Generation of isolated sub-40-as pulses from gas-phase CO molecules using an intense few-cycle chirped laser and a unipolar pulse

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We theoretically investigate the high-order-harmonic generation (HHG) from the oriented gas-phase CO molecule exposed to the combination of an intense few-cycle chirped laser and a unipolar pulse. We show that the HHG is dramatically extended and an ultrabroad extreme ultraviolet supercontinuum spectra in the plateau is generated by adding a unipolar pulse to the few-cycle chirped laser at a proper time. Both the classical trajectory simulation and quantum time-frequency analysis show that a dominant short quantum path is selected to contribute the HHG spectra in the plateau. By superposing the supercontinuum harmonics centered at several different central frequencies, we can directly produce several isolated pulses in which the shortest one achieves 38 as.

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High-order-harmonic generation (HHG) has been intensively studied for more than two decades because of its potential applications for generating coherent extreme ultraviolet (XUV) light sources [1,2] and producing attosecond (as) pulses [3–6]. The isolated attosecond pulse (IAP) provides a powerful tool for the real-time observation of ultrafast electron dynamics [7–9] and for probing the motion of an electron wave packet [10] and nuclear dynamics [11].

The HHG procedure has been well understood by the semiclassical three-step model [12,13]. First, the electron tunnels through the potential barrier formed by the Coulomb force and the laser field, then it oscillates quasifreely driven by the laser field and acquires additional kinetic energy. Finally, it can recombine with the parent ion and emit high-energy photons. The HHG can be efficiently controlled by modulating different steps of the harmonic processes to generate a broadband supercontinuum based on the three-step model. So far, three main techniques have been introduced to experimentally generate an IAP, namely, few-cycle-laser driving [14–16], polarization gating [17], and double optical gating (DOG) [18]. Very recently, Zhao *et al.* [19] successfully created the shortest 67-as pulse with the DOG technique. On the theoretical side, many different schemes have been proposed to produce the IAP, such as the few-cycle pulse [20], polarization gating [21–23], two-color field [24–35], and so on.

Attosecond pulses have been experimentally created by using the HHG from atomic targets. Thanks to the recent developments in field-free molecular alignment and orientation techniques [36], alignment- (or orientation-) dependent tunneling ionization and the HHG attracted a great deal of interest both experimentally and theoretically [37]. Furthermore, the HHG plateau can be dramatically extended with molecules at large internuclear distances [38] or stretched molecules [39,40]. Recently, the HHG from the simplest diatomic molecule H_2^+ fixed in space has been used to produce attosecond pulses [41,42]. Since the inversion symmetry of the aligned symmetric molecule, the HHG contributions from one half-cycle and the adjacent half-cycle are comparable such that the strong interference of the HHG does exist. This interference of the HHG is not beneficial to produce a broader supercontinuum. However, for an oriented polar molecule, the target molecule itself can serve as a gate (so-called system-induced gating) due to an increased (decreased) ionization rate [43]. Therefore, the oriented asymmetric molecule provides a promising target to generate a broadband supercontinuum spectra for producing ultrashort attosecond pulses [44–46]. Very recently, an isolated elliptically polarized 297-as pulse was produced using the HHG after propagation from the oriented polar molecule CO driven by a few-cycle-laser pulse [47].

In this Brief Report, we propose an efficient method to generate an isolated 38-as pulse with the HHG from the oriented molecule CO exposed to a combined field of a few-cycle femtosecond (fs) chirped laser and a unipolar pulse in the microscopic level. Atomic units are used throughout this paper unless otherwise stated.

