Multi-photon resonance enhanced super high-order harmonic generation*

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(Received 12 February 2010; revised manuscript received 17 March 2010)

This paper proposes highly charged ions pumped by intense laser to produce very high order harmonics. Numerical simulations and full quantum theory of Ne^{9+} ions driven by laser pulses at 1064 nm in the power range of 10^9 W/cm^2 $\sim$ 10^{15} W/cm^2 show that the emission spectrum corresponds to the electronic transitions from the excited states to the ground state, which is very different from the spectrum of general high-order harmonic generation. In such situation, harmonic order as high as 1000 can be obtained without producing lower order harmonics and the energy conversion efficiency is close to general high order harmonic generation of hydrogen atom in the same laser field.

Keywords: multi-photon excitation, high-order harmonic generation

PACC: 3280K

1. Introduction

The high-order harmonic generation (HHG)[1−20] is one of the nonlinear phenomena that atoms or molecules pumped by strong laser field radiate coherent photons with frequency from several to more than one hundred times of the incident photon. The HHG generally presents a characteristic distribution. For low laser intensity, it quickly decreases with the increase of the harmonic order, while for stronger fields, it shows three distinct regions: a rapid decrease from several to tens harmonic, a long plateau where the harmonics have almost constant amplitudes and a sharp cutoff at high order.[20−22] Considering that HHG spectrum contains a great deal of lower frequency radiation, we concern if the low-frequency radiation can be removed and a specific high order harmonics can be enhanced by multi-photon resonance via the bounded states of highly charged ions. If a harmonic order of several hundreds or even above one thousand can be enhanced, we can obtain coherent x-ray from infrared or visible light source. This may be implemented in a system composed of Ne^{9+} ions exposed to laser pulse at 1064 nm. Here, the transition from the ground state to the first excited state needs 875 photons of the laser, and the 875th harmonic, corresponding to 1.2 nm radiation, should be enhanced by the multi-photon resonance.

Nowdays, highly charged ions can be produced by intense laser ablation of solid targets,[23−25] laser induced cluster explosion[26,27] or electron beam ion trap (EBIT).[28−35] Laser induced cluster explosion is a technology highly approved in recent years to produce abundant highly charged ions of a specific pure element. These ion sources may be used to produce coherent monochromatic x-ray via multi-photon resonance enhanced HHG. In addition, as professional equipment for the research of highly charged ions, the spectrum detection of highly charged ions in the EBIT is a conspicuous problem. Multi-photon resonance enhanced HHG may be a technique better than other spectra methods. Because the energy difference between the bounded states is much larger than the photon energy of general light source, absorption spectrum is hardly used to detect the electronic structures. So the emission spectrum induced by collisions between the ions and electron beams is commonly employed in such study. However the detection efficiency is low because the collision cross section is small (10^{-18} cm^2 or below) and the spontaneous radiation is spatially isotropic. Clearly, it would be an effective approach to the problem that a specific order of HHG is enhanced by bounded states of the highly charged ions, producing coherent radiation that can be detected efficiently. This method can also be applied to plasma diagnoses and other situations of detecting

*Project supported by the National Natural Science Foundation of China (Grant No. 10574030).
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highly charged ions.

To explore the possibility of the multi-photon enhanced HHG, we employ the numerical resolution of the time-dependent Schrödinger equation (TDSE) for ions coupled with laser fields, and make a theoretical analysis based on full quantum theory. Since the solution of three-dimensional (3D) wave equation costs huge computational hours,[36−38] we first work with a one-dimensional (1D) model for obtaining some raw information. To test this information further in a 3D situation, the wave function is expanded by finite eigenstates of the ions. And then a full-quantum theoretical calculation is made to explain the mechanism. We focus on the HHG of hydrogen-like Ne^{9+} and Ca^{19+} ions pumped by laser pulse with its electric field much weaker than the ionic internal Coulomb field. Our calculations show that the emission spectrum presents more than ten specific resonant lines, precisely corresponding to the transitions from the excited states to the ground state. This phenomenon happens at laser strength of non-relativistic regime, and the harmonic order as high as 1000 can be easily obtained.

2. One-dimensional hydrogen-like ion

The Hamiltonian for 1D hydrogen-like ion is

$$\hat{H}_0 = -\frac{\mathrm{d}^2}{2\mathrm{d}x^2} - \frac{Z}{|x|}, \tag{1}$$

where Z is the charge of nucleus and x is the electronic coordinate. We use atomic unit through out this paper. The spectrum of $\hat{H}_0$ is given by

$$E_n = -\frac{Z^2}{2n^2} \quad (n = 1, 2, 3\ldots), \tag{2}$$

and each eigenenergy corresponds to a wave function with even parity

$$\psi_{n+}(x) = |x|\ \mathrm{e}^{-x/Zn} F(1-n,\ 2,\ 2\,|x|\,/Zn)/Z, \tag{3}$$

and a wave function with odd parity

$$\psi_{n-}(x) = \begin{cases} \psi_{n+}(x), & x \geq 0, \\ -\psi_{n+}(x), & x < 0, \end{cases} \tag{4}$$

where

$$F(\alpha,\ \gamma,\ z) = \sum_{k=0}^{\infty} \frac{\alpha(\alpha+1)\cdots(\alpha+k-1)}{\gamma(\gamma+1)\cdots(\gamma+k-1)} z^k. \tag{5}$$

At the singular point $x = 0$, the wave function is always zero, both in the stationary states and the dynamic progress shown below. Clearly, this 1D model has the same energy spectrum as the 3D one and therefore we may expect that the two models behave similarly in response to laser pulses. Generally, the electric field of laser pulses polarised along the x direction can be expressed by

$$E = E_0 F(t) \sin(\omega t), \tag{6}$$

where $F(t)$ is the amplitude profile. Under dipole approximation, the total Hamiltonian of the ion coupled with the laser field is

$$\hat{H} = \hat{H}_0 + x E_0 F(t) \sin(\omega t), \tag{7}$$

and the TDSE is

$$\left[-\frac{\mathrm{d}^2}{2\mathrm{d}x^2} - \frac{Z}{|x|} + x E_0 F(t)\sin(\omega t)\right]\psi = \mathrm{i}\frac{\mathrm{d}\psi}{\mathrm{d}t}. \tag{8}$$

