Isolated-attosecond-pulse generation from asymmetric molecules with an $\omega + 2\omega/3$ multicycle two-color field

Qianguang Li,[1,2] Weiyi Hong,[1] Qingbin Zhang,[1] Shaoyi Wang,[1] and Peixiang Lu[1,*]

[1]*Wuhan National Laboratory for Optoelectronics, Huazhong University of Science and Technology, Wuhan 430074, People's Republic of China*

[2]*Department of Physics, Xiaogan University, Xiaogan 432000, People's Republic of China*

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The high harmonic generation from asymmetric molecules with an $\omega + 2\omega/3$ multicycle bichromatic laser pulse has been investigated. It is shown that the ionization asymmetry in consecutive half optical cycles for asymmetric molecules is further enhanced since the $2\omega/3$ control laser pulse further enhances the amplitude of the ionization peak at the center of the laser pulse. The $2\omega/3$ control laser pulse also significantly enlarges the difference of the photon energies emitted from the ejected electron in the half optical cycle at the central laser pulse and its next half optical cycle. In addition, a broadband supercontinuum is produced in the plateau of the spectrum, from which an isolated 90-as pulse can be directly obtained.

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I. INTRODUCTION

The availability of isolated attosecond (as) pulses has paved the way to what has been called attoscience [1,2]. Currently, the production of isolated attosecond pulses is mainly based on high harmonic generation (HHG) in atomic gases [3–5]. With a few-cycle laser pulse interacting with a jet of neon atoms, an isolated attosecond pulse has been firstly produced by filtering the harmonics in the cutoff of the spectrum [3]. Further decreasing the duration of the laser pulse to sub-1.5-cycle, isolated sub-100-as pulses have been generated in experiment [5]. However, such driving pulse can be achieved only with a state-of-the-art laser system.

HHG can be described by the three-step model [6]. In the first step, the electron tunnels through the barrier formed by the Coulomb potential and the laser pulse, then gains kinetic energy moving in the laser ponderomotive potential, and finally recombines with the parent ion and releases its kinetic energy by emitting high harmonics. For atoms, the laser-dressed potential is reverse symmetric when the laser field changes its sign in consecutive half optical cycles. Thus, the process of HHG is reverse symmetric in consecutive half optical cycles. Consequently, only odd harmonics are generated in the frequency domain and attosecond pulses are produced periodically every half optical cycle of the fundamental field in the time domain. Since molecules have additional degrees of freedom and more complicated structure, the physical phenomena of molecular HHG are richer and have recently attracted more and more interest [7–10]. It can be divided into two categories: one is the symmetric molecule, e.g., H_2^+, O_2, N_2, etc., and the other is the asymmetric molecule, e.g., HeH^{2+}, HCl^+, LiH^+, etc. For symmetric molecules, the harmonics is similar to atoms, i.e., only odd harmonics, since symmetric molecules have an inversion symmetry similar to that of atoms. Since symmetric molecules have more than one ionization and emission atomic center, the interferences between these centers may produce some new phenomena in the ionization and HHG of symmetric molecules. For example, the interferences may suppress the ionization [7] and generate a pronounced minimum in the harmonic spectrum [8,9], which also exhibits traces of the molecular structure. For asymmetric molecules, the interferences are negligible since the electric wave packets are mainly located around one atomic center. Since the electron prefers locating at one nucleus, the ionization process is strikingly different in consecutive half optical cycles, and the inversion symmetry in the process of HHG has been broken for asymmetric molecules [11]. Then both odd and even harmonics can be generated in the frequency domain and attosecond pulses are produced every full optical cycle of the fundamental field rather than each half optical cycle in the time domain [12–15]. This asymmetry for asymmetric molecules enables the generation of isolated attosecond pulses with multicycle laser pulse [14] or broadband isolated attosecond pulses with a few-cycle laser pulse [15].

