Ellipticity-tunable attosecond XUV pulse generation with a rotating bichromatic circularly polarized laser field

XIAOFAN ZHANG,† XIANGSONG ZHU,†,‡ XI LIU,† DIAN WANG,† QINGBIN ZHANG,† PENGFEI LAN,† AND PEIXIANG LU†,‡,†

†Wuhan National Laboratory for Optoelectronics and School of Physics, Huazhong University of Science and Technology, Wuhan 430074, China
‡Laboratory of Optical Information Technology, Wuhan Institute of Technology, Wuhan 430205, China

†E-mail: lupeixiang@mail.hust.edu.cn

Received 9 December 2016; revised 6 February 2017; accepted 9 February 2017; posted 10 February 2017 (Doc. ID 282413); published 3 March 2017

We propose and theoretically demonstrate a method to generate attosecond XUV pulses with tunable ellipticity from aligned molecules irradiated by a bichromatic counterrotating circularly polarized (BCCP) driving laser field. By rotating the BCCP field, the attosecond XUV pulse varies from being left elliptically polarized to right elliptically polarized. The rotation of the BCCP field can be easily achieved by adjusting the relative phases between the two circularly polarized components. This scheme will benefit a broad range of applications, including the exploration of chiral-sensitive properties of the light–matter interaction and time-resolved imaging of magnetic structures.

OCIS codes: (020.2649) Strong field laser physics; (320.7090) Ultrafast lasers; (340.7480) X-rays, soft x-rays, extreme ultraviolet (EUV).

https://doi.org/10.1364/OL.42.001027

High harmonic generation (HHG) in atoms and molecules is a highly nonlinear phenomenon in strong field light–matter interactions [1]. In the HHG process, electrons are ionized into the continuum and, subsequently, accelerated under the influence of the driving electric field [2]. When recombining with the parent ion, the electrons liberate their accumulated kinetic energy, and HHG occurs [3]. The HHG has been an attractive topic for its preeminent applications such as the coherent attosecond pulse generation in the XUV regime. In the past decades, the generation of linearly polarized attosecond XUV pulse has been studied in the vast majority of works [4–7]. A linearly polarized attosecond pulse is one-dimensional, and the range of its applications is greatly restricted, while attosecond pulse with polarization other than linear polarization provides an additional degree of freedom and is very important for a wide range of applications [8–10] such as the study of ultrafast chiral-specific dynamics in molecules and x-ray magnetic circular dichroism spectroscopy. Therefore, the study of the generation of non-linearly polarized attosecond pulse has been a hot spot in recent years [11–15].

Very recently, the so-called bichromatic counterrotating circularly polarized (BCCP) field has attracted a great deal of attention [16–23]. A BCCP laser field consists of two coplanar counter-rotating circularly polarized fields having the angular frequencies $\omega$ and $2\omega$ [22]. The time-dependent electric field performs a Lissajous figure on its polarization plane with a three-fold symmetry [23,24], i.e., it consists of three lobes per cycle. HHG with the BCCP laser field is very important for applications [25,26]. There are two main advantages to generating high harmonics with this field: (1) the generated individual harmonics are circularly polarized, and (2) the conversion efficiency of HHG needs to not be compromised [18]. In parallel with the realization of generation of circularly polarized harmonics, it was shown that the polarization state of individual harmonics can be controlled in a recent experiment [18]. This controllability was achieved by continuously changing the ellipticity of one of the bichromatic field components. In practical applications, the controllability of the polarization state of isolated attosecond pulses synthesized by a broadband harmonics is more widely demanded. However, a robust and practical scheme for controlling the polarization state of the attosecond pulse is very challenging.

