Phase-Controlled Single-Cycle Strong-Field Photoionization


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Abstract

The evolution of the electric field of laser pulses consisting of a few optical cycles depends on the so-called absolute phase. Strong-field photoionization not only provides means to measure the absolute phase. Rather phase-controlled few-cycle pulses allow investigating photoionization on the attosecond time scale as the sub-cycle ionization dynamics becomes manifest in the photoelectron spectra. Few-cycle pulses thus can serve as a time-domain microscope for investigation of electronic transitions.

1. Introduction

The typical wavelength of Ti:Sapphire femtosecond lasers is $\approx 800\,\text{nm}$ and corresponds to an optical period $T$ of $\approx 2.5\,\text{fs}$. One of the most remarkable breakthroughs in femtosecond laser technology has been the generation of powerful pulses with a duration (FWHM) around 5 fs, i.e. pulses consisting of just 2 optical cycles (few-cycle pulses) [1]. This leads to the new situation that the pulse envelope $E(t)$ varies on the same time-scale as the electric field $E$. More importantly, the temporal variation of the electric field depends on the phase $\varphi$ of the carrier wave with respect to the envelope. This can be seen by expressing the field as a product of envelope and carrier:

$$E(t) = E(0) \cos(\omega t + \varphi).$$

Choosing the convention that $t = 0$ at the maximum of the envelope leads to “cosine-like” pulses for $\varphi = 0$ and “sine-like” pulses for $\varphi = -\pi/2$. As $\varphi$ characterizes a global property of a few-cycle pulse, it is commonly referred to as the “absolute” phase.

The significance of the absolute phase comes from the fact that it determines by definition the field at and around the pulse maximum. Therefore, any application taking advantage of few-cycle pulses will depend on the absolute phase. So far, generation of soft-X-ray radiation and isolated attosecond pulses have taken advantage of control of the absolute phase [2]. It is foreseeable that coherent control of atomic and molecular behavior will be one of the next fields in this respect: absolute-phase-control together with control of spectral phase, amplitude and eventually polarization will allow generating tailored variations of strong electric fields. These will deform molecular potential surfaces in a virtually arbitrary way and should make possible to steer chemical reactions with unprecedented precision. An even nearer application could be electron-ion collisions: Atoms or ions exposed to intense laser pulses are photoionized. During the ionization process, an appreciable fraction of the electron wave packet can return to the vicinity of the core with considerable kinetic energy. The returning electron wave packet corresponds to enormously high current densities otherwise only available in the most advanced accelerators. Using few-cycle pulses and controlling the absolute phase, it is possible to adjust such conditions such that the electrons return at a zero crossing of the field and thus probe the ion core.

It is likely that the list of single or sub-cycle applications can be almost endlessly continued thus touching biophotonics, solid-state, nuclear, and plasma physics.

The absolute phase (or, to be precise, the rate with which it changes) has attracted attention in a field of physics hardly brought into connection with strong-field physics so far: Controlling the absolute phase [3–5] allows using femtosecond lasers as optical clock-works [6,7]. In fact, the technical realization of phase-stabilization revolutionized optical frequency metrology in the past years and at the same time launched the field of phase-dependent strong-field physics outlined above [8]. Meanwhile it is possible to generate powerful few-cycle pulses with stabilized absolute phase [2]. However, measurement of the absolute phase has been achieved only very recently by photoionization [9] and by photoemission experiments [10].