Further enhancing the asymmetry in the process of HHG for asymmetric molecules with a wave-form-controlled driving laser pulse, it is possible to generate broadband isolated attosecond pulses from the harmonics in the plateau of the spectrum with a multicycle laser pulse. It has been proposed that the wave-form-controlled two-color field can significantly enlarge the difference between the highest and the second highest half-cycle photon energies and form a broadband supercontinuum near the cutoff, from which a broadband isolated attosecond pulse can be generated [16,17]. On the other hand, a few-cycle wave-form-controlled two-color field can also control the tunnel ionization in the HHG [18]. The two-color field can restrict the ionization peak within one half optical cycle and also enhance its amplitude, then a broadband isolated attosecond pulse can be generated from the harmonics in the plateau of the spectrum. In this article, we introduce a $2\omega/3$ control laser pulse to further control the ionization and acceleration processes in HHG for asymmetric molecules. It is shown that the ionization of the asymmetric molecules within the half optical cycle around the central electric field is largely enhanced and the photon energies emitted from the ejected electron in this half optical cycle also enhanced significantly by the $2\omega/3$ control laser pulse. Therefore, the asymmetry in the process of HHG for asymmetric molecules is further enhanced by the $2\omega/3$ control laser pulse, which enables the

*Corresponding author: lupeixiang@mail.hust.edu.cn

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generation of broadband isolated attosecond pulses from the harmonics in the harmonic plateau in multicycle regime.

II. RESULT AND DISCUSSION

The laser-molecule interaction is studied by numerically solving the time-dependent Schrödinger equation (TDSE). The two-color field used in this work is synthesized by a 10-fs linearly polarized driving pulse with a wavelength of 800 nm and a 15-fs linearly polarized control pulse with a wavelength of 1200 nm. The peak intensities of the driving and the control pulses are 3.5×10^{14} and 7.0×10^{13} W/cm^2, respectively. The electric field of the synthesized laser pulse is expressed by $E = E_0 f_0(t)\cos[\omega_0(t - T/2)] + E_1 f_1(t)\cos[\omega_1(t - T/2) + \phi]$. Here E_0, $f_0(t)$ and ω_0 are the amplitude, envelope, and frequency of the driving pulse, respectively. E_1, $f_1(t)$, and ω_1 are the amplitude, envelope, and frequency of the control pulse, respectively. The driving and control pulses are all centered at $T/2$ and are given by $f_0(t) = \cos^2[1.5\pi(t - T/2)/T)]$ with $2.5T_0 \leqslant t \leqslant 12.5T_0$($T_0$: optical cycle of the driving pulse), $f_0(t) = 0$ with $0 \leqslant t \leqslant 2.5T_0$ and $12.5 \leqslant t \leqslant 15T_0$, and $f_1(t) = \sin^2(\pi t/T)$ with $0 \leqslant t \leqslant T = 15T_0$. ϕ is the relative phase between the driving and the control pulses and is chosen to be 0.1π to generate broadband supercontinuum in the plateau. The molecule is aligned along the laser polarization direction, which can be achieved with a prepulse or static electric field [19,20]. Thus the electron predominantly oscillates in the laser polarization direction, and the one-dimensional model will produce quite reliable results, which has been widely used in previous works. The Hamiltonian is given by $H = [-\partial^2/2\partial x^2 + V(x) - xE(t)]$. $V(x)$ is the Coulomb potential, which can be described by [15, 21] $V(x) = -Z_1/\sqrt{1 + (x + R/2)^2} - Z_2/\sqrt{1 + (x - R/2)^2}$, where Z_1 and Z_2 are the electric charges of the two nuclei and R is the internuclear distance. Here we have assumed that the nuclei are fixed at $\pm R/2$ since the molecular vibrational period is longer than the laser pulse duration [11,15]. In the calculation, we adopt $R = 4$ a.u., $Z_1 = 1$ a.u., and $Z_2 = 2$ a.u. to simulate the asymmetric molecule HeH^{2+} [12,15]. Figure 1 shows the spectrum of the high harmonics generated from asymmetric molecules in the multicycle two-color field (bold blue line). For comparison, the spectrum of the high harmonics generated from asymmetric molecules in the fundamental field alone (thin gray line) is also presented in Fig. 1. As shown

