Effect of electron correlation on high-order harmonic generation in helium model atom*

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High-order harmonic generation (HHG) of a helium model atom in an intense laser field has been numerically investigated. The influence of electron correlation on HHG is analysed by changing the strength between the electrons. The numerical results show that as the electron interaction strength becomes small, the first ionization energy increases rapidly, which results in the decrease in ionization. So the conversion efficiency of the high harmonic lying in the plateau decreases greatly, while the cutoff harmonic order in the harmonic spectrum increases.

Keywords: electron correlation, high-order harmonic generation, B-spline

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1. Introduction

High-order harmonic generation (HHG) has been one of the most intensively investigated subjects for more than twenty years because of its significance in applications.[1,2] In addition to producing attosecond pulses,[3–5] HHG also opens up great opportunities for resolving microscopic processes with angstrom precision.[6] The typical harmonic spectrum decreases for the first few harmonics, then displays a long plateau with almost the same conversion efficiency, and ends up with a rapid cutoff. If the keldysh parameter $\gamma = \sqrt{I_p/2U_p} \ll 1$ (where I_p is the ionization energy of the active electron, and $U_p = I/4\omega^2$ is its ponderomotive energy in the laser field with intensity I and frequency ω), the ionization process is described by tunneling, and the physics picture of HHG can be understood by using the widely accepted three-step model.[7,8] Firstly, the active electron tunnels through the barrier formed by the Coulomb potential and the laser field into the continuum with zero velocity; secondly, the electron is driven away from or back to the parent ion by the laser field; finally, the electron recombines with the parent ion, returns to the ground state, and emits a harmonic photon. The maximum kinetic energy of an ionized electron acquired from the oscillating laser field when it is drawn back to the parent ion is $3.17U_p$. Thus, the maximal photon energy emitted is $I_p + 3.17U_p$, which determines the cutoff location of the harmonic spectrum. If $\gamma \gg 1$, the ionization process is described by multiphotons.

In the past, HGG has always been investigated using the single-active-electron (SAE) model,[9] in which only one active electron can be excited or ionized, while the other electrons are frozen in their ground state orbitals. In the SAE model, both the electron correlation and the role of double excited state are neglected. Thus, comparing the HHG calculation adopting the hyperspherical coordinate theory with that of the SAE approach, Guan *et al.*[10] attributed the difference to the electron correlation. Furthermore, the same harmonic strength is predicted for all noble gases in the SAE approach,[11] while the experiments show that the heavier noble gases produce more intense harmonics than the light ones,[12] so the behavior of many-electron dynamics manifests in the HHG.[11,13] It is difficult to study the effect of electron correlation on the laser–atom processes treating every electron with equal footing in three dimensions using the *ab initio* methods. Thus, a number of models considering the electron correlation are proposed. One partially correlated helium model atom model describes the outer electron with a constant effective potential of the atomic core and the inner electron with the time-dependent potential of the outer electron.[14] A shortcoming of the model is that the dynamic correlation experienced by the outer electron due to the inner one is neglected. One improved two-electron model has been proposed by Hu *et al.*,[15,16] in which each electron is affected by the time-dependent Coulomb potential of the other electron. Another model was developed by Liu *et al.* for the study of nonsequential double ionization of a helium atom in an intense laser field,[17] in which the interaction strength between electrons can be adjusted by changing the repulsion charge.

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In this paper, we theoretically investigate the effect of electron correlation on the HHG by adjusting the interaction strength between electrons in the model.[17] The results show that as the interaction between electrons becomes small, the first ionization energy increases, which results in the extended harmonic plateau and the decrease in the harmonic conversion efficiency. The organization of this paper is as follows. Firstly, the method for solving the time-dependent Schrödinger equation (TDSE) with B-spline[18] and split-operator approach[19,20] is described briefly in Section 2. Secondly, the numerical results and analysis are displayed in Section 3. Finally, our conclusions are presented in Section 4. Atomic units are used throughout this paper.

