Attosecond Double-Slit Experiment

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A new scheme for a double-slit experiment in the time domain is presented. Phase-stabilized few-cycle laser pulses open one to two windows (slits) of attosecond duration for photoionization. Fringes in the angle-resolved energy spectrum of varying visibility depending on the degree of which-way information are measured. A situation in which one and the same electron encounters a single and a double slit at the same time is observed. The investigation of the fringes makes possible interferometry on the attosecond time scale. From the number of visible fringes, for example, one derives that the slits are extended over about 500 as.

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The conceptually most important interference experiment is the double-slit scheme, which has played a pivotal role in the development of optics and quantum mechanics. In optics its history goes back to Young's double-slit experiment. Its scope was greatly expanded by Zernike's work and continues to deliver new insights into coherence to the present day [1]. One of the key postulates of quantum theory is interference of matter waves, experimentally confirmed by electron diffraction [2,3]. More than 30 years later, Jönsson was the first to perform a double-slit experiment with electrons [4]. Of particular importance for interpreting quantum mechanics have been experiments with a single particle at any given time in the apparatus [5,6]. More recent work has illuminated the fundamental importance of complementarity in which-way experiments [7] and of quantum information in quantum-eraser schemes [8].

In this Letter a novel realization of the double-slit experiment is described. It is distinguished from conventional schemes by a combination of characteristics: (i) The double slit is realized not in position-momentum but in time-energy domain. (ii) The role of the slits is played by windows in time of attosecond duration. (iii) These ''slits'' can be opened or closed by changing the temporal evolution of the field of a few-cycle laser pulse. (iv) At any given time there is only a single electron in the double-slit arrangement. (v) The presence and absence of interference are observed for the same electron at the same time.

Interference experiments in the time-energy domain are not entirely new. Interfering electron wave packets were created by femtosecond laser pulses [9]. Accordingly, the windows in time (or temporal slits) during which these wave packets are launched were comparable to the pulse duration. In the present experiment, in contrast, the slits are open during a small fraction of an optical cycle, which gives the attosecond width. A number of experiments, in particular, in intense-laser atom physics but also for Rydberg atoms and microwaves [10], can and have been interpreted in this spirit (for a review see, for example, [11]). In this Letter, however, the optical cycles are precisely tailored by controlling the phase of few-cycle laser pulses (also known as absolute or carrier-envelope phase). This provides an unprecedented degree of control for the double-slit arrangement. Not only are the principles of quantum mechanics beautifully demonstrated, it is also likely that applications exploiting interferometric techniques for measuring attosecond dynamics will emerge.

In the present experiment, argon atoms are ionized by intense (1×10^{14} W/cm^2) few-cycle 760 nm laser pulses. Photoelectrons emitted in opposite directions (''left'' and ''right'') are detected by two opposing electron detectors placed symmetric to the laser focus. The laser field is horizontally polarized, i.e., parallel to an axis defined by the electron detectors. Interference of temporally separated wave packets leads to a fringe pattern in the energy domain because time and energy are conjugated variables. Therefore, the electron kinetic energy needs to be measured, in our case via the time-of-flight method. The carrier-envelope phase of the field and thus its temporal evolution can be controlled by delaying the envelope of the pulse with respect to the carrier. This is accomplished by shifting a glass wedge into or out of the beam. The phase of the field is measured as described in [12].

Photoionization of atoms with an ionization threshold much greater than the photon energy is a highly nonlinear process. For intense fields, the first step can be described by optical field ionization. This immediately explains the generation of one attosecond window (or slit) in time per

half-cycle close to its extremum; see Fig. 1. By using phase-controlled few-cycle laser pulses [13], it is possible to manipulate the temporal evolution of the field, thus gradually opening or closing the slits, and controlling which-way information. Depending on the field, one or two half-cycles (or anything in between) contribute to the electron amplitude for a given direction and electron energy. This corresponds to a varying degree of which-way information and, accordingly, to a varying contrast of the interference fringes. The temporal slits leading to electrons of given final momentum are spaced by approximately the optical period. This results in a fringe spacing close to the photon energy.

Figure 2 displays measured electron spectra. In Fig. 2(a) the spectra recorded at the left and the right detectors are shown for $\pm$ cos-like and $\pm$ sin-like pulses as defined in Fig. 1. A problem in presenting such spectra is that they quickly roll off with increasing electron energy. This roll-off was eliminated by dividing the spectra by the average of all spectra over the pulse's phase. Clear interference fringes with varying visibility are observed as expected from the discussion above. The highest visibility is observed for $-$ sin-like pulses in the positive (right) direction. For the same pulses, the visibility is very low in the opposite direction. Changing the phase by π interchanges the role of left and right as expected. The most straightforward explanation—which will be detailed by a simple model below—is to assume that, for $-$ sin-like pulses, there are two slits and no which-way information for the positive direction and just one slit and (almost) complete which-way information in the negative direction. The fact that the interference pattern does not entirely disappear is caused by the pulse duration, which is still slightly too long to create a perfect single slit.

