The quantitative analysis of enhancement of high-order harmonics in two-color intense laser fields

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The time-dependent Schrödinger equation of the interaction of laser pulse with He⁺ is solved by using the asymptotic boundary condition and symplectic algorithm in fundamental laser-field and two-color laser fields. We find that the conversion efficiency of high-order harmonic generation (HHG) is higher in the two-color laser fields than in the fundamental laser field, especially for the combination of $\omega_0 - 19 \omega_0$. To explain these phenomena, the ionization probability, the average distance, the probability of first excited state, and the transition probability are calculated. We give the qualitative and quantitative analysis for the enhancement of conversion efficiency of HHG.

KEY WORDS: High-order harmonic generation, two-color laser field, symplectic algorithm, transition probability, quantitative analysis

1. Introduction

Recently, with the development of the ultra-short intense laser technology, the short-pulse laser with the maximal energy about 10^{21} W/cm² can be obtained in experiments, which prompt the research on the interaction between materials and the intense laser field. Those surprising developments make the research on atom, molecule, and cluster physics in intense laser field become very interesting and excited topics in the basic theory research [1–3].

The high-order harmonic generation (HHG) spectra have a generic shape [4]. Quantum theory of HHG as a three-step process had been presented and accepted [5]. Recently, the investigation of HHG has become an active field. For example, the clustering environment is responsible for the enhancement of harmonic emission [3]; high-order harmonic emission from He⁺ is enhanced by many orders of magnitude in the two-color laser field compared with the case of the fundamental pulse alone [6]; the efficient HHG in a two-color laser field had been observed in the experiment [7]; highly efficient HHG in Helium had

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achieved by using an orthogonally polarized two-color laser field [8], and efficient HHG was obtained below 10 nm using a linearly polarized two-color laser field [9], etc.

The time-dependent Schrödinger equation (TDSE) of the interaction of the laser with atom can be solved numerically by Floquet theory method [10], R-matrix method [11], split-operator technique [12], etc. The infinite space has to be truncated into a finite space in numerical solving, since the electron has the population in the large distance, the values of the wave-functions at the finite boundary cannot be simply taken as zero. So the artificial boundary conditions, e.g. absorber or mask functions [13], are often applied to solve the TDSE so as to eliminate the reflection of the wave-functions on the boundaries.

The numerical results and the analysis will be demonstrated in section 2, and the conclusion will be given in section 3.

2. Numerical results and discussions

In this paper, the asymptotic boundary condition (ABC) [14, 15] and symplectic algorithm [16–19] are applied to solve the 1-dimensional TDSE. The softpotential of He⁺

$$V_0(x) = -\frac{2}{\sqrt{x^2 + 0.5}}$$

is considered in the computation.

The laser field was chosen to be

$$\varepsilon(t) = \varepsilon_0 \sin^2(\omega_0 t/2N)(\sin(\omega_0 t) + r\sin(n\omega_0 t)), \quad 0 \le t \le NT_0,$$

where $\varepsilon_0 = 0.3 \text{ a.u.}$ and $\omega_0 = 0.056 \text{ a.u.}$ are the peak amplitude and the frequency of fundamental laser field, respectively. *n* is the ratio of high frequency to fundamental frequency, N = 5 is the number of optical period, $T_0 = 2\pi/\omega_0$ is the optical period. And the space boundary *R* is set as 500 a.u.

The ionization probability, the average distance, the first excited state probability, the transition probability and the harmonic spectrum are shown in figures 1–5 under the fundamental laser field (case 1: r = 0) and the two-color laser fields with combination of fundamental laser field (ω_0) and its *n*th harmonic field (case 2: r = 0.1 and n = 2; case 3: r = 0.1 and n = 19; case 4: r = 0.1 and n = 30).

As shown in figure 1, the ionization probability in the two-color laser fields is higher than the case in the fundamental field, but the ionization probability is not linear increasing with the increasing of the value of n. The yield of ionization is most when n = 19, the mixing of 2nd harmonic and 30th harmonic did not obviously affect the changes of ionization. This reveals that the main electron wave packet is bounded for the cases 1, 2, and 4 but some probability of the electron wave packet is unbounded for the case 3.



