Intense isolated attosecond pulse generation in pre-excited medium

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Abstract: We present a theoretical study of isolated attosecond pulse generation in pre-excited medium driven by an intense few-cycle laser pulse. We show that the generation of the macroscopic xuv supercontinuum is governed by the initial population of the excited state. Large initial population of the excited state leads to the high density of the free electrons in the media and strongly changes macroscopic properties of the driving pulse and the supercontinuum, and diminishes the isolated attosecond pulse generation. Instead, small initial population of 5% in the pre-excited media subjected to the few-cycle driving pulse can produce well phase-matched xuv supercontinuum, and a pure intense isolated attosecond pulse with the pulse duration of approximately 150 as and the pulse energy up to 0.5 nJ is directly obtained.

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1. Introduction

Fascinating developments in attoscience and technology lead to the generation of isolated xuv pulses as short as one hundred attoseconds [1]. This paved the way to some dramatic applications to the study of the ultrafast processes with extremely short time-scales, such as the electronic dynamics inside atom [2] and on the surface of metal nanostructure [3]. Nowadays, high-order harmonic generation (HHG) is the unique way to produce isolated attosecond pulses in experiment. In the past few years, much effort has been expended to broaden the bandwidth and shorten the duration of the isolated attosecond pulse for the higher time resolution in its potential applications [4–10]. The 100-as-barrier has been first brought through by Goulielmakis et al [4]. In their experiment, a sub-4-fs near-single-cycle driving pulse has been employed to generate a 40-eV supercontinuum and a 80-as pulse with the pulse energy of 0.5 pJ has been filtered out. Recently, a very broadband xuv continuous spectrum, which supports 16-as isolated pulse generation, has been produced with double-optical-gating technique (DOG) [5]. On the other hand, the energy of the attosecond pulse is crucial since the extremely low pulse energy of the available attosecond sources would prevent the demonstration of attosecond pump-probe spectroscopy. Very recently, an isolated attosecond source with the pulse energy that reaches...
nanojoule-level has been achieved by Ferrari et al [11].

Microscopically, the HHG process can be depicted by the classical three-step model [12]: ionization, acceleration and recombination of the electrons in the intense laser field. This classical picture not only well explains the time-frequency properties of HHG, but also suggests that HHG process can be manipulated via modulating the external field or the target to broaden the bandwidth of the generated attosecond pulses or enhance the pulse efficiency. Two-color and multi-color schemes have been introduced to control electron dynamics or confine the ionization process within a short time to generate broadband attosecond pulses [5, 6, 8–10, 13, 14]. Asymmetric molecules have also been suggested to work as a time gating for broadband attosecond pulse generation [15, 16]. The harmonic efficiency is mainly associated to the ionization rate when the intensity of the driving field is far below the saturation intensity of the target [17]. It has been proposed that the ionization rate can be significantly enhanced by synchronizing a xuv field [18–22]. This xuv-assisted ionization enhancement can also gate the ionization times to produce efficient isolated attosecond pulse [22]. Another alternative way is to prepare a coherent superposition state of the target, i.e., to populate part of the electron wave packet (EWP) at the excited state [23, 24]. This part of EWP can be easily projected to the continuum, while the population of EWP at the ground state with much larger ionization potential is hardly ionized and prevents the saturation of HHG. Then, such pre-excited system with large ground-state potential can experience very intense driving field and emits harmonic photons with high energy and considerable efficiency, which serves as a good candidate to achieve intense isolated attosecond pulse generation.

As in many other nonlinear processes, HHG and attosecond pulse generation also intensively depend on the propagation and phase-matching in the macroscopic media [25, 26]. For a pre-excited system, the population of EWP on the excited state would be completely depleted after the strong driving field. Then the plasma dispersion due to the highly-ionized media significantly influences the propagation of HHG. In other words, the macroscopic characteristics of the generated attosecond pulses, including the phase-matching, temporal, spectral and spatial distributions are associated to the initial population of the excited state of the medium. To the best of our knowledge, the propagation effects of attosecond pulse generation in the pre-excited system have never been reported. In the paper, we theoretically investigate the macroscopic effect of the intense isolated attosecond pulse generation in the pre-excited medium with a strong few-cycle driving field. It is found that the initial population of the excited state of the medium significantly influences the macroscopic xuv supercontinuum. Small population of the excited state of 5% can lead to good phase-matching of the xuv supercontinuum, and a pure isolated 150-as pulse with the pulse energy up to 0.5 nJ is obtained.

