Efficient enhancement of below-threshold harmonic generation by laser-driven excited states of Cs atom

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\textbf{A B S T R A C T}

We propose an efficient method for the enhancement of below-threshold harmonic generation (BTHG) by mid-infrared laser-driven excited states of a Cs atom. The BTHG is calculated by solving three-dimensional time-dependent Schrödinger equation accurately and efficiently using the time-dependent generalized pseudospectral method. We adopt an excited state as the initial state of a Cs atom. As a result, the BTHG is significantly enhanced by two orders of magnitude compared with the case of the initial ground state. Furthermore, we find that a single vacuum-ultraviolet pulse can be generated by mid-infrared laser-driven excited states by superposing several below-threshold harmonics of a Cs atom. Our finding suggests that the generation of below-threshold harmonics by laser-driven excited states of an atom can provide a powerful methodology for the production of intense vacuum-ultraviolet pulses.

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1. Introduction

High-order harmonic generation (HHG) by atomic and molecular systems caused by intense laser fields provides a powerful tool to produce an ultrashort attosecond pulse, leading to many physical applications, such as attosecond time-resolved spectroscopy [1,2] and the direct probing of electronic dynamic behavior in atoms, molecules, and condensed matter [3–11]. The HHG spectrum is characterized by a rapid drop at low orders followed by a broad plateau where all harmonics have similar amplitudes, and finally a sharp cutoff, beyond which no further harmonic emission is observed. The HHG process can be well described by the semiclassical three-step model [12,13]: first, the electron is ionized by tunneling, then accelerated, and finally driven back towards the parent core to recombine into the ground state and emit harmonic photons. The highest harmonic photon energy is located approximately at the cutoff energy $E_c = 3.89 \text{ eV}$, which allows us to obtain the harmonic spectrum characterized by the ponderomotive potential ($U_p$) and the atomic ionization potential ($I_p$). The enhancement of the HHG through increasing the ionization probability at the peak intensity of the laser field is one of the most promising routes. To achieve higher ionization probability, the initial state as a preexcited electronic state [18,19] or a coherent superposition of two bound states [20–26] is used. However, the conversion efficiency of above-threshold harmonic generation (photon energy above the ionization potential) is generally much lower than that of below-threshold harmonic generation (BTHG). The enhancement of the BTHG by a preexcited electronic state has not been studied much. More recently, the BTHG has attracted considerable attention [27–32] and has become one of the most active research directions in vacuum-ultraviolet (VUV) frequency comb generation through below-threshold harmonics.

In this paper, we propose a method to enhance the BTHG using a mid-infrared laser to drive the excited states of a Cs atom. The three-dimensional (3D) time-dependent Schrödinger equation (TDSE) is solved accurately and efficiently using the time-dependent generalized pseudospectral method [33]. The Cs atom has been chosen particularly as the atomic target for the below-threshold harmonic generation due to the following two reasons: (i) The Cs atom possesses a lower ionization potential ($I_p=3.89 \text{ eV}$), which allows us to obtain the harmonic generation by using the lower laser intensity, and (ii) Power et al. [27] have pointed out that the Cs atoms as the target replace the inert gas atoms, which provides an opportunity to shift the harmonic spectrum and $I_p$ from the VUV into a spectral region where well-developed temporal metrology can be used to characterize the harmonic radiation near threshold. The results show that the generation of below-threshold harmonics is enhanced owing to the contribution of the excited states.
and the efficiency of the BTHG is significantly enhanced by two orders of magnitude compared with the case when a Cs atom is in the initial ground state. In addition, a single VUV pulse is produced by mid-infrared laser-driven excited states through superposing several below-threshold harmonics of a Cs atom. To probe the origin of the enhancement of the BTHG, we perform the calculations of time-dependent excited state population and the time-frequency transform of the BTHG. We find that the excited states of a Cs atom play an important role in enhancing the efficiency of the BTHG.

