

Pulse-duration dependence of high-order harmonic generation with coherent superposition state

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(Received 15 September 2005; published 30 December 2005)

We make a systematic study of high-order harmonic generation (HHG) in a He^+ -like model ion when the initial state is prepared as a coherent superposition of the ground state and an excited state. It is found that, according to the degree of the ionization of the excited state, the laser intensity can be divided into three regimes in which HHG spectra exhibit different characteristics. The pulse-duration dependence of the HHG spectra in these regimes is studied. We also demonstrate evident advantages of using coherent superposition state to obtain high conversion efficiency. The conversion efficiency can be increased further if ultrashort laser pulses are employed.

DOI: 10.1103/PhysRevA.72.063412

PACS number(s): 42.50.Hz, 42.65.-k, 32.80.Rm

I. INTRODUCTION

High-order harmonic generation (HHG) is a very useful source of coherent light in the extreme ultraviolet and soft x-ray regions of the spectrum [1–4]. HHG occurs when atomic systems interact with intense laser fields. There are two important aspects we need to consider in HHG, i.e., the cutoff frequency of the harmonic spectrum and the conversion efficiency of the harmonic generation. The cutoff frequency is predicted by the cutoff law [5,6], and the conversion efficiency is determined by the ionization of atoms. Many methods have been proposed to increase the cutoff frequency and the conversion efficiency, such as using ultrashort pulses [7,8]. Recently, ions have been used to extend the HHG spectrum cutoff [9–11]. However, the HHG conversion efficiency is usually very low because the ionization probability is low due to the large I_p . Increasing the harmonic conversion efficiency by preparing the initial state as a coherent superposition of two bound states was first proposed by Gauthey *et al.* [12]. Burnett and co-workers demonstrated that a harmonic spectrum with distinct plateaus could be obtained by such superposition states. Ishikawa [11] showed that the conversion efficiency of HHG by He^+ ions can be increased effectively by applying an additional harmonic pulse to populate one of the excited states. More recently, Averbukh [13] investigated the atomic polarization effects on HHG by preparing the initial state as a coherent superposition of one S state and one P state of atoms. The superposition state can be obtained by multiphoton resonant excitation [14] or using one harmonic pulse with the frequency corresponding to the energy difference between the two bound states before the fundamental laser pulse [11].

The idea of preparing the initial state as a coherent superposition of the ground state and an excited state is that it can induce dipole transitions between the continuum and the ground state via the excited state responsible for the ioniza-

tion. This process depends on the degree of the ionization of the excited state. In this paper, we will make a systematic study of the HHG with coherent superposition state in a He^+ -like model ion. It is found that, according to the degree of the ionization of the excited state, the laser intensity can be divided into three regimes in which HHG spectra exhibit different characteristics. The pulse-duration dependence of the HHG spectra in these regimes is studied. We also demonstrate evident advantages of using coherent superposition state to obtain high conversion efficiency. The conversion efficiency can be increased further if ultrashort laser pulses are employed.

II. NUMERICAL METHOD

Our theory is based on solving the one-dimensional time-dependent Schrödinger equation for a hydrogenlike ion in a laser pulse, which can be expressed as (atomic units are used throughout)

$$i \frac{\partial \psi(x,t)}{\partial t} = \left[-\frac{1}{2} \nabla^2 - \frac{a}{\sqrt{b+x^2}} - xE(t) \right] \psi(x,t), \quad (1)$$

where a and b are the parameters describing different ions. We set $a=2$ and $b=0.5$ in order to get the same ground state binding energy of an He^+ ion, i.e., 2 a.u., and the second excited state binding energy is 0.53 a.u. in this one-dimensional case. We consider the second excited state rather than the first excited state because it has the same symmetry of the ground state and has approximately the same binding energy as the first excited state of the real He^+ ion. $E(t) = F(t)\sin(\omega t + \phi)$ is the electric field of the pulse. Here, we choose $\omega=0.056$ (wavelength 800 nm) and $\phi=0$ in the calculations. $F(t)$ is the pulse envelope, which equals $\sin(\pi t/T)^2$ for 10 fs pulse, while

