

# Measurement of the carrier-envelope phase of few-cycle laser pulses by use of asymmetric photoionization

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Using numerical solutions of the time-dependent Schrödinger equation for a hydrogen and a helium atom in a linearly polarized, few-cycle laser field, we calculate the photoelectron left–right asymmetry measured by two opposing detectors placed along the laser polarization vector, with the laser focus in the center. We find a simple dependence of this asymmetry on carrier-envelope (CE) phase  $\phi$  for laser intensities slightly below the tunneling regime, which may allow us to measure (or to calibrate) and to stabilize the CE phase. In particular, we suggest that the condition of zero asymmetry for few-cycle pulses may be useful for both these goals. © 2004 Optical Society of America

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High-power ultrashort laser pulses with durations as short as a few optical cycles are now available as research tools.<sup>1,2</sup> Although long monochromatic pulses are completely characterized by their polarization, the frequency, and the temporal shape of their envelope, short pulses require at least one additional parameter, since the electric field envelope of a few-cycle pulse varies significantly during one cycle. Typically the temporal shape of the laser electric field is represented as a product of a Gaussian-like envelope times a trigonometric function. The phase of this function becomes a physically important parameter for pulses shorter than 4 cycles and is called the carrier-envelope (CE) phase  $\phi$  or the absolute phase. Recently, significant progress was achieved in stabilizing the CE phase.<sup>2</sup> When such a few-cycle laser pulse interacts with atomic gas, its CE phase will significantly affect various physical processes,<sup>1</sup> but it leaves a particular simple signature in angular distributions of photoionized electrons. Both linearly<sup>3–7</sup> and circularly<sup>8</sup> polarized laser pulses lead to strong left–right asymmetries of photoelectrons, which are sensitive to the value of the CE phase. The effects of CE phase, induced by phase-stabilized few-cycle laser pulses, were also recently seen in the photoelectron signal from a metal surface illuminated by phase-stabilized few-cycle laser pulses<sup>9</sup> as well as in photocurrents in semiconductors, through interference between one- and two-photon absorption paths,<sup>10</sup> which is possible because of the huge bandwidth of few-cycle pulses. Clearly, the CE phase has become an important new laser parameter that shows up in various laser-induced phenomena.

For long monochromatic laser pulses the CE phase of the laser pulse does not induce any measurable effect: For example, angular photoelectron distributions  $f(\theta)$  are symmetric; i.e.,  $f(\theta) = f(\pi - \theta)$  (where  $\theta$  is the angle between the photoelectron momentum and the laser polarization vector) because of the symmetry of the monochromatic electric field and of the atom. Considerable variation of the field envelope during one cycle and the nonlinear response of an

atom lead to an asymmetry in photoelectron angular distributions, which can be used as a measurable signature of the absolute phase of an ultrashort laser pulse. Our previous numerical calculations<sup>3,4</sup> based on the time-dependent Schrödinger equation (TDSE) have shown that the ratio  $P_+/P_-$ , where  $P_+$  and  $P_-$  are photoelectron signals (from a hydrogen atom) measured by two opposing detectors placed along the laser polarization (with the laser focus in the center), is sensitive to the CE phase of a few-cycle laser pulse. In this Letter we investigate the dependence of these asymmetries on laser intensity, central wavelength, pulse duration, and the atom's ionization potential in view of possible calibration of the CE phase as well as the use of asymmetries as a tool for CE phase stabilization. In particular, we show that the dependence of asymmetry on CE phase becomes particularly simple in the subtunneling regime, i.e., for Keldysh parameter value  $\gamma = (I_p/2U_p)^{1/2} \approx 1.5$ , where  $I_p$  is the atom's ionization potential and  $U_p = (eE_0)^2/(4m_e\omega^2)$  is the ponderomotive energy of the electron in the laser of amplitude  $E_0$  [the tunneling regime is defined via inequalities  $\gamma < 1$ ,  $\omega \ll I_p$  (Ref. 1)].