Strong-field approximation theory (the so-called Lewenstein model) has been widely used to investigate the HHG of atoms [48] and of molecules fixed in space [49–51]. We calculate the HHG from the oriented molecule CO using the extended Lewenstein model [43], in which the linear Stark shift of the highest occupied molecular orbital (HOMO) is taken into account. The time-dependent induced dipole moment can be expressed as

$$\begin{aligned} x(t) = i\int_0^\infty d\tau \left(\frac{\pi}{\epsilon + i\tau/2}\right)^{3/2} & [d_y^*(t)\sin\theta + d_z^*(t)\cos\theta] \\ & \times [d_y(t-\tau)\sin\theta + d_z(t-\tau)\cos\theta]E(t-\tau) \\ & \times e^{-iS_{\rm st}(t,\tau)} g^*(t)g(t-\tau) + \text{c.c.}, \end{aligned} \tag{1}$$

where $d_y(t)$ and $d_z(t)$ are the y and z components of the transition dipole moment between the ground state and the continuum state. We mention that the laser's polarization direction is defined along the positive z direction ($\theta = 0^\circ$) in the present calculations. $E(t)$ is the electric field of the laser pulse, and ϵ is a positive regularization constant. The quasiclassical action at the stationary points τ is written as

$$S_{\rm st}(t,\tau) = \int_{t-\tau}^{t} I_p(t')dt' - \frac{1}{2}p_{\rm st}^2(t,\tau)\tau + \frac{1}{2}\int_{t-\tau}^{t} A^2(t')dt', \tag{2}$$

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where $A(t)$ is the vector potential. The canonical momentum at the stationary points is given by

$$p_{\rm st}(t,\tau) = \frac{1}{\tau}\int_{t-\tau}^{t} A(t')dt'. \quad (3)$$

The time-dependent ionization potential is determined by [43]

$$I_p(t) = I_{p0} + \boldsymbol{\mu}_h \cdot \boldsymbol{E}(t), \quad (4)$$

where I_{p0} is the field-free ionization potential, and $\boldsymbol{\mu}_h$ is the permanent dipole of the HOMO of CO which points from the C end to the O end. The permanent dipole of the HOMO is calculated to be $\boldsymbol{\mu}_h = 1.72$ a.u.$(4.365D)z$ by

$$\boldsymbol{\mu}_h = -\int d\boldsymbol{r}\boldsymbol{r}\rho^H(\boldsymbol{r}), \quad (5)$$

where $\rho^H(\boldsymbol{r})$ is the electron density of the HOMO:

$$\rho^H(\boldsymbol{r}) = \Psi^*(\boldsymbol{r})\Psi(\boldsymbol{r}). \quad (6)$$

In Eq. (1), $g(t)$ is introduced to account for the depletion of the ground state, which is approximated by

$$g(t) = \exp\left(-\int_{-\infty}^{t} W(t')/2dt'\right), \quad (7)$$

where $W(t')$ is the ionization rate obtained from the molecular Ammosov-Delone-Krainov (MO-ADK) theory [52–54]. In the present calculations, the ground-state electronic wave function of CO at the equilibrium distance of 2.132 a.u. is obtained from the GAUSSIAN package [55] using the Hartree-Fock method in conjunction with the aug-cc-pVQZ basis sets. The calculated HOMO energy of CO is −0.554 36 a.u.. The continuum state is approximated by the plane wave.

The HHG spectra can be obtained by

$$P_A(\omega) = |\, a_q(\omega)\,|^2\,, \quad (8)$$

where

$$a_q(\omega) = \frac{1}{\sqrt{2\pi}}\int_{-\infty}^{\infty} a(t)\exp(-iq\omega t)dt \quad (9)$$

is the Fourier transformation of the time-dependent dipole acceleration $a(t) = \ddot{x}(t)$, ω is the central frequency of the infrared (IR) laser pulse, and q corresponds to the harmonic order. By superposing a supercontinuum HHG spectra, an IAP can be produced by

$$I(t) = \left|\sum_q a_q(\omega)\exp(iq\omega t)\right|^2. \quad (10)$$

In our calculations, the oriented molecule CO is driven by an intense chirped IR laser in combination with a unipolar pulse. The electric field of the combined pulse is taken to have the form

$$E(t) = E_0\exp[-2\ln 2(t/\tau')^2]\cos[\omega t + \phi(t)] + E_{\rm uni}(t), \quad (11)$$

where E_0 and τ' are the peak strength and the pulse duration (FWHM) of the IR laser pulse, respectively. The time-varying carrier envelope phase $\phi(t)$ has the form [31]

