Methods for the measurement of the duration of high-harmonic pulses

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We present two methods for measuring the duration of ultrashort high-harmonic pulses produced by an intense laser pulse propagating in an atomic gas. Both methods rely on the nonlinearity of ionization of atomic gases by optical laser pulses. In the first method, the same medium is used for the dual purpose of generating and measuring the high harmonics. The second method uses atomic photoionization in an intense low-frequency laser field to produce the optical analog of a ''streak camera'' capable of subfemtosecond resolution. [S1050-2947(97)03810-9]

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I. INTRODUCTION

With ultrashort pulses from conventional lasers already approaching the duration of a single cycle of visible light, obtaining shorter pulse durations requires shorter wavelengths and new nonlinear media. High-harmonic generation in ionizing gases promises to provide both requirements at once. Controlling the high-harmonic generation process has been proposed as a way to generate attosecond pulses [1–5]. Although the wavelengths emitted through high-harmonic generation are currently limited to the soft-x-ray range, high harmonics are already used [6] as an alternative to synchrotrons. This coherent extreme ultraviolet (XUV) source also promises a high temporal resolution and easy synchronization with short visible pulses.

Pulse duration measurement at these wavelengths requires a new technology especially for the subfemtosecond durations. The standard techniques of autocorrelation are currently impossible to use because of the lack of suitable optical materials at these wavelengths and of efficient nonlinear processes at the intensities expected. Cross correlations [7–11] are limited to a resolution of the order of the duration of the reference pulse. Conventional XUV streak cameras have a resolution of ~ 1 ps [12–14] and cannot be much improved without drastic modifications.

We propose two methods for measuring the duration of high-harmonic pulses. Both rely on basic characteristics of strong-field laser-atom interactions, summarized in Sec. II.

In Sec. III we present a measurement method that uses the same nonlinear medium for the high-harmonic generation and for the simultaneous measurement of the duration of the high-harmonic pulse. This can be done by controlling the polarization state of the fundamental laser field. Section III deals with three different issues. First, we present the effect of a modulation of the ellipticity of the fundamental pulse on the high-harmonic generation and show how it allows us to extract the duration of the high-harmonic pulse. Second, we present the resolution of the technique and its limitations. Third, we consider how the method could be used experimentally.

The second method, presented in Sec. IV, can be used to measure ultrashort pulses created by any method. It relies on the principle of the streak camera, but with two important modifications. First, the standard photocathode is replaced by an atomic gas [15,16] placed directly in the region where the measurement occurs. Second, the fast deflecting field, used for the measurement, is a strong laser field. After a general presentation of the method, we consider the measurement of pulses that are (i) shorter and (ii) longer than the optical period of the deflecting field. We then investigate the resolution and limitations of the technique and illustrate its capabilities by considering typical experimental conditions.

Section V concludes the paper by summarizing its main results and highlighting some promising implications.

II. STRONG-FIELD–ATOM INTERACTION AND HIGH-HARMONIC GENERATION

The field-atom interactions that are of interest in this paper can be described by three main steps [17–19] in the low-frequency–strong-field limit. This limit is defined by $U_p \gg I_p \gg \hbar\omega$, where $U_p = e^2 E_{\max}^2 / 4 m_e \omega^2$ is the average kinetic energy (i.e., ponderomotive energy) of an electron (mass m_e and charge $-e$) oscillating in a strong field (of frequency ω and maximum amplitude $E_{\max}$) and I_p is the ionization potential of the atom. These three steps form the foundations of strong-field high-harmonic generation.

In the first step, the atom is separated into an ionic core and a quasifree electron. This step occurs if the electron tunnels through the potential barrier formed by the superposition of the Coulombic field of the core with the laser electric field. Tunnel ionization occurs with an instantaneous rate determined by the amplitude of the field and is quantitatively described by the Ammosov-Delone-Krainov (ADK) theory [20,21]. Strong-field tunnel ionization is the first step of the high-harmonic generation process.

In the second step, the electron is accelerated by the strong electric field (also called the driving field) and gains energy. The laser electric field has the general form $\vec{E}(t) = E_0(t)\{\cos(\omega t)\vec{x} + \varepsilon(t)\sin(\omega t)\vec{y}\}$, where ε is a parameter describing the field ellipticity ($\varepsilon = 1$ for circularly polarized light and 0 for linearly polarized light) and $E_0(t)$ is the envelope of the field. This plane-wave description of the laser

field is only valid in the short-pulse limit where the electron motion in the field is confined to a small volume in which the laser field is uniform. To first order, the electron motion can be described classically [18]. The corresponding expressions for the electron velocity $\vec{V}(V_x(t),V_y(t))$ and position $\vec{R}(X(t),Y(t))$ are