Based on the solution of Eq. (8), the power L emitted from the ion during the dynamic progress can be obtained by

$$L = \int_0^T \frac{2}{3c^3}\left(\frac{\mathrm{d}^2 P(t)}{\mathrm{d}t^2}\right)^2 \mathrm{d}t, \tag{9}$$

where $\mathrm{d}^2P/\mathrm{d}t^2$ is the dipole acceleration and P is the dipole moment,

$$\begin{aligned} P(t) &= -\int_{-\infty}^{+\infty} \psi^+ x \psi \mathrm{d}x \\ \frac{\mathrm{d}^2 P}{\mathrm{d}t^2} &= \int_{-\infty}^{+\infty} \psi^+ \left(-\frac{Z}{x|x|} - E_0 F(t)\sin(\omega t)\right) \\ &\quad \times \psi \mathrm{d}x. \end{aligned} \tag{10}$$

By Fourier transformation, the emission intensity at ω during the interaction period from 0 to T is expressed as

$$L = \int_0^{\infty} \frac{\mathrm{d}L}{\mathrm{d}\omega}(\omega)\mathrm{d}\omega. \tag{11}$$

By Pasaval theorem, the Fourier transformation of dipole acceleration can be expressed by the Fourier coefficients of dipole moment:

$$\frac{\mathrm{d}L}{\mathrm{d}\omega}(\omega) = \frac{2\pi\omega^4}{3c^3}[P_a^2(\omega) + P_b^2(\omega)], \tag{12}$$

where

$$P_a(\omega) = \frac{1}{\pi}\int_0^T P(t)\cos(\omega t)\mathrm{d}t, \tag{13}$$

and

$$P_b(\omega) = \frac{1}{\pi}\int_0^T P(t)\sin(\omega t)\mathrm{d}t. \tag{14}$$

We investigate the radiation of 1D Ne^{9+} ion pumped by laser pulse at 1064 nm (photon energy 1.166 eV, YAG laser). The energy difference between the ground state and the first excited state is 37.5 a.u. (1020 eV) and the corresponding transition needs about 875 photons. Obviously, intense laser pulse should be employed for such multi-photon transition. When the peak power of the laser pulse is as high as 3.5×10^{20} W/cm^2, the corresponding electric field E_0 is about 5.2×10^{11} V/cm (100 a.u.), which is about one tenth of the Coulomb field of Ne^{9+}, 5.2×10^{12} V/cm (1000 a.u.), seen by the electron at the ground state. As shown in Fig. 1, as the electric field strength weaker than 100 a.u., the electron can be well bounded in a potential trap. Accordingly, in our simulations the laser field strength E_0 is selected as 0.0001, 0.001, and up to 100 a.u. (corresponding peak power 3.5×10^8, 3.5×10^{10} and up to 3.5×10^{20} W/cm^2). The spatial integral range for the TDSE should be much larger than the ionic radius 0.1 a.u. (0.053 Å) (1 Å = 0.1 nm) and enough for motion of the electron driven by the laser oscillation. So we perform calculations with absorption boundary condition of 20–150 a.u. and show that the spectrum for $E_0 < 10$ a.u. does not change with the increase of the spatial size while the spectrum for $E_0 \geq 10$ a.u. changes considerably when the spatial size increases from $20 \sim 100$ a.u. and becomes stable when the length is large than 100 a.u.. So a spatial range covered 150 a.u. (79.35 Å) will be enough. The integral period is 12000 a.u. (290.4 fs), which is about 82 cycles of the laser wave. The TDSE is solved by a method due to Goldberg *et al.*[39] with space and time integral step size to be 1/1000 a.u. and 1/3000 a.u., respectively.

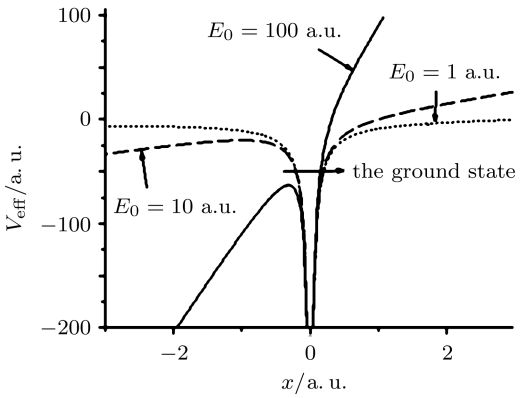

Fig. 1. Effective potential $V_{\rm eff} = -10/|x| + E_0 x$ for the electron of Ne^{9+} in a laser field with electric field amplitude E_0.

The laser pulses employed in our simulations are either square profile or Gaussian packages (Fig. 2). Nevertheless, the emission spectrum of the ion is nearly the same for both the profiles. Thus, the results presented below are all about the Gaussian wave. As shown in Figs. 3(a) and 3(b), when the strength of laser field is much lower than that of the ionic Coulomb field (1000 a.u. for the Bohr orbit of the ground state), the narrow peaks of the emission spectrum precisely locate at the frequencies of the ionic transitions from the excited states to the ground state, corresponding to the energy of 875.3, 1037.4, 1094.2 photons of the incident laser and so on. Notice that this short wavelength emission is coherent, just like the general HHG, but the frequencies are non-integral harmonics of the pump laser. In such a sense, we call this radiation as super high-order harmonic generation (SHHG). When the field strength E_0 increases from 0.0001 a.u. to 0.001 and 0.01 a.u., the corresponding SHHG emission power increases by 100 and 10000 times. We see a relation of $w \propto I$ between the power of every SHHG peak w and the incident laser intensity $I = cE_0^2/8\pi$. Specifically, $w = 48.6I$ for the first SHHG peak corresponding to the 1s–2p transition. As the laser gets stronger, the emission spectrum splits into many closely arranged peaks with their amplitudes becoming small and small (Fig. 3(b)), and when E_0 gets up to 0.1 a.u., most peaks overlap together. Hence, $E_0 = 0.1$ a.u. (1/10000 of the ionic Coulomb field) is regarded as an up-limit for SHHG. At the laser strength below this up-limit, the ionic dynamic progress can be well expressed under non-relativistic dipole approximation[40] and the electronic intermediate states remain bound because the tunneling effect is very low in highly charged ions.[41] So the SHHG spectrum should be brought out from tightly bound electron. When E_0 reaches 100 a.u., the SHHG is completely replaced by the normal HHG, as shown in Fig. 3(c). Here, $E_0 = 100$ a.u. is a threshold for general HHG. The highest harmonic order of the HHG, referred to as non-tunneling harmonic generation,[41,42] is about 100th, which is much lower than the lowest harmonic order 875.3 of the SHHG. Note, pumped by E_0 of 100 a.u., the photon number emitted in the highest harmonic range of the HHG is about 1.3×10^{-4}, while for the lowest harmonic emission of the SHHG, 2.63×10^{-4} photons can be obtained with $E_0 = 0.01$ a.u.. Obviously, the energy conversion efficiency of SHHG is about eight magnitude orders higher than that of general HHG.