We solve numerically the three-dimensional TDSE (using spherical coordinates  $r$ ,  $\theta$ ) that describes the interaction of a hydrogen or a helium atom (single active electron approximation) with a linearly polarized laser field  $E(t)$  along the  $z$  axis.<sup>3,11,12</sup> We use well-established numerical methods<sup>11,12</sup> and electric field  $E(t)$  defined via the vector potential, as in Ref. 3 except that here we use a Gaussian envelope instead of a  $\sin^2$  envelope. Figure 1 of Ref. 3 illustrates our definition of the absolute phase  $\phi$ : for  $\phi = 0$  the maxima of field and envelope coincide. The photoelectron signal measured by two opposing detectors  $P_{\pm}$  is obtained by integrating the radial component of probability current  $j_r(\theta, t)$  (Refs. 3 and 12) over time and over the detector angle  $\theta < \theta_0$  and  $180^\circ - \theta < \theta_0$ , where we use  $\theta_0 = 15^\circ$ .  $j_r(\theta, t)$  was calculated near the absorbing boundaries at  $r = 128$  bohr. Next we calculated the normalized asymmetry coefficient  $a = (P_+ - P_-)/(P_+ + P_-)$ , which is a convenient measure of the asymmetry

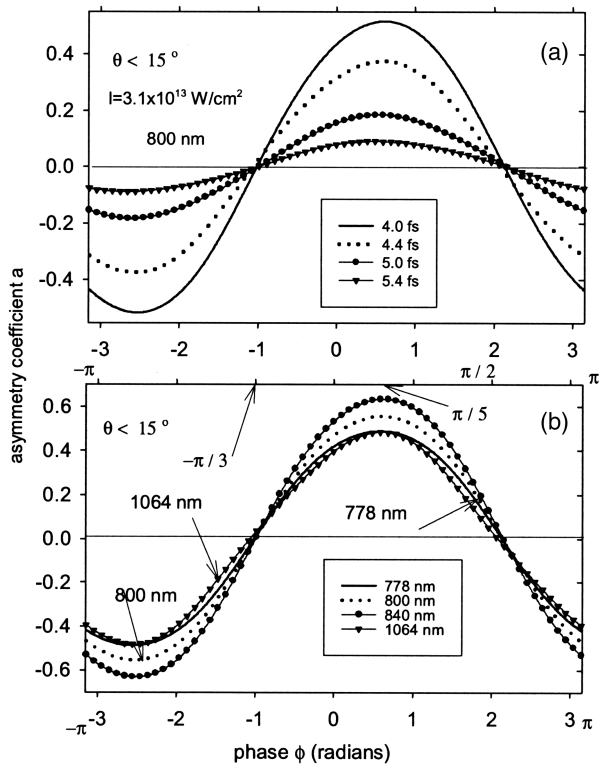


Fig. 1. Asymmetry coefficient  $a$  as a function of the CE phase for laser intensity  $I = 3.1 \times 10^{13} \text{ W/cm}^2$  and  $\lambda = 800 \text{ nm}$  for (a) various pulse durations ranging from  $\tau_p = 4.0 \text{ fs}$  to  $\tau_p = 5.4 \text{ fs}$  and (b) four central wavelengths  $\lambda$ : 778 nm ( $I = 3.7 \times 10^{13} \text{ W/cm}^2$ ), 800 nm, 840 nm ( $2.6 \times 10^{13} \text{ W/cm}^2$ ), and 1064 nm.  $\tau_p = 3.9 \text{ fs}$  was used for all central wavelengths except  $\lambda = 1064 \text{ nm}$ , for which  $\tau_p = 5.2 \text{ fs}$  was used to compensate for a longer period.

since  $a = 0$  for long many-cycle pulses. We show in Fig. 1(a) the asymmetries as a function of CE phase  $\phi$  at intensity  $I = 3.1 \times 10^{13} \text{ W/cm}^2$  and for the pulse duration (FWHM)  $\tau_p = 4 \text{ fs}$ . Surprisingly, we get simple sinelike curves for functions that are precisely fitted by a function  $\text{const} \times \sin(\phi - \phi_0)$ , where  $\phi_0 = -0.32 \approx -\pi/3$ . Thus asymmetry is zero for phases  $\phi_0 = -0.32\pi, \phi_0 \pm \pi, \dots$  etc. and is maximum at  $\phi = \phi_M = 0.19\pi \pm \pi \dots$  etc. Remarkably, at the chosen intensity (which is the intensity at which asymmetry is maximal) similar simple functions  $a(\phi)$  occur for various pulse durations,  $4 \text{ fs} < \tau_p < 5.4 \text{ fs}$  [Fig. 1(a)], and wavelengths as seen in Fig. 1(b), which shows the coefficient  $a$  for various central wavelengths: 778, 800, 840, and 1064 nm. For each wavelength the pulse duration was  $\tau_p = 1.5$  cycles (FWHM). We note that similar asymmetries occur for all these wavelengths, which proves that the effect seen for 800 nm is not related to accidental resonances in the hydrogen atom as might have been the case for this particular wavelength.