2. Photoionization by few-cycle pulses

The purpose of this paper is to review basic aspects of photoionization with few-cycle pulses and its phase dependence. At near-infrared wavelengths ionization is a multiphoton process. This holds in particular for rare gas atoms as considered here. Multiphoton ionization at high field strength can cause atoms to absorb more photons than necessary for ionization leading to photo-electron energies considerably higher than the photon energy (above-threshold ionization, ATI) [11]. In addition, the photoelectron can undergo a scattering process during ionization and thus acquire much higher kinetic energy [12]. The latter process is highly non-perturbative and leads to photoelectron spectra (ATI spectra) characteristic for intense-field photoionization: The ionization probability as a function of kinetic photoelectron energy decreases steeply for low energies, stays almost constant for energies beyond 20 to 30 eV, and rolls off after this so-called plateau-region (cutoff), see Fig. 1. This general behavior is well understood and in fact can be explained with simple classical arguments [13]. For a recent review see [14].
Coming back to few-cycle pulses, it is quite natural to predict a non-inversion symmetric electron yield from the asymmetric and phase-dependent shape of the field [15]. Therefore, an approach to detect and eventually measure the absolute phase is to record electrons emitted in opposite directions. The first work investigating this problem theoretically was published by Cormier and Lambropoulos [16]. They considered alkali atoms ionized with cosine-like pulses of moderate intensity, i.e. in the perturbative regime. The phase-effects predicted under these conditions were discouragingly small. Later work by Dietrich et al. [17], Christov [18], and Chelkowski and Bandrauk [19] used higher intensities and also higher ionization thresholds. However, only the total electron yield was considered. The asymmetries predicted for electron emission in opposite directions are in the 10% region for 5 fs-pulses and thus in principle readily observable. Only recently, Milošević et al. [20] calculated electron spectra for strong-field ionization and obtained orders of magnitude stronger phase-effects for high-energy photoelectrons.

On the experimental side, the first absolute-phase effect was observed in 2001 [21], although the absolute phase was randomly varying from pulse to pulse in that experiment. The key idea had been that the asymmetry in electron emission translates into an anti-correlation of electrons emitted in opposite direction if investigated shot by shot. Very recently, ATI has been studied using phase-stabilized few-cycle pulses which, as indicated above, yielded the first measurement of the absolute phase. Phase stabilization made possible to record photoelectron spectra (and not just total rates) as a function of the absolute phase. The experiment agreed with the theoretical finding that the phase-dependence of high-energy (i.e. plateau) electrons exceeds that of low-energy electrons (which determine also the total rate) by almost two orders of magnitude. It is therefore interesting to discuss the origin of this eye-catching difference in phase sensitivity.

3. Low-energy (“direct”) photoelectrons

Atomic behavior in intense laser field can—at least in a qualitative way—be well described within the strong-field approximation (SFA). Its classical version [22–24] assumes that an electron is lifted into the continuum at some time \( t_0 \). Assuming the initial conditions \( x(t_0) = 0 \) and \( \dot{x}(t_0) = 0 \) [25,26] the electron trajectory \( x(t) \) can be calculated by solving the equation of motion

\[
\dot{\mathbf{x}} = \mathbf{F} = e \cdot \mathbf{E}(t),
\]

where \( m \) is the electron mass and \( e \) the elementary charge. Apparently, the atomic potential \( V \) is neglected hereby. \( V \) enters only by determining the ionization rate \( R \) together with the field strength \( \mathbf{E}(t_0) \). Obviously, a highly non-linear function of both \( V \) and \( \mathbf{E} \) has to be assumed. This situation is shown in Fig. 2 for a sine- and cosine-like pulse with linear polarization being assumed. It is obvious that the asymmetry of \( R \) exceeds that of the field by far due to the non-linearity of ionization.

Now it is most important to realize that an electron lifted into the continuum at \( t = t_f \) will not necessarily fly in the (negative) direction of the electric field \( \mathbf{E}(t = t_0) \) at that time \( t_0 \) of ionization. Rather from conservation of the canonical momentum

\[
\mathbf{p}_\text{can} = m \dot{x}(t) + \frac{e}{c} \mathbf{A}(t)
\]

together with the initial condition \( \dot{x}(t_0) = 0 \) it follows that a photoelectron created at \( t = t_0 \) will have a drift momentum proportional to the vector potential \( \mathbf{A}(t = t_0) \). The vector potential however has just the opposite parity than the field. A cosine-like field for example has a sine-like vector potential. Therefore, the ionization probability in positive direction is given by the integral of the ionization rate \( R(t) \)

