All optical method for measuring the carrier envelope phase from half-cycle cutoffs*

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An all optical method is demonstrated for measuring the carrier-envelope phase (CEP) of few-cycle laser pulses. It is found that, in the few-cycle regime, the high harmonic spectrum generated from asymmetric molecules shows several half-cycle cutoffs that change their positions as the CEP varies. Such half-cycle cutoffs represent the fingerprint of different quantum trajectories and the waveform of the driving pulse. In this case, the CEP can be accurately measured from the half-cycle cutoffs.

Keywords: high harmonic generation, carrier-envelope phase, measurement

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1. Introduction

In the last decade, the development of laser technology makes it possible to generate intense, near single-cycle infrared (IR) light pulses.[1–3] For such few-cycle laser pulses, the waveform of the laser field depends sensitively on the phase shift between the carrier wave and envelope, which is usually called the carrier-envelope phase (CEP) or the absolute phase.[1] Since the electronic motions in atoms and molecules, which are of the attosecond scale, can directly respond to the instantaneous field of the driving pulse, the emergence of few-cycle intense laser pulses opens up a new horizon for time-resolved studying and controlling the ultrafast laser–matter interaction processes, such as high harmonic and attosecond pulse generation,[1,4] above threshold ionization,[5–7] double ionization,[8,9] Thomson scattering[10–12] and so on.[13,14] Therefore, precise CEP measurement becomes an essential issue.

At present, the f-to-2f scheme is the most widespread technique for phase stabilization and measurement of a few-cycle femtosecond laser.[15–17] However, the f-to-2f interferometer only measures the change rate of the CEP and thus is blind to the absolute phase. Therefore, the measurement of the absolute CEP requires a fundamentally different approach. To this end, a CEP meter based on measuring the strong left–right asymmetry[5,18–20] of rescattering photoelectrons in above threshold ionization (ATI) process was demonstrated by Paulus *et al.* This method, however, has to face the difficulty of measuring the rescattering photoelectron spectrum which consists of only a tiny fraction (1%) of the total photoelectron yield.[21,22] Usually, due to the space charge effect of time-of-flight and velocity map imaging spectrometers, thousands of consecutive pulse shots are required to acquire enough signal-to-noise ratio. Consequently, it requires lengthy measurement time and relatively complex experimental setups. Another approach for CEP measurement is based on the high harmonic generation (HHG) from atomic gases. It has been reported that the spectral structure in the cutoff region[1,23,24] [e.g., to be discrete or continuum as shown in Figs. 1(c) and 1(d)] and the cutoff position[25–28] depend on the CEP of the driving laser pulse. The signature of CEP can also be found in the interference pattern in the plateau region.[29–32] Compared with ATI, the HHG approach is an all optical method that can be more easily realized. The high harmonic spectrum can be routinely measured with the conventional flat field X-ray spectrometer, which facilitates the single-shot CEP measurement, i.e., measures the CEP for each laser shot in parallel to another pulse shot.[33] However, the disadvantage is that the CEP dependence of the spectral structure or cutoff frequency shift is very subtle, which makes it difficult to retrieve the exact CEP values. In this letter, we propose and demonstrate an efficient all optical method that enables one to evidently distinguish the CEP dependence of HHG.

Different from previous methods,[1,23,24] our technique relies on high harmonics generated from asymmetric molecules. Due to the additional freedoms and more complicated structures, the molecules provide a new route to steer the HHG, which has attracted more and more interest in recent years.[34–38] In this work, by simulating the HHG from the interaction of a diatomic asymmetric molecule with few-cycle pulse, we report that the harmonic spectrum exhibits more than two cutoffs. With the change of CEP, the cutoffs change their positions by as much as several tens of electronvolts in frequency and two or three orders in amplitude. Such a signifi-

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cant change represents an evident fingerprint of CEP, allowing the precise measurement of CEP.