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$$F(t) = \begin{cases} \sin(\pi t/\tau)^2 & \text{if } 0 < t < \tau/2, \\ 1 & \text{if } \tau/2 < t < T - \tau/2, \\ 1 - \sin[\pi(T-t)/\tau]^2 & \text{if } T - \tau/2 < t < T, \end{cases}$$

for 100 fs pulse, where τ is the period of the optical cycle and T is the laser pulse duration. Equation (1) is integrated numerically with the help of fast Fourier transform algorithm [15], where the length of the integration grid is 800, the spatial step is $dx=0.1$ and the time increment is 0.0125. To avoid reflection of the wave packet from the boundary after each time increment, the wave function is multiplied by a $\cos^{1/8}$ mask function that varies from 1 to 0 over a range from $|x|=300$ to 400.

III. HHG WITH COHERENT SUPERPOSITION STATE

The HHG spectrum can be obtained from the Fourier transform of the time-dependent dipole acceleration $D(t) = \langle \psi(x, t) | \nabla | \psi(x, t) \rangle$, which can be written as

$$D(t) \propto \langle \psi_{\text{bound}}(x, t) | \nabla | \psi_{\text{continuum}}(x, t) \rangle + \text{c.c.} \quad (2)$$

Here, we neglect the continuum-continuum transitions because they have no significant influence on the harmonic generation. We prepare the initial state in a superposition of the ground state $|g\rangle$ and some excited state denoted by $|e\rangle$, i.e.,

$$\psi(x, t \rightarrow -\infty) = \frac{1}{\sqrt{2}}(|g\rangle + |e\rangle), \quad (3)$$

where the phase difference between these states is set to zero for simplicity. If we assume that the ground and excited states are not coupled to any other bound state during the pulse, then the time-dependent wave function can be written in the form

$$\psi(x, t) = \alpha(t)e^{-i\omega_g t}|g\rangle + \beta(t)e^{-i\omega_e t}|e\rangle + \int dk \gamma_k(t)e^{-i\omega_k t}|\phi_k(t)\rangle. \quad (4)$$

In this expression, $|\phi_k(t)\rangle$ is the continuum state characterized by the momentum k , and $\alpha(t)$, $\beta(t)$, and $\gamma_k(t)$ are the time-dependent amplitudes of the ground, excited, and continuum states, respectively. Here, we have factorized out the energy dependence of the states. Accordingly, the temporal evolution of the bound state is

$$\psi_{\text{bound}}(x, t) = \alpha(t)e^{-i\omega_g t}|g\rangle + \beta(t)e^{-i\omega_e t}|e\rangle, \quad (5)$$

and we have the time-dependent dipole moment

$$D(t) = D_{gg}(t) + D_{ee}(t) + D_{ge}(t) + D_{eg}(t), \quad (6)$$

where

$$D_{gg}(t) \propto \int dk \alpha(t) \gamma_k^g(t) e^{-i(\omega_g - \omega_k)t} \langle g | \nabla | \phi_k(t) \rangle + \text{c.c.}, \quad (7)$$

$$D_{ee}(t) \propto \int dk \beta(t) \gamma_k^e(t) e^{-i(\omega_e - \omega_k)t} \langle e | \nabla | \phi_k(t) \rangle + \text{c.c.}, \quad (8)$$

$$D_{ge}(t) \propto \int dk \alpha(t) \gamma_k^e(t) e^{-i(\omega_g - \omega_k)t} \langle g | \nabla | \phi_k(t) \rangle + \text{c.c.}, \quad (9)$$

and

$$D_{eg}(t) \propto \int dk \beta(t) \gamma_k^g(t) e^{-i(\omega_e - \omega_k)t} \langle e | \nabla | \phi_k(t) \rangle + \text{c.c.}, \quad (10)$$

where $\gamma_k^g(t)$ and $\gamma_k^e(t)$ are the amplitudes of the continuum state $|\phi_k(t)\rangle$ originated from the ionization of the ground and excited states, respectively, which, in the strong-field approximation [16], can be written as