2. Model and numerical methods

We consider a one-dimensional helium model atom, each electron is allowed to move along the polarization direction. The time-dependent Schrödinger equation with dipole approximation in the length gauge is written as

$$\mathrm{i}\frac{\partial\Psi(Z_1,Z_2,t)}{\partial t} = \left[\sum_{j=1}^{2}\left(-\frac{1}{2}\frac{\partial^2}{\partial Z_j^2} - V(Z_j) + Z_jE(t)\right) + \frac{\alpha}{\sqrt{(Z_1-Z_2)^2+b}}\right]\Psi(Z_1,Z_2,t), \quad (1)$$

where Z_1 and Z_2 are the electronic coordinates parallel to the laser polarization axis; $V(Z_j) = -2/\sqrt{Z_j^2+a}$ is the potential between the nuclear and the electron; the softening parameters a and b are equal to 0.55;[21] and the repulsion charge α denotes the interaction strength between the electrons,[17] $\alpha = 1$ unless stated otherwise. The laser field $E(t)$ is equal to $E_0 f(t)\cos(\omega t)$, where $E_0 = 0.14$ a.u. is the amplitude of the electric field, $\omega = 0.0857$ a.u. is the laser frequency, $T = 2\pi/\omega$ is the period, and $f(t)$ has the form

$$f(t) = \begin{cases} \dfrac{t}{2T}, & 0 \le t \le 2T, \\ 1, & 2T \le t \le 10T, \\ -\dfrac{t}{2T}+6, & 10T \le t \le 12T. \end{cases} \quad (2)$$

At $t = 0$, the wave function is expanded by B-spline

$$\Psi(Z_1,Z_2,t=0) = \sum_{i,j=1}^{N} C_{ij}B_i^k(Z_1)B_j^k(Z_2), \quad (3)$$

where B_i^k is the B-spline function of order $k = 7$ and $N = 50$. We obtain the ground state energy and the corresponding wave function by using the linear variation method. Then, we distribute the ground wave function in a two-dimensional space Z_1 and Z_2. The time-dependent wave function can be obtained by using the split-operator method.[19,20]

In our calculation, we employ a box of size $|Z_j| \le 100$ a.u. ($j = 1,2$), containing 1000 equal grid points. The propagation time is 12 optical cycles, and the time step is $\delta t = 0.01$ a.u. In order to avoid the reflection of wave function from the boundaries, a $\cos^{(1/8)}$ mask function in $|Z_j| = 70$–100 a.u. ($j = 1,2$) is multiplied after every time step δt.[22]

As mentioned above, the outgoing wave packet is absorbed with a mask function. So the ionization probability is time-dependent

$$P_0(t) = 1 - \int_{-Z_{1\max}}^{Z_{1\max}}\int_{-Z_{2\max}}^{Z_{2\max}} \left|\Psi(Z_1,Z_2,t)\right|^2 \mathrm{d}Z_1\,\mathrm{d}Z_2. \quad (4)$$

In order to properly consider the contributions from single and double ionizations, the calculating area is divided into three parts for the two electrons:[19] bound state area $P_\mathrm{b}(t)$, $\max(|Z_j|) \le R$; double ionization area $P_\mathrm{d}(t)$, $\min(|Z_j|) > R$; single ionization area $P_\mathrm{s}(t)$, $\min(|Z_j|) > R$ and $\max(|Z_i|) < R$ ($i \ne j$), j (i) $= 1,2$. Here, R is equal to 5 a.u. (we also let R be 8 a.u., and the results remain almost the same).

Then, we can calculate the field-induced dipole moment in the acceleration form

$$a(t) = -\left\langle \Psi(Z_1,Z_2,t)\left|\sum_{j=1}^{2}\frac{\partial V(Z_j)}{\partial Z_j}\right|\Psi(Z_1,Z_2,t)\right\rangle - 2E(t). \quad (5)$$

The high-order harmonic power spectrum can be evaluated by the modulus squared of the Fourier transform of the dipole moment in time span $[t_\mathrm{i}, t_\mathrm{f}]$[23]

$$P(w) = \left|\frac{1}{t_\mathrm{f}-t_\mathrm{i}}\frac{1}{\omega^2}\int_{t_\mathrm{i}}^{t_\mathrm{f}} a(t)\,\mathrm{e}^{-\mathrm{i}\omega t}\,\mathrm{d}t\right|^2. \quad (6)$$

3. Numerical results and analysis

In Table 1, we illustrate the first and the second ionization potentials. As α decreases, the first ionization potential increases, while the second ionization potential remains the same. The ponderomotive energy U_p is equal to 0.671 a.u.

