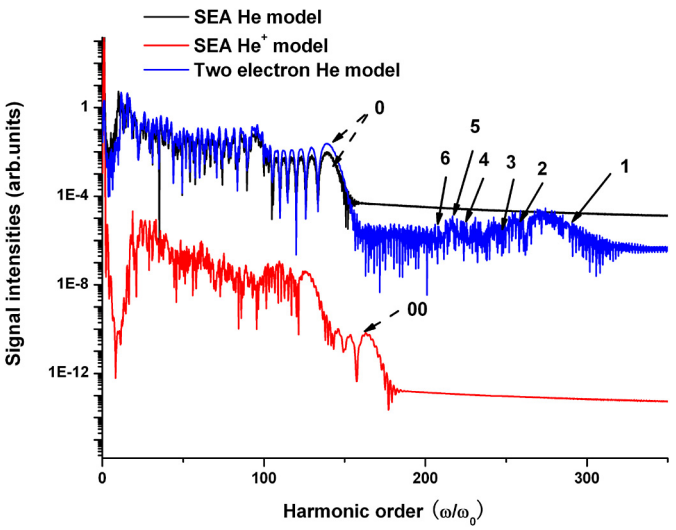

Fig. 1. HHG spectra calculated from the two-electron model for the He model (solid blue line) as well as from the SEA model for the He atom (solid black line) and the He^+ ion (solid red line). The laser field here is 5 fs/800 nm, $I = 1.0 \times 10^{15}$ W/cm^2. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

$$i\frac{\partial\varphi(x_1,x_2,t)}{\partial t} = \Big[-\frac{1}{2}\frac{\partial^2}{\partial x_1^2} - \frac{1}{2}\frac{\partial^2}{\partial x_2^2} - \frac{2}{\sqrt{x_1^2+a}} - \frac{2}{\sqrt{x_2^2+a}} + \frac{1}{\sqrt{(x_1-x_2)^2+b}} + (x_1+x_2)E(t)\Big]\varphi(x_1,x_2,t), \quad (1)$$

where the electron–nucleus interaction $V(x_i) = -\frac{2}{\sqrt{x_i^2+a}}$ $(i = 1, 2)$ and the electron–electron interaction $\frac{1}{\sqrt{(x_1-x_2)^2+b}}$ are modeled by the usual smoothed Coulomb potential with shielding parameter a and b. In our work, we study the He atom with the parameter $a = 0.5$ and $b = 0.329$ [19,20]. Propagation of the time-dependent electronic wave function $\varphi(x_1, x_2, t)$ can be carried out using the standard second-order split-operator method [21–26].

In the calculation, the linear polarization laser field can be expressed as,

$$E(t) = E\exp\big[-4\ln(2)t^2/\tau^2\big]\cos(\omega_0 t), \quad (2)$$

where E, ω_0, and τ are the amplitude, the frequency, and the pulse duration of the 5 fs/800 nm pulse.

According to the Ehrenfest theorem [27], the time-dependent dipole acceleration is calculated by,

$$a(t) = -\big\langle\varphi(x_1,x_2,t)\big|\frac{\partial V(x_1)}{\partial x_1} + \frac{\partial V(x_2)}{\partial x_2}\big|\varphi(x_1,x_2,t)\big\rangle - 2E(t). \quad (3)$$

By Fourier transforming the time-dependent dipole acceleration $a(t)$, the HHG spectrum is obtained as follows,

