Isolated attosecond pulse generation with the stability against the carrier-envelope phase shift and with the high-beam quality from CO gas medium

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Abstract: We theoretically investigate the isolated attosecond pulse generation with a 1300-nm infrared few-cycle laser pulse from the CO medium. It is found that the supercontinuum in the plateau from CO in the microscopic level can be generated for nearly all the carrier-envelope phase (CEP) of the driving pulse. The macroscopic investigation shows that when the molecular axis is parallel to the electric field, the supercontinuum can be phase-matched in a broader spectral range than the antiparallel case and achieve a good beam quality with the divergence angle of 0.2 mrad, which benefits for some potential applications of ultrafast detections with high spatial and temporal resolutions.

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References and links

1. Introduction

The appearance and development of the attosecond pulses pave the way for the study and control of the ultrafast microscopic dynamics with unprecedented resolutions [1, 2]. Nowadays, attosecond pulses based on the high-order harmonic generation (HHG) have been successfully produced in experiment [3]. It has been demonstrated that the harmonic signal bursts at the periodicity of half an optical cycle of the driving laser, resulting in an attosecond pulse train (APT) [4]. The isolated attosecond pulse can be obtained only if the harmonic process is confined within half a cycle.

Microscopically, the HHG process can be well depicted by the well-known three-step model [5]: ionization, acceleration and recombination in the external laser field. This classical picture not only well explains the time-frequency properties of the high-order harmonics, but also sug-
gests that the harmonic process can be controlled in both time and frequency domains to extract an isolated harmonic burst and further shorten the pulse duration. The few-cycle-pulse driving scheme [6] and the polarization gating technique [7] have been introduced in the earlier stage to generate an isolated attosecond pulse. However, the bandwidths of the generated xuv supercontinuum are limited to 20 eV. Recently, it has been proposed that wave-form-controlled two-color fields can significantly modulate the acceleration step of the harmonic processes and the form a broadband supercontinuum with bandwidth of several tens eV near the cutoff [8, 9, 10]. However, the yields of the supercontinuum are much lower than that of the harmonics in the plateau, due to the intrinsic properties of the corresponding electron dynamics of the harmonic process [11]. An isolated attosecond pulse can also be generated by restricting the high harmonic generation within half an optical cycle of the driving laser, and the harmonics in the plateau merge to a supercontinuum [9, 12, 13]. The restriction of the ionization, so-call “ionization gating”, can be realized by inducing the asymmetry of the ionization process in the adjacent half cycle. Such a gating is directly formed by synthesizing a second harmonic field [9, 12] or a very short uv pulse [13] to the fundamental field. It has also been found that the supercontinuum with the ionization gating maintain the efficiency after propagation [9].

Another alternative way to form the ionization gating is to adopt a target with asymmetric structure [14]. Asymmetric molecules, due to their intrinsic dipole, have been widely investigated in laser-matter interaction, including ionization [15] and non-sequential double ionization [16], high harmonic generation [14] and molecular dissociation [17], and so on. The laser-dress potentials of such molecules (for proper laser intensities) are reverse asymmetric, and the electron preferentially located in the deeper well forming an intrinsic dipole, which leads to the strikingly difference of the ionization processes in consecutive half cycles of the driving laser. When the electric field of the laser is parallel to the intrinsic dipole of the molecule, the electron energy decreases by the downshifting action of the laser field, which suppresses the ionization probability and further lower the harmonic yields, and vice versa. Therefore, the intensity of the attosecond pulse in the half cycle where the laser field is antiparallel to the intrinsic dipole of the molecule is much higher than that in adjacent cycle, resulting in an attosecond pulse train with the periodicity of a full optical cycle. Due to the inverse asymmetry of the HHG process, the asymmetric molecules have been introduced to loose the requirement of the driving-pulse duration for isolated attosecond pulse generation and to further shorten the pulse duration [14] via controlling the acceleration [18] or recombination [19] steps by the external fields. The previous works almost focused on the asymmetric molecules such like HeH\(^+\), of which the asymmetric structure can afford very intense driving pulse and emit the harmonics with very high photon-energies for broadband supercontinuum generation. However, the orientation of such molecule still remains challenging. The real asymmetric molecules, such as CO, can be oriented by the current laser techniques [20], which is much more practical in the HHG experiment. However, the attosecond pulse generation from the real asymmetric molecules has seldom been investigated, to the best of our knowledge.