$$X(t)=X_m\{-\cos(\omega t)\}+V_x^0 t+X_0, \tag{1a}$$

$$Y(t)=\varepsilon X_m\{-\sin(\omega t)\}+\varepsilon V_y^0 t+Y_0, \tag{1b}$$

$$V_x(t)=V_m\sin(\omega t)+V_x^0, \tag{1c}$$

$$V_y(t)=-\varepsilon V_m\cos(\omega t)+V_y^0, \tag{1d}$$

where $V_m=-eE_0/m_e\omega$, $X_m=-eE_0/m_e\omega^2$, and V_x^0, V_y^0, X_0, and Y_0 can be determined by assuming that $\vec{V}(t_0)=0$ and $\vec{R}(t_0)=0$, with t_0 being the time of ionization. The use of these classical equations and initial conditions is justified by good agreement with experiments [22–24] and for high-harmonic generation they are consistent with the results of a fully quantum mechanical treatment [25]. The quantities $V_x^0=-V_m\sin(\omega t_0)$ and $V_y^0=\varepsilon V_m\cos(\omega t_0)$ are directly measured in short-pulse above-threshold ionization experiments with low-frequency circularly polarized light as the final electron velocities [22,26]. Measuring V_x^0 and V_y^0 can form the basis of a measurement method with suboptical cycle resolution since they are a signature of the laser phase at the time of ionization.

A third step in the strong-field–atom interaction can occur if the electron is driven back into the vicinity of the parent ion. There the electron may recombine to the ground state, emitting a high-frequency photon. These three successive steps are the basis of high-harmonic generation. The periodicity of this coherent process permits emission of odd high harmonics of the fundamental only.

For efficient high-harmonic generation, other conditions such as phase matching [27,28] or depletion of the atomic medium [29,30] are very important. These conditions suggest a number of approaches to generating very short high-harmonic pulses. (i) High-harmonic generation requires ionization that is impossible until the laser intensity passes the ionization threshold intensity. It also becomes impossible once every atom is ionized (unless the field is intense enough to ionize the ions). Since tunnel ionization is a highly nonlinear process, the rapid turn-on (beginning of the ionization) and turn-off ($\sim$100% probability of ionization) of the single-atom response calculated in a one-dimensional configuration can lead to high-harmonic pulses significantly shorter than the fundamental pulse duration. (ii) Kan *et al.* [31] proposed to further reduce the high-harmonic duration by use of dispersion due to free electrons. This dispersion induces a phase mismatch that turns off the high-harmonic generation when the electron density is too high. The rapid turn-on of the atomic susceptibility (beginning of the ionization) combined with the rapid turnoff due to dispersion should squeeze the duration of high-harmonic pulses to approximately one optical period of the fundamental. (iii) High-harmonic emission requires electron-ion recollision and the probability of recollision can be controlled through the polarization of the fundamental. During the second step, the electron travels several nanometers away from the core before the field reverses its direction driving the electron back toward the ionic core. Compared to atomic dimensions, this distance is huge and a slight ellipticity in the driving field is sufficient to deflect the electron in the transverse direction. This deflection prevents recollision and reduces the emission of high harmonics [23,32–34]. This crucial ellipticity dependence of high-harmonic generation has been proposed as a way to create attosecond pulses [1]. The central purpose of this paper is to present methods for measuring the short pulses that should be obtained through high-harmonic generation.

III. A MULTISHOT MEASUREMENT METHOD BASED ON THE TEMPORAL CONTROL OF THE DRIVING PULSE ELLIPTICITY

Any measurement method must be able to determine the duration that a light pulse would have in the absence of measurement. Usually, the measurement is separated from the production process to prevent perturbing the production by the measurement. The following method relies on a very different approach since the measurement occurs during the light-emission process. It can be used for measuring the duration of high-harmonic pulses produced by a linearly polarized fundamental pulse.

A. Ellipticity modulation and measurement of the high-harmonic pulse duration

High-harmonic generation is only possible if three conditions are simultaneously fulfilled: (i) Ionization of atoms can occur, (ii) phase-matching conditions are fulfilled, and (iii) electron-ion recollision is possible. The high sensitivity of the third condition to the ellipticity of the polarization of the fundamental provides a way to introduce a time scale in the high-harmonic emission process by temporally modulating the ellipticity of the driving field. Provided that the maximum ellipticity is small ($\varepsilon=E_{0y}/E_{0x}\sim 0.1$), modulating the ellipticity can be done without affecting the conditions (i) and (ii) that determine the duration τ_{harm} of the high-harmonic pulse obtained with the linearly polarized fundamental.