2. Model

The theoretical investigation of HHG in the medium involves calculation of the dipole response in single-atom level induced by the laser field and simulation of the copropagation of the driving laser and harmonic beams. At the single-atom level, the initial state of the pre-excited system can be written as

$$\Psi(r,t_0) = \alpha |g\rangle + \beta |e\rangle,$$

where the ground state $|g\rangle$ and excited state $|e\rangle$ are chosen to be the 1s and 2s states of the $He^+$ ions with the binding energies of 54.4 eV and 13.6 eV, respectively. $\alpha$ and $\beta$ are the amplitudes of the ground and excited states, and $\alpha^2 + \beta^2 = 1$. The initial population of the excited state can be expressed as $p = \beta^2$. The instantaneous dipole moment of the system in single-atom response is

$$d_{nl}(t) = \langle \Psi(r,t) | \hat{x} | \Psi(r,t) \rangle,$$
which is obtained by numerically solving the time-dependent Schrödinger equation. The calculation details can be found in [22]. The harmonic spectrum is then obtained by Fourier transforming the time-dependent dipole acceleration $\ddot{a}(t)$:

$$a_q = \frac{1}{T} \int_0^T \dot{a}(t) \exp(-iq\omega t) dt,$$

(3)

where $\dot{a}(t) = \ddot{a}(t)$, $T$ and $\omega$ are the duration and frequency of the driving pulse, respectively. $q$ corresponds to the harmonic order.

The collective response of the macroscopic medium is described by the copropagation of the laser and the high harmonic field, which can be written separately by [27]

$$\nabla^2 E_f(\rho, z, t) - \frac{1}{c^2} \frac{\partial^2 E_f(\rho, z, t)}{\partial t^2} = \frac{\omega_p^2(\rho, z, t)}{c^2} E_f(\rho, z, t)$$

(4)

$$\nabla^2 E_h(\rho, z, t) - \frac{1}{c^2} \frac{\partial^2 E_h(\rho, z, t)}{\partial t^2} = \frac{\omega_p^2(\rho, z, t)}{c^2} E_h(\rho, z, t) + \mu_0 \frac{\partial^2 P_{nl}(\rho, z, t)}{\partial t^2}$$

(5)

where $E_f$ and $E_h$ are laser and harmonic fields, $\omega_p = e\sqrt{4\pi n_e(\rho, z, t)/m_e}$ is the plasma frequency, and $P_{nl}(\rho, z, t) = [n_0 - n_e(\rho, z, t)] d_{nl}(\rho, z, t)$ is the nonlinear polarization generated by the medium. $n_0$ is the gas density and $n_e(t) = n_0 [1 - \exp\left(-\int_{-\infty}^{t} W(t') dt'\right)]$ is the free-electron density in the gas. The equations here take into account both temporal plasma induced phase modulation and the spatial plasma lensing effects on the driving field. They do not consider the linear gas dispersion and the depletion of the fundamental beam during the HHG process, which is due to the low gas density (75 torr at room temperature in our scheme) [27]. Then the induced refractive index $n$ can be approximately described by the refractive index in vacuum ($n=1$). Equation (4) and (5) can be numerically solved using the Crank-Nicholson method. The calculation details can be found in [28].

3. Result and discussion

In our investigation, a 4-fs 800-nm few-cycle driving pulse with the intensity of $8 \times 10^{14} W/cm^2$ is used. The electric field of the laser pulse is expressed as

$$\vec{E}(t) = E_0 \cos(\omega(t - T_0)) + \phi \sin^2\left(\frac{\pi t}{T}\right) \vec{x},$$

(6)

where $E_0$, $T_0$ and $\phi$ are the amplitude, optical cycle and carrier-envelope phase of the driving field, respectively. For this choice of wavelength and laser intensity, the excited state is responsible for ionization during the laser-matter interaction, while the ionization of the ground state is negligible. Figure 1 shows the sketch of our scheme. The yellow and grey regions in Fig. 1(a) present the normalized populations of the excited and ground states as a function of time during the laser field (red line). The tunnelling ionization of the electron at the excited state is much easier to take place than that at the ground state. As shown in Fig. 1(a), the population of the ground state is approximately constant during the laser field, while the population of the excited state steeply decreases to zero between 0.8$T_0$ and 1.1$T_0$, which indicates that the ionization rate in this time interval is significantly higher than others. According to the three-step model, the electrons ionized in this time interval may return to the ground state and emit harmonic photons with high efficiency. It is important to mention that the electron ionized in this time interval cannot return to the excited state since there is nearly no population at the excited state after the steep depletion and the excited state cannot be coupled to the continuum to induce dipole momentum. Figure 1(b) is the corresponding classical electron trajectories in
the driving pulse. The ionization and recombination times are shown by red and blue dots. Here we only consider the electrons return to the ground state. As shown in this figure, there are three main peaks of the return electrons with the maximum harmonic orders of 115th, 135th and 105th, which are marked as $P_1$, $P_2$ and $P_3$, respectively. The electrons of $P_1$ are ionized near $1T_0$, and the ionization times of $P_1$ are totally within the time interval where the population of the excited state is steeply depleted and dominates the ionization of the system. This is the reason that the driving pulse duration is chosen to be 4 fs. Therefore, the harmonics contributed from $P_1$ would be much stronger than others. Note that the rapid depletion of the excited state takes place in a very short time, which also works as a temporal gating for isolated attosecond pulse generation.