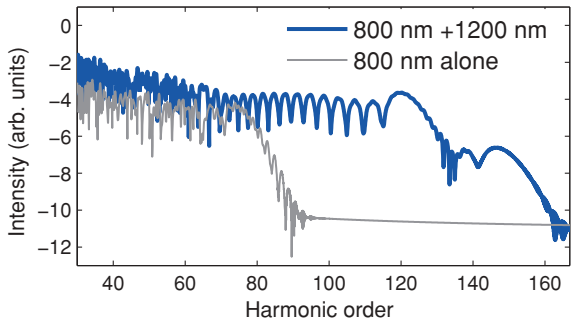

FIG. 1. (Color online) The spectra of high harmonics generated from asymmetric molecules in a two-color field (bold blue line) and in a fundamental field alone (thin gray line).

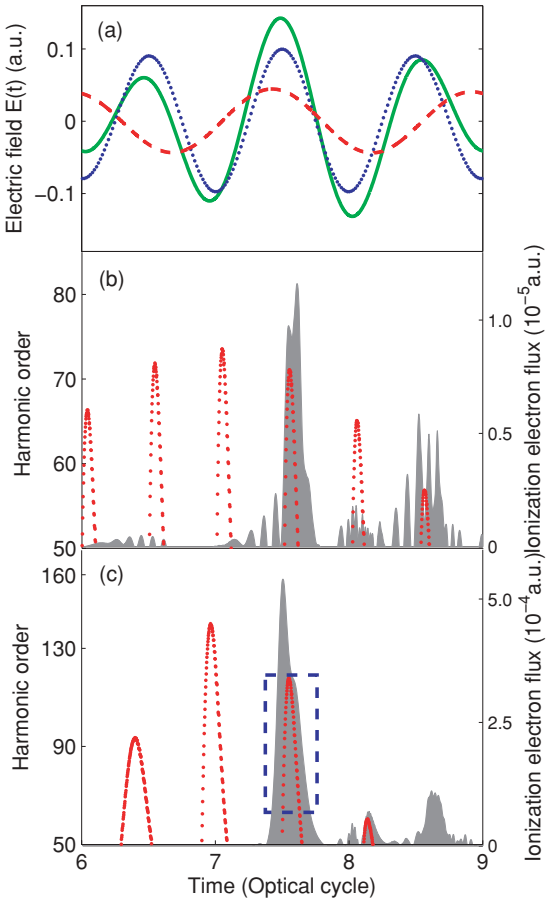

FIG. 2. (Color online) (a) The electric fields of the fundamental field (blue dotted line), control field (red dashed line), and synthesized two-color field (green solid line). (b) The dependence of the classical electron energy on the ionization time (red dots), and the ionized electron flux (filled line) in the fundamental field. (c) The dependence of the classical electron energy on the ionization time (red dots), and the ionized electron flux (filled line) in the two-color field.

in Fig. 1, the harmonics above $60\omega_0$ (ω_0: frequency of the fundamental field) in the two-color field become regular and continuous and form a supercontinuum with a bandwidth of about 100 eV in the plateau, while most harmonics in the fundamental field alone are chaotic (irregular in the whole plateau).