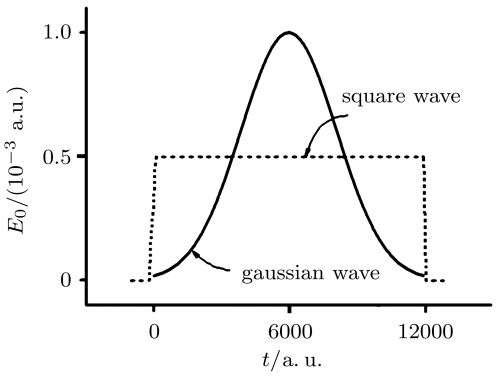

Fig. 2. A sketch for the electric field amplitude of square wave (dashed line) and Gaussian wave (solid line).

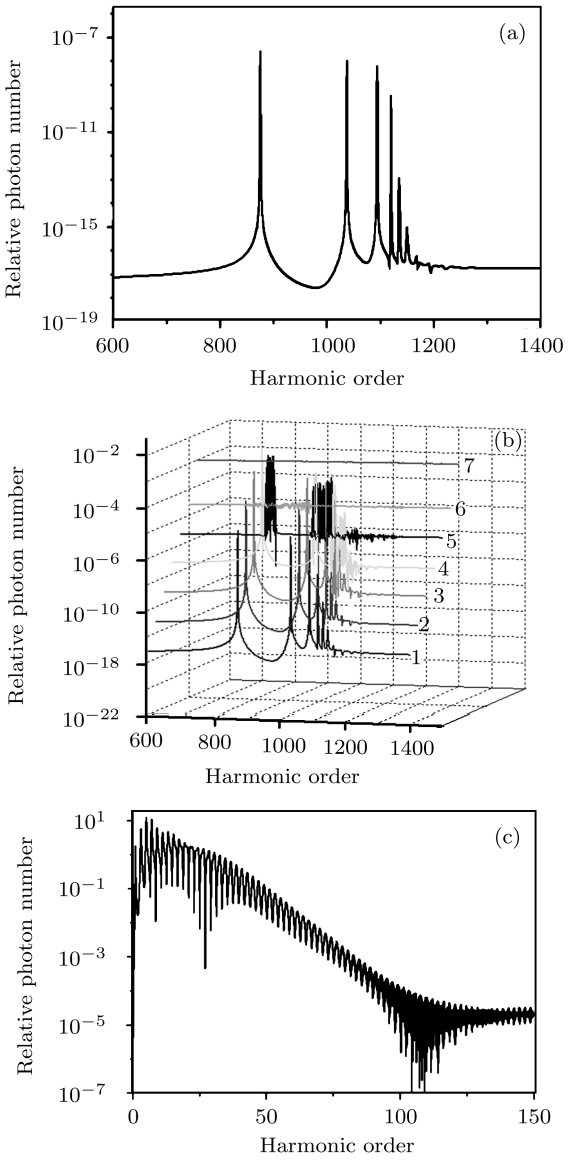

Fig. 3. Emission spectrum of 1D Ne^{9+} ion pumped by a laser pulse at 1064 nm: (a) the SHHG spectrum at $E_0 = 0.0001$ a.u.; (b) the SHHG spectrum marked by 1, 2, 3, 4, 5, 6 and 7 corresponding to $E_0 = 0.0001$, 0.001, 0.01, 0.1, 1, 10 and 100 a.u., respectively; (c) the general HHG at $E_0 = 100$ a.u..

To explore whether the SHHG can be further enhanced by exact resonance with the ionic bound states, we let the incident photon frequency to be exact integral fraction of a specific transition. Considering that odd and even photon number may have different effects, we let the pump photon energy equal either 1/875 or 1/876 of the energy difference between the first excited state and the ground state, and find no obvious enhancement effects in the SHHG spectrum.

The absolute intensity of the SHHG emission is a key for its application. In order to address this issue, we repeat the above simulations with Ne^{9+} replaced by 1D hydrogen atom. Because the electron is less tightly bounded than that in the Ne^{9+} ion, we do not obtain the SHHG spectrum but a general HHG instead. The corresponding HHG spectrum at $E_0 = 0.01$ a.u. is shown in Fig. 4. At such laser strength, HHG for the hydrogen atom can reach 25th order with a relative emission photon number of 1.67×10^{-4}. With the same laser strength, the emitted photon number of Ne^{9+} is 2.63×10^{-4} for the 875.3th order of the SHHG. That is, the SHHG of Ne^{9+} generates as many photons as the HHG of hydrogen pumped by the same laser pulse, indicating that the emission of SHHG is intense enough for common detection since the HHG emission of hydrogen can be detected.

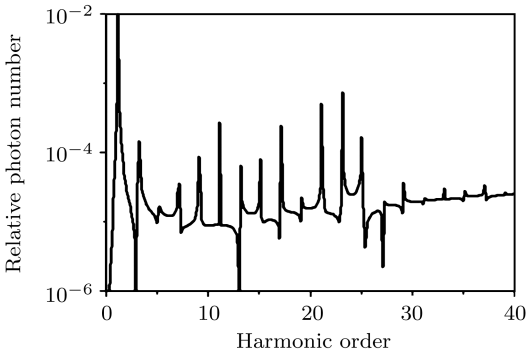

Fig. 4. The HHG spectrum of 1D hydrogen atom pumped by a laser pulse at 1064 nm with $E_0 = 0.01$ a.u.