![Fig. 2. Explanation for the weak phase dependence of low-energy ("direct") electrons. The thin solid curve represents the electric field \( \mathbf{E}(t) \) of a few-cycle pulse for absolute phase \( \varphi = 0 \) (cosine-like, left) and \( \varphi = -\pi/2 \) (sine-like, right). The instantaneous ionization rate \( R(t) \) (bold curve) depends in a highly non-linear way on the magnitude of the electric field \( \mathbf{E} \) and thus amplifies the asymmetry. The electrons however eventually drift in the direction which the vector potential had at the time \( t_0 \) when the electron entered the continuum. Therefore, the number of electrons flying in positive direction is given by the integral of \( R \) over all times where \( A \) is positive and vice versa. One immediately sees that the phase effect is strongly reduced due to the deflection of the electrons by the oscillating field.](image-url)
over those times $t$ for which $A(t)$ is positive and vice versa. This is also depicted in Fig. 2. Two features following from this discussion are noteworthy.

First, the most asymmetric field variation (i.e., the ± cosine-like pulse) leads to equal electron yield in opposite directions, while the most symmetric field leads to the most asymmetric electron emission. This surprising consequence of the strong-field approximation, however, should not be over-emphasized as the atomic potential neglected in the framework of the SFA might influence the electron trajectories due to the low kinetic energy of the majority of the photoelectrons [27].

The other consequence is less susceptible to details of the ionization process: The asymmetry in photoionization, which one might have expected to be very pronounced because of the non-linear dependence of ionization on field strength, in fact is extensively canceled by the deflection of the photoelectrons in the oscillating laser field.

For circular polarization the situation is different. There the electric field of a few-cycle pulse can be visualized as a screw the radius of which increases and eventually decreases within a few rotations. The absolute phase can be identified with the direction to which the field at the maximum of the pulse points. Changing the absolute phase therefore just changes the direction to which the field points at the maximum of the pulse. Analogous to linear polarization, the momenta of the photoelectrons will be determined by the vector potential and the Coulomb potential. In contrast to linear polarization, however, electrons are emitted in all directions perpendicular to the laser beam and the spatial asymmetry in ionization, which is of course due to the spatial asymmetry of the field, is not cancelled by subsequent deflection of the photoelectrons in the field [17]. Circular polarization thus seems to offer advantages for phase measurement as compared to linear polarization. However, detailed calculations [28] unveil that the improvement is hardly bigger than a factor of 2. Under most circumstances this will not outweigh the disadvantages of circular polarization: Femtosecond lasers by construction produce linear polarization. Conversion to circular polarization is not trivial considering the tremendous bandwidth associated with few-cycle pulses [29]. More importantly, the processes underlying present approaches to attosecond physics [30–32] require linear polarization.

4. High-energy (“plateau”) photoelectrons

So far, only low-energy electrons were considered. From a technical point of view these (as compared to plateau electrons) appear to be much more interesting for a measurement of the absolute phase because of their higher abundance. However, recalling the mechanism of generation of plateau electrons leads to the conjecture that plateau electrons are much more sensitive to the absolute phase. This has in fact been proven valid in a recent experiment [9].

The plateau electrons originate from electron trajectories which return to the vicinity of the parent atom typically within less than one optical cycle. Such trajectories are only possible for linear polarization to which we come back now. If a returning electron is elastically back-scattered upon its revisit to the core at time $t = t_1$, it will be accelerated by the laser field due to the change in sign of the electron velocity at $t = t_1$. The maximum drift energy the electrons may acquire via this process is $10U_p$, where $U_p$ denotes the ponderomotive potential [33]. Rescattering certainly is not the only option the returning electron has. Rather, scattering might also be inelastic. In the case of recombination this leads to the production of soft X-rays [34,35] and attosecond pulses [31,32], while non-sequential (i.e. correlated) multiple ionization is observed in the case of impact ionization [24,36,37]. Thus, all prominent effects of atoms in strong fields find a simple and intuitive explanation. However, one should not fail to mention highly visible effects which seem to be beyond this simple classical model as well as its quantum version [38–40].