To give a deeper insight into the high harmonic features, we analyzed the electron dynamics of the HHG processes in the fundamental field and in the two-color field, and the results were shown in Fig. 2. In Fig. 2(a), we present the electric fields of the fundamental field (blue dotted line), control field (red dashed line), and synthesized two-color field (green solid line). In Figs. 2(b) and 2(c), we present the dependence of the classical electron energy on the ionization time (red dots) in the fundamental field and in the two-color field, respectively. The ionized electron fluxes (filled line) in the fundamental field and in the two-color field are also presented in Figs. 2(b) and 2(c). Here, the ionized electron flux is calculated by $\Gamma(t) = -d_t[\ln N(t)]$, where $N(t)$ is the norm of the wave function in the bound states [22]. As shown in Fig. 2(b), the asymmetric molecule is predominantly ionized around the

electric peaks at $t = 7.5, 8.0$, and $8.5T_0$ (T_0: the optical cycle of the fundamental field), and the ionized electron flux is highest around the electric peak at $t = 7.5T_0$ and lowest around the electric peak at $t = 8.0T_0$. This is induced by the electric field of the fundamental field and the ionization characteristic of the asymmetric molecule. The ionization process of asymmetric molecules is strikingly different in consecutive half optical cycles. In the half optical cycle, when the electric field is parallel to the permanent dipole of the molecule (PDM), the ionization is suppressed. In the following half optical cycle, the electric field changes its sign and is antiparallel to the PDM. Then the ionization is enhanced [11,12,15]. Therefore, the ionized electron flux is highest around the electric peak at $t = 7.5T_0$ since the electric field is antiparallel to the PDM and its intensity is strongest. The ionized electron fluxes around the electric peaks at $t = 7.0$ and $8.0T_0$ are lower than that around the electric peak at $t = 8.5T_0$ though the intensities of the electric peaks at $t = 7.0$ and $8.0T_0$ are higher than that at $t = 8.5T_0$. The intensity of the fundamental field is much lower than the saturation intensity of the HeH^{2+} molecule. According to the three-step model, the harmonic efficiency is mainly determined by the first step, i.e., the ionization rate. Thus the high harmonics in the fundamental field are mainly emitted from the ejected electron in the half optical cycles around $t = 7.5$, 8.0, and $8.5T_0$. Moreover, the difference between the consecutive half optical cycle photon energies is very small since the laser intensity varies slow in multicycle regime. Thus, the harmonics in the plateau originate from the emission in three different half optical cycles. Since the harmonics emitted in different half optical cycle have different phase, the superposition of these harmonics makes the spectrum chaotic (irregular in the whole plateau).

In the following, we analyze how we can further steer HHG to produce a broadband supercontinuum in the plateau from asymmetric molecules by adding a $2\omega/3$ control field to the multicycle fundamental field. As shown in Fig. 2(a), the intensity of the synthesized field is significantly enhanced at the peaks around $t = 7.5T_0$ and $t = 8.0T_0$ and is dramatically suppressed at the peak around $t = 6.5T_0$ by adding a $2\omega/3$ control field. As a result, the intensity of the multicycle synthesized two-color field varies fast near the pulse center, and the difference between the consecutive half optical cycle photon energies is significantly enlarged compared with that in the multicycle fundamental field. Particularly, the cutoff frequency of the harmonics emitted from the ejected electron in the half optical cycle around $t = 7.5T_0$ is enhanced from $70\omega_0$ to $120\omega_0$ in the presence of the $2\omega/3$ control laser pulse. However, the cutoff frequency of the harmonics in the next half optical cycle is reduced a little, making the difference of the photon energies originated from these two consecutive optical cycles is about 100 eV. Simultaneously, the ionized electron flux in the half optical cycle around $t = 7.5T_0$ is largely enhanced, which makes the ionized electron flux in this half optical cycle is much higher than that in other optical cycles, as shown in Fig. 2(c). Therefore, the high harmonics above $60\omega_0$ in the two-color field, i.e., the high harmonics in the blue dashed rectangle in Fig. 2(c), form a supercontinuum with a bandwidth of about 100 eV at the end of the plateau, as shown in Fig. 1 (blue bold line). As the analysis above, the generation of the broadband supercontinuum in the plateau of the harmonic spectrum with a multicycle driving pulse is attributed from the $2\omega/3$ control field, which further enhance the asymmetry in the process of HHG for asymmetric molecules through simultaneously enhancing the ionization rate in the half optical cycle around the center of the laser pulse and the photon energies emitted from the ejected electron in this half optical cycle. It must be noted that the ionization asymmetry for asymmetric molecules is also a key role for the generation of the supercontinuum. Without the ionization asymmetry, the ionization around the electric peak just before the central electric peak will not be suppressed. Then, the harmonics emitted from the ejected electron around this electric peak will make the supercontinuum disappear. Therefore, our scheme is based on the ionization asymmetry for asymmetric molecules and is not useful for symmetric molecules, such as $H_2{}^+$.