In this paper, we theoretically study the isolated attosecond pulse generation from the CO gas with a 1300-nm infrared few-cycle driving pulse. Here the infrared driving laser with longer wavelength is adopted to extend the harmonic cutoff and avoid breaking the inverse asymmetry of the CO molecule in the laser field. The ionization properties for the HHG in the single-atom response are presented. It is found that the generation of the isolated attosecond pulse from CO is robust against the carrier-envelop phase(CEP) of the driving pulse. The simulation of the 3-dimensional propagation for the supercontinuum generated from CO has been also carried out to investigate the spectral, temporal and spacial characteristics of the generated attosecond pulse.
2. Theoretical model

We employ the Lewenstein model [21] to calculate the harmonic radiation. The time-dependent dipole moment is given by

\[ d_{nl}(t) = i \int_{-\infty}^{t} dt' \left[ \frac{\pi}{e + i(t - t')/2} \right]^{3/2} \times d_{rec} \left[ p_{nl}(t') - A(t') \right] d_{ion} \left[ p_{nl}(t') - A(t') \right] \times \exp \left[ -iS_{nl}(t', t) \right] E(t')g(t') + c.c. \] (1)

In this equation, \( E(t) \) is the electric field of the laser pulse, \( A(t) \) is the corresponding vector potential, \( \epsilon \) is a positive regularization constant. \( p_{nl} \) and \( S_{nl} \) are the stationary momentum and quasiclassical action, which are given by

\[ p_{nl}(t', t) = \frac{1}{t - t'} \int_{t'}^{t} A(t'')dt'', \] (2)

\[ S_{nl}(t', t) = (t - t')I_p - \frac{1}{2} p_{nl}^2(t', t)(t - t') + \frac{1}{2} \int_{t'}^{t} A^2(t'')dt'', \] (3)

where \( I_p \) is the ionization energy. \( g(t') \) in the equation(1) represents the ground state amplitude:

\[ g(t') = E_f(t') \exp \left[ -\int_{-\infty}^{t'} w(t')dt' \right]. \] (4)

\( w(t') \) is the ionization rate, which is calculated by the molecular Ammosov-Delone-Krainov (MO-ADK) tunnelling model [22]:

\[ w(E; R) = \sum_{m'} \frac{B^2(m')}{2m''|m''|!} \frac{1}{\kappa^{2Z_c/\kappa-1}} \left( \frac{2\kappa^3}{E} \right)^{2Z_c/\kappa - |m'| - 1} e^{-2\kappa^3/3E}, \] (5)

where

\[ B(m') = \sum_l C_l D_{m', m_l}(R)Q(l, m'), \] (6)

\[ Q(l, m) = (-1)^m \sqrt{\frac{(2l + 1)(l + |m|)!}{2(l - |m|)!}} \] (7)

with \( Z_c \) being the effective Coulomb charge, \( \kappa = \sqrt{2\mu_p} \), \( R \) being the Euler angles between the molecular axis and the field direction, and \( D_{m', m_l}(R) \) the rotation matrix. The coefficients \( C_l \) for CO are calculated by the multiple-scattering method [22, 23].

The transition dipole moment between the ground and the continuum state is calculated by

\[ \tilde{d}_{CO}(\beta) = \langle \Psi_{CO}(\beta) | \hat{r} | e^{-i\beta F} \rangle, \] (8)

where \( \Psi_{CO} \) is the HOMO of CO obtained with the Gaussian 03 \textit{ab initio} code [24].

In this model, the linear Stark shift of HOMO is considered, which will lead to the time-dependent ionization energy [25, 26]

\[ I_p(t) = I_{p0} + \mu_h \cdot E(t), \] (9)

where \( I_{p0} \) is the field free ionization energy, \( \mu_h \) is the permanent dipole of HOMO of CO and \( E(t) \) is the external field. The permanent dipole of HOMO is calculated to be 1.72 a.u. (4.37 D) by [25]

\[ \mu_h = - \int drr\rho^H(r), \] (10)
where $\rho^H(r)$ is the electron density of the HOMO calculated by $\rho^H(r) = \int dr \Psi^*(r)\Psi(r)$.

