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Absolute-phase phenomena in photoionization with few-cycle laser pulses

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Currently, the shortest laser pulses[1] that can be generated in the visible spectrum consist of fewer than two optical cycles (measured at the full-width at half-maximum of the pulse's envelope). The time variation of the electric field in such a pulse depends on the phase of the carrier frequency with respect to the envelope—the absolute phase. Because intense laser–matter interactions generally depend on the electric field of the pulse, the absolute phase is important for a number of nonlinear processes[2–8]. But clear evidence of absolute-phase effects has yet to be detected experimentally, largely because of the difficulty of stabilizing the absolute phase in powerful laser pulses. Here we use a technique that does not require phase stabilization to demonstrate experimentally the influence of the absolute phase of a short laser pulse on the emission of photoelectrons. Atoms are ionized by a short laser pulse, and the photoelectrons are recorded with two opposing detectors in a plane perpendicular to the laser beam. We detect an anticorrelation in the shot-to-shot analysis of the electron yield.

Investigation of ultrafast processes driven by femtosecond laser pulses has acquired significance in many fields of the natural sciences. In addition, the short pulses provide access to coherent light sources with spectrally broad bandwidth and generate extremely high intensities with table-top laser systems. The key to some of these developments was the invention of laser systems delivering pulses in the range of 5 fs with energies higher than 100 µJ (ref. 1). The fields generated by these pulses are comparable to, or even higher than, the internal fields of atoms ($>10^{14}\,\mathrm{W\,cm^{-2}}$). Atoms are therefore easily ionized. Ultrashort pulses have the advantage that the highest pulse intensity is reached in a time shorter than that which the electron needs to escape from an atom, allowing the use of much higher effective field strengths. These facts and the need for even higher time resolution and broader bandwidth are promoting the development of shorter and shorter pulses in fields as varied as particle acceleration, plasma physics, coherent control of chemical reactions[4], frequency metrology[5–7], generation of coherent soft-X-ray radiation[9,10], and generation of attosecond pulses[8,11].

Until now, the only proven way to produce powerful laser pulses in the 5-fs regime has relied on the hollow-fibre technique[1,12]: pulses from a femtosecond oscillator are amplified to the millijoule level, compressed to their bandwidth limit of about 20 fs, and then fed into a hollow gas-filled fibre, where they broaden spectrally by self-phase modulation. Residual phase errors ('chirps') are then compensated when the pulses are reflected from specifically designed dielectric mirrors. The pulse repetition rate of such systems is determined by that of the pump laser of the amplifier, and is typically 1 kHz. Probably the most noteworthy property of such short pulses is that the duration of the oscillation period of the radiation (2.6 fs, corresponding to the typical Ti : sapphire femtosecond laser central wavelength of 800 nm) approaches the pulse duration. This means that such a laser pulse consists of just a few cycles, causing a new quantity to become important: the phase φ of the carrier with respect to the envelope of the pulse—the so-called absolute phase of the pulse. In the few-cycle regime, the electric field E should be written as

$$E(t) = E_0(t)\sin(\omega t + \varphi)$$

where $E_0(t)$ denotes the envelope of the pulse. Examples of such pulses are shown in Fig. 1a–c. It is evident that the electric field as a function of time depends on the absolute phase, although the envelope is the same for all pulses.

Most effects in intense-field atom interaction—for example, the photoionization discussed here—depend on the time dependence of the electric field. These effects must be expected likewise to depend on the absolute phase[2,3]. A particularly important example is the generation of isolated attosecond pulses in the soft-X-ray spectral region[8]. For photoionization, a vivid picture can be obtained of the processes taking place: photoionization can be regarded as the consequence of an oscillating electric field that shakes the electrons of an atom until one or more of them gain so much energy that they can leave the atom. It is clear that the absolute phase should have an influence on this process: for very short pulses, it makes a difference whether the atom receives the first blow from one side or from the other. Unfortunately, no way has so