2. Results and discussion

In this work, the HHG is simulated by numerically solving the time-dependent Schrödinger equation (TDSE). A linearly polarized Ti:sapphire laser pulse with wavelength of 800 nm is adopted. The electric field can be expressed by[5] $E(t) = E_0 \sin^2(t\pi/T_p)\cos[\omega(t - T_p/2) + \phi_0]$, where $0 \le t \le T_p$ and E_0, ω, T_p, ϕ_0 denote the amplitude, angular frequency, full pulse duration, and CEP, respectively. To meet the zero-net-force condition, which any real laser pulse satisfies, the total duration equals an integer number of optical cycles, i.e., $T_p = NT_0$ where N is an integer number and $T_0 = 2.7$ fs. We assume that the molecule is aligned along the laser polarization direction, which can be effectively realized with a pre-pump pulse.[39,40] Then, the electron dominantly moves in the laser polarization direction, and we adopt a one-dimensional model which has been widely used and produced quite reliable results in previous works.[35,41,42] The Hamiltonian is given by

$$H = -\frac{\partial^2}{2\partial x^2} + V(x) - E(t)x,$$

where $V(x)$ is the Coulomb potential which can be described by the soft-core potential[43,44]

$$V(x) = -Z_1/\sqrt{1 + (x + R/2)^2} - Z_2/\sqrt{1 + (x - R/2)^2},$$

where R is the internuclear distance, Z_1 and Z_2 are the electric charges of the two nuclei. Atomic units are used with $m = \hbar = -e = 1$ throughout this article unless otherwise indicated. In the calculation, $Z_1 = 2$, $Z_2 = 1$ and the internuclear distance $R = 3.9$ a.u. (atomic unit), which is consistent with the property of HeH^{++}.[45,46] Since the molecular vibrational period is much longer than the laser pulse duration, the nuclei are considered to be fixed at $\pm R/2$, respectively. For comparison, we also carry out the simulation for the He atom, where the Coulomb potential is expressed by $V(x) = -1/\sqrt{0.485 + x^2}$. The time-dependent Schrödinger equation is solved with the split-operator method.[47,48] The initial wavefunction is chosen to be the field-free ground state which is obtained by propagation in imaginary time. The ionization energy is calculated to be 47.6 eV for HeH^{++} and 24.5 eV for He. The dipole acceleration $a(t)$ is given by

$$a(t) = -\langle \Psi(x,t)|\frac{\partial V(x)}{\partial x}|\Psi(x,t)\rangle,$$

where $\Psi(x,t)$ is the wavefunction. The harmonic spectrum is calculated by Fourier transforming the dipole acceleration.

Figures 1(a) and 1(b) show the high harmonic spectra generated from the asymmetric molecule HeH^{++} in a few-cycle laser pulse with $\phi_0 = \pi/2$ and 0, respectively. The total pulse duration is $5T_0$, which corresponds to a duration of 5 fs defined by the full width of half maximum (FWHM) of laser intensity. The peak intensity is 6×10^{14} W/cm^2. For comparison, HHG for He is shown in Fig. 1(c) and 1(d) at the same conditions. One can see that, for the commonly used atom medium, discrete harmonics are generated at the cutoff ($\simeq 138$ eV) for $\phi_0 = \pi/2$. If the CEP is changed to 0, continuum harmonics are generated at the cutoff ($\simeq 140$ eV). However, the frequency shift of the cutoff is only 2 eV. The difference between other CEPs is even smaller. Consequently, it is difficult to retrieve the CEP values, particularly for the values between 0 and $\pi/2$, even though such spectral structure and frequency shift at the cutoff can be observed in experiment. In contrast, one can observe a pronounced difference in the HHG spectra of asymmetric molecules. As shown in Fig. 1(a), the harmonic spectrum shows two cutoffs at $\phi_0 = \pi/2$, the first one is located at 120 eV and the second one is located at 160 eV. The intensity of the second cutoff is about two orders of magnitude lower than that of the first one. As the CEP is changed to 0, three cutoffs can be observed in Fig. 1(b). The first cutoff shifts to 95 eV, the second cutoff shifts to 145 eV and an additional cutoff appears at 162 eV. The intensity of the second cutoff is about two orders of magnitude lower than that of the first one, but one order higher than the third one. The high contrast of cutoff intensities allows one to easily distinguish each cutoff. Moreover, the cutoff changes its position for more than 20 eV as CEP varying from 0 to $\pi/2$, which facilitates to precisely retrieve the CEP value.