$$\phi(t) = \beta\left(\frac{t-t_0}{\tau_0}\right)^2. \quad (12)$$

Here, β and τ_0 are two adjustable parameters for controlling the chirp form, and t_0 is used to adjust the frequency sweeping range. The chirped parameters β, τ_0, and t_0 are taken to be $\beta = 7.1$, $t_0 = 216$, and $\tau_0 = 210$, respectively. Note that the laser chirp form in Eq. (12) has already been employed in the HHG experiment [56]. The unipolar pulse is defined as [57]

$$E_{\rm uni}(t) = k\theta(t-t_0')E_0\left[\frac{a(t-t_0')^3\exp[-8(t-t_0')/\tau'']}{\tau''^3} - \frac{b(t-t_0')^5\exp[-(t-t_0')/\tau'']}{\tau''^5}\right] \quad (13)$$

where $\theta(t)$ is the step function, the unipolar pulse starts at t_0', and τ'' is the pulse duration of the unipolar pulse. We choose $t_0' = -0.06(2\pi/\omega)$ and $\tau'' = 7$ fs in our simulations. The dimensionless parameters a, b are chosen as $a = 400$, $b = 10^{-5}a$, respectively, k is a real number that controls the peak intensity of the unipolar pulse, and kE_0 is approximately equal to the electric field strength of the unipolar pulse.

In Fig. 1(a) we show a chirp-free IR laser, a chirped IR laser, a unipolar pulse, and the combined field of the chirped laser and the unipolar pulse, respectively. We choose a 7-fs IR laser pulse with a peak intensity of 2.5×10^{14} W/cm^2

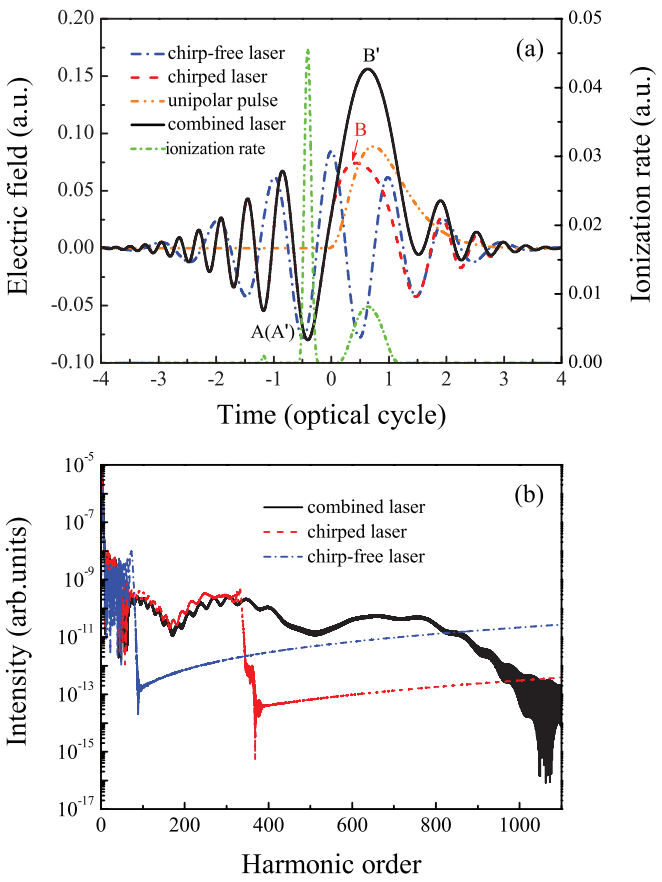

FIG. 1. (Color online) (a) a chirp-free laser (dashed blue line), a chirped laser (dashed red line), a unipolar pulse (dashed orange line), a combined field (solid black line), and ionization rate (dashed green line). (b) High-order harmonic spectra of the oriented CO in the chirp-free laser (dashed blue line), the chirped laser (dashed red line), and the combined laser fields (solid black line), respectively.