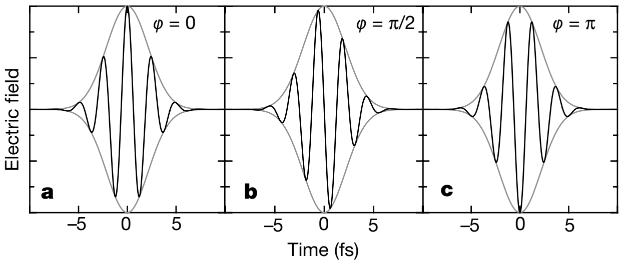

Figure 1 The time variation of the electric field of laser pulses consisting of very few optical cycles depends on the phase φ of the carrier frequency with respect to the pulse's envelope. The maximum of the electric field points in opposite directions at $t = 0$ for $\varphi = 0$ and $\varphi = \pi$. The grey curves indicate the envelope of the pulses. **a**, $\varphi = 0$; **b**, $\varphi = \pi/2$; **c**, $\varphi = \pi$.

far been found to control the absolute phase in laser systems as outlined above. Rather, it must be expected to fluctuate randomly from pulse to pulse. In contrast, for femtosecond laser oscillators—that is, for pulses in the nanojoule region at repetition rates of tens of megahertz—techniques have been invented that stabilize at least the phase difference between consecutive pulses[5–7,13]. This is of importance to optical frequency metrology[14,15].

Preliminary evidence of a regime of influence of the absolute phase on the emission of high harmonics of the laser radiation (that is, high-harmonic generation) has been reported[16]. Pulses of 20 fs duration were used, for which no effects of the absolute phase can usually be expected. However, high-harmonic generation is strongly governed by phase-matching effects that may increase the sensitivity under appropriate circumstances.

We present here a demonstration of the influence of the absolute phase on intense-field photoionization, investigated with a 5-fs laser system. Photoionization of atoms in intense laser fields is characterized by above-threshold ionization (ATI)[17]. ATI means that an atom may absorb more photons than are necessary for its ionization. This leads to photoelectron energies considerably higher than the photon energy. The effects of the absolute phase on ATI spectra (and the related phenomenon of high-harmonic generation) have been studied theoretically[18–22]. Typically, ATI is investigated by means of time-of-flight spectroscopy (Fig. 2).

If the interaction of atoms with long pulses is investigated, it is found that the intensity of photoelectrons emitted to opposite sides is identical, thus ensuring inversion symmetry. This is because the electric field of a long pulse (as well as the atom) exhibits inversion symmetry. This holds for any laser polarization. For short pulses, however, it can be seen from Fig. 1 that, depending on the absolute phase, the creation of photoelectrons will violate inversion symmetry. The nonlinear character of strong-field photoionization, for which the photon energy is much smaller than the ionization threshold of the atom, supports this assertion. So the absolute phase determines whether more electrons are emitted to the one or the other side. For example, a pulse as depicted in Fig. 1b can be expected to lead to an equal number of photoelectrons escaping to the left and to the right, whereas pulses like those in Fig. 1a and c will lead to asymmetric yields. Altogether, the absolute phase is expected to cause an anticorrelation in the number of electrons escaping in opposite directions.