The paper is organized as follows: In Section 2 we introduce the theoretical method, namely, the solution to the 3D TDSE. In Section 3 we present and discuss the BTHG results, and propose a method to enhance the BTHG using a mid-infrared laser to drive the excited states and the coherent superposition states of a Cs atom. Conclusions are given in Section 4. Atomic units are used throughout the paper unless otherwise stated.

2. Numerical methods

The BTHG is produced by the interaction of an intense laser field with the atoms, which can be calculated by solving the following TDSE (in atomic units). In the length gauge, the TDSE in the dipole approximation for a single-atom interacting with a laser field is given by:

$$\frac{\partial}{\partial t} \psi(r, t) = \hat{H}_0 \psi(r, t) + \hat{V}(r, t) \psi(r, t),$$

where \(\hat{V}(r, t) = \hat{H}_0 \psi(r, t) + \hat{V}(r, t)\psi(r, t)\),

(1)

is the time-dependent atom–field interaction, it is given by

$$\hat{V}(r, t) = -E(t) \cdot r = -E_0 f(t) z \cos \omega t,$$

(2)

where \(E_0\) is the electric field amplitude, \(f(t)\) is the pulse envelope, and \(\omega\) is the field frequency. Here, \(\hat{H}_0\) represents the field-free Hamiltonian for a Cs atom, which can be written as

$$\hat{H}_0 = -\frac{1}{2} \overline{V} + |\Psi_0^\dagger \Psi_0|^2,$$

(3)

where \(V\) is the model potential for a Cs atom, and \(|\Psi_0^\dagger \Psi_0|^2\) is the spherical harmonic. The model potential method has been widely used to provide accurate descriptions of electron scattering, photoionization, and particle impact ionization processes, as well as for the calculations of atomic and molecular properties for one- and two-valence electron systems. To obtain an accurate calculation of the harmonic spectra of a Cs atom, an angular momentum-dependent model potential [32] is used, which is given by

$$V_f = -\frac{a}{2r^4} W_f(r) - \frac{1}{r} \left( \frac{N - S}{r} + A_1 \right) e^{-B_1 r} - \left( \frac{S}{r} + A_2 \right) e^{-B_2 r},$$

(4)

where \(a\) is the Cs core dipole polarizability, \(r_s\) is the effective Cs core radius, and the values of the parameters determined are listed in Table 1. \(W_f(x)\) is a core cutoff function, it is given by

$$W_f(x) = 1 - [1 + x + (2x)^2 + \cdots + (nx)^n],$$

(5)

The TDSE is solved accurately and efficiently using the time-dependent generalized pseudospectral method (TDGPS). The numerical scheme of the TDGPS method consists of two essential steps [33]. First, the spatial coordinates are optimally discretized in a nonuniform fashion using the generalized pseudospectral technique with denser grids near the nuclear origin and sparser grids for larger distances. Second, a second-order split-operator technique in energy representation, which allows the explicit elimination of undesirable fast-oscillating high-energy components, is used for the efficient and accurate time propagation of the wave function:

$$\psi(r, t + \Delta t) \approx \exp(-i\hat{H}_0 \Delta t/2) \exp[-i\hat{V}(r, \theta, t + \Delta t/2) \Delta t] \times \exp(-i\hat{H}_0 \Delta t/2) \psi(r, t) + O(\Delta t^3).$$

(6)

Once the time-dependent wave function \(\psi(r, t)\) is available, we can calculate the expectation value of the induced dipole moment in the length form:

$$d(t) = \langle \psi(r, t) | z | \psi(r, t) \rangle.$$  

(7)

The HHG power spectra in the length form can be obtained using the Fourier transformation of the time-dependent dipole moment \(d(t)\),

$$P(\omega) = \left| \int_{t_f}^{t_i} d(t)e^{-i\omega t} dt \right|^2.$$  

(8)

By superposing several harmonics, the VUV pulse can be calculated as follows:

$$I(t) = \sum_q a_q e^{i\omega_q t},$$  

(9)

where \(a_q = \int d(t)e^{-i\omega_q t} dt\).