$$\begin{aligned} \gamma_k^g(t) \approx & i \int_0^t dt' \alpha(t') e^{-i(\omega_g - \omega_k)t'} E(t') \langle k + A(t)/c - A(t')/c | x | g \rangle \\ & \times \exp \left[-i \int_{t'}^t dt'' \left(\frac{[k + A(t)/c - A(t'')/c]^2}{2} - \omega_k \right) \right], \end{aligned} \quad (11)$$

and

$$\begin{aligned} \gamma_k^e(t) \approx & i \int_0^t dt' \beta(t') e^{-i(\omega_e - \omega_k)t'} E(t') \langle k + A(t)/c - A(t')/c | x | e \rangle \\ & \times \exp \left[-i \int_{t'}^t dt'' \left(\frac{[k + A(t)/c - A(t'')/c]^2}{2} - \omega_k \right) \right], \end{aligned} \quad (12)$$

with $A(t)$ the vector potential of the laser pulse. Physically, $D_{gg}(t)$ and $D_{ee}(t)$ are simply the dipole moments one would obtain starting in the ground and excited states, respectively. On the other hand, $D_{ge}(t)$ [$D_{eg}(t)$] can be regarded as the interference term, where the excited state $|e\rangle$ (the ground state $|g\rangle$) is coupled to the continuum, inducing dipole moment between the continuum and the ground state $|g\rangle$ (the excited state $|e\rangle$). It is important to mention that the tunneling ionization is usually much easier for electrons at the excited state than at the ground state. On the other hand, the probability of transition from the continuum back to the ground state is higher than that to the excited state. Specifically, as discussed by Sanpera *et al.* [10], we have $|\langle e | \nabla | \phi_k(t) \rangle| / |\langle g | \nabla | \phi_k(t) \rangle| \approx (\omega_g / \omega_e)^{(5/2)}$, which equals approximately 30 in our case.

We are interested in producing high-energy harmonic photons with high conversion efficiency. In principle, ions can produce higher-energy harmonics due to their large ionization potentials. However, harmonic signal for ions has been shown to be very weak because the efficiency of the harmonic signal is directly proportional to the ionization rate. On the other hand, it is much easier to promote electrons into the continuum from excited states. As pointed out by Sanpera and co-workers [10], a possible way of increasing the harmonic efficiency is to prepare the initial state as a coherent superposition of the ground state and an excited state so that the dipole transition is induced between the continuum and the ground state, where the excited state is responsible for the ionization [i.e., $D_{ge}(t)$ term in Eq. (9)].