In order to explain the origin of plateau electrons more explicitly, we first consider long pulses, i.e. the time-dependence of the amplitude $\mathcal{E}_0$ is neglected. Always implying the well-established initial condition $\mathcal{E}(t_0) = 0$, returning trajectories are only possible if the initial ionization takes place between an extremum and the following zero crossing of the electric field, see Fig. 3. Electrons injected into the field at a time $t_0$ where the field assumes an extreme value will return to the core exactly one optical cycle later, i.e. $t_1 = t_0 + T$. Obviously, the velocity of the electron then is zero again, $\mathcal{E}(t_1) = 0$. The other limiting case is injection of the electron at the zero crossing. This is a trivial case as $t_1 = t_0$ and consequently again $\mathcal{E}(t_1) = 0$. As the travel time $t := t_1 - t_0$ changes from $T$ to 0 between these limiting cases, the velocity upon return $\mathcal{E}(t_1)$ cannot vanish for all $t_0$. Therefore, there must be a maximum for $\mathcal{E}(t_1)$ and thus also for the return energy $E_r$. Rescattering leads to a change in sign for $\mathcal{E}(t_1)$. This is the prerequisite for further acceleration of the electron in the field after $t = t_1$. It turns out that the maximum electron energy is reached for ionization times $t_0$ close to (but not at) extremal values of the field. The field strength at $t_0$ is very important because the probability for ionization is a strongly non-linear function of $E(t_0)$.

Usually, it is not explicitly mentioned that, besides a high field strength $E(t_0)$, the amplitude after rescattering at $t = t_1$ should be as high as possible, too. This after all is self-evident for long pulses. However, it is exactly at this point where there is a qualitative difference between “ultrashort” (meaning femtosecond) pulses and few-cycle. In order to get an appreciable yield of high-energy electrons, $E(t_0)$ and $E(t > t_1)$ should be as high as possible. For a few-cycle pulse these two conditions are hard to meet at the same time because $t_1 \approx t_0$ is of the order of the optical period within which the amplitude of the field necessarily changes appreciably. Changing the absolute phase affects the interplay between these competing requirements for production of plateau electrons. This is the main (although not the only) reason for the high sensitivity of plateau electrons on the absolute phase.

5. Time-domain information of electronic transitions

At this point, it might be instructive to realize another difference between low- and high-energy electrons. For the former, one can find an absolute phase for which both spectra corresponding to emission in opposite directions (parallel to the polarization) are the same. The simplest
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approach to explain this feature is to assume that ionization is an instantaneous process in the sense as outlined above: Electron yield and momentum are entirely determined by the field and the vector potential at $t = t_0$. Disregarding small effects from the non-vanishing derivative of the envelope, one can find an absolute phase ($\varphi = 0$ when neglecting the Coulomb potential, see above) for which the ionization rate $R(t)$ is inversion symmetric with respect to time. This together with instantaneous ionization leads immediately to symmetric electron emission.

The high-energetic plateau electrons, in contrast, are created through a more complex mechanism that involves two times, namely the time $t_0$ at which the electron enters the continuum and the time $t_1$ at which it returns to the ion core and scatters. Under these circumstances, a pulse for which ionization in opposite directions would be the same is difficult to envision. Inversion symmetry as in the case of instantaneous ionization is not possible: An electron injected into the field before the maximum of the pulse envelope can acquire a much higher drift momentum as an electron injected in the inversion-symmetric instant, which is after the pulse maximum (see Fig. 3). The built-in delay in the ionization process for plateau electrons breaks the symmetry discussed above.

This basic feature of photoionization as a function of the absolute phase is in beautiful agreement with the prevailing model of strong-field laser atom interaction. However, the experimental evidence also sticks a question mark on this model since symmetric ionization occurs at the “wrong” absolute phase. It is likely that this has to do with the Coulomb potential neglected in the strong-field approximation. Nevertheless, fundamental properties of phase-dependent photoionization spectra impose stringent conditions on possible alternatives and provide access to time-domain information on electronic transitions elapsing on the attosecond time scale.