In this Letter, a scheme to generate an attosecond XUV pulse with controllable ellipticity is demonstrated numerically. The controllability of the ellipticity is based on the dependence of the harmonic process on the angle between the driving field and the non-spherical targets. Specifically, we use a BCCP laser field interacting with aligned molecules to generate high harmonics. When the relative phase of the two components of the BCCP laser field is varied, the BCCP laser field is rotated relative to the aligned molecular axis. Consequently, the ellipticity of the high harmonics can be varied, and the ellipticity of the synthesized attosecond XUV pulse is tuned correspondingly. The relative phase can be easily adjusted in an experiment. Such a property makes our scheme extremely attractive for practical implementation.
The BCCP laser field vector polarizing in the \(x\)-\(y\) plane is defined by \(E(t) = E_0 f(t) \left[ (\cos(\omega t + \Delta \phi) + \cos(2\omega t))\hat{x} + \sin(\omega t + \Delta \phi) - \sin(2\omega t) \right] \) [27], where \(E_0\) is the amplitude of one component of the BCCP laser field. The corresponding intensity \(I\) is \(1 \times 10^{14}\ \text{W/cm}^2\). \(\omega\) is the frequency of the 1600 nm pulse. The \(\omega\) field rotates clockwise, while the second-harmonic field \(2\omega\) rotates counterclockwise. A 

\[ \sin^2(\xi) \]

is used to characterize the pulse profile \(f(t)\), in which we set \(T = 3T_0\), and \(T_0\) denotes the optical cycle of the \(\omega\) field. \(\Delta \phi\) is the relative phase of \(\omega\) and the \(2\omega\) field, which can be easily adjusted in experiment. For the BCCP laser field, the varying relative phase \(\Delta \phi\) will lead to a rotation of the field around the propagation direction by an angle \(\alpha = \Delta \phi/3\) [28]. This rotation can influence the response of the aligned molecule to the BCCP laser field and, thus, influence the polarization state of the generated high harmonics. This is the key feature of the BCCP laser field utilized in our scheme to control the polarization state of the attosecond XUV pulse. We first take the example of CO to demonstrate our method. The schematics for different \(\Delta \phi\) are shown in the insets of Figs. 1(a) and 1(b). We numerically solve the two-dimensional (2D) time-dependent Schrödinger equation (TDSE) for the interaction between the BCCP laser field and aligned molecules [29]. In addition, the dipole acceleration can be obtained [15]. The molecules are modeled with the effective 2D potential [30], and the ground state \(\Psi_0\) of the target molecule is obtained by solving TDSE with imaginary time propagation [31]. The ellipticity of high harmonics can be obtained as \(e = (|a_+| - |a_-|)/(|a_+| + |a_-|)\) [28], where \(a_\pm = \sqrt{\tau} (a_x \pm ia_y)\). \(a_x\) and \(a_y\) are the \(x\) and \(y\) components of dipole acceleration in the frequency domain, respectively. A harmonic spectrum can be projected into two counter-rotating components, which are right circularly polarized harmonics obtained as \(D_+ = |a_+|^2\) with \(e > 0\) and left circularly polarized harmonics obtained as \(D_- = |a_-|^2\) with \(e < 0\).

Here, we show that the relative intensity of the counter-rotating components \(D_\pm\) strongly depends on \(\Delta \phi\). In Figs. 1(a) and 1(b), for different \(\Delta \phi = 150^\circ\) and 240°, the intensity of right (red line) and left (blue line) circularly polarized components is presented. One can see that the intensity of the left circularly polarized component is higher than that of the right circularly polarized component at the range from 75th-order (labeled by the dashed-dotted line) to 105th-order (labeled by the dashed line) for \(\Delta \phi = 150^\circ\). However, for \(\Delta \phi = 240^\circ\), a reversed result is observed. According to the expression of \(e\), the whole harmonic spectrum at range from 75th-order to 105th-order, since \(\Delta \phi = 150^\circ\) is left elliptically polarized \((e < 0)\) while that for \(\Delta \phi = 240^\circ\) is right elliptically polarized \((e > 0)\). These results suggest that, with different \(\Delta \phi\), a different ellipticity of the harmonic emission can be obtained. Then we present the ellipticity distribution as a function of \(\Delta \phi\) and harmonic order in Fig. 1(c). The red regions represent positive ellipticity, which indicates that the high harmonics are right elliptically polarized. The blue regions represent negative ellipticity, which indicates that the high harmonics are left elliptically polarized. The ellipticity is almost zero in the green regions, which indicates that the emitted harmonic signal is almost linearly polarized. From this figure, it is shown that the ellipticity is obviously affected by \(\Delta \phi\), and it is continuously varied from –0.5 to 0.7 with different \(\Delta \phi\). To clearly exhibit the variation of the harmonic ellipticity, we plot the ellipticity curves as a function of \(\Delta \phi\) for the 80th, 90th, and 100th harmonic orders in Fig. 1(d). Note that, unlike the HHG from atoms with \(C_n\) symmetry or molecules with \(C_{3n}\) \((n\) is a positive integer) symmetry [19,23,32], selection rules \((3n \pm 1)\) are violated here for the linear molecules. Each order harmonic consists of both components \(D_\pm\), and, thus, is not circularly polarized.