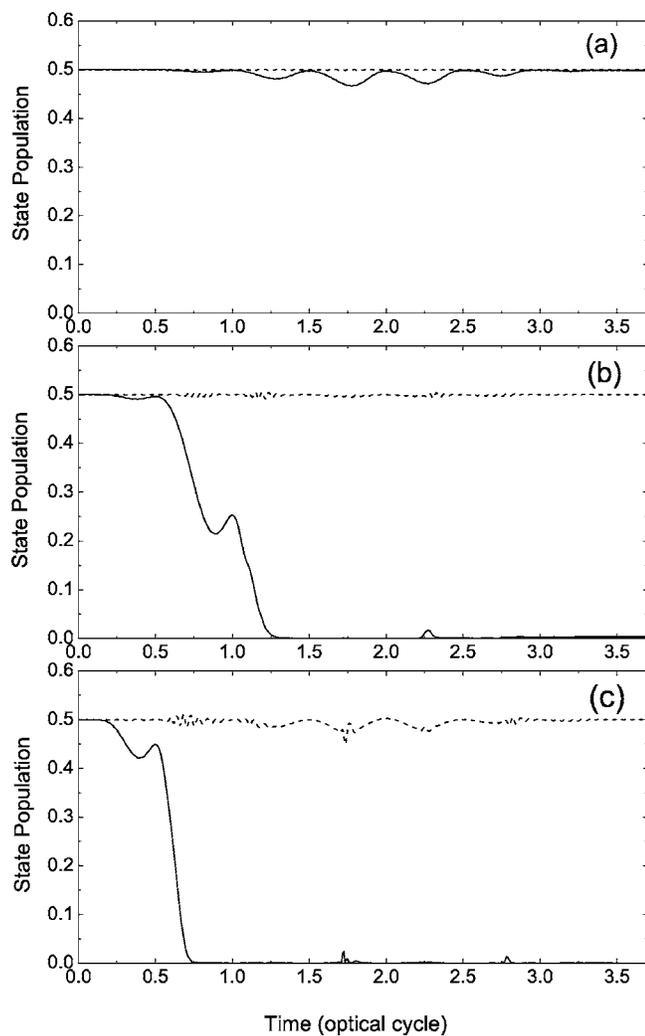


FIG. 1. Populations of the ground and second excited states as a function of time when the initial state is a coherent superposition of the ground and excited states with equally weighted populations. The laser pulse duration is 10 fs and intensities are $I =$ (a) 1×10^{13} W/cm², (b) 5×10^{14} W/cm², and (c) 4×10^{15} W/cm².

Equations (7)–(10) also indicate that dipole moments are directly related to the time-dependent amplitudes of the bound states. This is because harmonic generation originates from the coherent dipole transition between the continuum and the bound states. As a result, only those states that remain populated during the pulse will contribute to the harmonic generation [17].

IV. NUMERICAL RESULTS

We will divide the laser intensity into three regimes, according to the degree of the ionization of the excited state. Figure 1 presents the populations of the ground and second excited states as a function of time when the initial state is a coherent superposition of the ground and excited states with equally weighted population. The laser pulse duration is 10 fs and intensities are $I =$ (a) 1×10^{13} W/cm², (b) 5×10^{14} W/cm², and (c) 4×10^{15} W/cm². In the weak-field

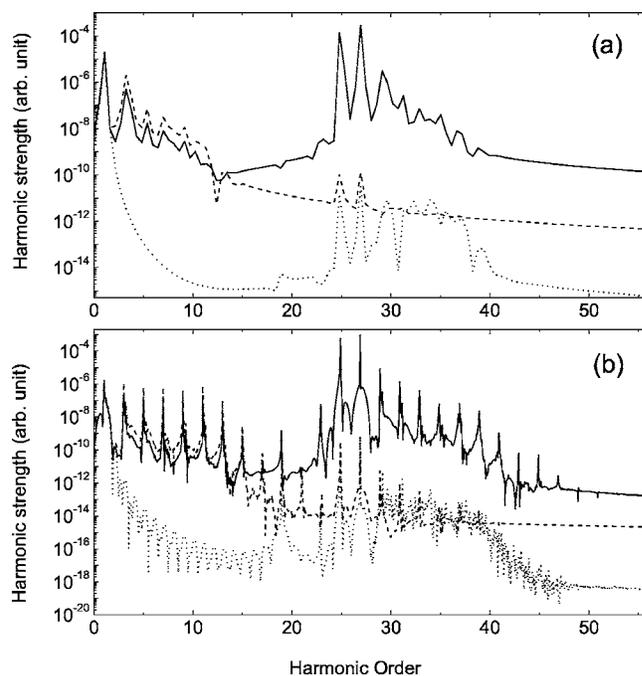


FIG. 2. Harmonic spectra of He⁺ ion with laser intensity 1×10^{13} W/cm² and pulse duration (a) 10 fs and (b) 100 fs when the initial states are superposition (solid curve), ground (dotted curve), and excited states (dashed curve).

regime [Fig. 1(a)] there is only small transference of population from the excited state to the continuum. In contrast, the population of the excited state decreases significantly within the first two optical cycles [from 0.5 to 0.01 within 1.5 optical cycles in Fig. 1(b)] in the intermediate-field regime; while, in the strong-field regime, the excited state is depleted almost completely before the peak of the laser pulse [Fig. 1(c)]. Since ionization plays a crucial role in the generation of harmonic photons, we will demonstrate that the HHG spectra show very different characteristics in different regimes. Furthermore, by comparing the HHG spectra for short and long laser pulses, we find that the spectra exhibit distinct pulse-duration effects, especially when the laser intensity is high.

