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Elliptically polarized high-order harmonic generation of Ar atom in an intense laser field

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High-order harmonic generation (HHG) of Ar atom in an elliptically polarized intense laser field is experimentally investigated in this work. Interestingly, the anomalous ellipticity dependence on the laser ellipticity (ε) in the lower-order harmonics is observed, specifically in the 13rd-order, which displays a maximal harmonic intensity at $\varepsilon \approx 0.1$, rather than at $\varepsilon = 0$ as expected. This contradicts the general trend of harmonic yield, which typically decreases with the increase of laser ellipticity. In this study, we attribute this phenomenon to the disruption of the symmetry of the wave function by the Coulomb effect, leading to the generation of a harmonic with high ellipticity. This finding provides valuable insights into the behavior of elliptically polarized harmonics and opens up a potential way for exploring new applications in ultrafast spectroscopy and light–matter interactions.

Keywords: high-order harmonic generation, Coulomb effect, elliptically polarized intense laser field

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1. Introduction

High-order harmonic generation (HHG) is a nonlinear process driven by the interaction of laser radiation with various media.[1,2] It has emerged as a promising technique for generating coherent extreme ultraviolet (XUV) light on a tabletop scale,[3] as well as for producing attosecond pulse trains or isolated attosecond pulses.[4,5] The advancements in HHG have provided new opportunities for investigating electronic dynamics in different systems, including gases,[6,7] solids,[8–11] liquids,[12] and plasmas.[13]

The HHG can be well described by the semi-classical three-step model (TSM).[14] According to this model, the high-order harmonic (HH) is generated through the following steps: (i) an electron is released from an atom through tunnel-ionization, (ii) the electron is accelerated away and can be driven back towards the parent ion core by the oscillating laser field, (iii) the electron may recombine with the parent ion core, leading to the emission of a coherent XUV photon. However, considering the significant influence of the Coulomb effect on electron dynamics, some researchers have proposed that there exists an additional step between tunneling-out and emission. This additional step has a non-negligible influence on the ionization time for HHG.[15,16]

Based on the picture of TSM, a linearly polarized driving laser can generate linearly polarized HH.[14] Additionally, HH can also be generated from an elliptically polarized driving laser.[17] The controlling of the laser ellipticity is a powerful method for manipulating electron trajectories[18,19] and has been widely investigated in HHG.[20,21] According to the prediction of a semiclassical model, the HH yields decrease with the increase of the driving laser ellipticity.[20] Furthermore, anomalous cases in which the maximum yield of harmonics shifted have been reported in several experiments[22–24] and theories studies[25–28] when the driving laser with high ellipticity, especially in near- or below-threshold harmonics.[23,24] The early experiments have measured the ellipticity dependence of lower-order harmonics which are slightly above the ionization threshold of Ar.[29] They have observed an offset angle between the major axes of the driving laser and the harmonic polarization, indicating the generation of elliptically polarized harmonics. These similar phenomena in Ref. [29] have also been reported in other rare gases.[30] However, the underlying physical origin of this anomalous phenomenon remains controversial.

Understanding the details of elliptically polarized harmonic generation is crucial for elucidating the physical mechanism of HHG.[31,32] Strelkov *et al.* have proposed a simple model to describe the harmonic ellipticity and the offset angle. They have attributed the origin of elliptically polarized harmonics to the quantum-mechanical uncertainty of electron motion.[31] According to a similar model from the Ref. [31], they have further described harmonic polarization properties with the different laser wavelengths and different valence orbitals.[32] The Coulomb effect was neglected in their model. However, more results have shown that the Coulomb effect plays an indispensable role in HHG.[15,16,33] The semiclas-

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sical Coulomb-modified strong-field approximation (MSFA) model has confirmed that the Coulomb effect changes the HHG emission time.[16] This effect is more pronounced for lower-order harmonics of short electron trajectories. Zhu *et al.*[33] have employed a theoretical model called the strong-field eikonal-Volkov approximation (SF-EVA) to elucidate the origin of elliptically polarized harmonics. They indicated that the Coulomb effect contributes to the generation of elliptically polarized harmonics for lower orders of short trajectories.