However, laser intensity variations have more effect in this simple pattern (see Fig. 2). We note that a simple sinelike function can be fitted only for intensities in the interval  $3 \times 10^{13} - 10^{14} \text{ W/cm}^2$  [see Fig. 2(a)], which corresponds to Keldysh parameter values  $1 < \gamma < 2$ , whereas at higher intensities significant changes occur. The asymmetry  $a$  obtained after

averaging of  $P_{\pm}$  over the distribution of peak intensities in the focus by use of maximum peak intensity  $10^{14} \text{ W/cm}^2$  is shown in Fig. 2(b). Clearly, averaging slightly increases asymmetry, since the asymmetries are larger at lower intensities, without significantly modifying the sinelike shape of asymmetries as a function of  $\phi$ . Using Fig. 2, for each laser intensity we define as  $\phi_0$  the phase at which the asymmetry is zero, and we show in Fig. 3 the dependence of  $\phi_0$  on laser intensity. We note that there exists an intensity range in which  $\phi_0$  depends little on the laser intensity. This means that, if the laser intensity is controlled well within the interval  $4 \times 10^{13} - 10^{14} \text{ W/cm}^2$  and the pulse duration is  $\tau_p$  is below 5 fs, the zero-asymmetry CE phase is nearly constant and is equal (on average) to  $-0.30\pi$  ( $-53.5^\circ$ ), displayed as a horizontal line in Fig. 3, with a precision of  $\pm 0.02\pi$  ( $3.8^\circ$ ). This stability of the zero-asymmetry CE phase  $\phi_0$  can also be experimentally useful for stabilizing the phase by just checking that the few-cycle pulse does not induce any asymmetry in the photoelectron signal. Thus one would select out pulses with phase  $\phi = \phi_0$ , using just the above criterion of zero asymmetry for each laser shot. Note that this stability of zero-asymmetry CE phase is lost at higher intensities, since  $\phi_0(I)$  increases abruptly at  $I > 10^{14} \text{ W/cm}^2$ .  $\phi_0$  becomes zero in the tunneling regime ( $\gamma < 1$ );  $I \approx 2 \times 10^{14} \text{ W/cm}^2$  and is equal to  $\pi/2$  at  $4 \times 10^{14}$ , i.e., in the barrier-suppression ionization regime. These properties of the zero-asymmetry phase can also be useful for

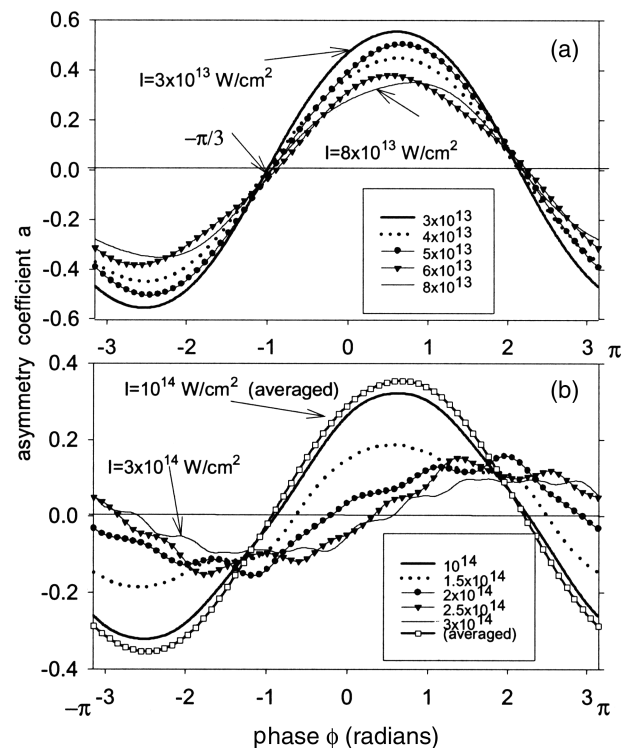


Fig. 2. Asymmetry coefficient  $a$  as a function of the CE phase for  $\lambda = 800 \text{ nm}$  and  $\tau_p = 3.9 \text{ fs}$  for various intensities ranging from  $I = 3.1 \times 10^{13}$  to  $3 \times 10^{14} \text{ W/cm}^2$ . The curve with squares shows the asymmetry  $a$  obtained after averaging of  $P_{\pm}$  over the distribution of peak intensities in the focus with maximum peak intensity  $10^{14} \text{ W/cm}^2$ .