and a central wavelength of 1000 nm. Parameter k is taken to be 1.0. By comparing with the chirp-free laser, we can see that the chirped laser is obviously broadened near peak B. Thus the ionized electron near peak A can experience a longer time acceleration such that the corresponding maximum kinetic energy gained by the electron is significantly increased to 17.05 U_p, where U_p is the ponderomotive energy. By adding a unipolar pulse to the chirped laser field at the proper time, a right-shifted combined driving pulse is synthesized (the peak B' is right-shifted relative to the peak B). The electron born at peak A' experiences a much longer time acceleration, and the corresponding maximum kinetic energy reaches 54.77 U_p. The ionization rates of the oriented CO in the combined laser field are also shown in Fig. 1(a). Since the linear Stark shift of the HOMO is considered [see Eq. (4)], the resulting ionization potential is larger (smaller) than the field-free value for the positive (negative) field values. Since strong-field ionization sensitively depends on the ionization potential [58], the Stark shift of the HOMO makes ionization is enhanced (suppressed) as the electric field points along the negative (positive) z direction [see Fig. 1(a)]. Therefore, a broadband supercontinuum spectra can be obtained due to the fact that the symmetry of the HHG emission is broken. Figure 1(b) shows the HHG spectra of the oriented CO in a field-free laser, a chirped laser, and the combined laser field, respectively. From Fig. 1(b) we can observe that the HHG cutoff is dramatically extended by at least 262 (973) harmonic orders by using the chirped (combined) laser pulses compared with the chirp-free laser case. Moreover, an ultrabroad XUV supercontinuum spectrum with a 930-eV spectral width is generated from the oriented CO driven by the combined laser field.

To facilitate the investigation of the underlying mechanism responsible for the HHG cutoff extension and an isolated attosecond pulse generation in detail, we perform a classical trajectory simulation and quantum time-frequency analysis, respectively. We mention that the quantum time-frequency analysis is performed by means of the Morlet wavelet transform [see Eqs. (8) and (9) in Ref. [30]] and the classical trajectory simulation of CO is carried out by solving Newton's equation, in which we assume that the active electron ionizes from one nucleus and then recombines with the same nucleus. For the chirped laser field, there are two major emission bursts marked by A and B with the maximum kinetic energies of $3.23U_p$ and $17.05U_p$, respectively [see Fig. 2(a)]. For a given electronic kinetic energy E_k, there are two quantum paths near the cutoff, namely, the long quantum path (earlier ionization, later recombination) and the short quantum path (later ionization, earlier recombination). The long quantum path is efficiently suppressed due to the introduction of the chirp. For the combined laser pulse, there are also two dominant emission events taking place, marked by A' and B' with the maximum kinetic energies of $3.23U_p$ and $54.77U_p$, respectively [see Fig. 2(b)]. By adding a unipolar pulse to the chirped laser field at the proper time, the long path is completely suppressed and the short quantum path is easily

