Measurement of the Phase of Few-Cycle Laser Pulses

G. G. Paulus,[1,2,3] F. Lindner,[2] H. Walther,[2,3] A. Baltuška,[4] E. Goulielmakis,[4] M. Lezius,[4,5] and F. Krausz[2,4]

[1]*Department of Physics, Texas A&M University, College Station, Texas 77843-4242, USA*
[2]*Max-Planck-Institut für Quantenoptik, 85748 Garching, Germany*
[3]*Ludwig-Maximilians-Universität München, 85748 Garching, Germany*
[4]*Institut für Photonik, Technische Universität Wien, Gusshausstrasse 27, A-1040 Wien, Austria*
[5]*Institut für Ionenphysik, Universität Innsbruck, Technikerstrasse 25, A-6020 Innsbruck, Austria*

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For the shortest pulses generated to date, the amplitude of the electromagnetic wave changes almost as rapidly as the field oscillates. The temporal variation of the field, which directly governs strong-field interactions, therefore depends on whether the maximum of the pulse amplitude coincides with that of the wave cycle or not, i.e., on the phase of the field with respect to the pulse envelope. It is demonstrated that the direction of electron emission from photoionized atoms can be controlled by varying the phase of the field, providing for the first time a tool for its accurate determination. Directing fast electron emission to the right or to the left with the light phase constitutes a new kind of coherent control.

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Generation of laser pulses with a pulse energy around 500 μJ and duration of 5 fs (measured at full width at half maximum) has become routine in past few years [1,2]. One of the most intriguing features of such pulses is that their envelope varies almost as fast as the electromagnetic field itself. Considering a typical wavelength of 760 nm, an optical cycle has a duration of 2.5 fs; i.e., the pulses consist of very few optical cycles only (few-cycle pulses). One of the most momentous consequences is that the time variation of the laser field will depend on the phase of the carrier frequency with respect to the envelope, the so-called carrier-envelope (CE) phase. Although few-cycle pulses have hitherto mainly been used for generating coherent soft x rays [3,4] and attosecond pulses [5], there is a high potential for applications in many other fields, ranging from coherent control of molecular dynamics to particle acceleration. All the processes involved depend on the time variation of the field and thus on the CE phase. The ability to measure and precisely stabilize the CE phase is therefore a crucial point for all applications. So far, clear evidence of phase effects has been seen in photoionization without phase control for the few-cycle pulses [6], in soft x-ray generation with phase-stabilized pulses [7], and virtually at the same time as the present experiment in photoemission from surfaces with non-amplified few-cycle pulses [8]. Phase-dependent effects in semiconductors have been predicted [9]. Here we present experimental results of nonlinear photoionization with intense phase-stabilized few-cycle visible light pulses. The spatially asymmetric field of few-cycle laser pulses leads to different photoionization rates for opposite directions. By detecting this difference it was possible for the first time to measure the CE phase of powerful few-cycle laser pulses.

The electric part $\mathcal{E}(t)$ of the field of a laser pulse can be written as

$$\vec{\mathcal{E}}(t) = \vec{e}_x \mathcal{E}_0(t) \cos(\omega t + \varphi), \quad (1)$$

where $\vec{e}_x$ denotes the axis of polarization, $\mathcal{E}_0(t)$ denotes the envelope of the pulse, $\omega/2\pi$ denotes the carrier frequency of the laser, and φ denotes the phase. The latter describes a general property of the pulse and is therefore also commonly referred to as the "absolute" phase. For a few-cycle pulse, the width of $\mathcal{E}_0(t)$ (measured at half the maximum) is about 2 times the optical period $2\pi/\omega$. With respect to φ, the convention of choosing $t = 0$ at the maximum of the envelope is used. Then, $\varphi = 0$ corresponds to a "cosine-like" pulse with the absolute maximum of the electric field pointing to the right. Accordingly, we speak of a sine-like pulse for $\varphi = -\pi/2$ (see Fig. 1). Evidently, it is possible to tailor the field of ultrashort pulses by controlling their absolute (CE) phase.