High-harmonic generation depends crucially on the ellipticity of the driving pulse, its efficiency falling by $\sim$50% for $\varepsilon\sim 0.1$. This dependence has been extensively studied [23,32–34] and shows good agreement with the semiclassical model [23]. For small ellipticity and for harmonics well above the ionization potential, the high-harmonic yield (number of photons of a given harmonic) fits a Gaussian

$$Y(\varepsilon)=Y_0\exp(-\gamma\varepsilon^2), \tag{2}$$

with $\gamma\sim 60$ [33,34]. The ellipticity dependence of the high-harmonic yield changes very slowly with the high-harmonic order [33,34].

Since the ion and the electron recombine within one laser period after ionization [35], Eq. (2) applies to a time-dependent ellipticity provided that the ellipticity is approximately constant over one optical cycle. This restriction still allows us to modulate the ellipticity on a time scale that can be much shorter than the envelope of the fundamental pulse.

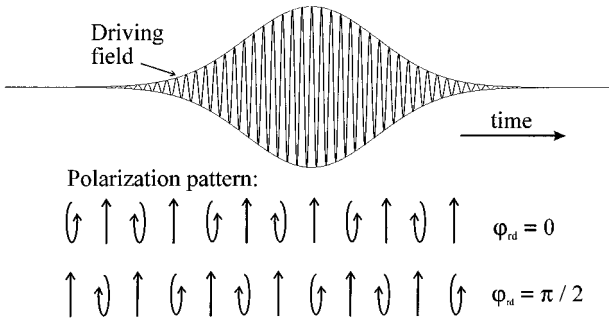

FIG. 1. Temporal evolution of the amplitude of the driving field and sketch of its polarization pattern. The position of the polarization pattern (relative to maximum of the envelope) is defined by the dephasing φ_{rd} between the two components of the driving field and is fixed for one shot. Shot-to-shot changes in the dephasing induce a global shift of the polarization pattern.

Perturbing the linearly polarized intense fundamental pulse (central frequency ω_0) with a weak perpendicularly polarized pulse (centered at a frequency $\omega_1 \neq \omega_0$, with $|\omega_1 - \omega_0| < \omega_0$, having an envelope similar to the envelope of the fundamental pulse and similar geometrical characteristics) leads to a total driving field with time-dependent polarization. The instantaneous ellipticity of the field is

$$\varepsilon(t,\varphi_{rd}) = \varepsilon_{\max} |\sin(\pi t/T_{\mathrm{mod}} + \varphi_{rd})|, \tag{3}$$

$T_{\mathrm{mod}} = \pi/(\omega_0 - \omega_1)$ being the (controllable) ellipticity modulation period and φ_{rd} the dephasing between the two colors (defined, for instance, at the maximum of the envelopes). In the following we consider the (experimentally simple) case of a phase φ_{rd} that is constant during each shot but changing randomly from shot to shot. Shot-to-shot changes of φ_{rd} shift the polarization pattern (Fig. 1).

In the measurement method studied in this section, we compare τ_{harm} (duration of the high-harmonic pulse obtained without the perturbing pulse) with T_{mod} via the effect of ellipticity modulation on the high-harmonic emission.

For $T_{\mathrm{mod}} \gg \tau_{\mathrm{harm}}$ the driving pulse ellipticity is quasiconstant during all the high-harmonic emission and the emitted signal depends on the instantaneous ellipticity at the time of high-harmonic emission. If the driving field is linearly polarized at that moment, then the high-harmonic signal is maximum. By contrast, the high-harmonic emission is reduced (by $\sim 50\%$ for $\varepsilon \sim 0.1$) if the driving field is elliptic at that time. Since the instantaneous ellipticity changes randomly from shot to shot, the high-harmonic signal fluctuates between these two extremes on a shot-to-shot basis.

For the other extreme ($T_{\mathrm{mod}} \ll \tau_{\mathrm{harm}}$), where the ellipticity oscillates many times during the high-harmonic emission, the high-harmonic signal is reduced for all φ_{rd}. In this case, the high-harmonic signal is stable (and attenuated) on a shot-to-shot basis. Therefore, recording the shot-to-shot fluctuations of the high-harmonic signal emitted by a perturbed pulse allows us to compare the periodicity of the ellipticity modulation with the duration of the high-harmonic signal emitted by the unperturbed fundamental.