Highly elliptically polarized harmonics have numerous applications, including magnetic interactions[34] and probing chiral interactions of molecules.[35] Several methods have been proposed to generate elliptically polarized harmonics.[36–39] Fleischer *et al.* have obtained harmonics with fully controlled polarization by using a dichromatic driving laser that has elliptical polarization with opposite helicity.[36] Dorney *et al.* have achieved bright, high-energy, and polarization-controlled HH by adjusting the intensity ratio of the dichromatic circularly polarized driving laser.[37] Recently, Huang *et al.* have demonstrated robust control of polarization for isolated XUV pulses by using two counter-rotating few-cycle laser beams while retaining the high conversion efficiency.[38] Lambert *et al.* have generated high ellipticity harmonics by using an orthogonal polarization two-color laser field with similar efficiencies to those of linearly polarized harmonics.[39]

In this work, we experimentally measure the HHG yields of Ar by varying the ellipticity and intensity of the driving laser field. We analyze the ellipticity dependence of HH ratios between low and high orders for both long trajectory and short trajectory, which is consistent with our previous studies.[40] Besides, we observe anomalous ellipticity dependence in the lower-order harmonic. Based on our experimental results and previous investigations,[33] we discuss the influence of the Coulomb effect on harmonic ellipticity.

2. Experiments

The experimental scheme is shown in Fig. 1. We use a driving laser (Ti: sapphire laser system, 1 kHz, 35 fs, 800 nm) to interact with the gas target and generate harmonics. Then a homemade XUV spectrometer is used to collect harmonics as described in our previous researches.[7,40,41] The intensity of the driving laser is controlled by a neutral density filter (ND). The elliptically polarized driving laser is adjusted by a zero-order half-wave plate (HWP) placed in front of a zero-order quarter-wave plate (QWP). The driving laser ellipticity (ε) is continuously adjustable between $\varepsilon = 0$ (linear polarization) and $\varepsilon = 1$ (circular polarization). To generate harmonics, the driving laser is focused onto the gas jet in the vacuum chamber by a convex lens ($f = 300$ mm). Due to the difference between the phase-matching mechanisms,[42] the long trajectory or short trajectory can be chosen experimentally by changing the relative position between the gas jet and the laser focus position. The generated harmonics are reflected by a gold mirror into the XUV spectrometer. The harmonic spectrum is resolved by using a spherical grating dispersion that has a groove density of 1200 grooves/mm. The grating can sensitively check the polarization direction of the HH.[23,43] Therefore, we set the fast axis of the QWP parallel to the groove direction of the grating. By adjusting the angle between the fast axis of HWP and QWP, the polarization direction and ellipticity of the driving laser field can be controlled. Then the harmonics are focused onto a microchannel plate (MCP) detector fitted with a phosphor screen. A CCD camera is used to image the phosphor screen and transfer it into a computer.

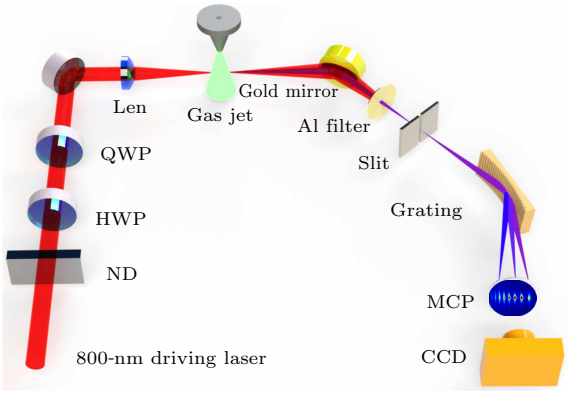


Fig. 1. Experimental scheme for high-harmonic generation driven by 800-nm laser field; see text for details.

3. Results and discussion

Firstly, we compare the ellipticity dependence of harmonic yields in low order and high order for different trajectories, respectively, which is consistent with our previous result.[40] We have plotted the normalized ratio of H17/H21 (Fig. 2(a)) and H17/H25 (Fig. 2(b)) as a function of driving laser ellipticity. The Hn denotes the n-th harmonic order, where n is the order of the harmonics. The result reveals that the ratio for both trajectories rises as the laser ellipticity increases. This trend can be explained well by a nonadiabatic semiclassical calculation, which indicates that the initial longitudinal momentum v_{par} and the initial transverse momentum v_{perp} have different weights for different order harmonics.[40] With the increase of laser ellipticity, the increment of v_{perp} for higher-order harmonics is particularly prominent. This causes the weights of low-order harmonics to be larger than the weights of higher ones, and the ellipticity dependence of H17/H21 ratio and H17/H25 ratio rise for both long trajectory and short trajectory. Comparing with short trajectory, the difference in v_{perp} between different harmonic orders is small for long trajectory, resulting in weaker dependence on laser ellipticity. Consequently, as the laser ellipticity increases, the ratio of low-order harmonics to high-order harmonics for short tra-

jectory rises faster than for long trajectory. These results show normal dependence on the laser ellipticity.

