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Intensity-dependent asymmetric photoionization in few-cycle laser pulses*

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The asymmetric photoionization of atoms irradiated by intense, few-cycle laser pulses is studied numerically. The results show that the pulse intensity affects the asymmetric photoionization in three aspects. First, at higher intensities, the asymmetry becomes distinctive for few-cycle pulses of longer durations. Second, as the laser intensity increases, the maximal asymmetry first decreases then increases after it has reached a minimal value. Last, the value of the carrier-envelope phase corresponding to the maximal asymmetry varies with the pulse intensity. This study reveals that the increasing of pulse intensity is helpful for observing the asymmetric photoionization.

Keywords: asymmetric photoionization, few-cycle laser pulses
PACC: 3280K, 3280F, 3280P

High-power ultrashort pulses with durations as short as only a few optical cycles are widely used in many fields.[1−7] The multi-cycle pulses can be characterized by frequency, intensity and polarization, while for few-cycle laser pulses, an additional parameter termed the carrier -envelope (CE) phase, which is the initial phase of the carrier wave for the pulse envelope, is required to specify a few-cycle pulse. The time variation of the electric field of laser pulses depends on the CE phase, and so do the physical processes induced by laser pulses.

In photoionization of atoms irradiated by few-cycle pulses, considerable variation of the field envelope during one cycle and the nonlinear response of the atom may lead to an asymmetry in photoelectron angular distributions. The numbers of ionized electrons emitted in a pair of opposite directions along the polarized direction are not always the same, which is termed inversion asymmetry. The inversion asymmetry has attracted much attention, both experimentally and theoretically.[8−17] Because the inversion asymmetry varies with the CE phase, the measurement of the asymmetric photoionization can manifest directly the CE phase of a short pulse.[13,18−20]

To quantitatively study the inversion asymmetry, an asymmetry parameter is introduced[15,16] and defined as

$$a = \frac{P_+ - P_-}{P_+ + P_-}, \tag{1}$$

where P_- and P_- are the energy-integrated partial ionization probabilities in a pair of opposite directions. The value of parameter a is zero for the case of symmetric ionization and reaches its extremum when the ionization occurs completely on one side.[15] Thus this parameter is referred to as asymmetry degree. The experimentally observed asymmetry in a radio-frequency domain is far larger than that in an infrared domain.[13,16]

In above-threshold ionization (ATI), the photoelectron probability depends nonlinearly on instantaneous strength of the electric-field, correspondingly, the asymmetry degree varies with CE phase, peak intensity and pulse duration of the few-cycle pulse. In Ref.[16], a distinctive asymmetry was demonstrated for the pulses maintained for up to eight cycles at a higher intensity, while the asymmetry decreases quickly with the increase of pulse duration at a lower intensity. The numerical study based on

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three-dimensional time-dependent Schrodinger equation (TDSE), performed in Ref.[15], indicated that the asymmetry varied distinctively with laser intensity when laser intensity $I > 10^{14}\,\mathrm{W/cm^2}$.

In this paper we perform a theoretical study on the ATI of hydrogen atoms in linearly polarized few-cycle laser pulses. We adopt the one-dimensional TDSE method, since it can disclose all the physical essences of photoionization. The ATI probabilities along the polarization vector are calculated separately, thereby we study the CE phase-dependent asymmetry. We focus our attention on the variation of the asymmetry with pulse intensity. Our study reveals that as the laser intensity increases, the maximal asymmetry first decreases then increases after it has reached a minimal value, and the corresponding value of the CE phase also changes. Although some results were also shown in Ref.[15], we seek for a physical insight into those phenomena in this paper. Furthermore, we find that the value of the CE phase corresponding to the maximal asymmetry also varies with the pulse duration and the ionization potential of the target atom. In the following, we first discuss the calculated results then seek for physical essences behind them.

The research is based on the numerical solution of TDSE for an atom interacting with an intense, linearly polarized few-cycle laser pulse.[12] The electric field $E(t)$ of an n-cycle pulse is determined by (throughout this paper, the atomic units (a.u.) are used)

$$A(t) = -\frac{E_0}{\omega}\cos^2(\pi t/T_\mathrm{p})\sin(\omega t+\varphi),$$
$$E(t) = -\mathrm{d}A(t)/\mathrm{d}t, \quad 0 \le t \le T_\mathrm{p}, \tag{2}$$

where φ is the CE phase, the pulse duration T_p is defined as $T_\mathrm{p} = 2n\pi/\omega$, which differs a little from the half width at half maximum (FWHM). The total ionization probability is calculated as the absorption in the calculation boundaries, and the absorption at one boundary is used to represent the partial ionization probability in the corresponding direction. The asymmetry in photoionization reflects the asymmetric electric-field distribution of laser pulses. The variation of the asymmetry degree with the CE phase depends on the pulse intensity and the pulse duration.