3. The 3D hydrogen-like ion

In virtue of the complexity for numerical solution of 3D wave equation, we perform the calculation in the matrix form by expanding the wave function to the eigenstates of the ion φ_1, φ_2, φ_3 ...:

$$\psi(t) = \sum C_i(t)\varphi_i, \quad (i = 1, 2, 3, 4 \ldots), \qquad (15)$$

and the corresponding TDSE is then given by

$$\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}t}\begin{pmatrix} C_1 \\ C_2 \\ \cdots \\ C_K \end{pmatrix} = H\begin{pmatrix} C_1 \\ C_2 \\ \cdots \\ C_K \end{pmatrix}, \tag{16}$$

where

$$\hat{H} = -\frac{1}{2}\nabla^2 - \frac{Z}{r} + zE_0F(t)\sin(\omega t). \tag{17}$$

When Ne^{9+} ion is pumped by a laser at a wavelength of 1064 nm, the emission spectrum shows similar features under either square wave profile or Gaussian package. So we also employ Gaussian wave package for this 3D simulation. Based on the discussion of last section, the SHHG generates from ions far away from ionization and closely relates to the excited bound states. Consequently, to examine how many eigenstates should be included in Eq. (15) for the SHHG simulations, we enlarge the expanding space by increasing the main quantum number n step by step, until the emission spectrum keeps unchanged with more eigenstates added. Because linear polarised laser is employed (Eq. (17)) and the ion initially lies in the ground state ($l = 0, m = 0$), we just select the eigenstates of quantum number $m = 0$. For Ne^{9+}, 210 states (maximum $n = 20$) are found to be enough to get the convergence.

When the peak power of the pump laser is in the range used in the 1D simulations, the main feature of the SHHG keeps unchanged, i.e. the narrow spectrum peaks precisely locate at the frequencies of the ionic transitions from the excited states to the ground state. The difference is that there are more resonant spectrum lines in the 3D model, corresponding to more excited states of the ion, as shown in Fig. 5. Although, the energy conversion efficiency for 3D model is significantly lower than that for 1D model, for weak pump laser intensity, the emission power of 3D model is proportional to $I^{2.3}$, specifically $w = 3.98I^{2.3}$ for the first SHHG peak (recall that the emission strength for the 1D model is proportional to I), and as the pump laser intensity increases up to 0.01 a.u., the SHHG strength of the 3D model catches up with the 1D model. It is worth to mention that the up-limit of SHHG for the 3D ions is the same as that for the 1D ion ($E_0 = 0.1$ a.u.). Slightly changing the wavelength of the pump laser to satisfy the resonant condition, e.g. the energy of the pump photon being 1/875 or 1/876 of the energy difference between the ground state and the first excited state, does not enhance the emission of SHHG.

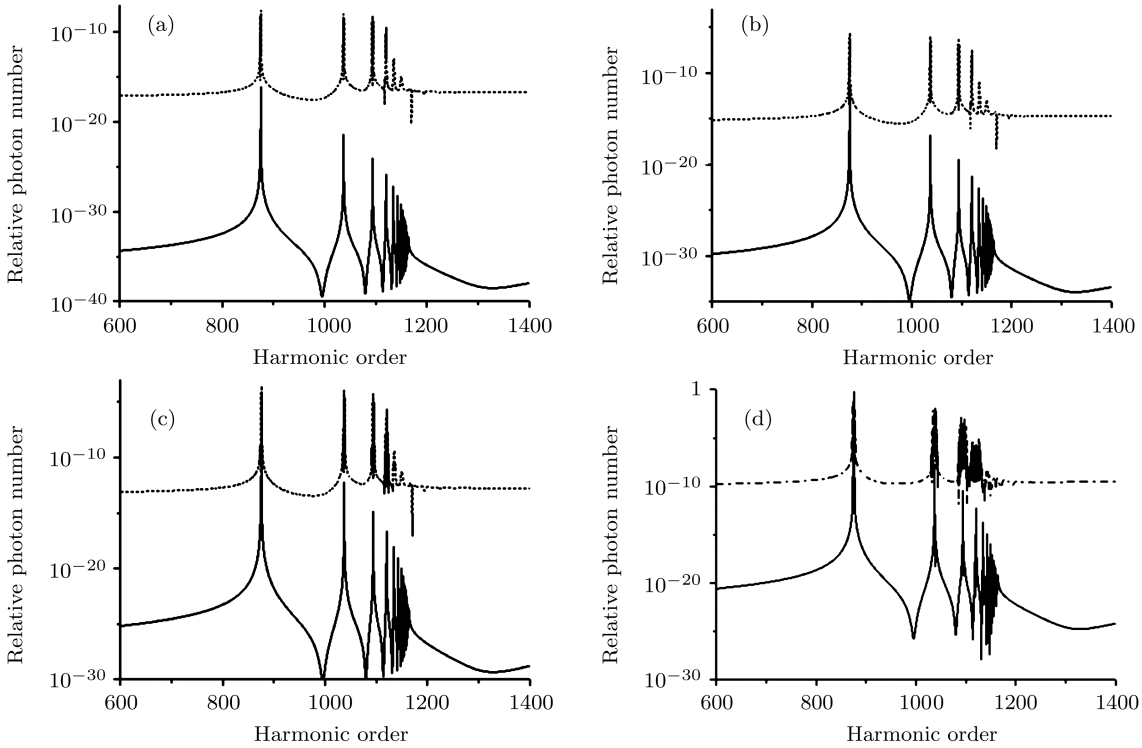

Fig. 5. The SHHG spectrum of Ne^{9+} ion pumped by a laser pulse at 1064 nm (solid line) compared with that for 1D Ne^{9+} in Fig. 3 (dotted line) : (a) $E_0 = 0.0001$ a.u.; (b) $E_0 = 0.001$ a.u.; (c) $E_0 = 0.01$ a.u.; (d) $E_0 = 0.1$ a.u.