Table 1. Comparison of the first ionization potential I_p1 and the second ionization potential I_p2 for different α.

α	1	0.8	0.5	0
I_p1	0.977	1.158	1.437	1.920
I_p2	1.920	1.920	1.920	1.920

In Fig. 1, we present the single-ionization probability, the double-ionization probability, and the bound-state probability for different α, where $P(t) \times 10^n$ means $P(t)$ multiplied by 10^n. In the laser–atom interaction process, the bound-state probability is dominant over the single-ionization and the double-ionization probabilities. For $\alpha = 1$, the single ionization occurs at $t = 2T$ and remains almost the same until after that. The double ionization occurs after $t = 4T$. At $t = 4T$,

the single-ionization probability is about 0.02, and the double-ionization probability is about 0.002. For $\alpha = 0.8$, the single ionization mainly occurs at $t = 3T$ and remains nearly the same after that, and the double ionization mostly takes place after $t = 4T$. At $t = 4T$, the single-ionization probability is about 0.004, and the double-ionization probability is about 0.0002. For other α, both single-ionization and double-ionization probabilities are very small. The results demonstrate that as α decreases, the single-ionization probability becomes smaller because of the increase of the first ionization potential as illustrated in Table 1. For the same α, the single-ionization probability is dominant over the double-ionization probability. Thus, we can neglect the contribution from the double ionization.

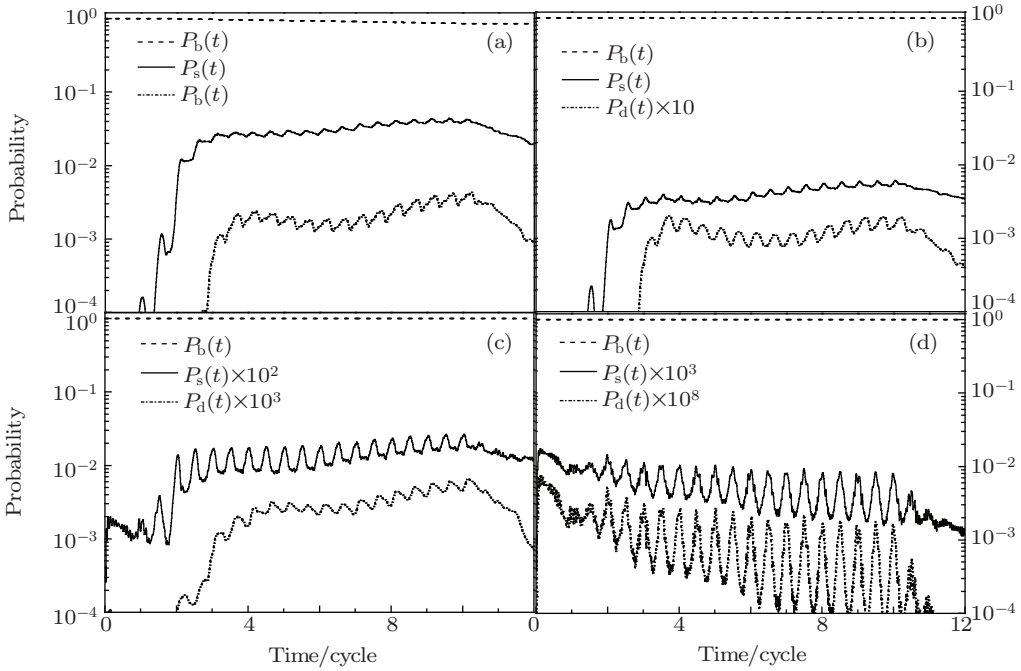


Fig. 1. Comparison of the time-dependent bound state probability, single ionization probability, and double ionization probability for (a) $\alpha = 1.0$, (b) $\alpha = 0.8$, (c) $\alpha = 0.5$, and (d) $\alpha = 0$.