To obtain deeper insight into the above multi-cutoff structure for asymmetric molecules, we analyze the electron trajectory in HHG, which can be well described by the ionization, propagation and recombination (IPR) three-step model.[49] The dashed line in Fig. 2(a) shows the return kinetic energies E_k of electron trajectories that start at different ionization times. The electric field is shown by the solid line in Fig. 2(a) for $\phi_0 = \pi/2$. One can see that ionization occurs at the peaks of the laser field, e.g., P_1, P_2, etc, from which the electron is freed and accelerated by the driving field. Finally the electron returns and recombines with the parent ion at $t = 2.5T_0$, $3T_0$, etc. As shown in Fig. 2(a), the maximum E_k for the trajectories P_1, P_2, and P_3 is about 111 eV, 110 eV, and 71 eV, respectively. According to the IPR three-step model, the electron releases its kinetic energy in the recombination process by emitting high harmonics with a photon energy of $I_p + E_k$, where I_p is the ionization energy of the target medium. Therefore, the cutoff energy of HHG for trajectories P_1, P_2, and P_3 is 160 eV, 158 eV, and 119 eV, respectively. In comparison with Fig. 1(a), the first cutoff at 120 eV originates from the trajectory P_3 and the second cutoff at 160 eV corresponds to the trajectories P_1 and P_2, whose maxima are too close to be distinguished from the spectrum. Note that these trajectories occur at a different half optical cycle of the laser field, we name these cutoffs as half optical cycle cutoffs (HOCs).[21] Similarly in the case of $\phi_0 = 0$, the HOCs at 95 eV, 145 eV, and 162 eV originate from the trajectories P_3, P_2, and P_1, respectively.