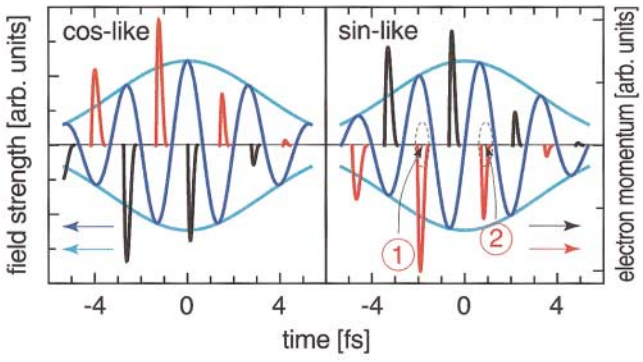

FIG. 1 (color). Electric field (blue) and envelope (light blue) of a 5 fs laser pulse for CE phase 0 (left panel) and $-\pi/2$ (right panel). The drift momentum of high-energy photoelectrons as a function of the ionization time t_0 is shown in black (emission to the right) and red (emission to the left). It was calculated with a classical model. Note that highest momentum does not necessarily coincide with highest electron yield: In the right panel the ionization probability at times labeled with 1 is considerably lower than those labeled with 2 because of lower field strength at t_0.

Conventional femtosecond lasers, however, generate pulses with more or less randomly changing phase. Its stabilization is routinely performed for femtosecond laser oscillators and has led to a breakthrough in frequency metrology [10–12]; for a recent review, see [13]. In contrast, phase stabilization for amplified pulses is a very recent achievement [7]. Considering the asymmetry of the field of few-cycle pulses [14], it appears that an obvious approach is to look for the signatures of the CE phase in nonlinear photoionization. Particular emphasis is given to noninversion symmetric emission of the photoelectrons, i.e., a different count rate for electrons emitted to the right and to the left. A number of theoretical investigations have been devoted to the problem in the past years [15–20]. Most of them concentrate on total yields of photoelectrons.

In our experiments xenon atoms were ionized with 5 fs long laser pulse of peak intensities of nearly 10^{14} W/cm^2. Absorption of eight photons is necessary in order to subdue the ionization threshold. As a matter of fact, processes of much higher order, i.e., absorption of photons in excess of those necessary for ionization, can be observed, and thus correspondingly high photoelectron energies. This phenomenon is known as above-threshold ionization (ATI) [21]. Our experimental approach to analyzing this phenomenon relies on time-of-flight spectroscopy; see Fig. 2.

Figure 3(a) shows a series of ATI spectra corresponding to different CE phases. Pulses with known phase differences $\Delta\varphi$ can be realized by delaying the envelope with respect to the carrier. Since the envelope propagates with the group velocity and the carrier with the phase velocity of light, glass dispersion can be used to shift the CE phase. At a central wavelength of 760 nm adding 52 μm of fused silica will change φ by 2π without distinctly affecting the pulse duration. Thus, two glass wedges which can be shifted with respect to each other allow any phase to be adjusted (see Fig. 2). Simply by inspecting the high-energy part (> 20 eV) of the spectra one can verify that $\Delta\varphi = \pi$ corresponds to a change from left to right, while $\Delta\varphi = 2\pi$ faithfully reproduces the spectra. For high electron energies, the ratio of left/right electron yield approaches 1 order of magnitude and can be controlled by adjusting the CE phase. This constitutes a new kind of *coherent control* and, to our knowledge, is the highest contrast observed so far.

Figure 3(b) displays the ratio of photoemission to the left and to the right for different spectral regions. The ratio of high- and low-energy electrons oscillates with a periodicity of $\Delta\varphi$ in excellent agreement with the periodicity expected from glass dispersion. Besides the much higher contrast for high-energy electrons, a phase shift of both sets of data is also apparent. To determine the CE phase we have to establish the connection between the phase of the field and the ATI signal. This is done by using the high-energy (> 20 eV) electrons. One reason certainly is their higher sensitivity to the CE