The asymmetry degree varies with CE phase in a sine-like pattern with a period of 2π. Here the variation of sine-like pattern means the regular oscillation of asymmetry degree from its positive maximum to its negative maximum as the CE phase changes. The variation pattern is determined by the pulse envelope and accords with the recent experimental observation.[13] The 2π-modulation pattern is a direct sequence of the electric field since $E(\varphi) = E(\varphi + 2\pi)$.

The asymmetry degree varies with pulse duration, and its maximum decreases when the pulse duration increases as shown in Fig.1 for $E_0 = 0.12\,\mathrm{a.u.}$. The maximal values of asymmetry are about 35%, 20% and 10% in four, five and six-cycle pulses, respectively. A similar change appears for other intensities. Since the inversion asymmetry reflects the asymmetric distribution of the electric field, and the electric field in few-cycle pulses with a shorter duration varies more dramatically than that in few-cycle pulses with a longer duration. Therefore, the asymmetry naturally becomes more prominent in shorter few-cycle pulses. Meanwhile, we note that the CE phase corresponding to the maximal asymmetry varies with pulse duration. For example, the asymmetry degree reaches its maximum at $\varphi = 1.1\pi$ for $n = 6$ but at $\varphi = 0.05\pi$ for $n = 4$. Owing to the sine-like pattern, we term this change a phase shift.

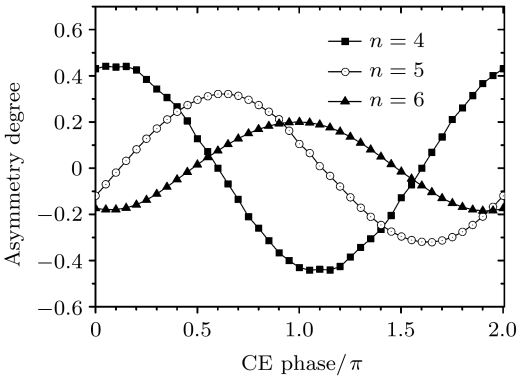


Fig.1. Variations of the asymmetry degree with CE phase in laser pulse with different pulse lengths for peak intensity $I = 5.5 \times 10^{14}\,\mathrm{W/cm^2}$ ($E_0 = 0.12\,\mathrm{a.u.}$).

The laser intensity affects the asymmetry in three aspects. First, for few-cycle pulse with a relatively long duration, the asymmetry becomes notable at higher intensities. Take the asymmetry degree in eight-cycle pulses (with FWHM about 7.8 fs) as an example. The asymmetry is not appreciable if the pulse intensity is lower than 0.1 a.u.; as the pulse intensity increases, the asymmetry becomes more and more clear. The maximal asymmetry is less than 5% for $E_0 = 0.12\,\mathrm{a.u.}$, which is still hard to detect experimentally. The maximal asymmetry is about 7% for $E_0 = 0.15\,\mathrm{a.u}$, and it is larger than 20% for $E_0 = 0.2\,\mathrm{a.u.}$ which is easy to detect experimentally. Second, with pulse intensity increasing, the

maximal asymmetry degree first decreases then increases as shown in Fig.2 for four-cycle pulses (with FWHM 3.9 fs). The maximal asymmetry degree is larger than 50% for $E_0 = 0.05$ a.u. and about 40% for $E_0 = 0.07$ a.u.; the maximal asymmetry reaches a minimum about 30% for $E_0 = 0.09$ a.u.; for further increasing pulse intensities, the maximal asymmetry increases. The asymmetry degree reaches a maximum of about 45% for $E_0 = 0.12$ a.u., and larger than 60% for $E_0 = 0.15$ a.u.. Finally, we find a phase shift when the pulse intensity changes. For example, the value of the CE phase corresponding to the maximal asymmetry is not fixed. The maximal asymmetry is reached at $\varphi = 1.3\pi$ for $E_0 = 0.05$ a.u. but at $\varphi = 0.45\pi$ for $E_0 = 0.15$ a.u..