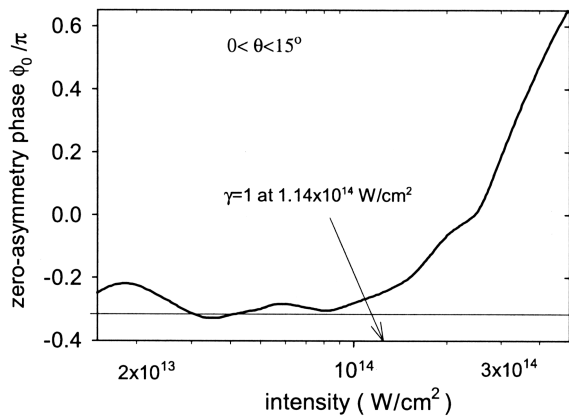


Fig. 3. Zero-asymmetry CE phase  $\phi_0$  as a function of laser intensity.

determining the CE phase of the pulse from experiments in which asymmetry is plotted as a function of thickness  $\Delta x$  of a material that is used as a phase shifter, as in a recent experiment<sup>6</sup> in which a simple classical model was used to find the relation between the CE phase and  $\Delta x$  (the thickness of the dispersive material). Note that Fig. 3 of Ref. 6 shows the experimental dependence on  $\Delta x$ , which is similar to that in our Fig. 2(a). However, to use our results directly for finding the relation between  $\Delta x$  and the absolute phase would require that this experiment be performed at several slightly lower intensities (the experiment was done at  $I = 10^{14}$  W/cm<sup>2</sup> with xenon, i.e., very close to the tunneling regime) to be sure that the regime of stable zero asymmetry, such as the plateau in Fig. 3, was indeed found. Then one can find the relation between  $\Delta x$  and  $\phi$  just by comparing experimental plot  $a(\Delta x)$  with theoretical  $a(\phi)$  shown in Fig. 2. This may be a more practical method of finding the relation between  $\Delta x$  and  $\phi$  than the method used in Ref. 6, which was based on asymmetry of fast electrons only, since their signal is many orders of magnitude weaker than the total photoelectron signal.

We also studied the time dependence of the asymmetry for a larger pulse duration. As in the case of function  $a(\phi)$ , we observed that dependence on pulse duration  $\tau_p$  is more regular at lower intensities,  $I = 3.1 \times 10^{13}$  W/cm<sup>2</sup>, than at  $I = 8 \times 10^{13}$  W/cm<sup>2</sup>, where the asymmetry persists for longer  $\tau_p$  and exhibits oscillatory behavior.<sup>12</sup> More detailed studies of these effects and the results for a helium atom as well as total ionization probabilities can be found in our longer paper,<sup>12</sup> in which we show that for a helium atom similar sinelike dependence on  $\phi$  occurs [as in Fig. 2(a)], again in the subtunneling regime for the value of parameter  $\gamma \approx 1.5$ , i.e., in the intensity range  $I = 8 \times 10^{13} - 2 \times 10^{14}$  W/cm<sup>2</sup>, for  $\lambda = 800$  nm.

We have shown that at intensities corresponding to the regime between the multiphoton-perturbative and the tunneling regimes ( $1 < \gamma < 2$ ) the asymmetries exhibit stable simple patterns. These asymmetries originate from Coulomb attraction correction to the tunneling<sup>4</sup> or to the strong field approximation models.<sup>5</sup> The zero of asymmetry coefficient

$a(\phi)$  depends little on intensity in the interval  $3 \times 10^{13} < I < 10^{14}$  W/cm<sup>2</sup>. It also depends little on various laser pulse parameters such as pulse duration, wavelength, detector angle  $\theta_0$  (Ref. 12) and on the atom's ionization potential  $I_p$ . We believe that the above stability of the zero-asymmetry phase may be useful in applications, e.g., for calibration of the CE phase value from measured asymmetry as a function of thickness of a phase shifter,<sup>6</sup> as well as for stabilizing the laser CE phase (when the source's phase fluctuates from shot to shot) by isolating laser shots that exhibit no asymmetry. Thus the pulses selected using the above criterion should have a fixed value of CE phase close to  $0.31\pi$  with high precision. Our study shows that higher intensities (at which  $\gamma < 1$ ) are much less convenient for the measurement of the CE phase because of strong dependence on laser intensity and irregular dependence on pulse duration and because of the smaller value of the asymmetry coefficient than at lower intensities. Therefore we suggest measurements of asymmetry coefficient  $a$  as a function of the laser intensity to determine experimentally this useful intensity regime.

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