3. Results and discussions

The BTHG power spectra of a Cs atom were calculated by solving the 3D TDSE with an accurate angular momentum-dependent model potential in an intense mid-infrared laser field. In our calculation, we use a cosine-squared shape and a duration of 20 optical cycles, and the laser intensity is \(I = 5 \times 10^{11} \text{W/cm}^2\). The atomic ionization potential of Cs is equal to 0.143099 a.u. [34], which coincides with 10 harmonic orders. The ionization energies of excited states 6s, 7s, 7p, 8s are 0.0904751, 0.0586446, 0.0433755, and 0.0323015 a.u., respectively.

These ionization energies are in good agreement with Ref. [34]. Fig. 1(a)–(d) show a comparison of the below-threshold harmonic spectra between ground state 6s and excited states 6p, 7s, 7p, 8s of a Cs atom driven by (a) 4325 nm, (b) 3235 nm, (c) 3196 nm, (d) 3288 nm laser pulse with the same laser intensity \(I = 5 \times 10^{11} \text{W/cm}^2\), respectively. The choice of the laser wavelength coincides with five-photon resonance transition 6s-6p (4325 nm), six-photon resonance transition 6s-7s (3235 nm), seven-photon resonance transition 6s-7p (3196 nm), and eight-photon resonance transition 6s-8s (3288 nm), respectively. In Fig. 1(a), a comparison of the below-threshold harmonic spectra between ground state 6s and excited state 6p is presented, the intensity of the harmonic spectra of excited state 6p is higher by about two orders of magnitude than those of ground state 6s alone. In Fig. 1(b), the intensity of the harmonic spectra of excited state 7s is higher by about 1 order of magnitude than those of ground state 6s alone. Furthermore, the intensities of the harmonic spectra of excited state 7p and excited state 8s are higher by about one or two orders of magnitude than that of ground state 6s alone as shown in Fig. 1(c) and (d), respectively.

To understand the enhancement of the below-threshold harmonic spectra by the excited state, Fig. 2(a)–(d) show the population of the ground state and excited states, and the time-dependent dipole moment of the excited states, respectively. In Fig. 2(a), the population of the ground state is very high, it implies that the ionization rate is small, which leads to weak harmonic radiation. However, the population of excited state 6p drops sharply and approaches zero in nine optical cycles, it indicates that large ionization occurs before the ninth optical cycle. In addition, we find that the magnitude of the time-dependent dipole moment of excited state 6p is increased due to ionization. In Fig. 2(b), the population of excited state 7s shows that electrons are almost completely ionized before the fourth optical cycle, and the corresponding magnitude of the time-dependent dipole moment of excited state 7s is increased largely, which implies that the emission of harmonics is enhanced. Similar results can be obtained from excited states 7p and 8s as shown in Fig. 2(c) and (d), respectively.

Next, to explore the role of the excited states on the enhancement of the below-threshold harmonics, we investigate the BTHG from Cs initially prepared in a coherent superposition of the ground state and an excited...
Table 1
Model potential parameters for Cs (in a.u.)[32].

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Fig. 1. Comparison of the below-threshold harmonic spectra of a Cs atom between ground state 6s and the excited states (a) 6p, (b) 7s, (c) 7p, and (d) 8s, respectively. Note that the choice of the laser wavelength coincides with the five-photon resonance transition 6s-6p (4325 nm), six-photon resonance transition 6s-7s (3235 nm), seven-photon resonance transition 6s-7p (3196 nm), and eight-photon resonance transition 6s-8s (3288 nm), respectively. The laser has a cosine-squared pulse and a duration of 20 optical cycles, and the intensity is $I = 5.0 \times 10^{11}$ W/cm$^2$.