To investigate the dependence of SHHG spectrum on the pump laser wavelengths and power, we further performed simulations with wavelength of 50 nm, 100 nm, 200 nm, 400 nm, 600 nm and 800 nm, without changing other conditions, and found that the SHHG emission only relates to laser power but not the laser wavelength. So we can derive the SHHG under a wide range of laser wavelength. Figure 6 gives the SHHG spectrum driven by 50 nm laser at different laser intensities. The SHHG emission power is proportional to $I^{2.3}$, the same as that at 1064 nm.

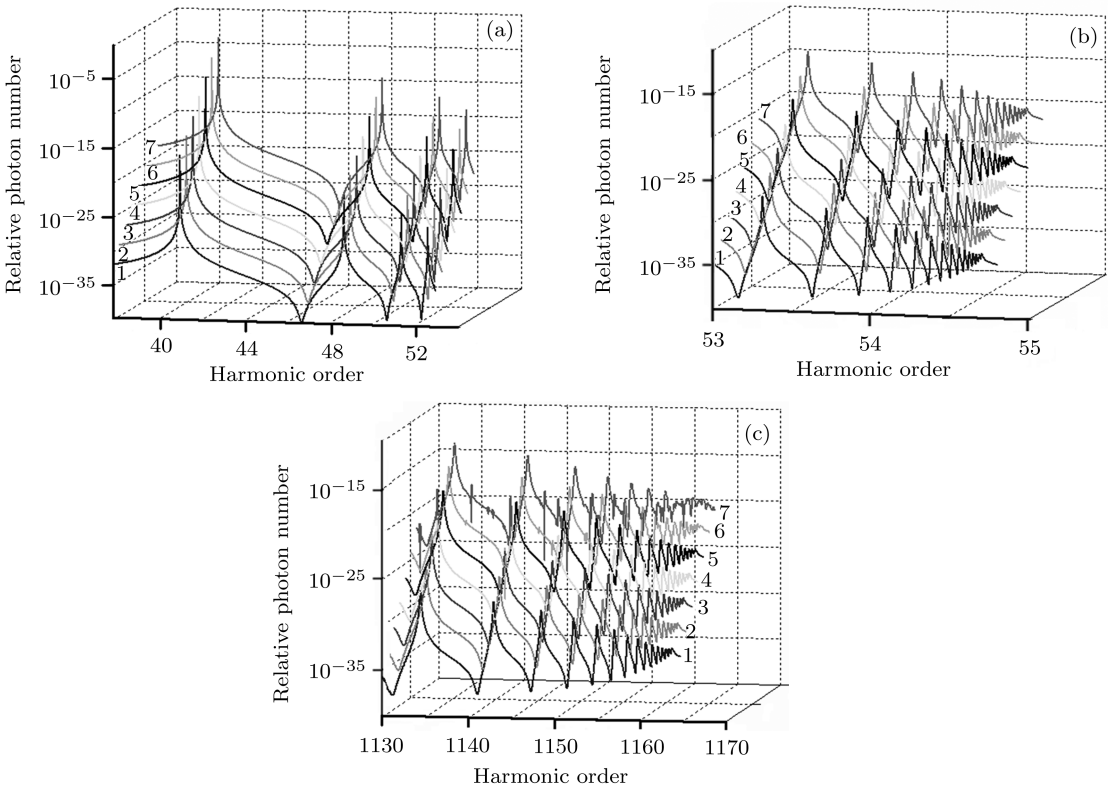

Fig. 6. The SHHG spectrum of Ne^{9+} ion pumped by a laser pulse at 50 nm. The marks 1, 2, 3, 4, 5, 6 and 7 correspond to the peak intensity of 10^9, 10^{10}, 10^{11}, 10^{12}, 10^{13}, 10^{14} and 10^{15} W/cm^2 respectively. Figures 6(a), (b) and (c) are in different scopes of harmonic order.

The SHHG emission at higher harmonic orders can be obtained from hydrogen-like ions with higher charge. For example, an SHHG spectrum of Ca^{19+} at an incident wavelength of 1064 nm is given by Fig. 7. Of course, the SHHG can also be extended to general neutral atoms by incident laser with very long wavelength. For hydrogen atom, we can select a laser pulse of Gaussian package at a frequency of 5×10^{-4} a.u. (about a wavelength of 91170 nm) with a peak intensity of 10^9 W/cm^2 and the emission of SHHG shown in Fig. 8 can be obtained.

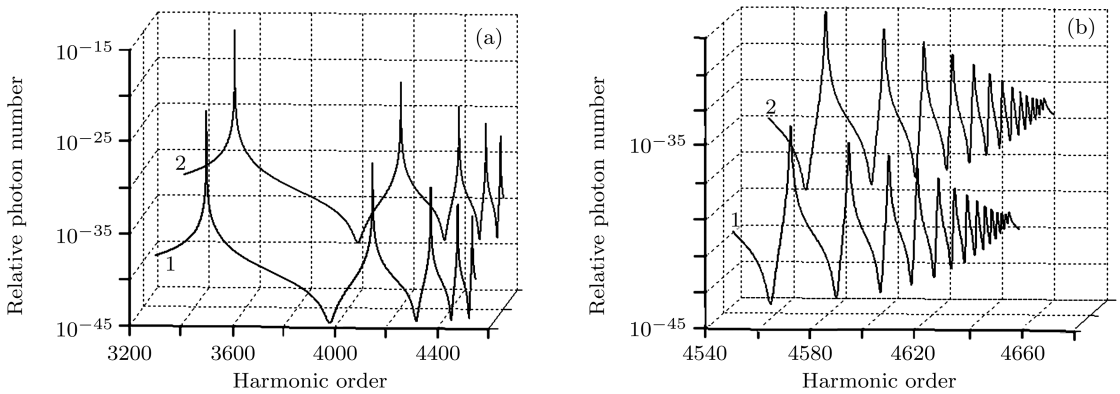

Fig. 7. The SHHG spectrum of Ca^{19+} ion pumped by a laser pulse at 1064 nm. The marks 1 and 2 correspond to the peak power densities of 10^9 and 10^{11} W/cm^2. Figures 7(a) and (b) are in different scopes of harmonic order.