A. Weak-field regime

We first study the harmonic generation in the weak-field regime in which there is only small ionization of the excited state [Fig. 1(a)]. The solid curves in Fig. 2 show the HHG spectra of He⁺ ion for a coherent superposition state with laser intensity 1×10^{13} W/cm² and pulse duration (a) 10 fs and (b) 100 fs. For comparison, we also present results when the initial state is the ground state (dotted curve), i.e., $\alpha(0) = 1$ and $\beta(0) = 0$, and the second excited state (dashed curve), i.e., $\alpha(0) = 0$ and $\beta(0) = 1$. The HHG spectra of the superposition state (solid curves) clearly show two different sets of harmonics. The first one agrees well with the spectrum of the excited state case (dashed curves), while the second one is about three orders of magnitude higher than that of the ground state case (dotted curve) with the same cutoff har-

monic frequency. We should mention that the harmonic spectra flatten out at the upper end, caused by the background numerical noise, has no physical meaning and does not affect the spectra results.

In the weak-field regime, the amplitudes of the ground and excited states are approximately constant during the laser pulse [see Fig. 1(a)]. On the other hand, it is much easier to ionize the excited state than the ground state; therefore, from Eqs. (7)–(10) we have $|D_{ee}(t)|$, $|D_{ge}(t)| \gg |D_{gg}(t)|$, $|D_{eg}(t)|$. In other words, harmonics of the superposition case originate from the recombination into the ground and excited states of the electron, where the excited state is responsible for the ionization. The maximum kinetic energy that the electron brings back equals $3.17U_p$; therefore, when it recombines into the ground state the energies of the emitted photons are between I_g and $I_g + 3.17U_p$. On the other hand, recombination into the excited state gives harmonics of energy between I_e and $I_e + 3.17U_p$. Two plateaus will be separated if $I_g - I_e > 3.17U_p$. In our system the corresponding laser intensity that two plateaus can be separated is lower than $1 \times 10^{14} \text{ W/cm}^2$.

We compare the HHG spectra of the short [Fig. 2(a)] and long [Fig. 2(b)] laser pulses. For long laser pulses, a short burst of radiation emits every half a laser cycle due to the scatter off the core of the continuum wave packet. As a result, the multicycle accumulation of the harmonic generation causes separate sharp peaks in each odd harmonic order. Besides, harmonics are usually more intense for long laser pulses, especially in the first plateau.

B. Intermediate-field regime

Now, let us consider HHG in the intermediate-field regime. Figure 3 presents the harmonic spectra of the He^+ ion with laser intensity $5 \times 10^{14} \text{ W/cm}^2$ for the pulse duration (a) 10 fs and (b) 100 fs. The HHG spectra of the superposition case (solid curves) show only one plateau, which are about six and five orders of magnitude higher than that of the ground state case (dotted curve) when pulse durations are 10 fs and 100 fs, respectively.

As shown in Fig. 1(b) the population of the excited state decreases significantly within the first two optical cycles in the intermediate-field regime. Since $|\alpha(t)| \gg |\beta(t)|$ at the time of recombination, we have from Eqs. (8) and (9) $|D_{ge}(t)| \gg |D_{ee}(t)|$. Therefore, in contrast to the weak-field case, where the recombination into the ground and excited states gives two plateaus in the HHG spectra, the main contribution to the harmonic generation in the intermediate-field regime is the transition from the continuum to the ground state. This fact is demonstrated further in Fig. 3 where the HHG spectra of the superposition case (solid curves) is about three and two orders of magnitude higher than that of the excited state case (dashed curves) when pulse durations are 10 fs and 100 fs, respectively.