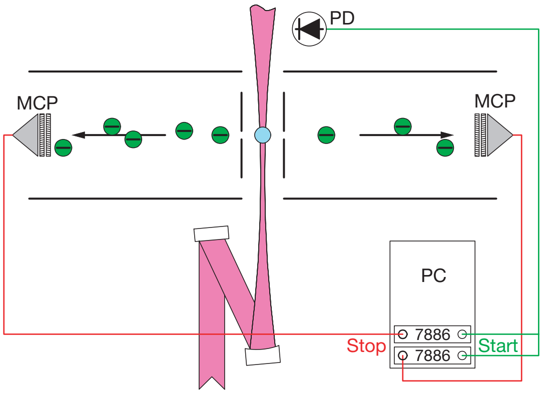

Figure 2 Experimental set-up. Laser pulses (6 fs full-width at half-maximum) are focused ($f = 1$ m) onto a gas jet (indicated in blue) in a vacuum apparatus. Slits with a width of 250 μm allow only photoelectrons originating from a predefined region in the laser focus to enter the shielded drift tubes. At the ends of the drift tubes the electrons are recorded by microchannel plates (MCP). The electrons' time of flight is measured by two computer-hosted multiscalers (FAST 7886, FAST ComTec, Oberhaching, Germany). The start signal is generated by a fast photodiode (PD), indicated on the upper part of the figure, and the stop signals are created by the MCPs. The electrons' time of flight can be used to calculate their kinetic energy. For the measurements presented in Fig. 3, the polarization of the laser pulses is changed to circular with an achromatic quarter-wave plate (B. Halle, Berlin). The waveplate is placed between the laser amplifier and the hollow-fibre compressor.

In line with the arguments given above, the idea behind our experiment is to place two electron detectors in opposite directions with respect to the laser focus (a 'stereo ATI experiment'; Fig. 2). For each laser pulse, the number of electrons detected with both detectors is recorded. (We also measured and stored the time of flight for each electron, but this information is not used here.) Accordingly, each laser pulse is characterized by two numbers: that is, the number of electrons detected in the left-hand and that in the right-hand arm of the ATI spectrometer. We then interpret these pairs of numbers as coordinates, and accumulate thus-characterized laser shots in a map (contingency map). As the signature of an effect from the absolute phase is an anticorrelation, it should appear as a structure inclined at −45° in the contingency map. This approach is expected to be very sensitive to correlations, because, in principle, an arbitrary number of laser shots can be used in order to make even small (anti)correlations statistically significant.

In implementing this idea, a number of details have to be considered in order to preserve the fragile effects of the absolute phase. This was pointed out in a theoretical analysis of the visibility of these effects in the angular distribution of the photoelectrons[19]. Because our correlation method is just a different—although very sensitive—way of looking at the angular distributions considered in that report, the results obtained there are applicable to our measurements. In ref. 19 it is also pointed out that the use of circularly polarized light is advantageous. The electric field of pulses with this polarization is reminiscent of a screw with a diameter that increases and decreases in accordance with the envelope of the pulse. The absolute phase determines the direction in which the maximum of the field points. For symmetry reasons, it also determines the direction in which most photoelectrons will fly. For linear polarization, the dynamics of the electrons is confined to the polarization axis, and the dependence on the absolute phase is more complicated. However, there are also disadvantages for circular polarization. The most severe one is that it is difficult to produce a circularly polarized beam, owing to the considerable bandwidth of the laser radiation. The benefits of circular polarization were therefore much smaller than expected. For circular polarization, the shortest pulse duration obtained in our experiment was 6 fs. A particularly difficult problem is presented by laser fluctuations, because they produce (positive) correlations: stronger pulses produce more electrons than weaker ones in a highly nonlinear dependence, irrespective of the emission angles of the electrons. This means that the anticorrelation produced by the absolute phase has to be stronger than the correlations due to laser-pulse fluctuations in order to be clearly measurable.

Figure 3 shows a contingency map obtained for krypton at an intensity of $5 \times 10^{13}\ \mathrm{W\,cm^{-2}}$ and circular laser polarization. The signature of anticorrelation—features inclined at approximately −45°—is clearly visible. In order to prove the anticorrelation and evaluate its strength and significance, we used statistical methods. One possibility is to calculate Kendall's τ (ref. 23). A positive value of τ indicates (positive) correlation, and a negative value, anticorrelation. Kendall's τ is the result of a non-parametric algorithm that relies on rank-ordering: essentially, the number of concordant and discordant events is subtracted. It should be noted that non-parametric procedures, in particular Kendall's τ, are conservative in the sense that if they indicate (anti)correlation, then it definitely exists. In our measurements with short pulses, we achieved small, negative values of τ, (typically $\tau \approx -0.05$) but of distinct significance (typically more than two standard deviations of the null hypothesis of no correlation). This was compared with the result of the same procedure for longer pulses (>7 fs). There, Kendall's τ had a positive sign with a significance of typically six standard deviations or more. This is