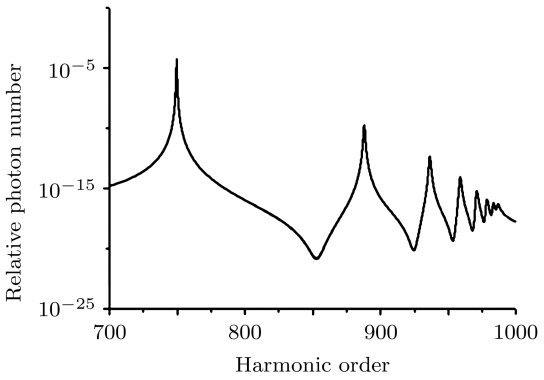

Fig. 8. The SHHG spectrum of hydrogen atom pumped by a laser pulse at a frequency of 5×10^{-4} a.u. and a peak intensity of 10^9 W/cm^2.

4. Full quantum theory

To further explain the mechanism of the SHHG phenomenon, we go into full quantum theory that involves both quantized electron and photon. In previous analytical models for HHG, the strong field approximation[43–49] assumes that the electron obtaining energy far above the ionization is generally used in both the semi-classical and full quantum theory.[48,50,51] Hence, these models can not be used to analyse the SHHG spectrum because the electronic energy is well below the ionization threshold.

In full quantum theory, the electronic states are described by φ_1, φ_2, φ_3 ..., which are the eigenstates of the ionic Hamiltonian $\hat{H}_{\rm atom} = -(1/2)\nabla^2 - Z/r$ and the laser field is quantized as a homochromatic mode with Hamiltonian expressed by

$$\hat{H}_{\rm photon} = \omega\left(a^+a + \frac{1}{2}\right), \tag{18}$$

where a^+ and a are generation and annihilation operators. Eigenstates of the Hamiltonian $\hat{H}_{\rm photon}$ are signed as $|0\rangle$, $|1\rangle$, ..., $|n\rangle$, ... and the corresponding electric field operator is

$$\hat{H} = \sqrt{\frac{4\pi\omega}{V}}(a^+ + a), \tag{19}$$

where V is normalization volume of the photon field. The pump laser field is described by either Fock state $|n\rangle$ or coherent state

$$|\alpha\rangle = \exp\left(-\frac{|\alpha|^2}{2}\right)\sum_{j=0}^{\infty}\frac{|\alpha|^j}{\sqrt{j!}}|j\rangle. \tag{20}$$

In this framework, the state of the electron and incident photon is expanded into basis as $|\varphi_i\rangle|j\rangle$. By assuming the polarisation of the pump laser in z-direction, the total Hamiltonian is now expressed as

$$\hat{H} = \hat{H}_{\rm atom} + \hat{H}_{\rm photon} + \sqrt{\frac{4\pi\omega}{V}}z(a^+ + a). \tag{21}$$

The solution of the TDSE will be presented by

$$\begin{aligned}|\psi(t)\rangle &= \sum_j k_j|\phi_j\rangle\exp(-{\rm i}E_jt)\\ &= \sum_j k_j\left(\sum_{k,l} f_{j,k,l}|\varphi_k\rangle|l\rangle\right)\exp(-{\rm i}E_jt).\end{aligned} \tag{22}$$

Here $|\phi_i\rangle$ (i = 1, 2, 3 ...) are the eigenstates of $\hat{H}$ with energy E_i respectively. Initially ($t = 0$) the electron stays in the ground state $|\varphi_{1\rm s}\rangle$ and the photons stay in a Fock state or a coherent state, i.e.

$$|\psi(0)\rangle = |\varphi_{1\rm s}\rangle|n\rangle, \tag{23}$$

or

$$|\psi(0)\rangle = |\varphi_{1\rm s}\rangle\exp\left(-\frac{|\alpha|^2}{2}\right)\sum_{j=0}^{\infty}\frac{|\alpha|^j}{\sqrt{j!}}|j\rangle, \tag{24}$$

and then the coefficients k_j are solved. A systematic method in full quantum theory was developed to describe the absorption and emission of photons in the HHG progress by a scattering theoretical approach.[52] Here, to simplify the calculation in full quantum approach we employ an approximate semi-classical calculation for the emission spectrum, which could be derived by the Fourier expansion of the electric field or the atomic electronic dipole acceleration. For arbitrary time t the dipole moment

$$\begin{aligned}P(t) &= \langle\psi(t)|\left[\sum_{m_1}|m_1\rangle\exp(-{\rm i}m_1\omega t)\right]z\left[\sum_{m_2}\langle m_2|\exp({\rm i}m_2\omega t)\right]|\psi(t)\rangle\\ &= \left\{\sum_{j_1}k_{j_1}\left(\sum_{k_1,m_1}f_{j_1,k_1,m_1}\langle\varphi_{k_1}|\right)\exp[{\rm i}(E_{j_1}-m_1\omega)t]\right\}\end{aligned}$$

$$\times z\left\{\sum_{j_2} k_{j_2}\left(\sum_{k_2,m_2} f_{j_2,k_2,m_2}|\varphi_{k_2}\rangle\right)\exp[-\mathrm{i}(E_{j_2}-m_2\omega)t]\right\}, \tag{25}$$

and the emission spectrum is calculated by Fourier expansion of dipole acceleration (Eqs. (12)–(14)). In strong field region, the quantum effect of laser field could be neglected and such approximation could be suitable.

The calculation is carried out for a basis set that involves electronic states of Ne^{9+} ion for $n \leq 10$ (where n is the main quantum number) at a laser strength of 10^{13} W/cm^2. In principle, countless Fock states of photons should be involved in the calculation but that leads to an unsolvable problem. In virtue of computation limit, we can only take in a series of Fock states near the average photon number $\bar{n}$ of the laser field. Since the bound–bound transitions in Ne^{9+} cover an energy range of 50 a.u., one hundred Fock states ranged from $|\bar{n}-50\rangle$ to $|\bar{n}+50\rangle$ should be enough to understand the main transition processes if the energy of single photon is larger than 0.5 a.u. (corresponding wavelength shorter than 91.2 nm). Thus, in our calculations the laser wavelength varies from 3 nm to 90 nm and the Fock states ranged from $|\bar{n}-50\rangle$ to $|\bar{n}+50\rangle$ keep unchanged. In such a wavelength range the SHHG can occur (at about 50 nm laser or longer wavelength) and the main features of SHHG for longer wavelength can be extrapolated from this result.