In order to analyze the physical reason of the controllability of the high harmonic polarization state by varying \(\Delta \phi\), we calculate the superposition of the three recombination dipole moments contributed from the three lobes of the BCCP laser field. Each recombination dipole moment is calculated as \(D = \Re [\Psi_0(r, \theta) \exp(\hat{\rho} \cdot \hat{r})] [30]\), in which \(\rho\) and \(\theta\) are the recombination momentum and the angle between the recombination direction and the molecular axis in cartesian coordinates. Then we present the phase difference \(\Delta \phi_{\text{HHG}}\) between the \(x\) and \(y\) components of the superposed dipole moment in a polar plot [see Fig. 2(a)]. Besides, the schematics for the relative orientation between the BCCP laser field and the aligned molecule for three typical \(\Delta \phi\) are presented in Figs. 2(b)–2(d). The molecule is aligned at the \(x\) axis. When \(\Delta \phi\) is changed, the driving field is rotated by an angle \(\alpha = 2 \Delta \phi/3\) relative to the aligned molecular axis. From Fig. 2, one can see that, at about \(\Delta \phi = 135^\circ\) (pointed by the blue arrow), the phase difference \(\Delta \phi_{\text{HHG}}\) is about \(-\pi/2\). Correspondingly, a negative ellipticity peak appears in Fig. 1(c). In this case, the driving field is rotated by \(\alpha = 90^\circ\), and the first field lobe is located at the \(y\) axis [as shown in Fig. 2(c)], which is perpendicular to the molecular axis. At about \(\Delta \phi = 225^\circ\) (pointed by the orange arrow), \(\Delta \phi_{\text{HHG}}\) is about \(\pi/2\). Similarly, a positive ellipticity peak is shown in Fig. 1(c). In this case, the driving field is rotated by \(\alpha = 150^\circ\) and the second field lobe is also perpendicular to the aligned molecular axis [as shown in Fig. 2(d)]. When \(\Delta \phi\) tends to 180°, \(\Delta \phi_{\text{HHG}}\) tends to zero or \(-\pi\) (as shown by the black and white arrows), and the ellipticity of the high harmonics tends to zero [see Fig. 1(c)]. When \(\Delta \phi\) right equals

---

**Fig. 1.** Polarization of HHG from a CO molecule. (a), (b) Harmonic intensity as a function of the harmonic order with \(\Delta \phi = 150^\circ\) and 240°. The red and blue curves correspond to the harmonic components with right elliptical polarization \(D_+\) and left elliptical polarization \(D_-\). The insets show the BCCP laser field and its rotation relative to the CO molecular axis. (c) Ellipticity distribution of high harmonics versus harmonic order and \(\Delta \phi\). (d) Ellipticity as a function of \(\Delta \phi\) for an 80th-order (green line), 90th-order (red line), and 100th-order (blue line) harmonic. The horizontal dotted line represents the zero ellipticity.