We can compute the emitted high-harmonic signal by using the Gaussian ellipticity dependence [Eq. (2)] of the high-harmonic emission and the known temporal evolution of the ellipticity. The number of detected photons in the high-harmonic pulse is

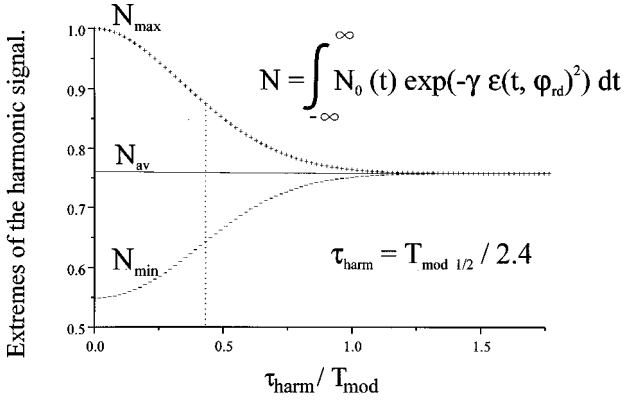

FIG. 2. High-harmonic signal emitted as a function of the ellipticity modulation period in units of the duration τ_{harm} of the unperturbed high-harmonic pulse. Depending on the initial phase φ_{rd} between the two fields, the high-harmonic signal is between the maximum (+) and the minimum (−) signal plotted here. The signal averaged over every phase (full line) is quasiconstant and must be kept constant in any experiment in order to ensure that the effective ellipticity is constant. A measurement of the ellipticity modulation period $T_{\mathrm{mod1/2}}$ that reduces the amplitude of the fluctuations to 50% of the maximal value allows us to obtain the high-harmonic pulse duration through the relation $\tau_{\mathrm{harm}} = T_{\mathrm{mod1/2}}/2.5$.

$$N(\varphi_{rd}) \propto \int_{-\infty}^{+\infty} Y_0(t) \exp[-\gamma \varepsilon(t,\varphi_{rd})^2]\, dt, \tag{4}$$

where $\varepsilon(t,\varphi_{rd})$ is given by Eq. (3), $Y_0(t)$ is the temporal envelope of the high-harmonic emission that would be obtained without the perturbing pulse, and φ_{rd} is the phase characteristic of one shot. In equation (4), we add the photons emitted at different times. This is justified in the time domain. In the frequency domain, the ellipticity modulation changes the spectrum and can transfer energy from one harmonic to another. A measurement of the number of photons emitted should therefore be done without selecting a well-defined frequency. However, recent calculations [36] have shown that even an ellipticity of 30% associated with an ellipticity modulation period of $1.2T_0$ (T_0 is the optical period of the fundamental) efficiently couples neighboring harmonics only. For smaller ellipticities and longer modulation periods the effect should be even smaller and therefore detecting simultaneously a few harmonics should be sufficient to justify this approximation.

For a given ellipticity modulation period T_{mod} and high-harmonic duration τ_{harm} the number of photons emitted [Eq. (4)] fluctuates from shot to shot as a function of φ_{rd}. These fluctuations are bounded by two extreme values $N_{\max}(T_{\mathrm{mod}}, \tau_{\mathrm{harm}})$ and $N_{\min}(T_{\mathrm{mod}}, \tau_{\mathrm{harm}})$. In Fig. 2 these extreme values of the high-harmonic signal are displayed as a function of $\tau_{\mathrm{harm}}/T_{\mathrm{mod}}$ assuming that the unperturbed high-harmonic pulse has a Gaussian temporal shape and that the maximal ellipticity is $\varepsilon_{\max} = 0.1$. The fluctuations of the signal are large for $T_{\mathrm{mod}} \gg \tau_{\mathrm{harm}}$. They decrease as T_{mod} decreases and disappear for $T_{\mathrm{mod}} \ll \tau_{\mathrm{harm}}$. The essential point is

TABLE I. Ratio of $T_{\mathrm{mod1/2}}$ to the high-harmonic pulse duration τ_{harm} for different high-harmonic pulse shapes.

Ratio	Lorentzian	sech^2	Gaussian	square
$T_{\mathrm{mod1/2}}/\tau_{\mathrm{harm}}$	3.7	2.6	2.4	1.7

that measuring $T_{\mathrm{mod1/2}}$, the modulation period for which the amplitude of the fluctuations is reduced to 50% of the maximal amplitude, gives the duration τ_{harm} of the high-harmonic pulse through the relation $\tau_{\mathrm{harm}} \sim T_{\mathrm{mod1/2}}/2.6$ (in Fig. 2, $T_{\mathrm{mod1/2}}$ is indicated by a dotted line).