Our calculations show similar results to those in Section 3. The SHHG spectrum is nearly independent of the initial state of the pump laser, a Fock state or a coherent state. So the results presented below are all concerned with the coherent state. For laser wavelength shorter than 7 nm, the electronic emission spectrum has only two sharp peaks (Fig. 9(a)), corresponding to the pump frequency and the 1s–2p transition, respectively. As the laser wavelength increases from 7 nm to 20 nm, the spectrum shows more peaks (Fig. 9(b)), corresponding to some lower electronic transitions from excited bound states to the ground state. When the laser wavelength exceeds 20nm, the spectrum turns into a typical SHHG spectrum with peaks corresponding to the electronic transitions from the 2p, 3p, 4p states and so on to the ground state (Figs. 9(c) and 9(d) for 35 nm and 75 nm respectively). Hence, 20 nm is regarded as a minimal wavelength for appearing SHHG. To pay attention to the change of spectra intensity from non-SHHG to SHHG, the spectra intensity corresponding to 1s–2p transition varied with increasing pump wavelength is plotted in Fig. 10. In the range of shorter wavelength it decays quickly with increasing wavelength. When the laser wavelength is longer than 20 nm, it gradually turns into a constant and meanwhile the intensity of all the SHHG spectrum peaks keeps nearly unchanged (e.g., comparing Figs. 9(c) and 9(d)).

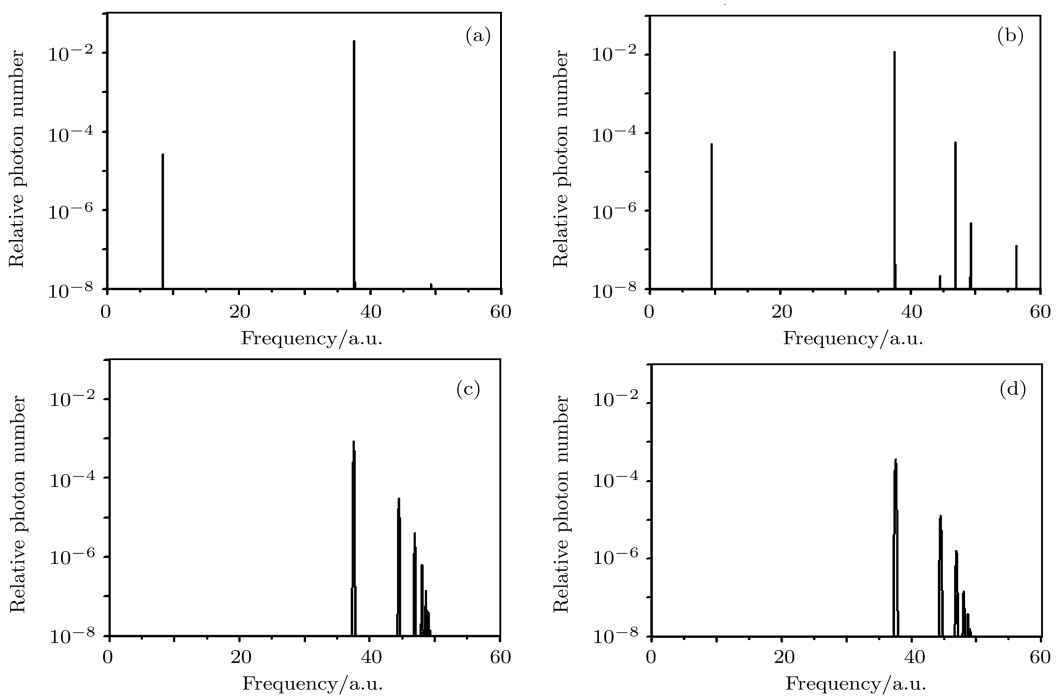

Fig. 9. The emission spectrum of Ne^{9+} pumped by laser at an intensity of 10^{13} W/cm^2 with different wavelengths: (a) 5.47 nm; (b) 9.72 nm; (c) 35.0 nm; (d) 75.0 nm.

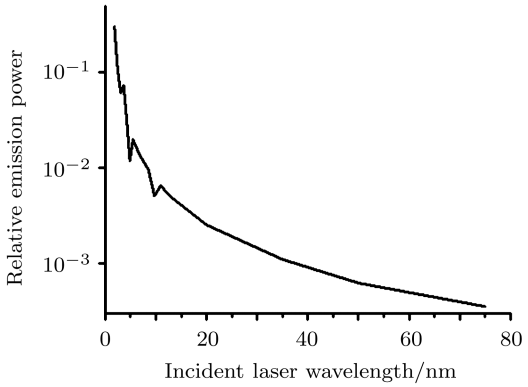

Fig. 10. Power of the emission spectrum peak corresponding to the transition from 1s to 2p for Ne^{9+} interacted with the laser at an intensity of 10^{13} W/cm^2 at different incident wavelengths.

5. Conclusions

The SHHG of hydrogen-like highly charged ions pumped with intense laser pulse is studied numerically in the 1D and 3D model. The two models provide similar results: when the laser field is much weaker than ionic Coulomb field (about 1/10000) and the carrier frequency is much lower than the Bohr frequency of the ion, the SHHG spectrum exhibits narrow lines precisely corresponding to the transitions from the excited states to the ground state, and the spectra intensity is independent of incident laser frequency. The SHHG spectrum is reckoned to be generated from highly bound states below the ionization threshold. These results can be understood qualitatively in full quantum theory. The emission of SHHG might be used as coherent x-ray source, or be employed to study the electronic state of the highly charged ions. If the wavelength of the pump laser is long enough, the spectrum of SHHG can be extended to study the electronic structure of neutral atoms and molecules.

Acknowledgments

We thank Professor Yang Fujia for his concering and supporting this study and acknowledge Professor Wang Yansen for helpful discussions. Some of the calculations are performed in the National High Performance Computing Center of Fudan University.