---
180° (pointed by the green arrow), the third lobe of the driving field is parallel to the molecular axis [as shown in Fig. 2(b)]. In this case, the y component of the superposed dipole moment is suppressed due to the symmetry of the molecule. As a result, the obtained phase of the y component is strongly modulated and disorder structures appear. However, the harmonic emission only contributed by the x component is actually linearly polarized, and the ellipticity is zero. From the results in Fig. 2, it is found that the ellipticity reaches the peak value when one laser lobe is perpendicular to the molecular axis (i.e., \( \Delta \phi = 45°, 135°, 225°, 315° \)). When one laser lobe is parallel to the molecular axis (i.e., \( \Delta \phi = 0°, 90°, 180°, 270° \)), the ellipticity is zero, and the high harmonics is linearly polarized. For other \( \Delta \phi \), for example between 135° and 225°, the phase difference \( \Delta \phi_{\text{HHG}} \) is varied from \(-\pi/2\) to \(+\pi/2\), and the ellipticity of high harmonics is varied correspondingly as shown in Fig. 1(c). All the results and discussions indicate that the variation of high harmonic ellipticity originates from the dependence of the HHG process on the angle between the driving field and the non-spherical molecules.

Next, we demonstrate that attosecond pulses with various ellipticities can be obtained from our scheme with various \( \Delta \phi \). The pulses synthesized by a series of high harmonics (from 75th-order to 105th-order) for \( \Delta \phi = 90°, 150°, \) and 240° are presented in Figs. 3(a)–3(c). It is shown that the duration of the main pulses is about 180 as. For clearly showing the polarization characteristic of the attosecond pulses, we present the corresponding electric field in Figs. 3(d)–3(f), i.e., the electric fields are elliptically polarized for both 150° and 240° and linearly polarized for 90°. The arrows in Figs. 3(d) and 3(f) label the rotating direction of the electric field. We also calculate the ratios of the minor axis to the major axis of the elliptical polarized attosecond fields in Figs. 3(d)–3(f) to evaluate the ellipticity of the attosecond pulses. The ratios are 0.52, 0, and 0.81 for Figs. 3(d)–3(f), respectively. These results obtained in Fig. 3 show that various ellipticities can be obtained with various \( \Delta \phi \), and the obtained ellipticity is in accordance with Fig. 1. Based on the ellipticity distribution of high harmonics in Fig. 1(c), the synthesized attosecond XUV pulse with different ellipticity between \(-0.52\) and 0.81 can be generated by adjusting the relative phase \( \Delta \phi \).

Finally, we demonstrate that the attosecond pulse with tunable ellipticity by varying \( \Delta \phi \) of the driving field can also be generated from other non-spherical molecules. We take N\(_2\) and CO\(_2\) as examples. The results are shown in Fig. 4. The ellipticity distributions of high harmonics for N\(_2\) and CO\(_2\) as a function of \( \Delta \phi \) and harmonic order are presented in Figs. 4(a) and 4(c) respectively. From these two figures, one can see that the ellipticity of high harmonics varies from more than 0.5 to \(-0.2\) for N\(_2\) and from 0.5 to \(-0.35\) for CO\(_2\) with varied \( \Delta \phi \). This phenomenon is consistent with the discussion about CO above. Then we present the electric field of the synthesized pulse from the 65th-order to 75th-order harmonic for N\(_2\) and from the 50th-order to 60th-order harmonic for CO\(_2\) on the \( E_x-E_y \) plane in Figs. 4(h)–4(d) and 4(f)–4(h) for different \( \Delta \phi \). The duration of the synthesized pulses is about 400 as. The arrows label the rotating direction of the electric field. One can clearly see that the attosecond pulses vary from right elliptically polarized to left elliptically polarized, and the ellipticity is controlled by varying \( \Delta \phi \). The controllability originates from the dependence of the HHG process on the angle between the driving field and the aligned molecules. Therefore, for different non-spherical molecules, the attosecond XUV pulse with tunable ellipticity can be obtained by varying \( \Delta \phi \) with our scheme.
In conclusion, we have demonstrated a scheme to generate an attosecond XUV pulse with tunable ellipticity from the interaction between a BCCP laser field and aligned molecules. It is shown that the attosecond XUV pulse with desirable ellipticity can be obtained by adjusting the relative phase $\Delta \phi$ of the two components of the BCCP laser field. Our scheme works for different non-spherical molecules. Since the tunability of the ellipticity can be achieved by only adjusting $\Delta \phi$, our scheme is easily implemented in current experiments.

**Funding.** National Natural Science Foundation of China (NSFC) (11234004, 11404123, 11422435, 11627809, 11574101).

**REFERENCES**