Under the conditions of this experiment, each argon atom emits at most one electron [14], whose various options of how to reach a given final state lead to interference. For sin-like pulses, these options correspond to a double slit in time in one direction and to a single slit in the other, and are created for each atom separately by the few-cycle laser pulse. Therefore, even though there is more than one argon atom in the laser focus, the experiment operates under single-electron conditions. On the scale of the electron's deBroglie wavelength, other atoms are far away and, moreover, randomly distributed. This is in contrast to the double slit in space where the beam has to be sufficiently dilute to ensure a one-electron measurement.

The fringe pattern exhibits an envelope. From Fig. 2 a width of this envelope of about 4 fringes is inferred. Just as for a double-slit experiment, the width of this envelope can

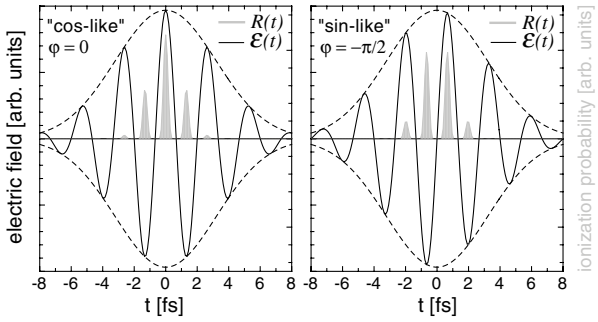

FIG. 1. Temporal variation of the electric field $\mathcal{E}(t) = \mathcal{E}_0(t) \times \cos(\omega t + \varphi)$ of few-cycle laser pulses with phase $\varphi = 0$ ("cos-like") and $\varphi = 3\pi/2$ ("sin-like"). In addition, the field ionization probability $R(t)$, calculated at the experimental parameters, is indicated. Note that an electron ionized at $t = t_0$ will not necessarily be detected in the opposite direction of the field $\mathcal{E}$ at time t_0 due to deflection in the oscillating field.

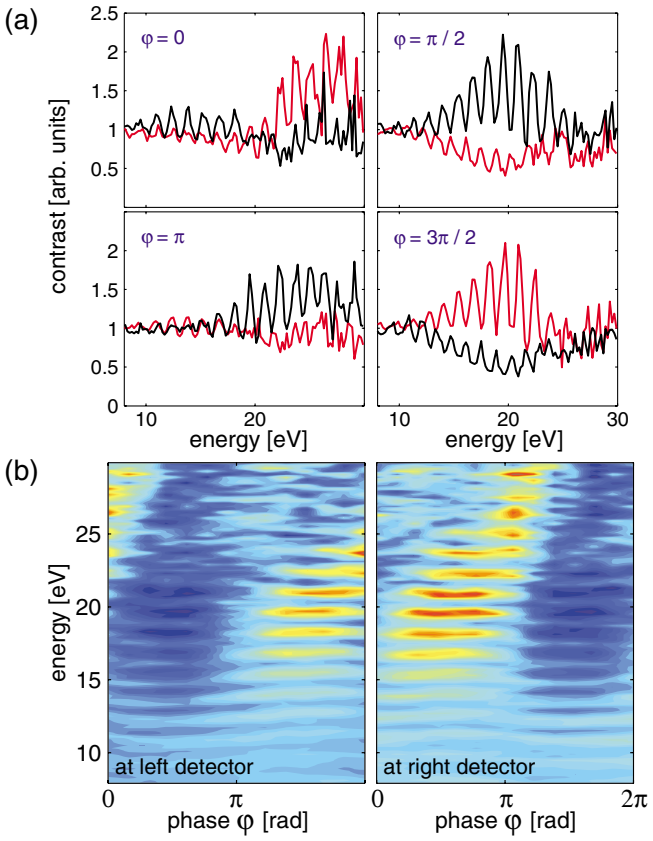

FIG. 2 (color). Photoelectron spectra of argon measured with 6 fs laser pulses for intensity 1×10^{14} W/cm^2 as a function of the phase. Panel (a) displays the spectra for $\pm$ sin- and $\pm$ cos-like laser fields. The red curves are spectra recorded with the left detector (negative direction), while the black curves relate to the positive direction. For $\varphi = \pi/2$ the fringes exhibit maximum visibility for electron emission to the right, while in the opposite direction minimum fringe visibility is observed. In addition, the fringe positions are shifted. Panel (b) displays the entire measurement where the fringe visibility is coded in false colors. The fringe positions vary as the phase φ of the pulse is changed. This causes the wavelike bending of the stripes in these figures. Both panels, in principle, show the same information because a phase shift of π mirrors the pulse field in space and thus reverses the role of positive and negative direction. However, the data shown were recorded simultaneously but independently as the phase φ was varied between 0 and 2π.

be associated with the width of the slits. It will turn out, however, that what is seen here is not the width of the slit. Rather, each slit can be resolved into a pair of slits whose separation is inversely proportional to the width of the envelope.