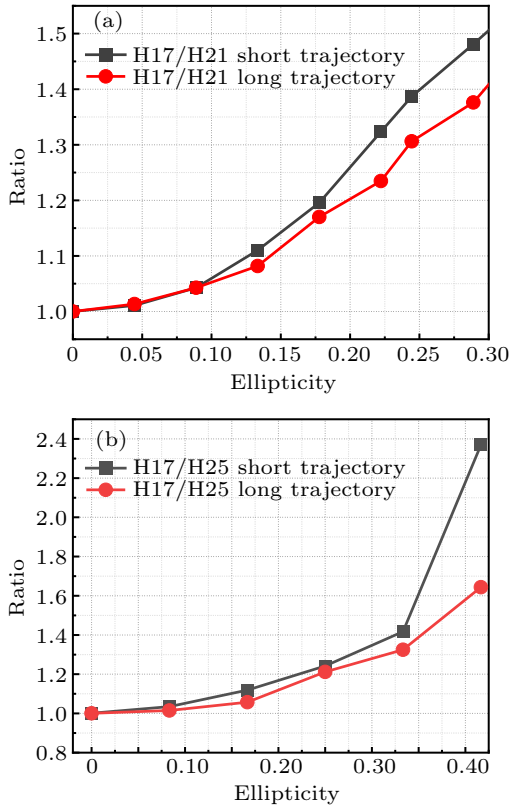


Fig. 2. Normalized experimental ellipticity dependence of (a) H17/H21 and (b) H17/H25 ratio of Ar for both short trajectory and long trajectory.

Next, we extend the measurement of the ellipticity dependence of HH yields for both short trajectory and long trajectory. Figure 3 shows the normalized ellipticity dependence of H13 and H19 for long trajectory and short trajectory, respectively. The intensity of the driving laser is approximately 3.66×10^{14} W/cm^2. Except H13 of short trajectory, we find that the overall trend of the harmonic ellipticity dependence decreases with driving laser ellipticity increasing. This phenomenon is similar to the finding of Ref. [18] and can be effectively explained by TSM.[14] The driving laser ellipticity triggers off a lateral shift of the electron wave-packet and reduces the probability of recombination between an electron and a parent ion. Interestingly, a striking difference appears in the case of H13 for the short trajectory as indicated by red circles in Fig. 3. The ellipticity dependence is more anomalously on the normalized harmonic yield, and the normalized harmonic yield reaches a maximum value at $\varepsilon \approx 0.1$ instead of $\varepsilon = 0$.

In order to intensively investigate the abnormal appearance, we observe the ellipticity dependence of harmonic yields for the short trajectory. Figure 4 displays the relationship between laser ellipticity and harmonic yields from H11 to H19, respectively. The harmonic yields of H15, H17, and H19 exhibit a decreasing trend as the laser ellipticity increases, which is consistent with the result of Ref. [44]. Previous studies have indicated that the sensitivity of laser ellipticity slightly rises with the increase of harmonic order.[45] The near-threshold harmonic H11 shows a weaker discrepancy than other orders. This can be attributed to the destructive interference between these trajectories and the influence of the Coulomb potential from the ion core.[24] Besides, the H13 exhibits an anomalous ellipticity dependence which is obviously different from other orders'. The experimental results are similar to those described in Ref. [23], that is, only certain harmonic orders are abnormal. The process of HHG in near- or below-threshold harmonics is complex due to the joint effect of various physical mechanisms,[46] which possibly causes abnormal results similar to our observation for H13.

According to the SF-EVA model, Zhu *et al.*[33] revealed the influence of the Coulomb effect on the harmonic ellipticity. When the model includes the Coulomb effect, the phase difference between two orthogonal components of the harmonic varies in the range $[0, 2\pi)$, resulting in the non-zero ellipticity of the harmonics. This effect is attributed to the broken symmetry of the wave function Ψ_k. According to the theoretical findings, the Coulomb effect has a greater influence for lower-order harmonics of short trajectory than for other harmonics. In addition, their results indicate that with the increase of the harmonic order, the harmonic ellipticity first increases briefly and then decreases sharply. When considering the short-range potential, the harmonic ellipticity drops to zero more quickly. The maximum of the harmonic ellipticity is obtained at photon energy of about 20.03 eV with using a 1300-nm driving laser field and is identified as H13 in our experiments using an 800-nm driving laser field. As described above, the ellipticity harmonic generation due to the Coulomb effect will induce an anomalous enhancement of the harmonic yield driven by a high ellipticity laser. Under the effect of this mechanism, H13 exhibits obviously abnormal ellipticity dependence with the maximum value of harmonic yield occurring at $\varepsilon \approx 0.1$ instead of $\varepsilon = 0$, as shown in Figs. 3 and 4.