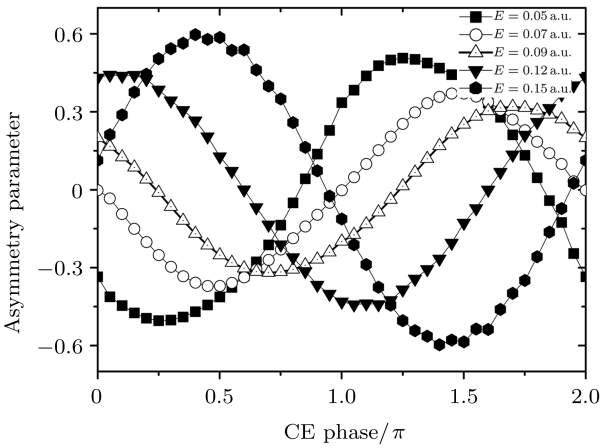


Fig.2. Comparison among the variations of asymmetry degree with CE phase in four-cycle pulses at several pulse intensities.

The phase shift induced by the pulse intensity was also found in Ref.[15]. It was shown that the CE phase corresponding to the zero-asymmetry varied greatly when the pulse intensity $I > 10^{14}$ W/cm^2. But the phase shift induced by the pulse duration was not observed in Ref.[15]. Moreover, we find that the phase shift may also be induced due to the variation of the atomic ionization potential as depicted in Fig.3. By changing these parameters in a special manner, the asymmetry degrees show the same variations, thus a scaling law of photoionization in few-cycle pulses is established.[12]

The nonlinear dependence of ionization on laser intensity determines the variation of the maximal asymmetry with pulse intensity and pulse duration. It is known that the ATI rate increases significantly when the laser intensity exceeds a threshold value. When the pulse intensity is low, the ionization occurs mainly in a pulse peak regime. The ionization probability in the strongest half-cycle is far larger than those in the sub-strongest half-cycles (the half-cycles that

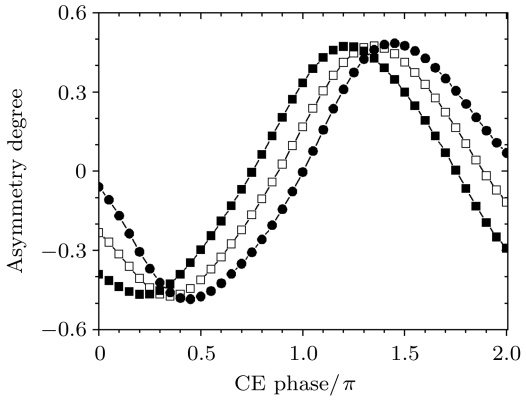


Fig.3. Variations of the asymmetry degree in three different binding potentials: 0.75 a.u. (solid circles), 0.80 a.u. (open circles), and 0.85 a.u. (solid squares), with $E_0 = 0.15$ a.u. and $n = 4$.

neighbour with the strongest one), thereby leading the ionization to occur mainly in one direction. According to expression (1), the asymmetry degree approaches to unity, which means that a larger asymmetry is reached at lower laser intensities. As the pulse intensity increases, the strengths of the sub-strongest half-cycles may finally exceed the threshold value, then the ionization probabilities in these half-cycles become notable. Although the ionization probability in the strongest half-cycle also increases, notable ionization occurs in both directions along the polarization vector and the correspondingly asymmetry degree becomes smaller and smaller with pulse intensity increasing. On the other hand, as the pulse intensity increases, the ionization on the leading edge of the pulse contributes more and more to the overall ionization probability and will finally become larger than 50% at an intensity. For further high intensity case, the ionization process occurs mainly on the leading edge of laser pulse, i.e. before the electric field in laser pulse reaching its maximum. Because the electric field on the leading edge varies more dramatically than that on the top of pulse envelope, the asymmetry degree reaches a larger maximum. As a result, the maximum of the asymmetry degree first decreases then increases as the laser intensity increases.

In order to verify the above conjecture, we calculate the time-resolved instantaneous ionization probabilities in four-cycle laser pulses of different intensities. Several numerical results are shown in Fig.4, from which we can clearly see how the principal ion-

ization moves to the leading edge of the laser pulse as the laser intensity increases. Figure 4(a) depicts the time-resolved instantaneous ionization probabilities for $E_0 = 0.05$ a.u.. The ionization probability on the right side, labelled as $P_+(t)$, shows two peaks and can be identified as the ionization in the strongest half-cycle and the following half-cycle in the same direction. The ionization probability on the left side, say $P_-(t)$, shows several peaks and the first two peaks can be identified as the ionization in the two sub-strongest half-cycles. Figure 4(b) depicts the time variations of $P_+(t)$ and $P_-(t)$ for $E_0 = 0.09$ a.u.. In addition to a large increase in ionization probability, the most notable difference between Fig.4(b) and Fig.4(a) is that the ionization in the first sub-strongest half-cycle contributes mainly to $P_-(t)$. Moreover, the difference between $P_+(t)$ and $P_-(t)$ decreases and the corresponding asymmetry degree is small. Figure 4(c) depicts $P_+(t)$ and $P_-(t)$ as functions of time for $E_0 = 0.15$ a.u.. It is clear that $P_+(t)$ comes mainly from the second half-cycle on the leading edge of the laser pulse and the ionization probability on the left side is larger than that on the right side, thus the corresponding asymmetry turns into that shown in Fig.4(a) (both for $\varphi = 0$). The asymmetry degree