Disregarding the changing visibility, the peaks observed in the spectra resemble the well-known above-threshold ionization (ATI) peak pattern and they are certainly related to them. However, the relationship is nontrivial: Besides the visibility of the fringes, their positions also change as the phase of the field is varied. Details of the fringe shifts can be seen in Fig. 2(b). For conventional ATI, one would try to explain this in terms of the ponderomotive potential U_P. This does not work here, because the concept of the ponderomotive potential, which is defined as the cycle-averaged kinetic energy of an electron quivering in an oscillating electric field, is questionable in the few-cycle regime.

In contrast, an interpretation based on the double-slit analogy is obvious. In a spatial double slit, the fringe pattern would shift if a phase shifter (for light, simply a glass plate) were placed in front of one of the slits. For nontrivial particle trajectories one needs to consider the action S along the particles' paths and use the fact that the particles' phases are given by $S/\hbar$.

In order to exclude other scenarios, we compare the experimental data with results obtained by numerically solving the time-dependent Schrödinger equation (TDSE) in three spatial dimensions. The calculation was done at the experimental conditions for an effective argon potential in the single active electron approximation [15]. The calculated spectra were divided by the phase-averaged spectrum, using the same procedure that was applied to the experimental data underlying Fig. 2. The numerical TDSE result for the right-going electrons is shown in Fig. 3, to be compared with the experimental result in the right panel of Fig. 2(b). Virtually all details found in the measurement can also be found in the calculation. This confirms that single-electron dynamics are sufficient to explain the fringes.

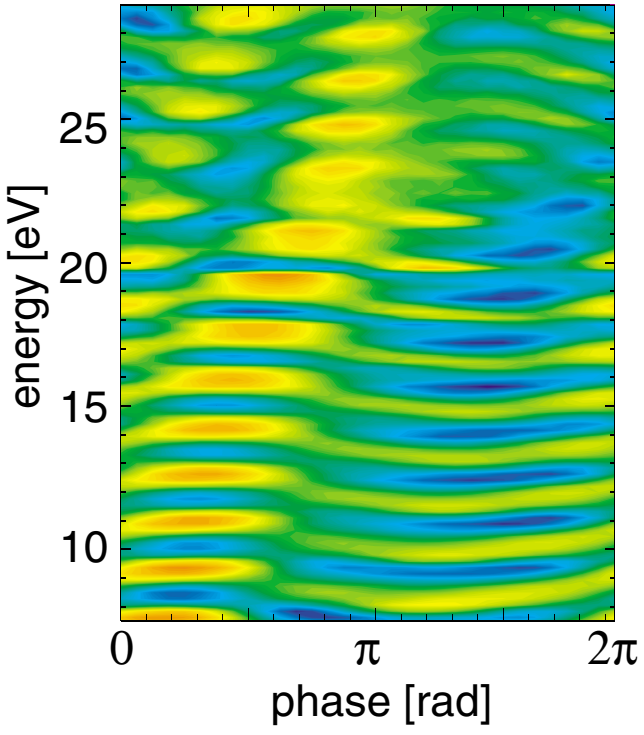

FIG. 3 (color). The result of a numerical solution of the time-dependent Schrödinger equation, as described in the text. This figure should be compared with the right panel of Fig. 2(b).

For an interpretation we resort to a classical model, the so-called simple-man's model [16], which—together with various extensions and modifications—has proven to be extremely helpful for understanding strong-field laser-atom interaction; for a review see, for example, [11]. Alternatively, Keldysh-type models, which can be interpreted as an approximation of Feynman's path integral [17], could be used. Respective results can be found in the literature: Ref. [18] predicts effects analogous to those described in this Letter for circular polarization. References [13,19,20] explain related classical effects for electromagnetic extreme ultraviolet radiation produced by high-harmonic generation. For the present problem, the classical and the quantum model lead to qualitatively the same results. The various assumptions of the model outlined in the following are fulfilled only approximately. Indeed, systematic deviations between the experiment and the model are the consequence.