The harmonic spectrum is then obtained by Fourier transforming the time-dependent dipole acceleration $\vec{a}(t)$:

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

where $\vec{a}(t) = d_{nl}(t)$, $T$ 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 fields, which can be written separately by [27]

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

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

where $E_f$ and $E_h$ are laser and harmonic fields, $\omega_p = e/\sqrt{4\pi\varepsilon_0 n_e(\rho, z, t)/m_e}$ 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 $n_e(t) = n_0 [1 - \exp (-\int_0^t w(t') dt')]$ 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 [7].

3. Result and discussion

We first consider the microscopic ionization dynamics for the HHG from the CO molecule with a 1300-nm 13-fs mid-infrared laser pulse. The laser intensity is chosen to be $1.5 \times 10^{14} W/cm^2$ to keep the molecule far from being highly ionized and avoid breaking the inverse asymmetry of the ionization process in the adjacent two half-cycles. The electric field of the driving pulse is expressed as

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

where $E_0$, $T_0$ and $\phi$ are the amplitude, optical cycle and carrier-envelope phase(CEP) of the driving field, respectively. First the CEP is set to zero. Figures 1(a)-(c) are the ionization rates of the CO molecules when the molecular axis is parallel, perpendicular and antiparallel to the electric field, respectively. For the parallel case, there are two main equivalent ionization peaks during the pulse. The electrons ionized in the peak would be accelerated in the next half-cycle with the strongest electric field and obtain the highest kinetic energy while the kinetic energy of the electrons for the other peak is much lower, leading to the continuous high-harmonic generation with a broad bandwidth. For the perpendicular case, the ionization property is nearly the same as the cases for the atoms or the symmetric molecules, then the supercontinuum can only be generated near the cutoff. For the antiparallel case, there is only one main ionization peak corresponding for the HHG, which forms a “standard” ionization gate and then the harmonics in the plateau would merge to a broadband supercontinuum. However, the electrons in this case are accelerated in the half-cycle with the second highest electric field, then the harmonic cutoff is lower than that of the parallel case.
In order to confirm the above predictions for the supercontinuum generation, we perform the quantum simulation by the MO-SFA to calculate the harmonic spectra from the CO molecule in the microscopic level. Figure 2(a) shows the high-energy part of the harmonic spectra for the parallel, antiparallel and perpendicular cases, depicted by the red, blue and black lines. One can clearly see that the harmonics from 55th to 100th become continuous for the parallel case, of which the bandwidth of the supercontinuum is 42 eV. The fringes in the supercontinuum is originated from the interference between the long and short quantum paths with nearly equivalent intensities. For the antiparallel case, the supercontinuum can also be observed in the plateau, but with lower cutoff energy and higher efficiency, due to the corresponding higher ionization rate and lower recollision energy of the electrons, as shown in Fig. 1(a). The harmonic spectrum for the perpendicular case is given for comparison. The harmonic yield in this case is 1 2 order lower than those in the parallel and antiparallel cases, and the supercontinuum can only be observed at the cutoff and the bandwidth is only 15 eV. A deeper insight is obtained by investigate the emission times of the harmonic spectra for the parallel and antiparallel cases in terms of the time-frequency analysis method, which is shown in Fig. 2(b) and (c), respectively. There are three main peaks corresponding to the harmonic generation during the pulse for both cases, marked by $P_1$, $P_2$ and $P_3$. When the molecular axis is parallel to the laser electric field, the harmonic yield for $P_2$ is much higher than those for $P_1$ and $P_3$ due to the ionization properties shown in Fig. 1(a). And for the antiparallel case, the harmonic yield for $P_3$ is dominant during the pulse since the electric field and the molecule gate the ionization process of the electrons.
Fig. 2. (a) The high-energy parts of the harmonic spectra for the parallel, antiparallel and perpendicular cases. (b) The time-frequency distribution for the parallel case. (c) The time-frequency distribution for the antiparallel case.

Corresponding for $P_3$.