![Normalized populations of the excited (yellow region) and ground states (grey region) as a function of time during the laser field (red line) and (b) the corresponding classical electron trajectories during the driving pulse.](image)

**Fig. 1.** (a) The normalized populations of the excited (yellow region) and ground states (grey region) as a function of time during the laser field (red line) and (b) the corresponding classical electron trajectories during the driving pulse.

In order to confirm the prediction of the sketch, we calculate the high-order harmonic generation in single-atom level. For convenience, we first consider that the populations of the ground and excited states are equally weighted. The harmonic spectrum is shown in Fig. 2(a) by red thick line. The harmonic spectrum when the initial state is the ground state is also presented for comparison (grey dotted line). The result agrees well with the prediction of the sketch in Fig. 1 that the harmonic efficiency from the superposition state is 6 orders higher than that from the ground state with the same cutoff harmonic order of approximately 135th. The two-plateau structure in both cases is due to the great difference of ionization rates between $P_1$ and $P_2$. The difference of the harmonic efficiency of the two plateaus for the case of the superposition state is much larger (up to 5 orders), since the ionization enhancement from the excited state is extremely prominent. In this case, the harmonics above 95th merge into supercontinuum, and
the modulation on the supercontinuum comes from the interference of short and long quantum paths with comparable efficiencies. A deeper insight is obtained by investigating the emission times of the harmonics in terms of the time-frequency analysis method, which is shown in Fig. 2(b). There are three bright peaks during the laser pulse with the maximum harmonic orders of 115th, 95th and 70th. The first peak agrees well with \( P_1 \) in Fig. 1(b), and the other two peaks come from the second and third return of the electrons from \( P_1 \) [not shown in Fig. 1(b)]. Since the ionization rate of \( P_1 \) is extremely high, the harmonic efficiencies from the second and third returns are still remarkable.

![Diagram](attachment:image.png)

**Fig. 2.** (a) The harmonic spectrum and (b) its time-frequency distribution from the pre-excited system with the few-cycle driving pulse.

The pulse duration of the driving field is 4 fs, which contains only 1.5 optical cycles, then the carrier-envelope phase (CEP) plays an important role in isolated attosecond pulse generation. We further investigate the xuv supercontinuum generation with different CEPs in single-atom level. Figure 3(a) shows the continuous parts of the harmonic spectra as a function of CEP. It is quite surprising that the efficiency and bandwidth of the generated xuv supercontinuum sensitively depend on the CEP of driving pulse. This phenomenon is quite different from that with the ground state, of which the CEP effect only influences the modulation of the supercontinuum and slightly changes its bandwidth and the harmonic cutoff [29]. This sensitive dependence on CEP can be associated with the variation of the population of the excited state, which is
presented in Fig. 3(b). As shown in this figure, the steep depletion time of the population shifts from $0.9T_0$ to $0.65T_0$ as the CEP changes, then the ionization times of $P_1$ would miss this steep depletion of the population in the cases of some CEP values. This sensitive dependence may be of potential to measure the CEP of few-cycle pulses.

![Fig. 3. (a)The harmonic spectrum as a function of CEP and (b) the variation of the population of the excited state as a function of CEP.](image)

The xuv supercontinuum generation is also associated with the initial population of the excited state. We first investigate the efficiency of the continuous part with different initial populations of the excited state $p$ in single-atom level. Figure 4 presents the emitted efficiency (shown by linear scale) of the supercontinuum as a function of $p$. Here the CEP is set to zero and the emitted efficiency of the supercontinuum is obtained by summing the harmonic intensities from 95th to 115th in the supercontinuum, of which the bandwidth is 31 eV. The result shows that the emitted efficiency increases almost linearly as $p$ increases from 0 to 0.25, and gradually become saturated as $p$ increases to 0.5. It should be mentioned that the variation of $p$ does not change cutoff and the spectral profile of the harmonics but only the efficiency.