Fig. 2. Population between ground state 6s and excited states (a) 6p, (b) 7s, (c) 7p, and (d) 8s, respectively. The red line indicates the time-dependent dipole moment of the excited states. The laser parameters used are the same as those in Fig. 1. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

state [21] driven by mid-infrared laser pulses. The harmonic spectrum and the atomic structure are closely related. Milošević [35] has indicated that one individual atom had enough time to interact with more pulses from the pulse train, so that a superposition of ground and excited states may be generated. We define the coherent superposition state as

$$\psi(r) = a|g\rangle + \beta|e\rangle,$$

where $a$ and $\beta$ are the amplitudes of the ground state and the excited states, and $a^2 + \beta^2 = 1$. According to previous study [21], low-energy harmonics scale as $\beta^4$ and high-energy ones scale as $a^2\beta^2$. We indicate
The intensity of the fifth harmonic (SH) as a function of the ratio between the amplitudes of the ground and excited states. Other laser parameters used are the same as those in Fig. 1.

Fig. 3. (a) Relative efficiencies of the HHG corresponding to coherent superposition states $\sqrt{1/10}|6s\rangle + \sqrt{2/10}|6p\rangle$, $\sqrt{1/10}|6s\rangle + \sqrt{1/10}|7p\rangle$, and $\sqrt{1/10}|6s\rangle + \sqrt{1/10}|6p\rangle$, respectively. (b) The intensity of the fifth harmonic (SH) as a function of the ratio between the amplitudes of the ground and excited states. Other laser parameters used are the same as those in Fig. 1.

Fig. 4. Comparison of the below-threshold harmonic spectra between excited states $6p$, $7s$, $7p$, and $8s$, alone, and coherent superposition states (a) $1/\sqrt{2}|6s\rangle + |6p\rangle$, (b) $1/\sqrt{2}|6s\rangle + |7p\rangle$, (c) $1/\sqrt{2}|6s\rangle + |7p\rangle$, and (d) $1/\sqrt{2}|6s\rangle + |8s\rangle$. The laser parameters used are the same as those in Fig. 1.

this result in Fig. 3(a), where we present the harmonic spectra for $a = \sqrt{1/10}$, $\beta = \sqrt{9/10}$ (black solid line), $a = \beta = 1/\sqrt{2}$ (red solid line), and $a = \sqrt{9/10}$, $\beta = \sqrt{1/10}$ (blue solid line), respectively. As predicted above, low-energy harmonics (the first harmonic and the third harmonic) are the highest, when the $a^2 : \beta^2$ ratio is 1:9, but a higher-energy harmonic (the fifth harmonic) has the highest intensity, when the ratio is 1:1 ($a = \beta = 1/\sqrt{2}$ ) as shown in Fig. 3(b). Therefore, we choose $a = \beta = 1/\sqrt{2}$ in calculation and adopt the laser wavelength suggested in Fig. 1.

Fig. 4(a)–(d) show a comparison of the below-threshold harmonic spectra between excited states $6p$, $7s$, $7p$, and $8s$, as well as the coherent superposition states of the excited states and ground state $6s$, respectively. The laser parameters used are the same as those in Fig. 1. In Fig. 4(a) (the coherent superposition of excited state $6p$ and ground state $6s$), the transition between the ground state and the excited state is dipole allowed. We find that the intensity of the fifth harmonic of the coherent superposition state is higher by about three orders of magnitude than that of excited state $6p$ alone. Namely, there is a huge enhancement of the fifth harmonic. Furthermore, we find there is a high-frequency tail of the HHG owing to high-energy harmonic scaling as $a^2\beta^2$ as shown in Fig. 3(a). In Fig. 4(b) (the coherent superposition of excited state $7s$ and ground state $6s$), the transition between the ground state and the excited state is dipole forbidden. However, we still find a small enhancement of the seventh harmonic. The reason is that the initially forbidden transition between the states becomes allowed when the laser field is strong enough to break the symmetry. Nevertheless, this enhancement of the HHG is smaller than the case shown in Fig. 4(a).