We are interested in producing harmonic photons with high conversion efficiency, which is directly proportional to the population of the continuum and the remaining population of the bound states. In the intermediate-field regime, the laser intensity is high enough to ionize the excited state

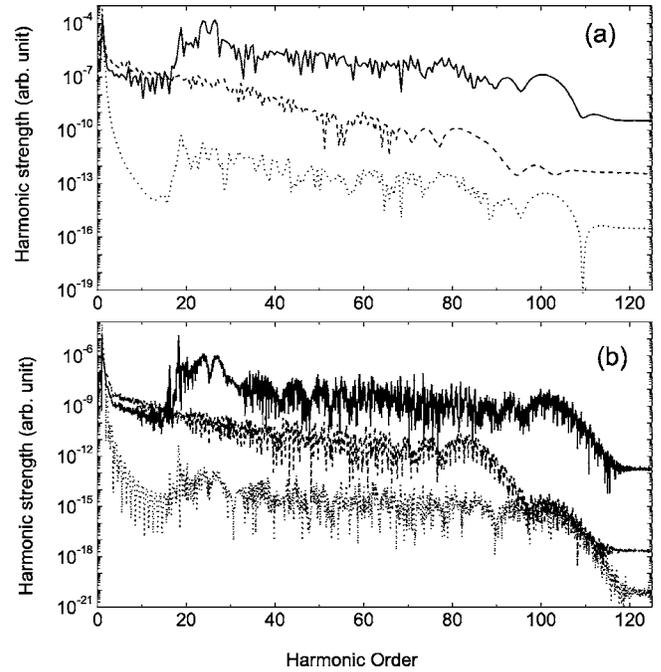


FIG. 3. Harmonic spectra of He^+ ion with laser intensity $5 \times 10^{14} \text{ W/cm}^2$ and pulse duration (a) 10 fs and (b) 100 fs when the initial states are superposition (solid curve), ground (dotted curve), and excited states (dashed curve).

within a few optical cycles, while too weak to directly ionize the ground state. Therefore, if the initial state is prepared as a coherent superposition of the ground state and an excited state, a large dipole transition will be induced between the continuum and the ground state, where the excited state is responsible for the ionization. In our system the intermediate-field regime is from $I \approx 1 \times 10^{14} \text{ W/cm}^2$ to about $1 \times 10^{15} \text{ W/cm}^2$. Moreover, Fig. 4 presents the temporal behavior of the harmonics of the 71st (dashed curve) and 91st (solid curve) harmonic orders for the superposition case when the laser intensity is $5 \times 10^{14} \text{ W/cm}^2$. It shows that harmonic photons emit mainly during the first few optical cycles in which the excited state ionizes efficiently. As a result, conversion efficiency can be increased further if short laser pulses are employed. For example, the HHG of 10 fs pulse is on an average one order of magnitude higher than that of the 100 fs pulse.

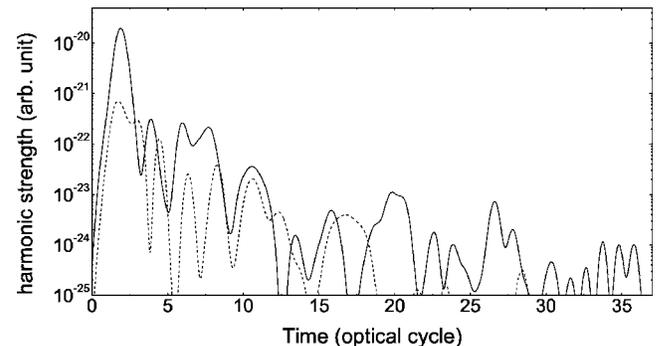


FIG. 4. Temporal behavior of the harmonics of the 71st (dashed curve) and 91st (solid curve) harmonic order for the superposition case when the laser intensity is $5 \times 10^{14} \text{ W/cm}^2$.