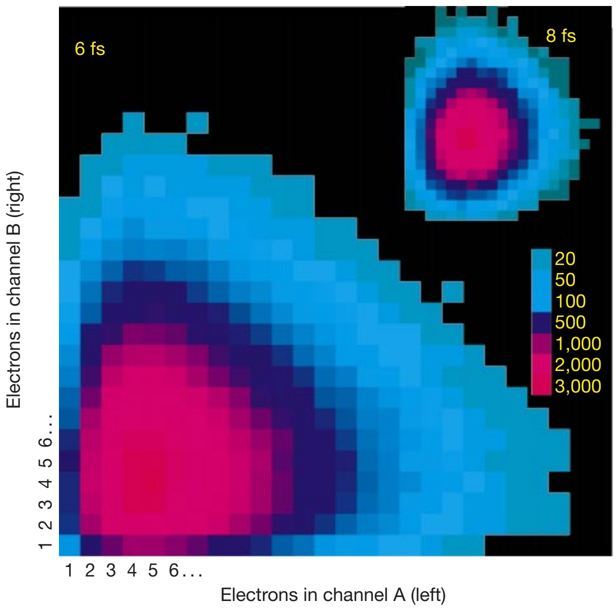

Figure 3 Contingency map. Every laser shot is recorded in this plot according to the number of electrons measured in the left (channel A) and the right (channel B) arm of the electron spectrometer. In the colour code, the pixel colours represent the number of laser shots with electron numbers given by the coordinates of the pixel. The signature of the absolute phase is an anticorrelation in the number of electrons recorded with the left and the right detector. In the contingency map they appear as features inclined at –45°. Shown here (main figure) is a typical measurement with krypton atoms for circular laser polarization, a pulse duration of 6 fs, and an intensity of 5×10^{13} W cm^{-2}. The data were taken over 200,000 laser shots, corresponding to a measuring time of 200 s. On average, each laser shot led to the recording of approximately 5 electrons in each arm of the spectrometer. Inset, the corresponding measurement for slightly longer pulses, showing no indication of anticorrelation.

expected because of the inevitable laser pulse fluctuations, as described above. In the light of this, the anticorrelation observed is a rather strong effect.

This is, to our knowledge, the first unambiguous measurement of an effect stemming from the absolute phase of an ultrashort pulse, and we believe that the method presented here is important for its potential applications. These include detailed investigations of the influence of the absolute phase on strong-field ionization. The close relationship of ATI and coherent soft-X-ray generation (high-harmonic generation) will also make such investigations instrumental in understanding absolute-phase effects in high-harmonic generation. One way to investigate ATI in detail is to measure the strength of the anticorrelation as a function of the photoelectron's kinetic energy, the laser intensity, and so on. The strength of the anticorrelation can also serve as a measure of the pulse duration, which is useful because such measurements become increasingly difficult for shorter and shorter pulses when conventional methods are used. Finally, with suitable refinements, our stereo ATI apparatus could be used to measure the absolute phase. The measured value could then be used to interpret other experiments, such as the generation of coherent soft X-rays, generation of isolated attosecond laser pulses, or coherent control of chemical reactions. ☐

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Sharper images by focusing soft X-rays with photon sieves

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Fresnel zone plates consisting of alternating transmissive and opaque circular rings can be used to focus X-rays[1]. The spatial resolution that can be achieved with these devices is of the order of the width of the outermost zone and is therefore limited by the smallest structure (20–40 nm) that can be fabricated by lithography today[2]. Here we show that a large number of pinholes distributed appropriately over the Fresnel zones make it possible to focus soft X-rays to spot sizes smaller than the diameter of the smallest pinhole. In addition, higher orders of diffraction and secondary maxima can be suppressed by several orders of magnitude. In combination with the next generation of synchrotron