$$S(\omega) \sim \left|\int \exp(-i\omega_0 t)a(t)dt\right|^2. \quad (4)$$

3. Results and discussion

Fig. 1 shows the two-electron harmonic spectrum with the He atom irradiated by a 5 fs/800 nm, $I = 1.0 \times 10^{15}$ W/cm^2 pulse (solid blue line). For comparison, the harmonic spectra from the single-electron-approximation (SEA) calculation are also shown in the figure for the He atom (solid black line) and the He^+ ion (solid red line). The SEA model can be found in our previous investigations [28–30]. It can be seen that for the SEA region ($E_{\text{harmonic}} \leq I_{p1} + 3.17U_p$, indicated by arrow 0 and $I_{p1} = 24.6$ eV, the first ionization potential of He atom), the harmonic spectrum from the single-electron model is in a good agreement with that from the two-electron model. Moreover, the harmonic intensity from the He atom, calculated with the two-electron model, is almost 4 orders of magnitude higher than that from the He^+ ion with the SEA model. This demonstrates that the outer electron plays an important role in the harmonic emission process. On the other side, for the harmonic region beyond the SEA region, there is a weaker extended plateau and many new cutoffs in the two-electron harmonic emission spectrum. Here we identified and showed six new cutoffs indicated by the arrows of 1–6 with $E_1 = 293\omega_0$, $E_2 = 259\omega_0$, $E_3 = 249\omega_0$, $E_4 = 225\omega_0$, $E_5 = 215\omega_0$, $E_6 = 205\omega_0$.

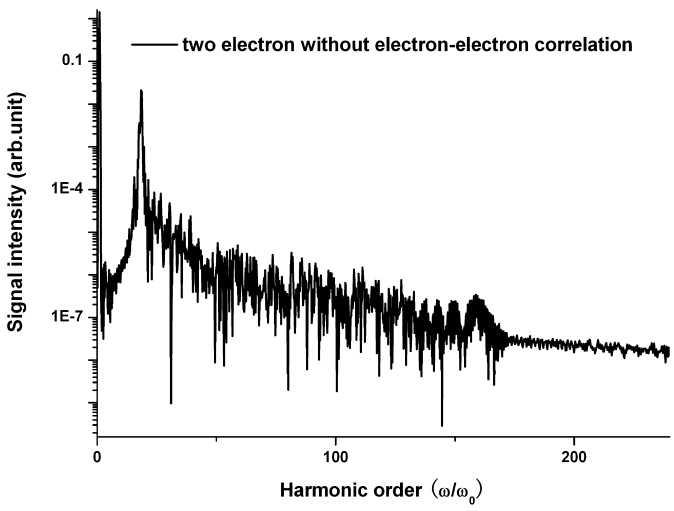

Fig. 2. HHG spectrum from the He atom calculated by the two-electron model but without the electron–electron correlation.

Fig. 2 shows the two-electron harmonic spectrum calculated without the electron–electron correlation term of $\frac{1}{\sqrt{(x_1-x_2)^2+b}}$. In this case, the two-electron model can be viewed as two independent He^+ ions. The laser pulse is the same as that in Fig. 1. Clearly, the extended new plateau is not shown up by switching off the electron–electron correlation, which means that the electron correlation is critical and determinant for the appearance of the extended new plateau in the harmonic spectrum.

To illuminate the two-electron harmonic emission process, in Fig. 3, we proposed a two-electron recombination model based on the nonsequential double recombination theory proposed by Koval et al. [18]. First, in the single-electron model, the electron follows the three steps of ionization–acceleration–recombination in every half of the laser cycle. The harmonic energy of each half cycle E_{HC} is estimated by $E_{HC} = \frac{1}{2}v^2$, $v = -\int E(t)dt$. If we consider a pulse with three half cycles, then, as shown in Fig. 3(a), the recombination of the single electron will occur three times (Fig. 3(a) right column, peaks 1–3) to produce three different cutoffs E_{HCi} $(i = 1–3)$ (Fig. 3(a) left column, three different color lines). In a two-electron model with the assumption that the two electrons are released by multi-photons sequential ionization, single recombination of each of the two electrons will occur also three times under the laser pulse of three half cycles to generate three different cutoffs E_{HCi} $(i = 1–3)$. Additionally, the double recombination of the two electrons arising from the electron–electron correlation, will cause emission of a single photon with higher kinetic energy (see Figs. 1 and 2), which is responsible for the extended new plateau on the two-electron harmonic spectrum [18]. First, if