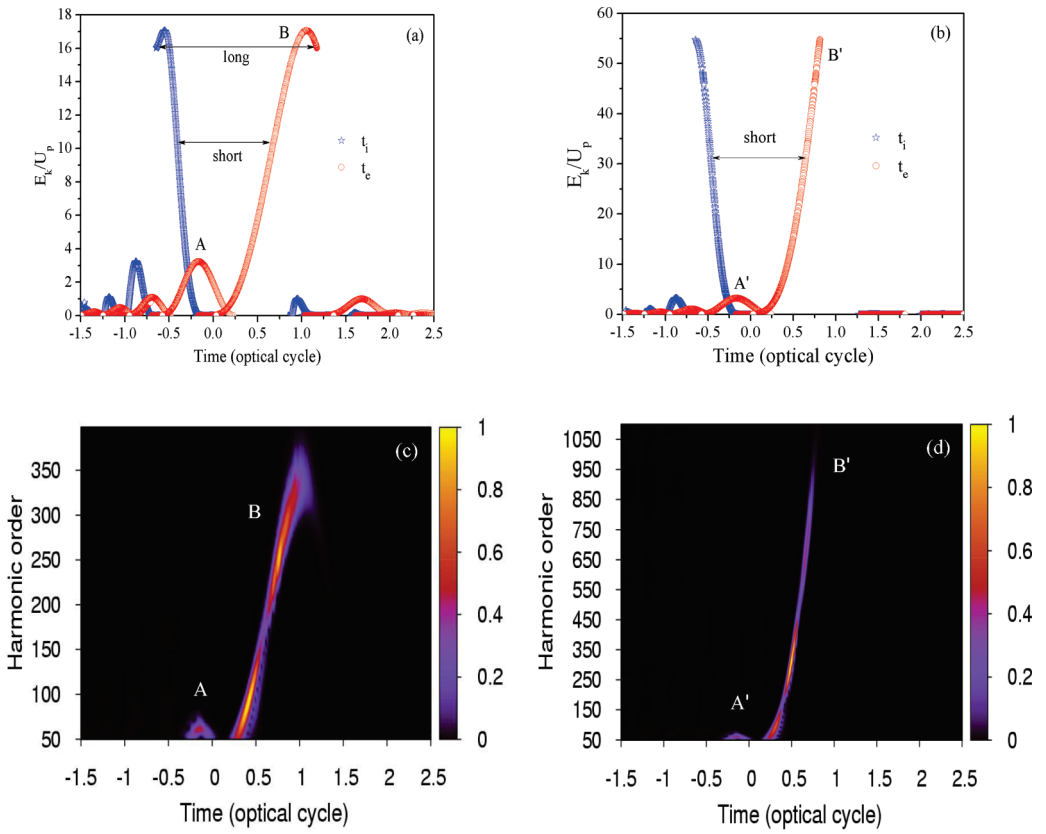

FIG. 2. (Color online) Dependence of the kinetic energy on the ionization times t_i and emission times t_e for the chirped (a) and combined (b) laser pulses. Wavelet time-frequency profiles of the HHG spectra from the oriented CO in the chirped (c) and combined (d) laser pulses. Laser parameters are the same as those in Fig. 1.

selected in the range of peak B' which is beneficial to generate the ultrabroad supercontinuum spectra [see Fig. 1(b)] and to produce the IAP. Furthermore, the slope of the kinetic energy with respect to the recombination times is remarkably increased compared with the chirped laser case [see Figs. 2(a) and 2(b)]. In Figs. 2(c) and 2(d), we show the quantum time-frequency profiles of the HHG power spectra from the oriented CO exposed to the chirped laser field and the combined laser pulse, respectively. Likewise, there are two major emission bursts marked by A and B for the chirped laser case [see Fig. 2(c)]. For the case of the combined laser field, there is one slender dominant emission burst near the peak B' as shown in Fig. 2(d). We note that each burst emission time agrees extremely well with the classical recollision time [see Figs. 2(a) and 2(b)].

Since the harmonics emit at different times (so-called harmonic chirp) [59], it is impractical to produce a ultrashort IAP by superposing an entire supercontinuum. Figure 3 shows the temporal profiles of the generated IAPs by superposing the harmonics centered at eight different central frequencies in the plateau for the combined laser case. A regular isolated pulse of 38, 39, 48, 48, 50, 56, 56 or 67 as has been obtained by selectively superposing the harmonics from 794ω to 879ω, from 712ω to 797ω, from 620ω to 685ω, from 520ω to 585ω, from 416ω to 476ω, from 320ω to 380ω, from 219ω to 279ω, or from 127ω to 187ω, respectively. By using the combined field, the pulse width of the IAP's is dramatically reduced compared with the 297-as pulse obtained from the oriented CO driven by a single few-cycle IR laser pulse [47].

Finally, we check how sensitive the IAP to the parameter k in Eq. (13). We found that the slope of the combined field with respect to time t in the raised edge near B' is gradually augmented as the k value increases. It predicts that the harmonics near the peak B' [see Fig. 1(a)] should be emitted in a more short time for the larger-k case. Figure 4 shows the temporal profiles of the selected shortest attosecond pulses by superposing the harmonics from 794ω to 879ω, from 756ω to 836ω, from 629ω to 704ω, or from 681ω to 751ω for the value of 1.0, 0.9, 0.8, or 0.7, respectively. It confirms that the pulse width of the generated IAP can be reduced by increasing the value.