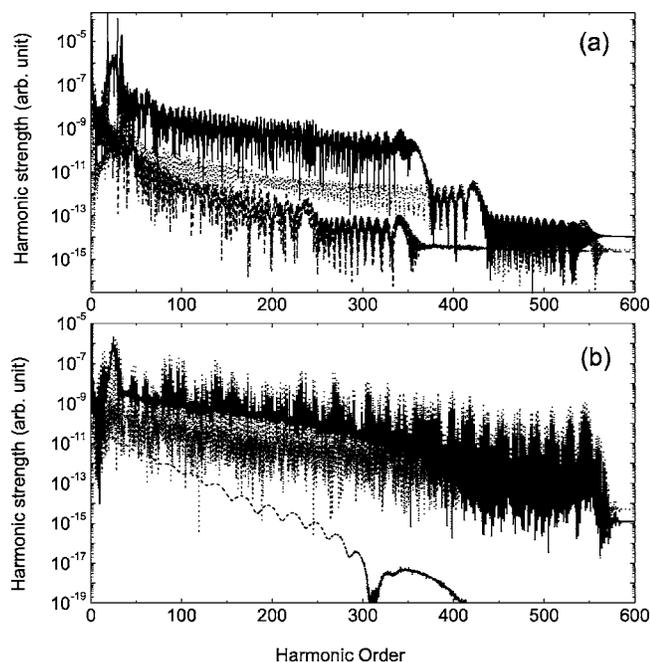


FIG. 5. Harmonic spectra of He^+ ion with laser intensity $4 \times 10^{15} \text{ W/cm}^2$ and pulse duration (a) 10 fs and (b) 100 fs when the initial states are superposition (solid curve), ground (dotted curve), and excited states (dashed curve).

Finally, as shown in Fig. 3(b) the HHG spectrum of the excited state case (dashed curve) exhibits two plateaus with the second cutoff consistent with that of the ground state case (dotted curve) when the laser pulse duration is 100 fs. This is because, under the intermediate laser power, there is population transfer from the excited to the ground states via multiphoton transition. Therefore, the dipole transition can be induced between the continuum and the ground state; even the atom is initially in the excited state.

C. Strong-field regime

We increase the laser intensity further to a point that there is a significant population depletion of the excited state within one optical cycle, and study how this population depletion affects the HHG spectra. Figure 5 presents the HHG spectra of the He^+ ion with $I=4 \times 10^{15} \text{ W/cm}^2$ for the pulse duration (a) 10 fs and (b) 100 fs when the initial states are the superposition state (solid curve), the ground state (dotted curve), and the excited state (dashed curve). Let us first consider the HHG spectra with short pulse duration [Fig. 5(a)]. It is found that the spectrum of the ground state case (dotted curve) exhibits a double-plateau structure. To understand this, we perform a wavelet time-frequency analysis [18] of the spectral and temporal structures of HHG. Figure 6(a) presents the time profile of the harmonics when the initial state is the ground state. It indicates that the cutoff at about the 551st harmonic emits at a time of approximately 1.8 optical cycles. On the other hand, there are at least four trajectories, which contribute to the harmonics below the 431st harmonic order, leading to another plateau with higher strength.

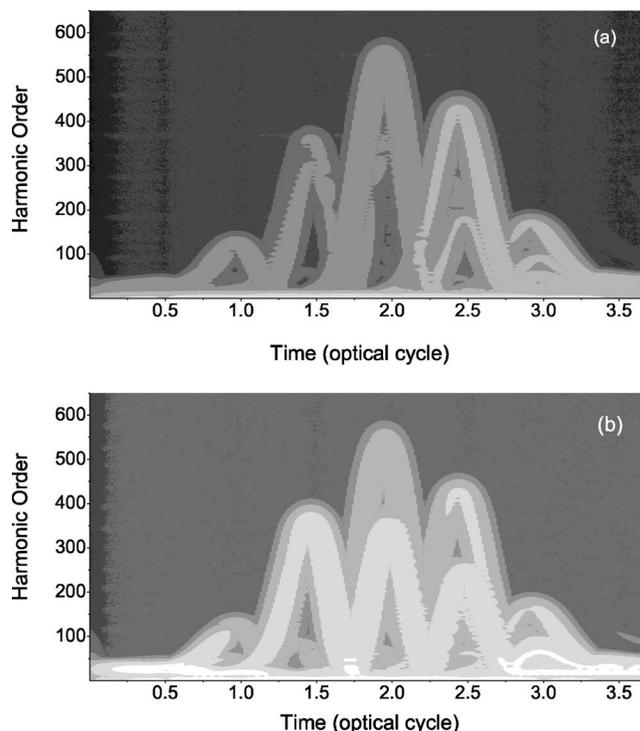


FIG. 6. Time profile of the harmonics when the initial state is (a) the ground state and (b) the coherent superposition state.