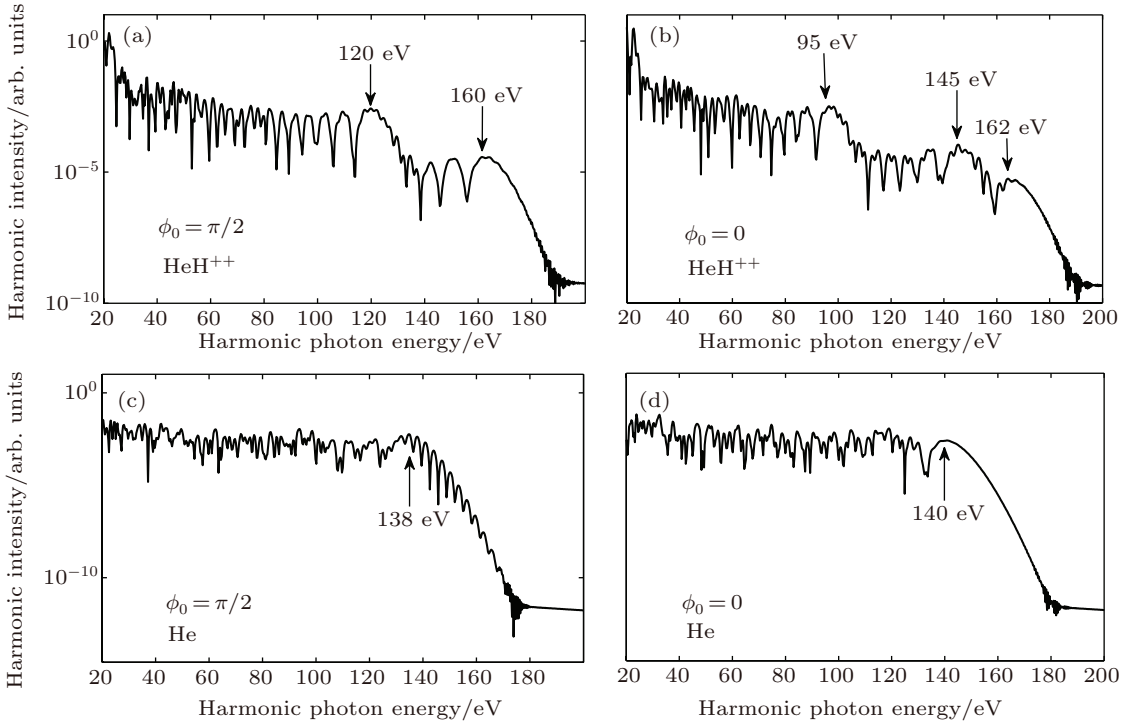


Fig. 1. High harmonic spectra for HeH^{++} [(a) $\phi_0 = \pi/2$, (b) $\phi_0 = 0$] and He [(c) $\phi_0 = \pi/2$, (d) $\phi_0 = 0$]. The laser intensity is 6×10^{14} W/cm^2, pulse duration is 5 fs.

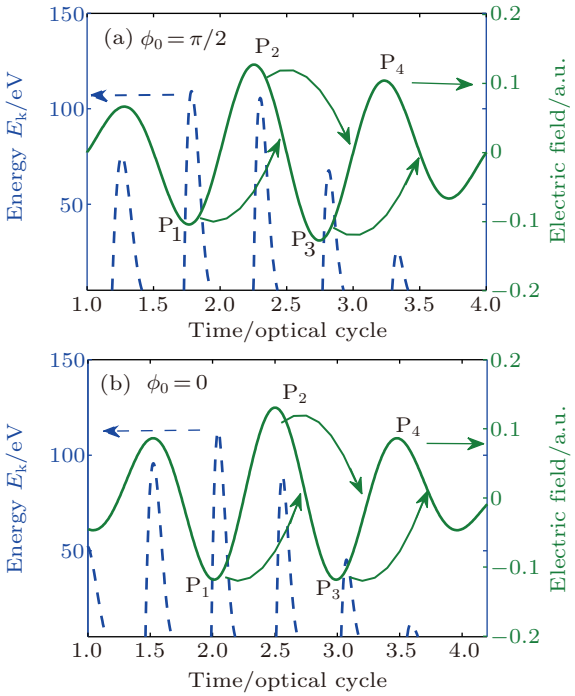


Fig. 2. (color online) Electric field (solid green line) and kinetic energy as a function of ionization time (dashed blue line). The CEP is (a) $\pi/2$ and (b) 0.

On the other hand, it should be noted that the HOC of asymmetric molecules changes its intensity for several orders of magnitude. This effect however can not be seen for He, which results in only one cutoff. This difference can be attributed to the influence of molecular structure to the ionization process of HHG. Figure 3 shows the Coulomb potential of asymmetric molecules in the dressing laser field, i.e., $V(x) - E_0 x$. The electric field is along the positive $\hat{x}$ (e.g., P_2) and negative $-\hat{x}$ directions (e.g., P_3) in Figs. 3(a) and 3(b), respectively. For asymmetric molecules, the electron is preferably located at the deeper well, i.e., the left core in Fig. 3. When the driving field is along the positive direction, as shown in Fig. 3(a), the left well potential is lifted up. Consequently, the electron can easily pass through the middle barrier and shift to the right core. However, because the right well potential is lowered by the driving field, the electron is hardly freed through directly tunneling the right well potential. Therefore, at P_2, the electron is likely captured by the right core and the ionization rate is significantly suppressed. In the subsequent half cycle (i.e., P_3), the electric field changes its sign. As shown in Fig. 3(b), the right well potential is lifted up, then the electron is easily freed, leading to an enhancement of ionization for one or two orders of magnitude at P_3. Consequently, as shown in Fig. 1(a), the intensity of HOC at 120 eV, which corresponds to the trajectory P_3, is about 100 times higher than that of the HOC at 160 eV. In contrast, for atom He, as the potential is reverse-symmetric in the neighboring half optical cycles, the ionization enhancement effect does not happen. Consequently, the HOCs of different trajectories are hardly distinguished in the spectrum and only one cutoff, which corresponds to the maximum kinetic energy of all the trajectories, is observed.