In Fig. 2, we illustrate the ionization probability for different α. As α decreases, the ionization probability decreases, since the first ionization potential increases. At the end of the pulse, the ionization probability is about 0.12 for $\alpha = 1$, 0.01 for $\alpha = 0.8$, and small for $\alpha = 0.5$ and $\alpha = 0$.

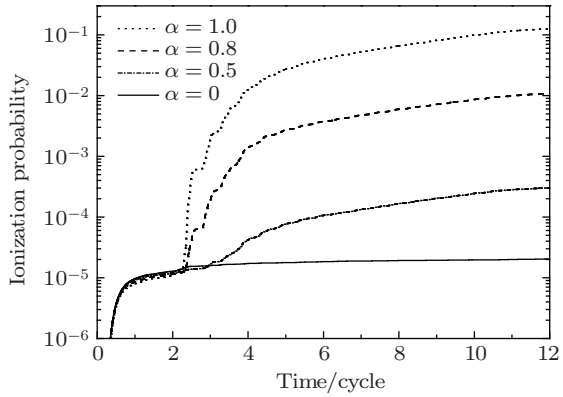


Fig. 2. Time-dependent ionization probability for different α.

In Fig. 3, we show the harmonic spectra for different α. As α gets smaller, the cutoff harmonic order of the harmonic spectrum becomes bigger, and the conversion efficiency of the plateau decreases. The reason for this is that the first ionization potential increases, which results in the decrease of the ionization probability. For $\alpha = 1.0$, the cutoff in the harmonic spectrum is around the 36th order. For $\alpha = 0.8$, it is about the 38th order. For $\alpha = 0.5$, there is a resonance around the 9th harmonic because the difference between the ground state energy and the first excited state energy is equal to about 9 photon energy displayed in Table 2. The cutoff in harmonic spectrum occurs at about the 41th order. For $\alpha = 0$, there is also a resonance around the 12th harmonic because the difference between the ground state energy and the first excited state energy is equal to about 12 photon energy as illustrated in Table 2. The cutoff in the harmonic spectrum occurs at around the 47th order.

Table 2. Comparison of the ground state energy E_0, the first excited state energy E_1, and the resonant harmonic order N_r for different α.