As with standard autocorrelation measurement, we assume a given shape for the high-harmonic pulse in order to extract its duration from the measurement. We calculated the measured duration for several pulse shapes $Y_0(t)$, all having a full width at half maximum of τ_{harm}. The different values obtained for $T_{\mathrm{mod1/2}}/\tau_{\mathrm{harm}}$, displayed in Table I, change only by a factor ~ 2 between the extreme cases where the pulse shape is square or Lorentzian. Without any precise knowledge on the high-harmonic pulse shape, we can estimate its duration through the relation

$$\tau_{\mathrm{harm}} = T_{\mathrm{mod1/2}}/2.5.$$

B. Resolution and limitations of the method

Three factors might impose fundamental limitations on the method. They are (i) perturbation of the ionization process, (ii) dispersion and other phase shifts during propagation of the driving pulse, and (iii) speed at which we can modulate the ellipticity.

Both the single-atom response and phase-matching conditions are sensitive to the ionization rate. Therefore, it is critical that the perturbing pulse be weak so that it does not significantly change the ionization rate. In the case where the maximal ellipticity is 0.1, the maximum amplitude of the total electric field lies between 1 and 1.005, increasing the ADK ionization rate [21] by only 5% of its value (calculated for helium and an intensity of 8×10^{14} W cm^{-2}). Still, $\varepsilon = 0.1$ is sufficient to reduce the high-harmonic signal by $\sim 50\%$ of its maximal value [Eq. (4)].

Equation (1) indicates that the transverse drift of the electron and therefore the probability of recombination depend on the frequencies. To ensure a constant maximum transverse deflection, the effective maximum ellipticity $\varepsilon_{\mathrm{eff}} = E_y(\omega_1)/E_x(\omega_0)(\omega_1/\omega_0)^2$ must be kept constant. For a perturbing pulse centered at 670 nm and a fundamental at 800 nm, $(\omega_1/\omega_0)^2$ is as big as 1.4. To ensure a constant effective ellipticity $\varepsilon_{\mathrm{eff}}=0.1$, the ellipticity must be $\varepsilon = E_y(\omega_1)/E_x(\omega_0) \sim 0.14$ and the weak field starts to perturb the ionization process. Measurement with a perturbing pulse on the infrared side of the fundamental lowers the required perturbing field strength and is preferred.

In principle, propagation of the laser pulses in the ionizing atomic medium could influence the measurement in a number of ways. For example, the polarization pattern must have a fixed position compared to the fundamental envelope for one laser shot. This imposes a fixed phase relation between the two perpendicular components of the driving field during all the high-harmonic emission process. However, different frequencies have different phase velocities in a dispersive medium and dephasing occurs through propagation. Dephasing also appears because of geometrical effects especially around the focus of a Gaussian beam.

In practice, however, no phase changes between the fundamental and measurement pulses are important provided that the phase-matching condition necessary for efficient high-harmonic generation is approximately fulfilled. In an ionizing atomic medium the propagation of the fundamental is more affected than the propagation of the high harmonics that is quasi-vacuum-like. The phase-matching conditions depend on the phase difference between fundamental and high harmonics and high-harmonic emission becomes very inefficient when the propagation of the fundamental differs significantly from a vacuumlike propagation in the volume where high harmonics are emitted [27]. Therefore, observing efficient high-harmonic generation implies that propagation is quasi-vacuum-like for both fundamental and perturbing pulses (which have similar propagation characteristics) and that dephasing between them can be ignored. As an example, when the phase-matching condition $L\Delta k = \pi$ (L is the length of the atomic medium where the dispersion is mainly due to free electron and Δk is the wave-vector mismatch between the fundamental and the high harmonics) is fulfilled, the error on the measurement due to the shift of the polarization pattern is approximately T_{mod}/q, where q is the order of the highest detected harmonic.

The main limitation of this measurement method arises from the requirement of slowly modulating the ellipticity since we have assumed that the ellipticity of the driving field is approximately constant over one optical period. In order to quantify the speed with which the ellipticity can change, we performed classical simulations of the electron trajectory. We found that the electron motion is not significantly perturbed by the temporal evolution of $\varepsilon(t)$ until the modulation period is as short as $2.5T_0$. Our criteria for a ''significant modification of the electron motion'' relies on the difference between the transverse electron deflection in a field with time-dependent ellipticity and in a field with constant ellipticity (the value of this ellipticity is the value of the time-dependent ellipticity at the moment of ionization). For $T_{\mathrm{mod}} \geqslant 2.5T_0$ this difference is less than 20% of the maximal deflection. In conclusion, the slow ellipticity variation allows us to measure high-harmonic pulses as short as one optical period of the fundamental. This is the basic resolution of this technique.

C. Experimental considerations

A weak tunable perturbing pulse, synchronized with the main intense pulse, is available in many laboratories. For instance, it can be obtained with continuum generation and subsequent reamplification or optical parametric amplification. Fundamental and perturbing pulses are then accurately synchronized and the phase between them is random.