References

[1] Burnett N H, Baldis H A, Richardson M C and Enright G D 1977 *Appl. Phys. Lett.* **31** 172
[2] McLean E A, Stamper J A, Ripin B H, Grie H R, McMahon F J M and Bodner S E 1977 *Appl. Phys. Lett.* **31** 825
[3] Carman R L, Forslund D W and Kindel J M 1981 *Phys. Rev. Lett.* **46** 29
[4] McPherson A, Gibson G, Jara H, Johann U, Luk T S, McIntyre I A, Boyer K and Rhodes C K 1987 *J. Opt. Soc. Am.* B **4** 595
[5] Li X F, L'Huillier A, Ferray M, Lompre L A and Mainfray G 1989 *Phys. Rev.* A **39** 751
[6] Ge Y C 2008 *Acta. Phys. Sin.* **57** 4091 (in Chinese)
[7] Preston S G, Sanpera A, Zepf M, Blyth W J, Smith C G, Wark J S, Key M H, Burnett K, Nakai M, Neely D and Offenberger A A 1996 *Phys. Rev.* A **53** R31
[8] Spielmann C, Burnett N H, Sartania S, Koppitsch R, Schnürer M, Kan C, Lenzner M, Wobrauschek P and Krausz F 1997 *Science* **278** 661
[9] Schnurer M, Spielmann C, Wobrauschek P, Streli C, Burnett N H, Kan C, Ferencz K, Koppitsch R, Cheng Z, Brabec T and Krausz F 1998 *Phys. Rev. Lett.* **80** 3236
[10] Zhang Y P, Zhang F S, Meng K L and Xiao G Q 2007 *Chin. Phys.* **16** 83
[11] Zhou Z Y and Yuan J M 2007 *Chin. Phys.* **16** 675
[12] Walser M W, Keitel C H, Scrinzi A and Brabec T 2000 *Phys. Rev. Lett.* **85** 5082
[13] Ge Y C 2008 *Acta. Phys. Sin.* **57** 2899 (in Chinese)
[14] Milosevic D B, Hu S and Becker W 2000 *Phys. Rev.* A **63** 011403(R)
[15] Hatsagortsyan K Z and Keitel C H 2001 *Phys. Rev. Lett.* **86** 2277
[16] Tempea G, Geissler M, Schnurer M and Brabec T 2000 *Phys. Rev. Lett.* **84** 4329
[17] Barfels R, Backus S, Zeek E, Misoguti L, Vdovin G, Christov I P, Murnane M M and Kapteyn H C 2000 *Nature* (London) **406** 164
[18] Mechain G, Couairon A, Franco M, Prade B and Mysyrowicz A 2004 *Phys. Rev. Lett.* **93** 035003
[19] Zhou Z Y and Yuan J M 2007 *Chin. Phys.* **16** 675
[20] Lai X Y, Cai Q Y and Zhan M S 2010 *Chin. Phys.* B **19** 020302
[21] Lewenstein M, Balcou P, Ivanov M Y, L'Huillier A and Corkum P B 1994 *Phys. Rev.* A **49** 2117
[22] L'Huillier A and Balcou Ph 1993 *Phys. Rev. Lett.* **70** 774
[23] Kwong V H S 1989 *Phys. Rev.* A **39** 4451
[24] Torrisia L and Gammino S 2002 *Methods Phys. Res. Sect.* B **209** 345
[25] Trinczek M, Werdich A and Mironov V 2006 *Nucl. Instrum. Methods Phys. Res. Sect.* B **251** 289
[26] Jungreuthmayer C, Geissler M, Zanghellini J and Brabec T 2004 *Phys. Rev. Lett.* **92** 133401
[27] Micheau S, Jouin H and Pons B 2008 *Phys. Rev.* A **77** 053201
[28] Church D 1991 *Nucl. Instrum. Methods Phys. Res. Sect.* B **53** 504
[29] Silver J, Varney A J, Margolis H S, Baird P, Grant I P, Groves P D and Hallett W A 1994 *Rev. Sci. Instrum.* **65** 1074

[30] Gillaspy J 1995 *Phys. Scripta* **T59** 392
[31] Beiersdorfer P 1996 *Rev. Sci. Instrum.* **67** 3818
[32] Donets E 1998 *Rev. Sci. Instrum.* **69** 614
[33] Gillaspy J 2001 *J. Phys.* B **34** R93
[34] Lopez-Urrutia J, Bapat B, Feuerstein B, Fischer D, Lörch H, Moshammer R and Ullrich J 2003 *Hyperfine Interact.* **146** 109
[35] Currell F, Aiken J, Dunn K F, Kavanagh T, Krastev V, Lu X, Matranga M, Morton A F, O'Rourke B E, Tawara H and Watanabe H 2005 *Phys. Scripta* **T120** 53
[36] Lorin E, Chelkowski S, Bandrauk A 2007 *Comp. Phys. Commum.* **177** 908
[37] Protopapas M, Keitel C H and Knight P L 1997 *Rep. Prog. Phys.* **60** 389
[38] Figueira de Morisson Faria C and Rotter I 2002 *Phys. Rev.* A **66** 013402
[39] Goldberg A, Schey H M and Schwartz J L 1967 *Am. J. Phys.* **35** 177
[40] Reiss H R 2008 *Progress in Ultrafast Intense Laser Science,* Volume III (Berlin: Springer) p.18
[41] Hu S X, Strace A F, Becker W, Sandner W and Milošević D B 2002 *J. Phys.* B **35** 627
[42] DiPiazza A, Fiordilino E and MittleMan M H 2001 *Phys. Rev.* A **64** 013414
[43] Keldysh L V 1964 *Sov. Phys. JETP* **20** 1307
[44] Faisal F H M 1973 *J. Phys.* B **6** L89
[45] Reiss H R 1980 *Phys. Rev.* A **22** 1786
[46] Reiss H R 1992 *Prog. Quantum Electron.* **16** 1
[47] Reiss H R 1990 *Phys. Rev.* A **42** 1476
[48] Reiss H R 1990 *J. Opt. Soc. Am.* B **7** 574
[49] Faisal F H M and Bhattacharyya S 2004 *Phys. Rev. Lett.* **93** 053002
[50] Klaiber M, Hatsagortsyan K Z and Keitel C H 2007 *Phys. Rev.* A **75** 063413
[51] Diestler D J 2008 *Phys. Rev.* A **78** 033814
[52] Gao L, Li X, Fu P, Freeman R R and Guo D S 2000 *Phys. Rev.* A **61** 063407