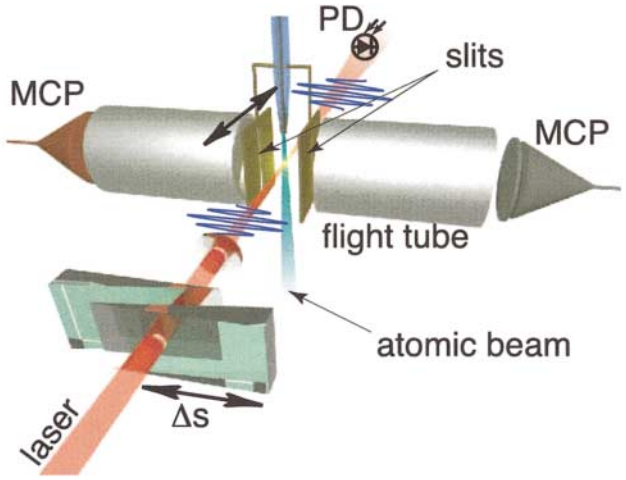

FIG. 2 (color). "Stereo-ATI" spectrometer. Two opposing electrically and magnetically shielded time-of-flight spectrometers are mounted in an ultrahigh vacuum apparatus. Xenon atoms fed in through a nozzle from the top are ionized in the focus of a few-cycle laser beam. The focal length is 250 mm (the lens shown in the sketch is in reality a concave mirror), and the pulse energy was attenuated to 20 μJ. The laser polarization is linear and parallel to the flight tubes. Note that the laser field changes sign while propagating through the focus. Slits with a width of 250 μm are used to discriminate electrons created outside the laser focus region chosen. The slits can be moved from outside the vacuum system. A photodiode (PD) and microchannel plates (MCP) detect the laser pulses and photoelectrons, respectively. Throughout this Letter, spectra corresponding to electrons detected with the left red-colored (right black-colored) detector are colored in red (black). The laser repetition rate is 1 kHz and 50 electrons per pulse are recorded at each MCP. A pair of glass wedges (apex angle 2.8°) is used to optimize dispersion and adjust the CE phase. Therefore, movement of one wedge by Δs introduces $\Delta x = \Delta s \tan(2.8°)$ glass.

phase. More importantly, the dominant overall features in this spectral region are known not to depend on specific properties of the atomic species or on intensity. Figure 4 shows a comparison between measurements and results from classical calculations, the latter taking into account only high-energy electrons. The concept of the calculation will be explained below. In the experimental as well as in the theoretical data the ratio of emission to the left and right depends in a characteristic way on electron energy and the CE phase. This leads to the inclined stripes in the false-color representations. Another common feature is that the asymmetry of left-right emission increases for increasing electron energy. It should be noted that, unlike the contrast of asymmetry, these characteristic features do *not* depend critically on pulse shape or pulse duration. The sharp edges visible in the calculation originate from classical energy conservation. This artifact is removed by energy-time uncertainty in quantum mechanics. Matching the characteristic features of the theoretical and experimental data leads

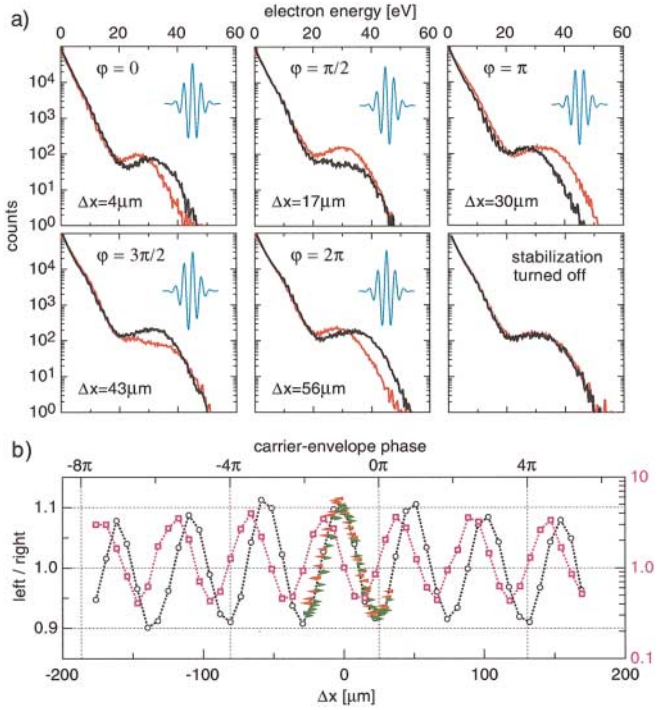

FIG. 3 (color). (a) Photoelectron spectra for different CE phases controlled by fine movement of one of the wedges. Δx indicates the glass added hereby. The black curves correspond to emission to the right (positive direction), the red ones to the opposite direction. The insets show the corresponding real time variation of the electric field, as deduced from the phase assignment shown in Fig. 4. Only without phase stabilization were identical spectra measured left and right as expected. (b) Left-right ratio of the total electron yield (circles) and high-energy electrons (squares) as a function of glass thickness Δx added or subtracted by moving one of the wedges. $\Delta x = 0$ corresponds to optimal dispersion compensation, i.e., the shortest pulses. Maximal left/right ratio for the total yield does not coincide with that for high-energy electrons. Note the different scales for low- and high-energy electrons. The upper x scale indicates the carrier-envelope phase of the pulse, as deduced from the comparison with theory shown in Fig. 4. The measurement of the CE phase drift was performed by a scan forth (triangles pointing to the right) and back (triangles pointing to the left) with finer variation of the glass thickness. The mismatch at the start and the end point indicates a phase drift of ≈ 50 mrad/min.

to an unambiguous determination of the CE phase with an estimated error of $\pi/10$.