Figure 1. The ionization probability in the one-color laser field and the two-color laser fields.



Figure 2. (a) The average distance in the one-color laser field (r = 0) and the two-color laser fields; (b) The enlarged picture of a part of (a).

Figure 2(a) shows the average distance of the electron for four cases; figure 2(b) is the enlarged picture of a part of figure 2(a). We can see from figure 2(a) and (b) that the amplitude of average distance and the vibrational frequency of electron are increased in the two-color laser fields comparing with fundamental laser field, but the maximums are largest when n = 19. Namely, the probability of electron returning to the core in the unit time can be greatly increased for the case 3.

The harmonic spectra are shown in figure 3(a); figure 3(b) is the enlarged picture of a part of figure 3(a). From figure 3(a) and figure 3(b) we can see that the conversion efficiency of HHG is enhanced when the two-color laser fields are applied. The values of conversion efficiency of HHG for cases 2, 3, and 4 are about zero-order, two-order, and one-order of magnitude, respectively, higher than that for the case 1.



Figure 3. (a) The Harmonic spectra in the one-color laser field and the two-color laser fields; (b) The enlarged picture of a part of (a).



Figure 4. The First excited state probability in the one-color laser field and the two-color laser fields.



Figure 5. The transition probability in the one-color laser field and the two-color laser fields.

Figures 1 and 2 shown that the vibration of the electron is quicker near the ion core and the amplitude is bigger in the two-color laser fields than the cases in the fundamental laser field and the effect is obvious for case 3. The quicker vibration leads to increase the probability of re-collision of electron with ion core so that more high-energy photons can be radiated. The effect on the ionization and the average distance with mixing of 19th harmonic is similar to the phenomenon in [14]. For the analysis of above the conversion efficiency of HHG should be enhanced in the two-color laser fields especially with the mixing of 19th harmonic.

Zeng et al. [20] had given that for a certain level structure, a specially chosen high frequency additional driving field can greatly enhance the efficiency of HHG. Because the energy of single-photon of the 19th harmonic (1.064 a.u.) is around the energy level difference between the ground state and the first excited state (1.068 a.u.). Thus the population of the first excited state will be increased by the single photon transition from the ground state to the first excited state. The first excited state probability in the fundamental laser field and the two-color laser fields as shown in figure 4 is consistent with our analysis. The first excited state probability is almost the same in the fundament laser field as in the twocolor laser fields with combination of $\omega_0 - 2\omega_0$ and $\omega_0 - 30\omega_0$, but it is enhanced obviously with combination of $\omega_0 - 19\omega_0$. At the same time the continuum state has definite population as shown in figure 1. Based on the selection rule of transition, channels of the transition from the bound state to the odd parity and even parity continuum are unobstructed when the 19th harmonic is mixed.

When the 19th harmonic is mixed, the electron leaves and returns to ion core quickly as shown in figure 2, at the same time the continuum state probability is increased, which provides the more probability of electron return and recollision with ion core. As shown in figure 3, the plateau of HHG is the highest when the 19th harmonic is mixed.

Above is the qualitative analysis for the enhancement of conversion efficiency of HHG. Next the corresponding quantitative analysis will be given by the transition probability with radiation 240th–260th harmonic of electron.

The transition probabilities with radiating 240th–260th harmonic are calculated and summed on the total pulse. The total transition probabilities are shown in figure 5. It reveals that the transition probability with radiation 240th–260th harmonic in the two-color fields with combination of $\omega_0 - 2\omega_0$, $\omega_0 - 19\omega_0$, and $\omega_0 - 30\omega_0$ is about zero-order, two-order, and one-order of magnitude, respectively, higher than that in the fundamental field. The relative difference of these values are consists with the order of magnitude of enhancement of conversion efficiencies of 240th–260th harmonic in figure 3. The results tell us that the order of magnitude of increasing of the transition probability is about equal to the order of magnitude of enhancement of conversion efficiency of HHG.