Very short ellipticity modulation periods are feasible. For instance, combining a fundamental pulse centered at 800 nm with a perturbing pulse centered at 1 μm or 670 nm leads to a modulation period $T_{\mathrm{mod}}=2.5T_0$, sufficient to measure high-harmonic pulses as short as one optical period of the fundamental.

Since the measurement method relies on recording the shot-to-shot fluctuations of the perturbed high-harmonic signal, it implies that the unperturbed signal must fluctuate very little. Statistical fluctuations of the unperturbed signal are reduced if no frequency selection is made since it allows an average over a larger number of XUV photons. Intensity fluctuations in the fundamental can also be reduced through a careful prefiltering of the laser shots.

Before concluding this section, we note that averaging over many shots of same phase φ_{rd} would greatly improve our measurement method. This requires knowledge of the phase φ_{rd}. Knowledge of φ_{rd} is possible either by controlling it or by measuring it. This technology is just emerging. A perturbing pulse and a driving pulse originating from the same ultrashort pulse and separately amplified [37] have a well-defined and controllable phase relation. A method to fully characterize the time-dependent polarization state of a pulse has also just been reported [38].

In summary, we have shown that time-dependent ellipticity can be used to measure the duration of the high-harmonic emission from experiments using a simple linearly polarized fundamental pulse. If a time-dependent ellipticity is used to reduce the duration of the high-harmonic pulses [1], an alternative method is necessary for the measurement. The next section presents such a method.

IV. A SUBFEMTOSECOND STREAK CAMERA

XUV streak cameras are useful for the characterization of XUV short pulses. Their resolution is presently limited to $\sim$1 ps [12–14]. The resolution limitations are given by the basic design of the streak cameras and a new design is required to overcome these limitations. Strong-field physics and atomic ionization give the opportunity for drastic improvement in time resolution.

A. Principle of a streak camera

The basic principle of a streak camera is to create an electron replica of a light pulse by illuminating a photocathode and to measure the duration of the electron pulse with electron optics [12]. Currently, there are two basic limitations to the resolution. The first comes from the spread in the initial electron velocities and the necessity of transporting the electron pulse to a deflection region without deformation. The second limitation is due to the long rise time ($\sim$100 ps) of the deflecting field used to characterize the temporal evolution of the electron pulse.

The two following modifications allow drastic improvement of the ultimate resolution of streak cameras. First, to overcome the limitations due to electron transport, we propose to use an atomic gas as the electron source [15,16]. The gas can be placed directly in the region where the fast deflection occurs. Second, to increase the sweep rate of the deflecting field, we propose to use an intense low-frequency laser field. With a characteristic sweep time of the order of several femtoseconds (for a wavelength of 10 μm the optical period is $\sim$33 fs), a low-frequency field allows us to measure the duration of subfemtosecond pulses. Furthermore, the low-frequency deflecting field and the ionizing field (the pulse that we want to measure) can copropagate. If they copropagate with the same velocity, a long interaction length

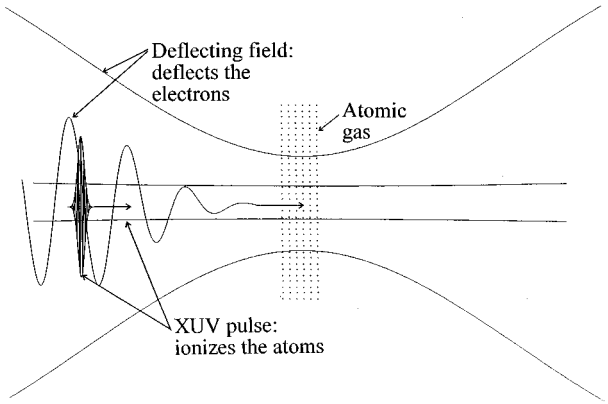

FIG. 3. Schematic of the subfemtosecond streak camera. An XUV pulse and a low-frequency field copropagate in an atomic gas. The XUV photons ionize the atoms and the low-frequency field deflects the electrons in a way that depends on the moment at which the atoms are ionized. Recording the distribution of electron velocities or energies gives access to the shape of the XUV pulse.

can be used for the measurement while simultaneously keeping a good resolution. Even with these important modifications, the basic principle of the measurement is similar to standard streak cameras (Fig. 3). A short ionizing pulse (XUV pulse) creates an electron pulse replica by single-photon ionization of the atoms and the duration of the electron replica is measured with a rapidly changing field (the low-frequency field). In the following, we assume that the continuum of states has no structure over the bandwidth of the ionizing pulse.