Figures 3(a) and 3(b) show the time-frequency spectrograms of the HHG from asymmetric molecule in the multicycle fundamental field and in the multicycle two-color field, respectively. As shown in Fig. 3(a), there are three dominant radiation peaks obviously corresponding to three dominant recollisions. According to the theory of HHG, the three dominant recollisions originate from the ejected electron around the electric peaks at $t = 7.5$, 8.0, and $8.5T_0$. In Fig. 3(b), only one dominant radiation peak is visible for the high harmonics above $60\omega_0$, corresponding to the dominant recollision originated from the ejected electron around the electric peak at $t = 7.5T_0$. Other radiation peaks disappeared since the cutoff frequency of these radiation peaks is lower than $60\omega_0$. The features consist with the results obtained in Fig. 2. Figures 3(c) and 3(d) show the attosecond pulses generated by filtering the 60th–70th harmonics in Fig. 3(a) and the 100th–110th harmonics in Fig. 3(b), respectively. There are many subpulses in Fig. 3(c), which are originated from different quantum paths. Each quantum path possesses a different phase, giving rise to a complex interference. Therefore, the spectrum is chaotic as shown in Fig. 1 (thin gray line). However, there are only two subpluses in Fig. 3(d), which are attributed to a pair of short and long quantum paths. The interference of this pair of short and long quantum paths leads to the regular modulations in the supercontinuum, as shown in Fig. 1 (bold bule line). The coexistence of the short and long quantum paths also prevents producing an isolated attosecond pulse from the supercontinuum.

To generate an isolated attosecnd pulse from the supercontinuum, one should eliminates one quantum path. This can be achieved by varying the configuration of the focus of the laser pulse relative to the gas jet since the short and long quantum paths have very different phase-match conditions [23,24]. In order to demonstrate this issue, we perform the nonadiabatic three-dimensional propagation simulations for both fundamental and harmonic fields in the gas target [15,25]. Figure 4(a) shows the on-axis macroscopic high harmonic spectrum generated from a 1.0-mm-long gas jet (solid line). The driving and control pulses are assumed as Gaussian beams and are focused 2 mm before the entrance of the gas jet. The beam waists at focus of the two pulses are all 25 μm. The density of the gas jet is isotropic and equal to $2.5 \times 10^{18}/\mathrm{cm}^3$. For comparison, the single-molecule high harmonic spectrum is also presented in Fig. 4(a) (dotted line).

