

Temporal Young's interference experiment by attosecond double and triple soft-x-ray pulses

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We study a temporal version of Young's interference experiment by attosecond soft-x-ray pulses. The photoelectron energy spectra by attosecond double pulses exhibit an interference pattern, since we have no information on which pulse has generated the electron. We can re-establish the "which-way" information and control the interference visibility by changing the electron's momentum with phase-stabilized laser pulses, by an amount depending on the time of ionization. Moreover, if we use a triple pulse, we can realize a situation where the electron passes through a single and a double slit simultaneously to the same direction and is observed by the same detector.

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One of the most fundamental and, simultaneously, mysterious concepts in quantum mechanics is the wave-particle duality [1], i.e., a wavelike nature of matter. The interference of electron de Broglie waves observed in a Young's double-slit experiment [2] is its most successful confirmation. The recent observation of subfemtosecond soft-x-ray pulses [3–7], based on high-order harmonic generation [8–11] (HHG), has opened a way to generate electron bursts, i.e., temporal slits of an attosecond time scale. A train of attosecond light pulses [12] can be obtained by superposing several high harmonics of an intense infrared femtosecond laser pulse. Such a pulse train is composed of light bursts repeated every half cycle of the laser optical field, with a discrete spectrum containing only odd multiples of the laser frequency. If the laser pulse is sufficiently short, the generated train contains only a few or even single attosecond pulse. When applied to atoms, such a pulse train produces periodic emission of ultrashort electron bursts through photoionization. The photoelectron energy spectrum by a double pulse exhibits discrete peaks corresponding to the harmonic components. These peaks can also be viewed as a quantum interference pattern [13], which appears because we do not know which of the two pulses has generated the observed electron (which-way information). Thus, the photoionization by a double pulse is nothing but a double-slit experiment in the time domain [13].

In this Rapid Communication, by direct solution of the time-dependent Schrödinger equation (TDSE) for a hydrogen atom, we show that we can re-establish which-way information by changing the momentum of each electron bunch by a different amount with a phase-stabilized laser pulse, and control the degree of the quantum interference of electron de Broglie waves through the phase of the laser pulse. Although femtosecond laser pulses were previously used to create interfering electron wave packets with a temporal width comparable to the laser pulse duration [13], the electron bursts or temporal slits considered in the present

study are of much shorter, sub-optical-cycle time scale.

To study the interaction of a hydrogen atom with soft-x-ray pulses $E_X(t)$ and a laser pulse $E_L(t)$, linearly polarized in the z direction, we numerically solve the TDSE in the length gauge,

$$i \frac{\partial \Phi(\mathbf{r}, t)}{\partial t} = \left[-\frac{1}{2} \nabla^2 - \frac{1}{r} + z[E_X(t) + E_L(t)] \right] \Phi(\mathbf{r}, t), \quad (1)$$

using the alternating direction implicit (Peaceman-Rachford) method [14]. To prevent reflection of the wave function from the grid boundary, the wave function is multiplied by a mask function after each time step [15]. In typical calculations, we use a grid with a maximum radius of 1125 a.u. and a maximum number of partial waves $l_{\max}=50$. The grid spacing is 0.25 a.u., and the time step is 8.0×10^{-3} a.u. The photoelectron energy spectra are determined with the help of a spectral analysis [16] of the atomic wave function.

The solid line of Fig. 1(a) shows an example of an attosecond soft-x-ray double pulse $E_X(t) = E_{X0}(t) \sum_{q(\text{odd})=23}^{31} f_q \cos[q\omega(t + \pi/2\omega)]$, composed of the 23rd to 31st harmonics of a laser pulse with a wavelength of 800 nm ($\hbar\omega=1.55$ eV). The harmonic mixing ratio is $(f_{23}, f_{25}, f_{27}, f_{29}, f_{31}) = (0.10, 0.24, 0.32, 0.24, 0.10)$, and the common amplitude envelope E_{X0} is assumed to be of a Gaussian temporal profile centered at $t=0$ with a full width at half maximum (FWHM) of 1 fs. The spectrum of the photoelectron by the double pulse is shown in Fig. 1(b). The spacing between adjacent interference fringes $\Delta E = 2\hbar\omega$ and the temporal "distance" between the two slits $\tau = \pi/\omega$ satisfy the relation $\Delta E\tau = \hbar$. Let us consider a situation in which a laser field $E_L(t) = E_0(t) \cos(\omega t + \phi)$ is superposed, where E_0 , ω , ϕ denote the amplitude envelope, carrier frequency, and carrier-envelope phase (CEP), respectively. We assume that the carrier frequency is the same as that of the fundamental laser light for the HHG. The presence of an intense light field affects the ejected electrons' motion and can be used to probe the emission time [17,18]. It follows from a simple classical analysis that the final momentum of an electron released at $t=t_r$ is changed by $\Delta p(t_r) = -eA_L(t_r)$ along the laser polariza-