α	E_0	E_1	N_r
0	–3.840	–2.829	12
0.5	–3.357	–2.552	9

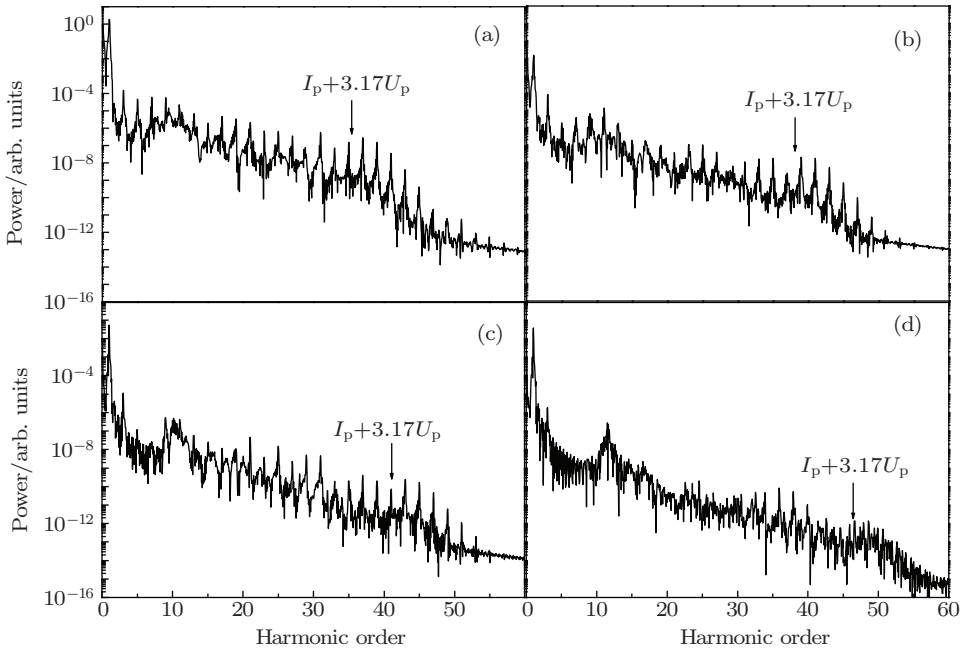


Fig. 3. The HHG in the intense laser field for (a) $\alpha = 1.0$, (b) $\alpha = 0.8$, (c) $\alpha = 0.5$, and (d) $\alpha = 0$.

4. Conclusion

We calculate the single-ionization probability, the double-ionization probability, the ionization probability, and the HHG for different interaction strength between electrons. The results demonstrate that as the electron correlation becomes small, the first-ionization energy increases, and the single-ionization probability is dominant over the double-ionization probability. Thus, the cutoff frequency of the HHG becomes bigger, and the conversion efficiency of HHG lying in the plateau decreases. For $\alpha = 0.5$ and $\alpha = 0$, there is a resonance in the harmonic spectrum because the difference between the ground state energy and the first excited state energy equals integer photon energy.

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References

[1] Brabec T and Krausz F 2000 *Rev. Mod. Phys.* **72** 545
[2] Krausz F and Ivanov M 2009 *Rev. Mod. Phys.* **81** 163
[3] Zou P, Li R X, Zeng Z N, Xiong H, Liu P, Leng Y X, Fan P Z and Xu Z Z 2010 *Chin. Phys.* B **19** 019501
[4] Wu J, Zhai Z and Liu X S 2010 *Chin. Phys.* B **19** 093201
[5] Zhang G T, Bai T T and Zhang M G 2012 *Chin. Phys.* B **21** 054214
[6] Niikura H and Corkum P B 2006 *Adv. At. Mol. Opt. Phys.* **54** 511
[7] Corkum P B 1993 *Phys. Rev. Lett.* **71** 1994
[8] Lewenstein M, Balcou P, Ivanov M Y, L'Huillier A and Corkum P B 1994 *Phys. Rev.* A **49** 2117
[9] Krause J L, Schafer K J and Kulander K C 1992 *Phys. Rev.* A **45** 4998
[10] Guan X X, Tong X M and Chu S I 2006 *Phys. Rev.* A **73** 023403
[11] Gordon A, Kärtner F X, Rohringer N and Santra R 2006 *Phys. Rev. Lett.* **96** 223902
[12] Wahlström C G, Larsson J, Persson A, Starczewski T, Svanberg S, Salières P, Balcou P and L'Huillier A 1993 *Phys. Rev.* A **48** 4709
[13] Sukiasyan S, McDonald C, Destefani C, Ivanov M Y and Brabec T 2009 *Phys. Rev. Lett.* **102** 223002
[14] Watson J B, Sanpera A, Lappas D G, Knight P L and Burnett K 1997 *Phys. Rev. Lett.* **78** 1884
[15] Hu S X, Qu W X and Xu Z Z 1998 *Phys. Rev.* A **57** 3770
[16] Prager J, Hu S X and Keitel C H 2001 *Phys. Rev.* A **64** 045402
[17] Liu W C, Eberly J H, Haan S L and Grobe R 1999 *Phys. Rev. Lett.* **83** 520
[18] Bachau H, Cormier E, Decleva P, Hansen J E and Martín F 2001 *Rep. Prog. Phys.* **64** 1815
[19] Liu C D, Zeng Z N, Wei P F, Liu P, Li R X and Xu Z Z 2010 *Phys. Rev.* A **81** 033426
[20] Cai J, Wang L M and Qiao H X 2009 *Chin. Phys.* B **18** 1458
[21] Bauer D 1997 *Phys. Rev.* A **56** 3028
[22] Yu H T and Bandrauk A D 1997 *Phys. Rev.* A **56** 685
[23] Bian X B, Qiao H X and Shi T Y 2006 *Chin. Phys. Lett.* **23** 2403