As mentioned above, the excited state is responsible for the ionization during the interplay between the laser-matter interaction. In this scheme, the population of the exited state is totally depleted after the laser pulse while that of the ground state is hardly ionized, then the population of the excited state directly determines the ionization probability of the system and the density of the free electrons in the medium. The presence of such high-density free electrons in the nonlinear medium strongly changes the spatiotemporal propagation dynamics of the laser field including the spatial defocusing of the beam and temporal distortion of the electric field, and
Fig. 4. The emitted efficiency of the supercontinuum as a function of the initial population of the excited state.

Fig. 5. The continuous parts of the macroscopic on-axis harmonic spectra in the pre-excited medium with different initial population of the excited state of $p = 0.05$ (red line), $p = 0.1$ (blue line), $p = 0.3$ (black line) and $p = 0.5$ (violet line).

d thereby the phase matching of HHG and the isolated attosecond pulse generation, which is a extremely complicated issue. In the following, we focus on the propagation effect of the xuv supercontinuum generation in such pre-excited medium with moderate length and gas density, and perform the nonadiabatic three-dimensional (3D) propagation simulations [28] for both fundamental and harmonic fields in the gas target. Here we consider focused laser beam with a beam waist of 45 $\mu$m and a 0.5-mm long gas jet with a density of $2.6 \times 10^{18}/cm^3$. The gas jet is placed 2 mm after the laser focus. In this configuration, the defocusing of the laser beam through the gas medium is inappreciable. Figure 5 presents the continuous parts of the macroscopic on-axis harmonic spectra in the pre-excited medium with different initial population of the excited state of $p = 0.05$ (red line), $p = 0.1$ (blue line), $p = 0.3$ (black line) and $p = 0.5$ (violet line). It is found that the efficiency of the xuv supercontinua in the cases of $p = 0.05$ and $p = 0.1$ is significantly enhanced after propagation, and the interference fringes through the plateau are
all removed, which implies that the continuous harmonics from a single path in these two cases are well phase-matched. On the contrary, the efficiency of the xuv supercontinua in the cases of $p = 0.3$ and $p = 0.5$ declines by several orders after propagation, and is 2 orders lower than that of $p = 0.05$ and $p = 0.1$, although it is one or more orders higher in single-atom level (See Fig. 4). Moreover, the supercontinua in these two cases are still deeply modulated.

![Electric field vs time plot](image)

**Fig. 6.** The electric fields of the driving pulses after propagation with the excited-state populations of $p=0.05$, $p=0.1$, $p=0.3$ and $p=0.5$.

The low efficiencies of the macroscopic xuv supercontinua in the cases of $p = 0.05$ and $p = 0.1$ can be attributed to two factors. The first one is the high density of the free electrons originating from the large population of the excited state. These high-density free electrons weaken the phase-match of the xuv supercontinuum. The other reason is distortion of the electric field after propagation. Figure 6 shows the on-axis electric field of the driving pulse at the end of the medium for the cases of $p = 0.05$, $p = 0.1$, $p = 0.3$ and $p = 0.5$. For comparison, the electric field of the driving pulse at entrance of the medium is also presented by the dashed line. As shown in this figure, the electric-field distortions for the cases of $p = 0.05$ and $p = 0.1$ are inappreciable. For the cases of $p = 0.3$ and $p = 0.5$, the electric distortions, which can be regarded as CEP shifts, are noticeable. As discussed above, the xuv supercontinuum strongly depends on the CEP of the driving pulse [See Fig. 3 (a)], and the CEP shifts as the laser pulse propagates, which also weaken the efficiencies of the macroscopic xuv supercontinua.

In the following, we investigate the temporal characteristics of the macroscopic attosecond pulse generation. The normalized temporal profile of the on-axis attosecond pulses in the case of $p = 0.05$, $p = 0.1$, $p = 0.3$ and $p = 0.5$ are shown in Fig. 7 (a)-(d), respectively. Here these pulses are obtained by applying a square window on the xuv supercontinuum from 95th to 115th. The bandwidth of the selected supercontinuum is 31 eV, corresponding to the Fourier-transform limit of 130 as. In order to clearly demonstrate the phase-matching features, the single-atom response is also presented for comparison (dashed line). For the cases of $p = 0.05$ and $p = 0.1$, it is clear that one pure attosecond pulse is filtered out after propagation from the double-peak structure, which indicates that the harmonics from one single quantum path (short or long) are well phase-matched while that from the other are suppressed. For the cases of $p = 0.3$ and $p = 0.5$, there are several peaks with irregular temporal structure after propagation. This phenomenon is not only due to the poor phase matching of the harmonics from both long and short quantum paths caused by the high-density free electrons, but also the significant
Fig. 7. Normalized temporal profiles of the attosecond pulses with the excited-state populations of (a) $p=0.05$, (b) $p=0.1$, (c) $p=0.3$ and (d) $p=0.5$. The dashed line is the case at single-atom level.