In Sec. II we discussed the motion of an electron released from rest in an intense electric field showing that the time of ionization can be determined through a measurement of the drift motion of the electron. An electron released at t_0 with an initial kinetic energy U_{k_0} $[U_{k_0}=\frac{1}{2}m(V_{0x}^2+V_{0y}^2)]$ will move with its drift velocity

$$V_{dx}=V_{0x}-V_m(t_0)\sin(\omega t_0), \tag{5a}$$

$$V_{dy}=V_{0y}+\varepsilon(t_0)V_m(t_0)\cos(\omega t_0) \tag{5b}$$

once the deflecting field is off.

This plane-wave description is valid in the short-pulse limit [39,40]. In this limit, the deflecting pulse is short enough (typically less than 1 ps) that the electron has insufficient time to drift by a significant fraction of the focal spot diameter while the pulse is on. In the opposite (long-pulse) limit, the electron leaves the high-intensity domain and therefore gains energy, the ponderomotive energy $U_p=e^2E_0^2/4m_e\omega^2$, by sliding over the field gradient [40]. Although the long-pulse limit can be used when the interaction volume is very confined, in this paper we consider only the short-pulse limit. The initial kinetic energy of the electron ($U_{k_0}=h\nu_{\mathrm{XUV}}-I_p$) determines the resolution of this measurement method and is explicitly considered in Sec. IV D. In the following, we assume that this energy is small compared to the drift energy that the electron acquires from the deflecting field.

Equations (5) show that the electron birth time t_0 determines three parameters [ωt_0, $V_m(t_0)$, and $\varepsilon(t_0)$]. Measuring

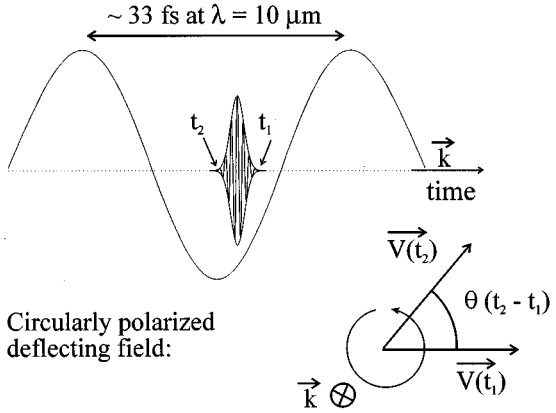

FIG. 4. For an XUV pulse shorter than the optical period of the deflecting field the angular spread of the electron velocity distribution depends on the duration of the ionizing pulse. For a deflecting pulse centered at $\lambda = 10\ \mu$m (CO_2 laser) a 2π angular deflection correspond to a duration of $\sim$33 fs.

distributions defined by each of these parameters provides three approaches for extracting the distribution of electron birth time and therefore the temporal shape of the XUV pulse. Exploiting these different parameters is the subject of the next two sections, which consider, successively, the cases where the XUV pulse is (i) shorter and (ii) longer than the optical period of the deflecting field.

B. Subcycle measurement: Use of the phase of the deflecting field

The kinetic energy of the photoelectron depends on the electron time of birth t_0 through the phase of the field (ωt_0). This phase dependence has been shown by Tate, Papaioannou, and Gallagher [41] in an experiment where they created free electrons at a controllable phase of a (long-pulse) low-frequency field. By changing the phase ωt_0 they changed the kinetic energy of the electrons. This phase dependence can be used with either a circularly or a linearly polarized deflecting field.

In a circularly polarized low-frequency deflecting field an electron released at t_0 is deflected in a direction $\theta(t_0)$ that is perpendicular to the direction of the field at t_0 [22] and in the plane of polarization. An electron released δt later drifts in a direction $\theta(t_0+\delta t)=\theta(t_0)+2\pi\delta t/T_0$, where T_0 is the optical period of the deflecting field. If an XUV pulse controls the electron release, the measurement of the distribution of angular deflection is a measurement of the pulse duration (Fig. 4). The optical period provides a direct time scale for the measurement. A 10-μm CO_2 deflecting field ($T_0 = 33$ fs) is well suited to the measurement of pulses of few femtoseconds or less since a 1-fs duration implies an $\sim 12°$ wide angular distribution. Measuring angular distributions allows us to do the measurement in a volume of gas where the amplitude of the deflecting field is not uniform.

A linearly polarized deflecting field is also interesting if the XUV pulse is shorter than a quarter of an optical period of the deflecting field. The electrons are primarily deflected in the direction of the polarization [42] and the shape of the ionizing pulse can be determined from the electron kinetic-energy distribution. A well-defined direction of deflection allows for good detection efficiency without using complex detectors. Provided that the energy of the XUV photons is well known, a linearly polarized deflecting field combined with a detector with spatial and temporal resolution allows us to determine the initial electron velocity V_0 and therefore to minimize the limitations due to the uncertainty on this velocity. However, there are major disadvantages of using a linearly polarized deflecting field. First, there is no automatic calibration of the time scale as opposed to the case of a circularly polarized deflecting field. A multipulse calibration is therefore necessary. Second, the velocity change is not linear with time and evolves as a sinusoidal function. This must be taken into account in the interpretation of the measurement and precludes the use of shots where the electrons have the maximal drift velocity.