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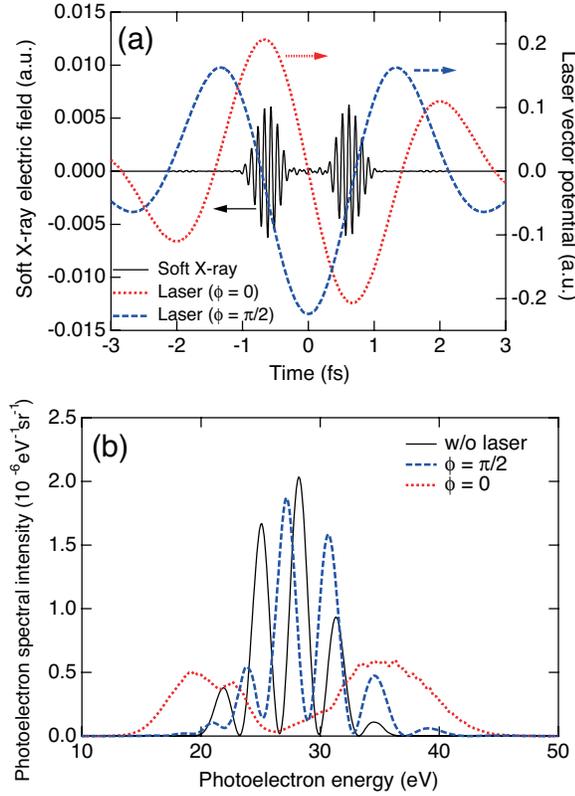


FIG. 1. (Color online) Visibility control in the attosecond double-slit experiment. (a) Temporal variation of an attosecond soft-x-ray double pulse (solid line), composed of the 23rd to 31st harmonics of a laser pulse with a wavelength of 800 nm. In addition, a temporal profile of the vector potential of the phase-stabilized laser electric field $E_L(t) = E_0(t)\cos(\omega t + \phi)$ with the CEP $\phi=0$ (dotted line) and $\pi/2$ (dashed line). The peak intensity is $5 \times 10^{12} \text{ W/cm}^2$. (b) The calculated kinetic energy spectra of photoelectrons ejected to the direction $\theta=0$ by the double pulse without (solid line) and with the energy-shearing laser pulse (dashed and dotted lines).

tion [17,18], with e and $A_L(t) = -(E_0(t)/\omega)\sin(\omega t + \phi)$ being the primitive charge and the vector potential in the Coulomb gauge, respectively. Correspondingly, the final kinetic energy is given by

$$W_f(t_r) \approx W_i - \sqrt{2e^2 W_i / m} A_L(t_r) \cos \theta, \quad (2)$$

where m and W_i are the electron's mass and initial energy, and θ is the angle between its final momentum vector and the laser polarization. Based on this principle, a series of elegant experiments have been done on time-resolved atomic inner-shell spectroscopy [19], atomic transient recorder [6], and direct measurement of light waves [20]. The energy shear induced by an applied laser field has also been proposed for the characterization of attosecond pulses through interferometry [21,22]. Since the two electron bursts are separated by half a cycle of the laser optical field, the energy of each wave packet is sheared by a different amount, in general. Therefore, the angle-resolved electron energy spectrum contains which-way information, in principle. Although Lindner *et al.* [23] has reported a similar idea, they have considered the

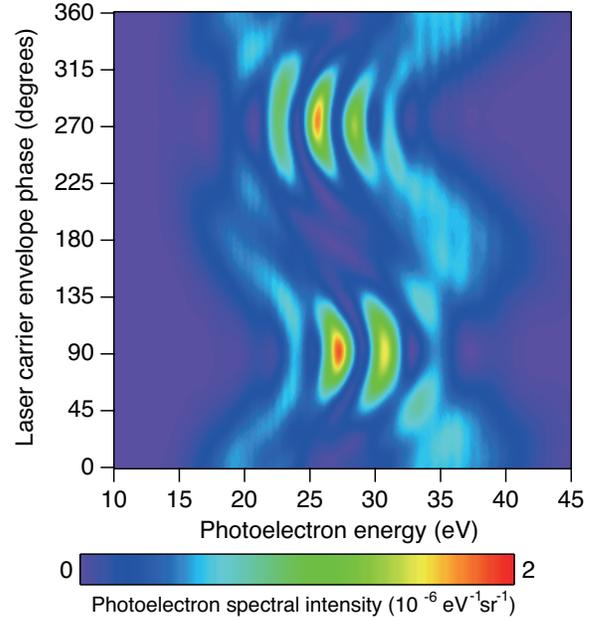


FIG. 2. (Color online) A series of calculated kinetic energy spectra of photoelectrons ejected to the direction $\theta=0$ by the attosecond soft-x-ray double pulse whose electric field is displayed in 1(a), as a function of the CEP ϕ of the energy-shearing laser pulse, in false-color representation.

same laser pulse both for electron burst generation and for momentum change. By contrast, in our case, electron bunches are produced by soft-x-ray pulses while the energy-shearing agent is a laser pulse, whose parameters such as the CEP can be manipulated independently of the soft-x-ray pulses. This is more analogous to the conventional Young's double-slit experiment.