Based on the theory of three-step process [1, 5], the cause of HHG is that the energetic photons are emitted while electron returns to bound states from

the continuum state, which is the transition between the continuum state and the bound states. So the more transition probability is, the higher conversion efficiency of HHG is. As it is single photon radiation in the process of the electron re-collision with the ion core from the continuum, the enhancement of transition rate should be equal to the enhancement of conversion efficiency of HHG. When the transition probability is increased, the conversion efficiency of HHG is enhanced, and the magnitudes of enhancement should be equal. The analysis is the same as the numerical results as shown in figures 3 and 5. Thus, we can analyze quantitatively the enhancement of conversion efficiency of HHG by the transition probability.

3. Conclusions

In this paper, the TDSE of the interaction of laser pulse with He⁺ is solved by using the asymptotic boundary condition and symplectic algorithm in fundamental laser field and two-color laser fields. The ionization probability, the average distance, the harmonic spectrum, the first excited state probability, and the transition probability are calculated.

The numerical results illustrate that the conversion efficiency of HHG can be enhanced effectively in the two-color laser fields especially for the combination of $\omega_0 - 19\omega_0$. The mechanism of the obvious enhancement of HHG in the two-color laser fields is analyzed qualitative and quantitative.

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References

- [1] P.B. Corkum, Phys. Rev. Lett. 71 (1993) 1994–1997.
- [2] M. Lewenstein, P. Balcou, M.Y. Ivanov, A. L'Huillier and P.B. Corkum, Phys. Rev. A 49 (1994) 2117–2132.
- [3] S.X. Hu and Z.Z. Xu, Appl. Phys. Lett. 71 (1997) 2605–2607.
- [4] J.L. Krause, K.J. Schafer and K.C. Kulander, Phys. Rev. Lett. 68 (1992) 3535-3538.
- [5] M.Y. Kuchiev and V.N. Ostrovsky, Phys. Rev. A 60 (1999) 3111-3124.
- [6] K. Ishikawa, Phys. Rev. Lett. 91 (2003) 043002.
- [7] I.J. Kim, H.T. Kim, C.M. Kim, et al., Appl. Phys. B 78 (2004) 859-861.
- [8] I.J. Kim, C.M. Kim, H.T. Kim, et al., Phys. Rev. Lett. 94 (2005) 243901.
- [9] T.T. Liu, T. Kanai, T. Sekikawa and S. Watanabe, Phys. Rev. A 73 (2006) 063823.
- [10] R.M. Potvliege and R. Shakeshaft, Phys. Rev. A 40 (1989) 3061-3079.
- [11] P.G. Burke, P. Francken and C.J. Joachain, J. Phys. B 24 (1991) 761-790.
- [12] T.F. Jiang and S.I. Chu, Phys. Rev. A 46 (1992) 7322-7324.

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- [13] S.X. Hu and Z.Z. Xu, Phys. Rev. A 56 (1997) 3916–3922.
- [14] C.L. Zhang, X.S. Liu, P.Z. Ding and Y.Y. Qi, J. Math. Chem. 39 (2006) 451-463.
- [15] Y.Y. Qi, X.S. Liu, X.Y. Liu and P.Z. Ding, J. Math. Chem. 39 (2006)133-149.
- [16] K. Feng, J. Comput. Math. 4 (1986) 279-289.
- [17] Th. Monovasilis and T.E. Simos, Chem. Phys. 313 (2005) 293–298.
- [18] G. Avdelas and T.E. Simos, Phys. Rev. E 62 (2000) 1375-1381.
- [19] K. Tselios and T.E. Simos, J. Math. Chem. 34 (2003) 83-94.
- [20] Z.N. Zeng, R.X. Li, Y. Cheng, W.X. Qu and Z.Z. Xu, Acta. Opt. Sin. (in Chinese) 21 (2001) 1153–1156.