The driving pulse duration in our simulation is 13 fs, which contains only 2 optical cycles, then the carrier-envelope phase (CEP) plays an important role in the isolated attosecond pulse generation. We further investigate the ionization dynamics and the corresponding high-harmonic generation with different CEPs in the microscopic level. Figure 3(a) and (b) shows the ionization rate and the continuous parts of the harmonic spectra as functions of the CEP, respectively. For comparison, those for the perpendicular case are also presented in Fig. 3(c) and (d). One can clearly see a significant difference in the ionization rates as functions of CEP between the parallel and perpendicular cases. As shown in Fig. 1(a), the ionization of the electrons corresponding to the high-harmonic generation is gated within a very short time for a wide range of CEP shift for the parallel case, leading to the supercontinuum generation in the plateau. For the CEP range in the vicinity of 0$\pi$, the two-peak structure of the ionization rate can also lead the the supercontinuum generation in the plateau, according to the above analysis. Then we can conclude that the supercontinuum can be produced in the plateau by using a two-cycle driving pulse with arbitrary CEPs. This can be confirmed by the result shown in Fig. 3(b), in which the harmonics in the plateau merge to supercontinua for nearly all the CEPs but different cutoff energies. For the perpendicular case, there are more than two ionization peaks during the pulse for all the CEPs, then the continuous harmonics can only be observed near the cutoff.
We further investigate the ionization properties of the CO molecule with different alignment angles. Figure 4 shows the ionization rate as a function of the alignment angle. Here the carrier-envelope phase is set to zero for convenience. As mentioned above, when the molecular axis of CO, i.e., the intrinsic dipole, is antiparallel to the electric field of the driving pulse, the ionization would be enhanced, and vice versa. This enhancement or suppression of the ionization by the intrinsic dipole is weakened as the alignment angle of the molecule tends from 0° or 180° to 90°, respectively. Therefore the inverse asymmetry of the high-harmonic generation in the adjacent two half-cycles is also diminished. Moreover, the ionization amplitude for the perpendicular case is much lower than those for other alignment angles, leading to the much lower harmonic.
Fig. 5. The high-energy parts of the harmonic spectra in microscopic level for the alignment angles of (a) 0°, 30°, 60°, and 120° and (b) 120°, 150°, 180°, respectively.

The harmonic spectra of the CO molecule for different alignment angles calculated by the MO-SFA are presented in Fig. 5. Figure 5(a) shows the continuous parts of the harmonic spectra for the angles of 0°, 30°, and 60°, respectively. One can clearly see that the harmonic yields of the generated supercontinuum decrease as the alignment angle of the molecule increases. Moreover, the supercontinuum becomes slightly irregular for the case of 60°. The supercontinua for the cases of 120°, 150°, and 180° are also presented in Fig. 5(b). On the contrary, the harmonic yields of the generated supercontinuum increase as the alignment angle of the molecule increases, and some small irregular modulations appear on the supercontinuum for the case of 120°. This can be contributed to the ionization properties shown in Fig. 4. We can conclude that the the harmonic yields of the generated supercontinuum decrease and the supercontinuum becomes irregular as the CO molecule rotates from parallel orientation to perpendicular orientation.

In our calculation, the 1300-nm mid-infrared laser pulse is adopted. Such mid-infrared pulse can produce harmonics in nearly nonionized medium. Here the calculated ionization probability is below 0.1%, then such neutral medium enables one to adopt the standard phase-matching technique to macroscopically enhance the harmonic yields of an individual quantum path in the supercontinuum to obtain a pure isolated attosecond pulse. In this technique, the phase-matching condition for either quantum path (short or long) can be fully satisfied by adjusting the gas pressure and the position of the laser focus. To demonstrate this issue, we perform the nonadiabatic three-dimensional(3D) propagation simulations for both fundamental and harmonic fields in the gas medium. In order to realize the phase-matching condition of an individual quantum path of the supercontinuum, we consider a focused laser beam with the beam waist of 50 μm and 0.5-mm long gas jet with the gas density of $2.6 \times 10^{18}/cm^3$. The gas jet is placed...
1 mm after the laser focus. Figure 6(a) and (b) show the spectra of the macroscopic harmonics for the alignment angles of 0°, 30°, 60°, and 120°, 150°, 180°, respectively. For comparison, the macroscopic spectrum for the perpendicular case is also presented in both Fig. 6(a) and (b) by the dashed lines. One can clearly see that the interference fringes through the plateau to the cutoff are mostly removed for all the angles. For the alignment angles of 0°, 30°, 60°, the supercontinua are well phase-matched from about 70th to the cutoff, while the phase-matched harmonics are only covered by about 20 harmonic orders for the angles of 120°, 150°, 180°. Moreover, there are some small irregular modulations in the supercontinuum for such angles.