It follows from Eq. (2) that the difference in the final energy between the electrons released by the first pulse and the one by the second pulse is given by

$$\Delta W = 2\sqrt{2e^2 W_i / m} A_L(t_1) \cos \theta \quad (3)$$

with t_1 being the arrival time of the first pulse. For a fixed value of t_1 , this takes the maximum value $|\Delta W| = 2\sqrt{2e^2 W_i / m} |A_L(t_1)|$ at an angle $\theta=0$ and π . When the CEP ϕ of the laser pulse is tuned in such a way that $\omega t_1 + \phi = \pi/2$, $|\Delta W|$ is the largest. If its value is larger than that of the spectral width of the electron energy, the two electron bunches are energetically separated, and, thus, we can tell which pulse has ejected the electron. The dotted curve in Fig. 1(b) demonstrates that this recovery of which-way information erases the interference patterns in the energy spectrum. For the case of $\omega t_1 + \phi = 0$, which gives $\Delta W = 0$, on the other hand, the interference fringes reappear as can be seen from the dashed curve in Fig. 1(b). It should be noted that the electron energy spectrum corresponds neither to the intensity spectrum of the combined soft-x-ray and laser electric field nor to two-photon two-color ionization as used for attosecond pulse characterization [4,24,25]. The ionization is solely due to the attosecond soft-x-ray pulses, and the laser pulse is an agent to shear the electron energy by its

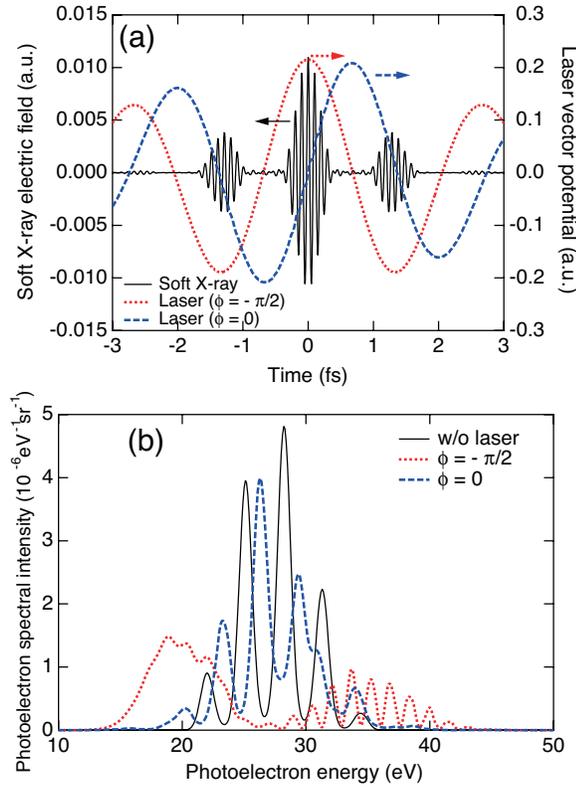


FIG. 3. (Color online) Visibility control in the attosecond triple-slit experiment. (a) Temporal variation of an attosecond soft-x-ray triple pulse (solid line), composed of the 23rd to 31st harmonics of a laser pulse with a wavelength of 800 nm. In addition, a temporal profile of the vector potential of the phase-stabilized laser electric field $E_L(t) = E_0(t)\cos(\omega t + \phi)$ with the CEP $\phi=0$ (dashed line) and $-\pi/2$ (dotted line). The peak intensity is $5 \times 10^{12} \text{ W/cm}^2$. (b) The calculated kinetic energy spectra of photoelectrons ejected to the direction $\theta=0$ by the triple pulse without (solid line) and with the energy-shearing laser pulse (dashed and dotted lines).

vector potential, not by the ponderomotive energy [26]. Since ΔW in Eq. (3) is a function of the CEP ϕ , the degree of the which-way information also depends on ϕ . Thus, the fringe visibility can be controlled by the CEP of the laser pulse as can be seen in Fig. 2.