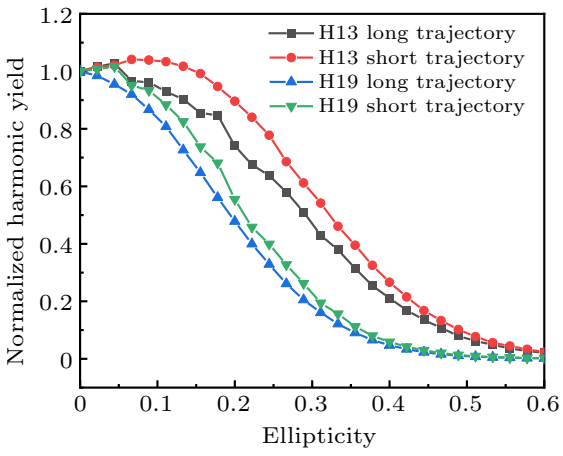


Fig. 3. Variations of normalized harmonic yield with ellipticity of H13 and H19 for long trajectory or short trajectory at laser intensity of 3.66×10^{14} W/cm^2.

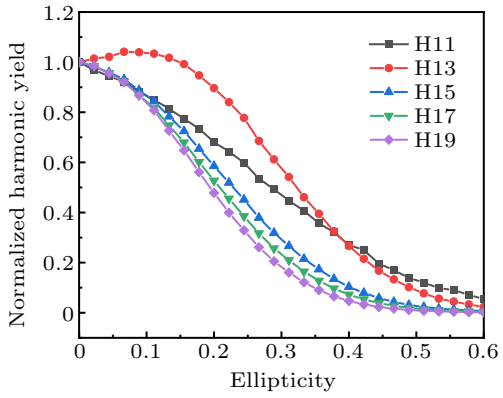


Fig. 4. Variations of normalized harmonic yield with ellipticity for H11, H13, H15, H17, and H19 at laser intensity of 3.66×10^{14} W/cm^2.

We further measure the influence of driving laser intensity for the anomalous ellipticity dependence of H13 in a range from 4.14×10^{13} W/cm^2 to 3.32×10^{14} W/cm^2. The results are presented in Fig. 5. At a lower intensity of 4.14×10^{13} W/cm^2, the H13 harmonic yield decreases with the increase of laser ellipticity, which is in contrast with the anomalous dependence observed at an intensity exceeding 1.24×10^{14} W/cm^2 (maximum harmonic yield occurs at $\varepsilon \approx 0.14$). Within a certain laser intensity range, the anomalous phenomenon changes obviously with the increase of the driving laser intensity. As mentioned above, the origin of elliptically polarized harmonics is influenced by the Coulomb effect, which will be affected by the intensity of the driving laser.[15] Therefore, this process will be influenced by laser intensity, thereby changing the tendency of ellipticity dependence of the harmonic.

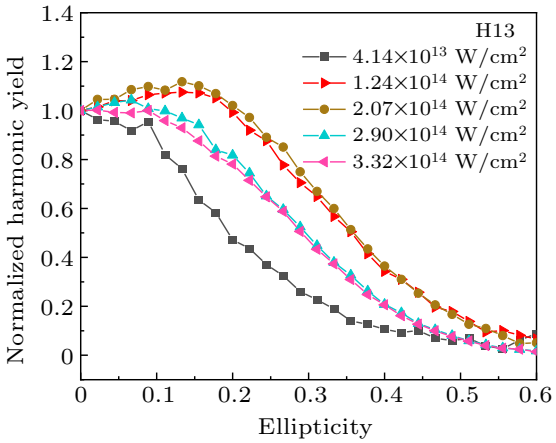


Fig. 5. Variations of normalized harmonic yield with ellipticity of H13 at different laser intensities.

4. Conclusions

In this study, we conducted a systematic investigation on the dependence of driving laser field ellipticity and intensity on the harmonic generation of Ar. Our results show that there is a corresponding increase in the normalized ratio of low-order harmonics to high-order harmonics with laser ellipticity increasing. This trend is more pronounced and faster in short trajectory than in long trajectory. Interestingly, we observe anomalous dependence of the H13 harmonic on ellipticity, which is strongly correlated with the intensity of the driving laser. To explain these observations, we consider that the break of symmetry in the continuum state, induced by the Coulomb effect, plays a crucial role. This explanation is based on SF-EVA model described in the literature. By considering the Coulomb effect, we can better understand the elliptically polarized harmonic generation process. Our findings open up new way for studying dynamics on an ultrashort time scale and offer more opportunities for exploring the behaviors of elliptically polarized harmonics.

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