We then consider the excited state case [dashed curve in Fig. 5(a)]. At intensity $I=4 \times 10^{15} \text{ W/cm}^2$ the excited state decreases from 0.5 to 0.01 within approximately 0.7 optical cycles [Fig. 1(c)]. As a consequence, the cutoff frequency (at about the 351st harmonic order) is much lower than that predicted by the three-step model, which equals the 521st harmonic order according to the $I_p + 3.17U_p$ law, because the excited state is depleted almost completely before the peak of the laser pulse. Moreover, in contrast to the previous cases, the high depletion of the excited state also causes the harmonic intensity of the excited state case much lower than that of the ground state case because $|\alpha(t)| \gg |\beta(t)|$ most of the time [see Eqs. (7) and (8)].

Now we consider the spectrum of the superposition case [solid curve in Fig. 5(a)], which exhibits a complex structure with three plateaus. The first plateau is about two orders of magnitude higher than that of the ground state case, while the other part of the spectrum agrees well with that of the ground state case. Physically, the HHG spectrum of the superposition case has two contributions: One originates from the dipole moment $D_{gg}(t)$, which gives spectrum above the 375th harmonic order and is consistent with that part of the ground state case. On the other hand, the first plateau in the spectrum is due to the interference term $D_{ge}(t)$. The strength of this plateau is about two orders of magnitude higher than that of the ground state case because of the large transition from the excited state to the continuum, demonstrating once again the advantages of using coherent superposition state to obtain high conversion efficiency. Also, from the wavelet time-frequency analysis [Fig. 6(b)] we find that harmonics at the cutoff of this plateau emit at time of approximately 1.4 optical cycles.

Finally, we consider the HHG spectra of the long pulse duration case [Fig. 5(b)]. First, there is almost no harmonic generation for the excited state case (dashed curve) because the excited state is depleted almost completely within one optical cycle. Second, since there is an effective transition from the ground state to the continuum with very little depletion of the ground state population, the conversion efficiency of the ground state case (dotted curve) is relatively high. Finally, the excited state plays no role in the harmonic generation when the laser has a long pulse duration; as a result, the HHG spectrum of the superposition case (solid curve) is consistent with that of the ground state case. It is worth mentioning that, because the population of the ground state is approximately constant during the laser pulse, there is no advantage of using short pulse in the strong-field regime. In contrast, the multicycle accumulation causes the conversion efficiency of the long pulse higher than that of the short pulse by about three orders of magnitude when the initial state is the ground state.

V. CONCLUSION

There are two factors which can affect the conversion efficiency of HHG, i.e., the ionization rate of the initial

bound states and the remained population of the bound states at the time of recombination. The advantage of using coherent superposition state is that it is possible to induce dipole transition between the continuum and the ground state, where the excited state is responsible for the ionization, thus, drastically increasing the conversion efficiency. In this paper, we have made a systematic study of HHG in a He⁺-like model ion when the initial state is prepared as a coherent superposition of the ground state and an excited state. Since the ionization plays a crucial role in the HHG with coherent initial state, the laser intensity can be divided into three regimes according to the degree of the ionization of the excited state. The HHG spectra exhibit different characteristics in these regimes. We have demonstrated evident advantages of using coherent superposition state to obtain high conversion efficiency. We have also found distinct pulse-duration effects in the intermediate- and strong-field regimes.

This research was supported by the National Natural Science Foundation of China under Grant No. 60478031, and the Climbing Programme of the Ministry of Science and Technology of China. B. Wang thanks Professor Qiren Zhu, Professor Armin Scrinzi, Dr. Jing Chen, and Dr. Jiangbin Gong for helpful discussions.

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