Fig. 8. Temporal profiles of emitted powers of the attosecond pulses with the excited-state populations of $p=0.05$, $p=0.1$, $p=0.3$ and $p=0.5$. The inset is the enlargement for the cases of $p=0.3$ and $p=0.5$.

distortion of the fundamental driving field (See Fig. 6).

We further calculate the emitted power of the attosecond pulses through the relationship

$$ P_r = \int_0^R \frac{1}{2} |E_0 c E_a|^2 2\pi r dr, \quad (7) $$

where $E_a$ is the electric field of the attosecond pulses at the end of the medium. The temporal profiles of the emitted power of the attosecond pulses in the case of $p=0.05$, $p=0.1$, $p=0.3$ and $p=0.5$ are shown in Fig. 8. A pure isolated attosecond pulse with the pulse during of
approximately 150 as and with the emitted power of \(1.4 \times 10^6\) W is directly obtained in the case \(p = 0.05\). The energy of this pulse is estimated to be approximately 0.5 nJ. For \(p = 0.1\), the emitted power of the isolated attosecond pulse is lower than that of \(p = 0.05\). The emitted powers of the attosecond pulses in the cases of \(p = 0.3\) and \(p = 0.5\) are two orders lower, and the enlargements of their temporal profiles are shown in the inset.

The characteristics of the macroscopic attosecond pulses also include the spatial properties. We further investigate the spatiotemporal profiles of the attosecond pulse generation in the cases of \(p = 0.05\), \(p = 0.1\), \(p = 0.3\) and \(p = 0.5\), which are shown in Fig. 9. For the case of \(p = 0.05\), the spot size at the end of the medium (or the divergence) is larger than that of others cases. The spatiotemporal distribution of the attosecond source is governed by the density of the free electrons in propagation. Therefore, for the case of \(p = 0.05\) of which the density of the free electrons is relatively low, the phase matching is easier to achieve in large radius region (or the large divergence). As the population of the excited state increases, high density of the free electrons reduces the radius region where the harmonics for the attosecond pulses are phase-matched, and also make the spatiotemporal distribution much more complicated, as shown in Fig. 9(c) and (d). The large phase-matching radius region in the case of \(p = 0.05\) leads to some slight spatial chirps, which are related to the geometry of the driving pulse.
4. Conclusion

In conclusion, we theoretically investigate the isolated attosecond pulse generation with an intense few-cycle driving pulse in the pre-excited medium. It is found that the temporal and spatial characteristics of the isolated attosecond pulse generation are governed by the initial population of the excited state, which is responsible of the ionization of the system and thereby the density of the free electrons in the medium. Large initial population of the excited states leads to high density of the free electrons. This high-ionized medium not only weakens the phase matching of the xuv supercontinuum but also shifts the CEP of the driving pulse, on which the generation of the xuv supercontinuum at the single-atom level sensitively depends, resulting in the lower emitted power and poor temporal and spatial properties of the attosecond pulse. Instead, small initial population of the excited state (5% in our calculation) in the pre-excited medium leads to the generation of well phase-matched xuv supercontinuum and a pure intense isolated attosecond pulse with the pulse duration of approximately 150 as and the pulse energy up to 0.5 nJ is directly obtained.

Finally, we would like to discuss the generality of this scheme. First, the laser intensity chosen in our scheme should be moderate to ensure that the excited state is rapidly depleted to enhance the ionization and also gate the harmonics, while the ground state is hardly ionized to provide the saturation of HHG. When the laser intensity is too low, for instance, $1 \times 10^{14}$ W/cm$^2$, the excited state cannot be rapidly depleted but only gradually ionized, then the broadband supercontinuum cannot be obtained. In this case the cutoff is only 47th and the bandwidth of the supercontinuum near the cutoff is inappreciable. On the other hand, when the laser intensity is sufficiently high to significantly ionize the ground state of He$^+$, for instance, $5 \times 10^{15}$ W/cm$^2$, the unexcited He$^+$ would be evidently preferable to produce broadband supercontinuum near the cutoff. Our calculation shows the this scheme works well when the laser intensity within $5 \times 10^{14}$ W/cm$^2$ and $2 \times 10^{15}$ W/cm$^2$. Second, the range of the excitation ratio $p$ is estimated to be within 4% – 6% for efficient single attosecond pulse generation by balancing the microscopic enhancement and the phase matching of the harmonics.

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