In conclusion, we present an efficient scheme to generate an isolated attosecond pulse with the HHG from the oriented molecule CO driven by a combined field of a few-cycle chirped laser and a unipolar pulse. By adding a unipolar pulse to the chirped laser field at a proper time, the HHG cutoff is dramatically extended and an ultrabroad XUV supercontinuum is obtained. Moreover, a dominant short quantum path is easily selected, and the corresponding long quantum path is completely suppressed, which is confirmed by the classical trajectory simulation and the quantum time-frequency analysis. By superposing the supercontinuum harmonics centered at several different central frequencies in the plateau, we directly produce several isolated pulses in which the shortest one achieves 38 as. We mention that the present scheme can also be used for any other oriented asymmetric molecules.

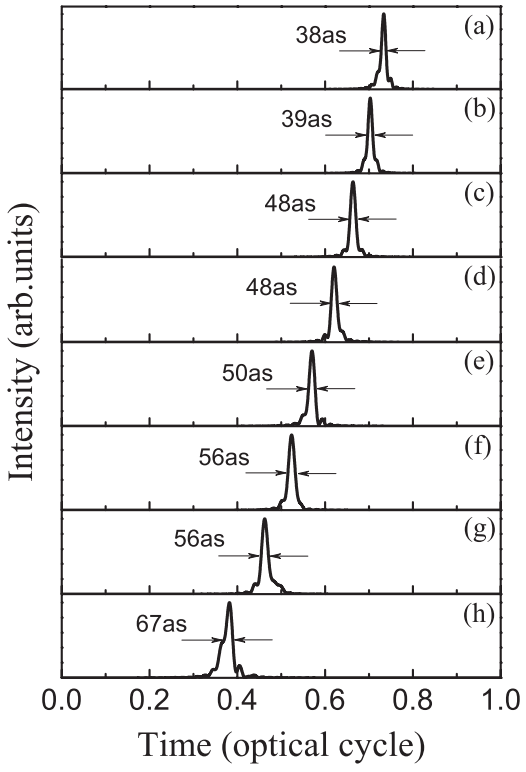

FIG. 3. The temporal profiles of the generated IAP's by superposing several harmonics: (a) from 794ω to 879ω, (b) from 712ω to 797ω, (c) from 620ω to 685ω, (d) from 520ω to 585ω, (e) from 416ω to 476ω, (f) from 320ω to 380ω, (g) from 219ω to 279ω, and (h) from 127ω to 187ω, for the combined laser field case. The corresponding HHG spectrum is the black solid line in Fig. 1(b).

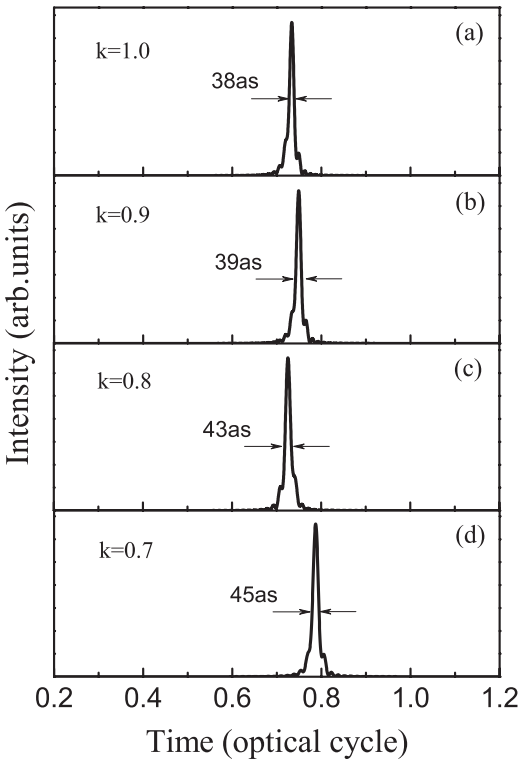

FIG. 4. The temporal profiles of the selected shortest attosecond pulses by superposing several harmonics from 794ω to 879ω, from 756ω to 836ω, from 629ω to 704ω, or from 681ω to 751ω for the value of 1.0, 0.9, 0.8, or 0.7, respectively.

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