In Fig. 4(c), the result shows an apparent enhancement of the seventh harmonic, this is similar to the one shown in Fig. 4(a). In Fig. 4(d), the result is similar to the one shown in Fig. 4(b). Therefore, these results indicate that the enhancement of the given below-threshold harmonics can be controlled by choosing the coherent superposition states as an initial state.

To explain the enhancement of the below-threshold harmonic spectra by the coherent superposition states, Fig. 5(a)–(d) show the population of the ground state and the excited states, and the corresponding time-dependent dipole moments of the coherent superposition states are also presented. In Fig. 5(a), the population of excited state $6p$ in the coherent superposition state shows an apparent decrease and is depleted in nine optical cycles, and the corresponding time-dependent dipole moment of the coherent superposition state oscillates sharply before the second optical cycle, a similar result is obtained in Fig. 5(c), but the oscillation amplitude is small owing to the small transition dipole matrix elements ($6s$→$7p$) [36] compared to the transition of $6s$→$6p$. However, there is no oscillation amplitude before the second optical cycle in Fig. 5(b) and (d) owing to them being dipole forbidden ($6s$→$7s$ and $6s$→$8s$).

To better understand the enhancement of the harmonic emission from the coherent superposition states, we perform the wavelet time-frequency analysis [37] of the below-threshold harmonic spectra of
Fig. 5. Population of ground state 6s and excited states (a) 6p, (b) 7s, (c) 7p, and (d) 8s, respectively, which are in coherent superposition states (a) $1/\sqrt{2}(|6s\rangle + |6p\rangle)$, (b) $1/\sqrt{2}(|6s\rangle + |7s\rangle)$, (c) $1/\sqrt{2}(|6s\rangle + |7p\rangle)$, and (d) $1/\sqrt{2}(|6s\rangle + |8s\rangle)$. The red line indicates the time-dependent dipole moment of the coherent superposition states, respectively. The other laser parameters used are the same as those in Fig. 1. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Fig. 6. The wavelet time–frequency analysis of the below-threshold harmonic spectra of coherent superposition states $1/\sqrt{2}(|6s\rangle + |6p\rangle)$ (a) and $1/\sqrt{2}(|6s\rangle + |7s\rangle)$ (b). The laser wavelengths are 4325 nm and 3235 nm, respectively, with the same intensity $I = 5.0 \times 10^{11}$ W/cm\(^2\).

Fig. 7. Physical process of the HHG.

coherent superposition states $1/\sqrt{2}(|6s\rangle + |6p\rangle)$ and $1/\sqrt{2}(|6s\rangle + |7s\rangle)$, the results are shown in Fig. 6(a) and (b), respectively. In Fig. 6(a), as the dipole allowed case, the dominant emission of the fifth harmonic is between the zeroth and tenth cycles, but the emissions of the third and seventh harmonics are between the second and tenth cycles. In fact, the contribution of the nonresonant emission of the below-threshold HHG comes from dressed states (see the physical process of the HHG shown in Fig. 7). Therefore, we find that the fifth harmonic emission clearly shows two components: one starts before the second optical cycle corresponding to the contribution of the resonance transition between 6s and 6p states; another one starts after two optical cycles corresponding to the contribution of dressed states (single states). Indeed, the intensity of the harmonics is many orders of magnitude higher than that of the harmonics generated from the ground state alone, and the HHG spectrum from a coherent superposition of state consists of two components [35]: the dressed state harmonic generation and resonant states. The dressed state harmonic generation corresponds to odd-order harmonics. However, resonant states, which correspond to the dipole transitions between the excited and ground states, lead to the emission of the harmonics with frequencies that are dependent on the energy interval of two states. Nevertheless, in Fig. 6(b), as the dipole forbidden case, time–frequency spectra only show the dressed states harmonic emission, and there is no resonant component.