The classical model assumes that an electron is launched into the continuum at some time t_0. Another crucial assumption of the model is that the electron's velocity is zero at $t = t_0$. This means that $p - eA(t_0) = 0$, where p is the momentum of the electron at the detector, $A(t)$ the vector potential of the field, and $e = -|e|$ the electron's charge. It is largely this relationship that explains the double-slit behavior of few-cycle photoionization.

The strength of the classical model is the intuitive insight it provides. In the following, hardly more than the number and position of the solutions of $p - eA(t_0) = 0$ for given p will be used in order to explain the double-slit behavior. The respective solutions $t_0(p)$ in a Keldysh-type model are complex, thus allowing access to classically forbidden electron energies. However, the symmetry of these solutions stays the same and so do the results qualitatively.

In Fig. 4 the vector potential $A(t)$ is drawn for a $-\sin$-like pulse. The solutions of $p - eA(t_0) = 0$ and thus all trajectories of momentum p that could interfere can be found by intersecting $A(t)$ with a horizontal line at p/e. It is now important to recall that a fringe pattern of maximal visibility requires *equally strong* slits, i.e., minimal which-way information. For a few-cycle pulse whose envelope is maximal at $t = 0$, the "strength" of a slit decreases very quickly with increasing $|t_0|$ and is essentially zero for $|t_0| > 2\pi/\omega$ because of the highly nonlinear dependence of photoionization on the field strength. As the

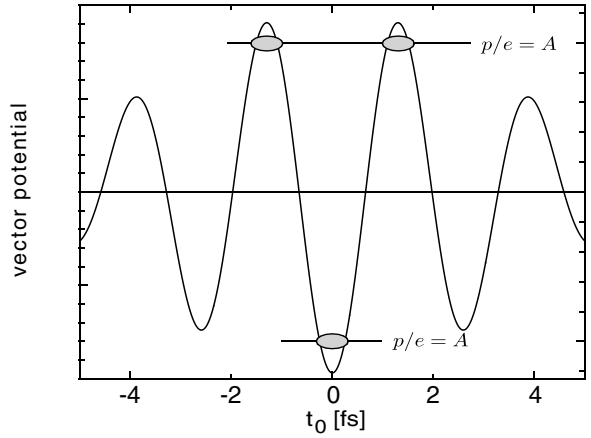

FIG. 4. Vector potential of a − sin-like few-cycle pulse. The temporal slits are given by the condition $p - eA(t_0) = 0$. For a − sin-like pulse, this leads to a double slit in the negative (since $e = -|e|$) direction and a single slit in the opposite direction. Each slit can be resolved into a pair of slits.

maximum of the pulse envelope was chosen to be at $t = 0$, the condition of equally strong slits is identical to requiring that the solutions of $p - eA(t_0) = 0$ be symmetric with respect to $t = 0$. This is the case for − sin-like pulses with electrons emitted in the negative direction and for + sin-like pulses with electrons emitted in the positive direction. For both cases, the respective opposite direction can be considered to act like a single slit as long as the pulse is short enough.

Figure 4 also shows that each slit is, on closer inspection, a pair of slits and that the temporal separation of these subslits depends on the electron energy [21]. The experimental data actually provide a measurement of the time difference of the two subslits, which is approximately 500 as. This is a first simple example for using interferometry on the attosecond time scale in order to investigate electronic dynamics. In addition, Fig. 2(a) shows that the relative phase of the subslits is different for sin-like and cos-like pulses, resulting in a shift of the fringe envelope.

It should be noted that the simple-man's model does not reproduce the dependence of the fringe visibility on electron energy as observed experimentally and in the solution of the TDSE. Therefore, the direction for which interference is predicted by the simple model may be wrong, depending on the energy. Using several theoretical models (3D TDSE, 1D TDSE, Keldysh-type, and classical), we were able to show that this is not a fundamental problem of the classical theory. Rather, it is an effect of the atomic binding potential, which obviously deflects the outgoing photoelectrons. The solution of the *one-dimensional* TDSE (which cannot deflect) with a soft-core potential, for example, agrees qualitatively very well with the classical and a Keldysh-type model. In particular, it does not show a pronounced energy dependence of the fringe visibility, and it predicts the interferences in the same direction as the simple models.

In conclusion, we have realized an intriguing implementation of the double slit in the time domain. The observation of interference and its absence at the same time for the same electron is a beautiful demonstration of the principles of quantum mechanics. It should also be noted that attosecond slits were used and that the interferograms reflect the attosecond dynamics of electronic transitions. Further experimental and theoretical progress should make it possible to use interferometric techniques for attosecond science.

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