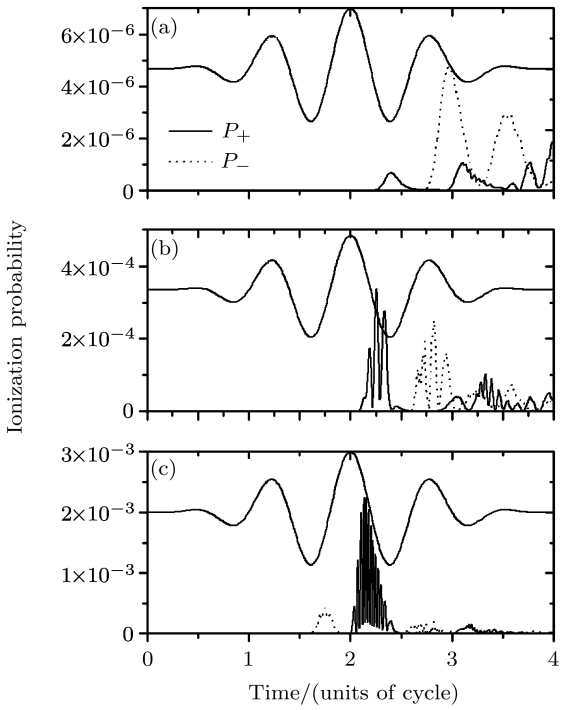


Fig.4. Time-resolved instantaneous ionization probabilities in four-cycle pulses with different peak intensities, where the CE phase is fixed at 0 and the dot lines show the time-variation of the electric field. (a) $E = 0.05$ a.u. (b) $E = 0.09$ a.u. (c) $E = 0.15$ a.u.

will be zero at intensities ranging from 0.09 a.u. to 0.15 a.u., but this does not mean that the maximal asymmetry is zero, since the maximal asymmetry is reached for another CE phase at that intensity.

The phase shift comes from the nonlinear feature of photoionization, and it appears together with the change of pulse intensity, pulse duration, and atomic ionization potential. From static viewpoint, the motion of an atomic electron in a laser field is governed by two forces: one is the Coulomb attraction of its parent core, and the other is the electric force of the laser field. Whether an atom can be ionized is dependent on whether the electric force can overcome the Coulomb attraction between the electron and the nucleus. An identical difference between the two forces can be reached for different CE phases when the pulse intensity is changed. Correspondingly, the same asymmetry degree corresponds to different CE phases when the pulse intensity changes. Thus, the phase shift appears when the pulse intensity changes. The variation of the electric field in the few-cycle pulses depends on pulse duration. For the same CE phase and the same peak intensity, the electric-field strength varies with pulse duration, so does the electric force of the pulse acting on the electron. This implies that for equal electric forces, the corresponding CE phases are not the same when the pulse duration changes. Thus, the phase shift appears when the pulse duration changes. The Coulomb force of the parent ion determines the ionization potential. Different ionization potentials indicate different Coulomb attractions of the parent core to the atomic electron. Thus, when the ionization potential changes, the phase shift also appears.

In summary, the laser intensity affects the asymmetric photoionization in three aspects. First, for longer few-cycle pulses, the asymmetry becomes distinctive at higher intensities, but not at lower intensities. Second, for fixed pulse durations, the maximal asymmetry first decreases then increases along with the increase of pulse intensity. Considering the dependence of the photoionization probability on laser intensity, we conclude that the adopting of higher pulse intensity is useful for showing and detecting the CE phase-dependence. Last, the change of the pulse intensity induces a phase shift, which stems from the nonlinear feature of photoionization and can also be induced by the pulse duration and the atomic ionization potential.

So far the phase shift has not been observed ex-

perimentally. In a recent experiment,[21] the modulation of the ionization yield in the CE phase in few-cycle pulses has been detected. The value of a relative phase corresponding to the maximal ionization yield is about 1.75π for the pulse of duration 5.81 fs, but it becomes 0.4π for pulse of duration 5.09 fs. Here the relative phase differs from the CE phase by a common phase offset. If the laser pulses, with the phase offset fixed, are used, the phase shift can be demonstrated experimentally.

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