Fig. 8 shows the time-domain synthesis of several below-threshold harmonics from the laser-driven ground state, superposition states, and excited states of a Cs atom. It is clear that the intensity of the VUV pulse generated from the excited states and the superposition states is many orders of magnitude higher than that of the harmonics generated in the presence of the ground state alone, note that the inset shows the case of the ground state alone. It is interesting that a single VUV pulse is produced through superposing several below-threshold harmonics from the laser-driven excited state.
Fig. 8. Time-domain synthesis of several below-threshold harmonics from laser-driven ground state 6s, excited states 6p, 7s, 7p, 8s, and superposition states $1/\sqrt{2}(|6s\rangle + |6p\rangle)$, $1/\sqrt{2}(|6s\rangle + |7s\rangle)$, $1/\sqrt{2}(|6s\rangle + |7p\rangle)$, $1/\sqrt{2}(|6s\rangle + |8s\rangle)$ of a Cs atom, respectively. The parameters used are the same as those in Fig. 3.

Fig. 9. Wavelet time–frequency analysis of the below-threshold harmonic spectra of ground state 6s (a), coherent superposition state $1/\sqrt{2}(|6s\rangle + |8s\rangle)$ (b), and excited state 8s (c). The laser wavelength is 3288 nm, and the intensity is $I = 5.0 \times 10^{11}$ W/cm$^2$.

To understand the generation of a single VUV pulse, we perform the wavelet time–frequency analysis of the HHG from ground state 6s, superposition state $1/\sqrt{2}(|6s\rangle + |8s\rangle)$, and excited state 8s. The results are shown in Fig. 9(a)–(c). In Fig. 9(a), the wavelet time–frequency analysis of the BTHG of ground state 6s shows that the main emissions of the below-threshold harmonics are between 4 and 16 optical cycles. In Fig. 9(b), the wavelet time–frequency analysis of the BTHG of coherent superposition state $1/\sqrt{2}(|6s\rangle + |8s\rangle)$ shows that the strong emission of the below-threshold harmonics are throughout the optical cycles, so we obtain the trains of the VUV pulse. In Fig. 9(c), the wavelet time–frequency analysis of the BTHG of excited state 8s shows that the strong emission of the below-threshold harmonics is located at a relatively short time duration. As discussed above, the emission rate of the below-threshold harmonics from the excited state is many orders of magnitude higher than that of the harmonics generated in the presence of the ground state alone due to the rapid ionization at the beginning of several cycles. So the emission of the harmonics allows to happen in a relatively short duration. As a result, a single VUV pulse is produced directly by superposing several below-threshold harmonics from the laser-driven excited states. Although the mechanism of the enhancement of the below-threshold harmonics from a coherent superposition state is
similar to that of the exited states, the emission of the harmonics from a coherent superposition state has a long duration due to the contribution of the resonant component.

4. Conclusions

We have presented an efficient method for the enhancement of the BTHG by the mid-infrared laser-driven excited states of a Cs atom. The BTHG is calculated by accurately solving the 3D TDSE. We propose an excited state as the initial state of time propagation, leading to a significantly enhanced emission of the BTHG. We find that the BTHG from the excited state is enhanced by two orders of magnitudes compared with the case of the ground state alone. Particularly, a single VUV pulse is produced by superposing several below-threshold harmonics from the laser-driven excited states of a Cs atom. In addition, to explore the role of the exited states in the enhancement of the below-threshold harmonics, we study the HHG from Cs initially prepared in a coherent superposition of the ground state and an excited state. We find that the enhancement of the given below-threshold harmonics can be controlled by choosing a coherent superposition state as an initial state of the Cs atom. To understand the enhancement of the below-threshold harmonic spectra, we perform the calculations of the populations of the excited state and the time–frequency transform of the BTHG. In addition, it is well know that the propagation effect is important to enhance harmonic generation, but the calculation of the propagation effect of Cs atom based on the TDSE is expensive, hopefully we can consider the propagation effects of below-threshold HHG of Cs atom in the future.

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