Moreover, as shown in Figs. 1(a) and 1(b), the first HOC at lower photon energy changes its position for more than 20 eV with the CEP varying from $\pi/2$ to 0. In contrast, the HOC with the highest photon energy just changes from 160 eV

to 162 eV, i.e., only 2 eV, which is similar to the HHG of an atom. This can be explained from the waveform of the laser field. As shown in Fig. 2, the HOC with the highest photon energy originates from the trajectory of P_1, for which the electron is ionized around P_1 and accelerated by the half-cycle around P_2 and recombines at around $2.5T_0$. As the CEP changes from 0 to $\pi/2$, the change of electric field amplitude in this half-cycle $|(E_{\phi_0=0}-E_{\phi_0=\pi/2})|/|(E_{\phi_0=0}+E_{\phi_0=\pi/2})|$ is only 1.1%. In contrast, for the trajectory P_3, which corresponds to the first HOC at lower photon energy, the high harmonics are generated at the tail of the laser field, where the electric field amplitude changes by about 13%, thus leading to a large frequency shift of HOC.

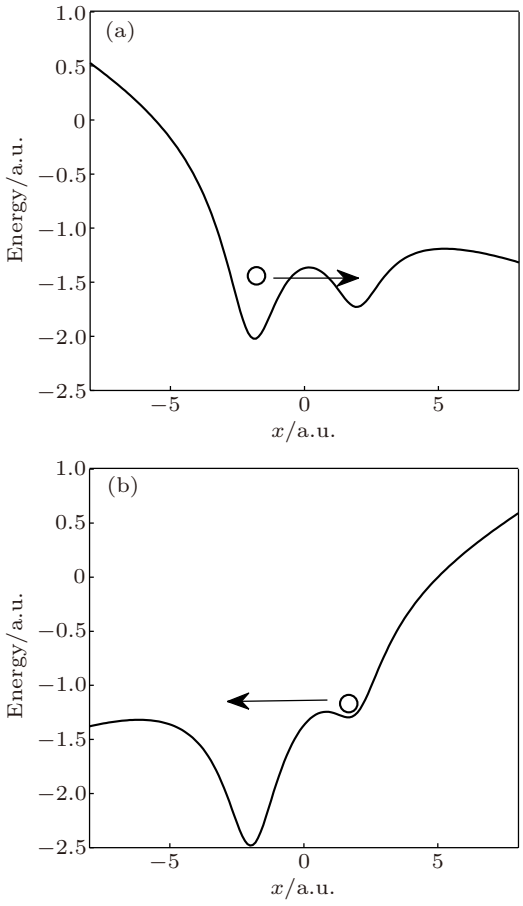


Fig. 3. Coulomb potential in the dressing laser field, i.e., $V(x)-E_0x$. The electric field is along the (a) $\hat{x}$ and (b) $-\hat{x}$ directions.

The large frequency shift of HOC facilitates the experimental measurement, and more importantly it represents an evident fingerprint of CEP. Figure 4 shows the high harmonic spectra generated from asymmetric molecules at different CEP values. One can see that the position of the most intense HOC varies from 95 eV to 155 eV as the CEP changes from 0 to 4.5 rad. Because of the competition between the trajectories P_2 and P_3, the most intense HOC jumps to 75 eV at $\phi_0 = 4.5$ rad and is gradually blue-shifted to 95 eV at $\phi_0 = 2\pi$. The CEP values are encoded in the frequency shift of HOC. To overcome the intricate simulation procedure for retrieving the CEP value, we consider to obtain an analytical formula. Following the above discussions and IPR three-step model, we can know that the kinetic energy of each trajectory in different half optical cycles is $3.17U_p(t_0)$ where $U_p(t_0)$ denotes the ponderomotive energy in the half optical cycle around t_0. Each HOC position can be expressed by $I_p+3.17U_p(t_0)$, which is rewritten as