The calculation used to determine the CE phase is based on the insight that, at high intensities, ATI can be understood in terms of quasistatic field ionization at some time t_0 and subsequent classical evolution of the electron in the laser field [22,23]. For a recent review, see [24]. A considerable fraction of the photoelectrons generated when atoms or molecules are exposed to intense laser pulses do not escape directly. Rather they return to the ion core within times of typically less than an optical cycle and in precise synchronization with the electric field of the laser pulse. The characteristics of returning electrons can be observed in the kinetic energy spectra of photoelectrons. The underlying mechanism is elastic scattering of the electrons when they return to the ion core at time $t = t_1$ and are subsequently accelerated by the laser field. As a consequence, photoelectrons with high kinetic energy can be observed. In the photoelectron spectra, they manifest themselves in the high-energy plateaulike annex to the exponentially decreasing count rate at low electron energies [25,26].

The kinetic energy spectrum of the plateau electrons is sensitive to the CE phase for several reasons. First of all, high-energy electrons returning to the ion core can be created only in subfemtosecond time intervals close to peaks of the electric field of the laser pulse (see Fig. 1). However, the probability that they tunnel through the atomic potential at t_0 depends exponentially on the field strength $\mathcal{E}(t_0)$ and as few-cycle pulses are involved it is likely only for those very few optical half-cycles close to the pulse maximum. Generally, the highest kinetic energies are reached for electrons returning to the core when the electric field becomes nearly zero [$\mathcal{E}(t_1) = 0$]. For few-cycle pulses in addition, the field amplitude $\mathcal{E}_0$ needs to be as high as possible for $t > t_1$ in order to allow efficient acceleration after rescattering. Since the start time t_0 and return time t_1 differ by almost one optical cycle, both conditions, namely, the highest possible field strength at t_0 and highest possible amplitude after t_1 are hard to meet and result in a strong dependence of photoionization on the CE phase. Number, strength, and timing of the wave packets lead to distinctive structures in the ATI spectra. Their analysis therefore provides detailed information about the key processes of attosecond science. Quantum mechanical calculations are in very good qualitative agreement with this classical treatment [20].

Having established that ATI can be used for precise determination of the CE phase, an example of the sensitivity of the instrument should be given. In Fig. 3(b) also a measurement with small steps $\Delta\varphi$ is shown. This measurement reveals small phase drifts, which have to be attributed to the laser system and its stabilization scheme. The phase drifts are of the order of 50 mrad/min, measured over more than 10 min. As the stereo-ATI phase meter not only can measure the CE phase and also detect small phase drifts, it clearly has the potential to be used in a feedback loop. In this respect it should be emphasized that the target gas pressure is so low ($< 10^{-4}$ mbar) that the ATI experiment does not affect the laser beam; i.e., a stereo-ATI phase meter can be placed anywhere in a laser beam line.

In summary, we have shown that strong-field photoionization provides a very efficient means of measuring the CE phase of few-cycle femtosecond laser pulses with *no* $\pm\pi$ phase ambiguity inherent in many other schemes. Conversely, the emission direction of photoelectrons can be steered by choosing the CE phase, establishing a new kind of coherent control scheme. Control of the CE phase allows one to tailor the field of a few-cycle pulse. A

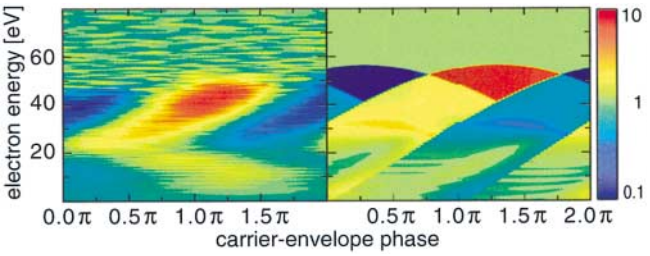

FIG. 4 (color). Determination of the CE phase. False-color representation (logarithmic scale) of the left-right ratio of photoelectrons as a function of the electron energy and the CE phase. Positive values (red) indicate dominant left emission, negative values (blue) dominant right emission. For the theoretical part (right panel), only rescattered electrons were taken into account. The parameters used are the same as in the experiment. This comparison allows precise carrier-envelope phase determination. With the convention of Eq. (1), the electric field is now uniquely characterized.

tailored field variation would deform molecular potentials in a virtually arbitrary way. This could help to induce or analyze exotic chemical reactions. In fact, it is possible to choose conditions such that the field vanishes upon the return of the wave packet. The very high current densities corresponding to the returning wave packets should facilitate electron-ion scattering experiments, hitherto hardly possible. Furthermore, investigation of interference patterns in ATI spectra will allow ionization to be studied in real time.

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