Even more detailed temporal information can be gained by considering interference effects. We start by explaining how to calculate the ATI plateau in the framework of the classical model and include quantum aspects thereafter. From Fig. 3 one can extract that, in general, electrons with a given drift momentum can be generated at several times $t_0$ within the pulse. In order to get the intensity of these electrons, the ionization probabilities at these times $t_0$ have to be added. A more complete model would also take into account the scattering cross section as a function of the impact energy.

Evidently, the “electrons” created at several instants $t_0$ within the laser pulse in reality are wave packets which may and, not surprisingly, which do interfere. For long pulses, in every optical cycle identical wave packets are created. This immediately leads to alternating constructive and destructive interference with a periodicity given by the photon energy. The interference pattern is nothing but the well-known ATI structure. For completeness we mention that two times within one optical (half-cycle) lead to electrons with the same momentum. Closer inspection shows that these two times correspond to a short and a long trajectory. Due to the smaller temporal distance, the resulting interference pattern has a longer periodicity in energy space. The interference of short and long trajectories thus leads to a modulation of the envelope of ATI spectra.

In order to quantify such interference effects, the phases of the electron wave packets need to be calculated. This again is quite simple as the phase is given by $S/h$, where $S$ is the classical action of the electron trajectories considered throughout this article. An example for such a calculation is shown in Fig. 4. ATI peak structure of high contrast needs at least two optical cycles which yield ionization of
Fig. 4. Calculated ATI spectra (rescattered electrons only) for a cosine-like 2-cycle pulse. Like in Fig. 3 black corresponds to emission in positive and grey to emission in negative direction. The strength of the electron signal and the contrast of the ATI peak structure is nicely explained by the analysis shown in Fig. 3. To this end, the respective curves of that figure were converted to energy and rotated by 90°, i.e. the right ordinate is a time axis. The cutoff energies exhibited by the spectra are faithfully reproduced. The strength of the spectra is determined by the electric field corresponding to the horizontal spikes. Note that the pulse maximum has been chosen to be at $t = 0$. Therefore, strongest emission at high energy is observed in negative direction. Interference with high contrast is only possible, if two of the horizontal spikes contribute and if the corresponding field strength is similar for both. This is the case below $\approx 15\times\omega_0$ for emission in positive direction. The sharp cutoffs are an artefact of the classical model and would be smoothed in a quantum mechanical description [20].

6. Conclusion

Fundamental aspects of laser of phase-dependent strong-field photoionization have been discussed. The different sensitivity of low- and high-energy electrons to the absolute phase has been explained employing simple classical arguments. Quantum mechanical extensions to the model allow explaining interference phenomena. All three phase effects discussed are suitable for measuring the absolute phase.

Fig. 5. Calculated ATI spectra (rescattered electrons only) for various absolute phases. Again, black (grey) curves correspond to spectra recorded in positive (negative) direction. The spectra are in good agreement with results from quantum calculations [20]. The intensity is $0.8 \times 10^{14}$ W/cm$^2$ and a logarithmic scale is used.
Less profane, phase phenomena provide a new gateway to unsolved problems in strong-field laser matter interaction, in particular to the role of the atomic or ionic potential. A particular exciting aspect is direct access to time-domain information for electronic transitions, photoionization being their prototype. Clearly, the vision to drive atomic and molecular behavior and reactions by single optical cycles and to steer them by controlling the absolute phase has come close to reality. The single-cycle aspect together with attosecond timing precision bears the character of time-domain microscopy of such reactions.

Acknowledgments

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References

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33. The ponderomotive potential is defined as the quiver energy of a free electron in an oscillating electric field. An useful formula is $U_p = 0.09337 \cdot f [W/cm^2] \cdot \lambda [\mu m]$. For a wavelength of $\lambda = 800 nm$ and an intensity of $I = 10^{15} W/cm^2$ the ponderomotive energy will be $U_p = 6 eV$ which is almost four times the photon energy.