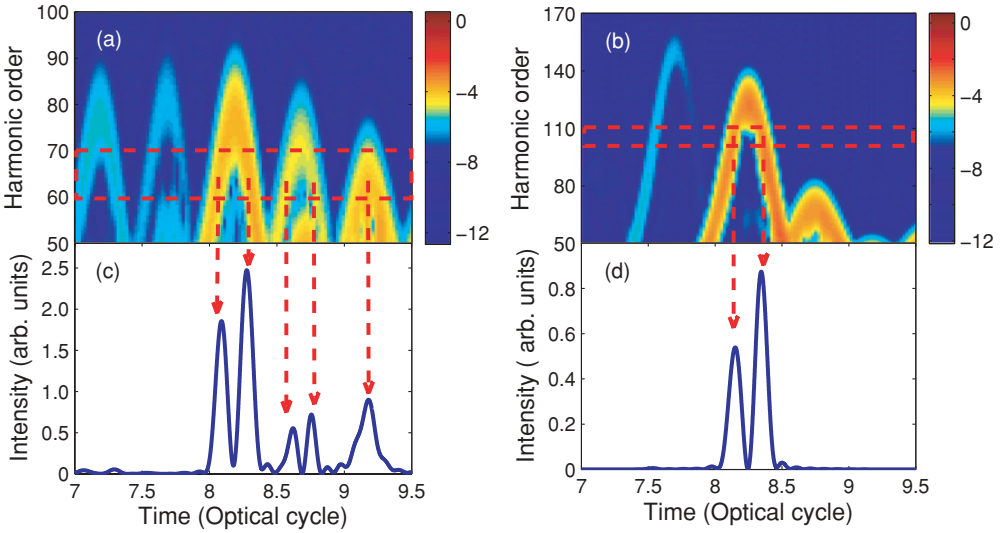

FIG. 3. (Color online) The time-frequency spectrograms of the HHG from asymmetric molecule in the multicycle fundamental field (a) and in the multicycle two-color field (b). [(c) and (d)] The attosecond pulses generated by filtering the 60th–70th harmonics in (a) and the 100th–110th harmonics in (b), respectively.

After propagation through the gas jet, the regular modulations in the supercontinuum are largely eliminated and a smooth broadband supercontinuum is obtained in the plateau. It is because the phase matching of the short quantum path is achieved for the harmonics in the supercontinuum and only the harmonics emitted from the short quantum path survive after propagation. Then the long quantum path is eliminated after propagation, and the interference is weakened as well. Figure 4(b) shows the intensity normalized harmonics pulses

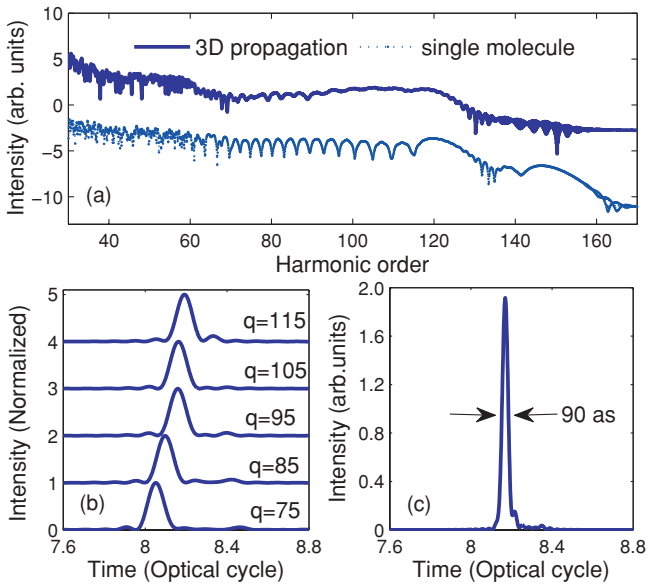

FIG. 4. (Color online) (a) The macroscopic (solid line) and single-molecule (dotted line) high harmonic spectra generated from asymmetric molecules. (b) The attosecond pulses centered at different frequencies generated by filtering 10 harmonics in the macroscopic high harmonic spectrum. (c) Isolated attosecond pulse generated by filtering the high harmonics from the 90th to 100th harmonics in the macroscopic high harmonic spectrum.

produced by superposing 10 harmonics at different orders. There is only one attosecond pulse along the time axis, which corresponds to the short quantum path as compared to Figs. 3(b) and 3(d). This validates that the phase matching of the short quantum path is well achieved for the harmonics in the supercontinuum after propagation through the gas jet. This also indicates that isolated attosecond pulses with tunable central wavelengths can be generated by filtering several harmonics in the supercontinuum. In addition, the duration of the isolated attosecond pulse can be further compressed by synthesizing more harmonics in the supercontinuum. As shown in Fig. 4(c), an efficient isolated 90-as pulse is generated by synthesizing the harmonics from 90th to 110th harmonics in the supercontinuum.