Fig. 6. The high-energy parts of the harmonic spectra after propagation for the alignment angles of (a) 0°, 30°, 60°, and 120° and (b) 120°, 150°, 180°, respectively. The dashed line in (a) and (b) is the case for the perpendicular cases.

Next, we investigate the spatiotemporal characteristics of the attosecond pulses from the CO gas medium. Here we only consider the parallel and antiparallel cases, since other cases for the angles smaller and larger than 90° are nearly the same with parallel and antiparallel cases, respectively. The temporal profile of the emitted power of the generated attosecond pulses can be calculated through the relationship

\[ P_τ = \int_0^R \left( \frac{1}{2} E_0 c E_a^2 \right) 2\pi r dr, \]

(15)

where \( E_a \) is the electric field of the attosecond pulses at the end of the medium. The temporal profile and the corresponding spatiotemporal distribution for the parallel and antiparallel cases are shown in Fig. 7(a) and (b), respectively. Here the attosecond pulses are obtained by applying square windows on the xuv supercontinua from 75th to 95th and 65th to 85th for the parallel and antiparallel cases, respectively. As shown in this figure, a pure isolated attosecond pulse with
the emitted power of $3.3 \times 10^5$ W is directly obtained. We can judge from the emitted time of the isolated attosecond pulses that the short path of the supercontinuum is well phase-matched after propagation. The spatiotemporal distribution for the parallel case shows a crescent-like structure, which can be related to the gaussian geometry of the driving pulse. For the antiparallel case, the resolution of the crescent-like distribution is slightly reduced and the radius region for the phase-matched harmonics of the attosecond pulses is also lessened. This may be related to the difference between the ionization amplitudes corresponding to the isolated attosecond pulse generation for the parallel and antiparallel cases, which has been shown in Fig. 4. Moreover, there is a satellite pulse with 5% intensity of the main pulse for the antiparallel case.

![Fig. 7.](image)

Fig. 7. (a) and (b) are the temporal profiles of the attosecond pulses for parallel and antiparallel cases, respectively. (c) and (d) are the corresponding spatiotemporal profiles.

Finally, we go further to the beam qualities of the isolated attosecond pulses generated from the CO medium. We calculate the far-field distributions of the generated isolated attosecond pulses by the Hankel transformation from the near-field harmonic emissions at the exit face of the medium. The calculation details can be found in Ref. [27]. The divergences of the isolated attosecond pulses for the parallel and antiparallel cases are presented in Fig. 8(a) and (b), respective. The insets show the spots with the distance of 1 m from the gas medium for these two case. It is clearly shown that the divergences for the parallel and antiparallel are both gaussian-like. The divergence angles for this two cases are 0.2 and 0.3 mrad, which indicates the better beam quality for the parallel case.

4. Conclusion

In conclusion, we have theoretically investigate the isolated attosecond pulse generation with a 1300-nm 13-fs infrared few-cycle laser pulse from the CO medium. It is found that the supercontinuum in the plateau from CO in the microscopic level can be generated for nearly all

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the carrier-envelop phase (CEP) of the driving pulse. The macroscopic investigation shows that
the well phase-matched spectral region of the generated supercontinuum for the 0° of alignment
angle is broader than that for the 180°. Further investigation implies that the spatiotemporal dis-
tribution structure of the isolated attosecond pulse for the parallel case is better than that for the
antiparallel case. Moreover, the divergence of the isolated attosecond pulse for the parallel case
is also smaller than that for the antiparallel case, indicating a better beam quality. Finally, we
would like to point out that the intensity of the driving pulse should be carefully chosen to avoid
breaking the inverse asymmetry and also producing high-density free electrons which distorts
the electric field of the few-cycle pulses and limits the harmonic yields macroscopically. For the
CO molecule, the intensity of the driving pulse should be less than $2 \times 10^{14} \text{W/cm}^2$ according
to our calculation.

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