$$I_p+3.17\frac{\{E_0\cos^2[(2\pi-\phi_0)/2N]\}^2}{4\omega_0^2}, \quad (1)$$

$$I_p+3.17\frac{\{E_0\cos^2[(4\pi-\phi_0)/2N]\}^2}{4\omega_0^2}. \quad (2)$$

The solid line and dashed line in Fig. 4 present the simulation results by using Eqs. (1) and (2), respectively. One can see that the simple formulas Eqs. (1) and (2) reproduce the HOC position quite well. Therefore, one can easily retrieve the CEP values without any intricate simulation by solving the TDSE. On the contrary, for the previous CEP measurement with photonionization,[5] intricate simulation procedures are required, which is very time consuming and sometimes gives different CEP values using different theoretical models. In addition, since the average ratio between the HOC position shift and the CEP is about 13 eV/rad, the CEP can be measured with an accuracy of 0.03 rad.[33]

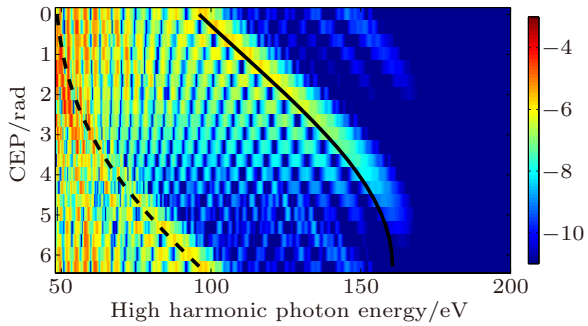


Fig. 4. (color online) High harmonic spectra as a function of CEP and photon energy. The solid and dashed lines show the simulations with Eqs. (1) and (2). The laser parameters are the same as those in Fig. 1.

It is well known macroscopic factors,[46] such as phase matching, and intra-atomic phase[50] may influence HHG intensely. However, the position and structure of cutoff may not be changed after propagation when the gas medium is not too long and dense.[51,52] In addition, since the harmonic in cutoff is only emitted once during the whole laser pulse, the intra-atomic phase has little influence on the harmonic in cutoff. To identify it, we perform the nonadiabatic three-dimensional propagation simulations for both fundamental and harmonic fields in the gas target.[46] Figure 5 shows the on-axis macroscopic high harmonic spectrum generated from a 0.5-mm-long gas jet. The laser pulse is assumed as a Gaussian beam and is focused 2 mm before the entrance of the gas jet. The beam waist at focus is 25 μm. The density of the gas jet is isotropic and equal to 2.5×10^{18} cm^{-3}. Other laser parameters are the same as these in Fig. 1(a). For comparison, the single-molecule high harmonic spectrum is also presented in Fig. 5.

After propagation through the gas jet, the positions and structures of the HOCs all are unchanged. It shows the CEP measurement from HOCs is a sound method.

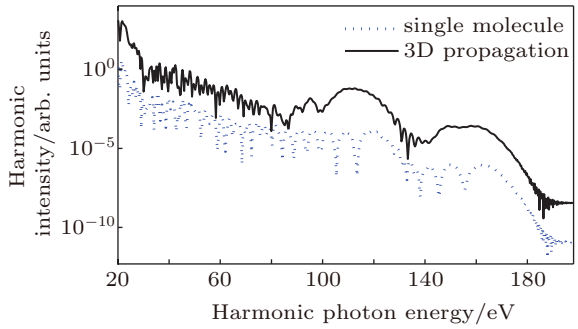


Fig. 5. (color online) The macroscopic (solid line) and single-molecule (dotted line) high harmonic spectra.

3. Conclusion

The HHG of asymmetric molecules is investigated in the few-cycle regime. The asymmetric molecular structure strongly modulates HHG and several HOCs are clearly observed from the harmonic spectra. By changing the CEP, the HOCs change their positions for several tens of electronvolts in frequency and two or three orders in amplitude. Moreover, we have obtained an analytical formula, which well predicts the position of HOC and enables one to precisely measure the CEP value in a very simple way. Since our scheme is an all optical method, we believe that this method will facilitate the experiments.

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