Let us next consider, as the simplest but intriguing extension, photoionization by a train of three attosecond soft-x-ray pulses $E_X(t) = E_{X0}(t)\sum_{q(\text{odd})=23}^{31} f_q \cos q\omega t$, whose temporal profile is shown in Fig. 3(a) in the presence of the energy-shearing laser pulse whose vector potential is also shown in Fig. 3(a) for the CEP $\phi=-\pi/2$ and 0. $E_{X0}(t)$ is assumed to have a FWHM of 1.5 fs. In the case of $\phi=-\pi/2$, the situation is the copresence of a single-slit scheme, in which the second electron wave packet receive a negative energy, and a double-slit scheme, in which the first and third wave packets receives the same amount of positive energy. The resulting photoelectron spectrum in the direction $\theta=0$, shown as a dotted curve in Fig. 3(b), is composed of two distinct parts: the lower energy part without interference, and the higher energy part with interference fringes. An interpretation based on which-way information is rather obvious. If the observed electron has an energy around 20 eV, we can tell that it is

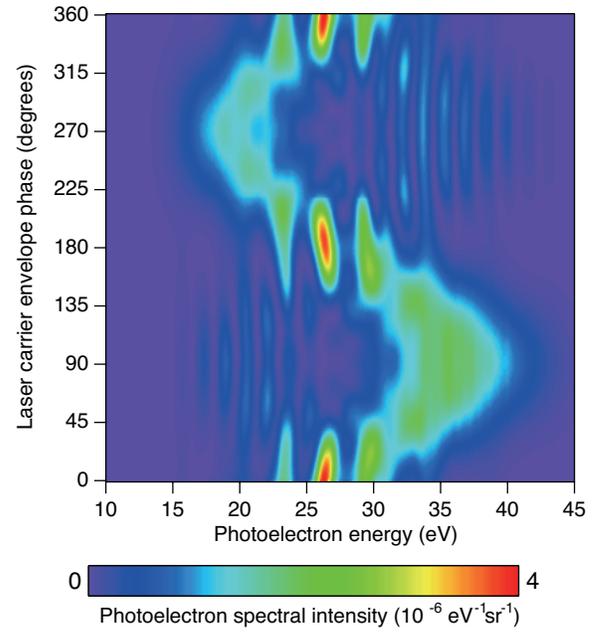


FIG. 4. (Color online) A series of calculated kinetic energy spectra of photoelectrons ejected to the direction $\theta=0$ by the attosecond soft-x-ray triple pulse whose electric field is displayed in 3(a), as a function of the CEP ϕ of the energy-shearing laser pulse, in false-color representation.

released by the second pulse. The spectrum contains, therefore, no interference pattern. If the electron energy is observed to be around 35 eV, on the other hand, it means that it is knocked free either by the first pulse or by the third. We cannot specify, however, which. The situation corresponds to a double-slit experiment in the time domain, and thus, an interference pattern is present. Since the interval between the first and third pulses is a laser optical cycle $\tau=2\pi/\omega$, the interference fringes are separated by $\Delta E = h/\tau = \hbar\omega$, rather than $2\hbar\omega$ for the case of double pulse. It should be stressed that this corresponds to a unique situation in which the same electron encounters a single and a double slit simultaneously. The copresence of a single- and double-slit experiments has also been reported in Ref. [23]. In the scheme of Ref. [23], however, electrons that encounter single- and double slits are emitted in opposite directions. In the present scheme, on the other hand, the results of both single and double-slit schemes can be recorded as a single photoelectron energy spectrum detected in the same direction by the same detector. Let us now turn to the case of $\phi=0$. In this case, the vector potential of the laser electric field nearly vanishes at all the three photoelectron bursts. Thus, the situation is similar to the case without the laser field, and the photoelectron spectrum exhibits interference fringes with a spacing of $2\hbar\omega$ [dashed curve in Fig. 3(b)]. Figure 4 illustrates how interference patterns in the photoelectron spectrum detected in direction $\theta=0$ vary as a function of the laser CEP ϕ . The transition between a triple-slit scheme and the copresence of a single- and a double-slit schemes is beautifully displayed.

In conclusion, we have presented a theoretical analysis of photoionization by attosecond soft-x-ray pulses as a temporal version of the double-slit and triple-slit experiment. The vis-

ibility of interference fringes, i.e., discrete peaks in the photoelectron energy spectrum can be controlled by varying the magnitude of which-way information through momentum change with a phase-controlled laser pulse. Moreover the simultaneous presence of single- and double-pulse schemes in the same spectrum for the case of the triple-pulse scheme is a remarkable manifestation of the wave-particle duality of the electron. The present results suggest that the combination

of state-of-the-art ultrashort soft-x-ray pulse generation and laser control techniques would become a new tool to manipulate attosecond dynamics of the electron, not only as a particle but also as an interfering quantum wave.

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