It must be noted that the ionization asymmetry in consecutive half optical cycles for asymmetric molecules becomes weak when the intensity of the driving laser pulse increases. Therefore, the intensity of the synthesized laser pulse cannot be too high though the bandwidth of the supercontinuum can be broadened by increasing the intensity of the fundamental and the control fields. Keeping the intensity rate between the control and the fundamental fields constant, i.e., 0.2, the intensity of the fundamental field needs to be lower than 4.5×10^{14} W/cm^2. The intensity of the control field also needs to be lower than 1.4×10^{14} W/cm^2 when the intensity of the fundamental field is fixed at 3.5×10^{14} W/cm^2. In addition, the supercontinuum will become narrow when the duration of the driving laser pulse increases. The bandwidth of the supercontinuum is only 25 eV when the duration of the driving laser pulse is 20 fs. Continuously increasing the duration of the driving laser pulse, the supercontinuum in the plateau will disappear.

There are several other techniques which can produce isolated attosecond pulses from muticycle pulses, such as two-color fields [15,16,26,27], polarization gating [4,28,29], and double optical gating (DOG) [30,31]. In the polarization gating and DOG techniques, part of laser energy is lost, which

is profitless for improving the attosecond pulse yield. In the technique of multicycle two-color field, the isolated attosecond pulse are generated from the continuous harmonics near the cutoff, of which the efficiency is much lower than that in the plateau due to the intrinsic dynamic properties of electron wavepacket in the two-color field [32]. In our scheme, besides the technique of two-color field, the ionization asymmetry for asymmetric molecules is also used to control the HHG. Therefore, isolated attosecond pulses can be generated from the harmonics in the first plateau.

III. CONCLUSION

In conclusion, the HHG from asymmetry molecules in the presence of a $2\omega/3$ control field has been investigated and isolated attosecond pulses are generated from the harmonics in the plateau of the spectrum with a multicycle laser pulse. With a $2\omega/3$ control field, the ionization of the asymmetric molecules in the half optical cycle around the central electric peak is largely enhanced, which further enhances the ionization asymmetry of asymmetric molecules and makes the ionization rate in this half optical cycle is much higher than that in other optical cycles. The $2\omega/3$ control laser pulse also significantly enlarges the difference of the photon energies emitted from the ejected electron in the half optical cycle around the central electric peak and its next half optical cycle. Therefore, the harmonics above $60\omega_0$ are mainly emitted from the ejected electron in the half optical cycle around the central electric peak, forming a broadband supercontinuum in the plateau of the spectrum. Since the harmonics in the supercontinuum are attributed to a pair of short and long quantum paths, there are regular modulations in the supercontinuum. The modulations can be eliminated after propagation through the gas jet. Then isolated attosecond pulses with tunable central wavelengths or an isolated attosecond pulse with a duration of 90 as can be directly filtered from the harmonics in the supercontinuum. Moreover, the supercontinuum is in the plateau of the spectrum and covers a very broad bandwidth, which allows one produce an isolated attosecond pulse shorter than 50 as in the transform limited case. In addition, the multicycle laser pulse is more easily available than a few-cycle laser pulse, which allows the scheme to be implemented in experiment relatively easily. At the same time, many asymmetric molecules, such as alkali hydrides (LiH^+, etc.), which are stable molecular systems with the similar ionization asymmetry as HeH^{2+} and a single valence electron, are possible candidates for future experiments. Therefore, even though our results are obtained with HeH^{2+}, which is unstable molecular system, it is expected that isolated attosecond pulses still can be produced with other asymmetric molecules.

ACKNOWLEDGMENTS

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