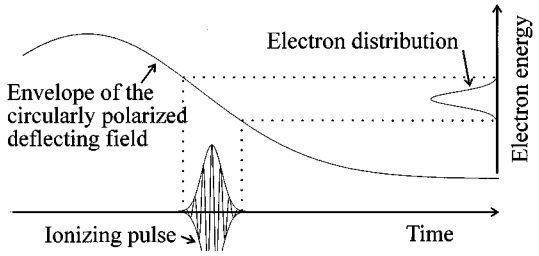

FIG. 5. For an XUV pulse longer than the optical period of the deflecting field, the envelope of the deflecting field allows us to obtain the duration of the pulse. The kinetic energies of the electrons depend on the amplitude of the deflecting field at the moment of ionization and the distribution of kinetic energies gives the shape of the ionizing pulse.

Both approaches are single-shot measurements if the phase of the deflecting field is not fixed compared to the envelope of the short pulse on a shot-to-shot basis. Locking the phase of an infrared field with the envelope of a femtosecond pulse is now becoming feasible [43] and could allow multishot measurements.

C. Multicycle measurement

XUV pulses longer than the optical period of the deflecting field can also be measured by using changes in the envelope of the deflecting field. The measurement is then based on $V_m(t_0)$ or $\varepsilon(t_0)$ [Eq. (5)].

For a circularly polarized deflecting field, the final drift energy of an electron released at t_0 is $U_d(t_0) = \frac{1}{2}e^2E_0^2(t_0)/m_e\omega^2 = \frac{1}{2}m_eV_m(t_0)^2$. Assuming a well-known rise time of the deflecting field (CO_2 pulses having an $\sim$2-ps rise time are possible to create and amplify [22,44]), a measurement of the distribution of kinetic energies of the electrons is a measurement of the duration of the ionizing pulse (Fig. 5). For measurement methods relying on electron kinetic energy, all electrons must be produced in a volume where the deflecting field is uniform.

In a linearly polarized deflecting field, electrons are primarily ejected along the polarization direction. However, it is possible to create a linearly polarized field with a time-dependent direction of polarization. Recording the angular distribution of photoelectrons gives access to the duration of the photoionizing pulse through the rotation rate of the polarization axis. One way to create a deflecting linearly polarized pulse with a time-dependent polarization axis is to split

the initial pulse in two pulses perpendicularly polarized, to delay one of them by approximately the duration of the pulse, and to adjust their relative phase so that the two perpendicular fields oscillate in phase. This can be done with a Michelson interferometer or simply by using a birefringent crystal [45]. The direction of the linear polarization at a given time is then defined by the ratio of the two pulse envelopes.

Short intense low-frequency pulses can be created with optical ultrashort pulses [44]. Their envelopes are then naturally accurately synchronized with ultrashort pulses and methods relying on changes in the envelope of the deflecting field can be used for multishot measurements.

D. Resolution limits

The subfemtosecond streak camera requires that (i) the XUV pulse creates a perfect electron replica of itself, that is, the atomic ionization rate must be constant over the entire bandwidth of the ionizing pulse, and (ii) since propagating fields are used, the phase (subcycle measurement) or group (multicycle measurement) velocities of the deflecting field must be the same as the group velocity of the XUV pulse, that is, phase advances (dispersion) must be considered. However, the typical necessary gas pressure (tens of mTorr), propagation length, and low fraction of ionization ensure that the light propagation in the measurement medium is very close to the ideal vacuum propagation.

The use of a short deflecting pulse implies that the bandwidth of this pulse is large. However, this measurement method requires that the frequency ω of the deflecting pulse is well defined since the measured drift velocity depends on ω. An uncertainty $\delta\omega$ on the frequency implies an uncertainty $\delta V_m = V_m(\delta\omega/\omega)$ for the photoelectron drift velocity. Since the bandwidth $\delta\omega$ of a Fourier transform pulse is linked to its duration τ by $\delta\omega = 0.315(2\pi/\tau)$, using a deflecting pulse longer than 30 optical period of the deflecting field implies a relative uncertainty $\delta V_m/V_m$ smaller than 1%. Such a pulse duration is compatible with the short-pulse limit and allows an accurate measurement.

We now consider the influence of V_0, the initial velocity of the electron at t_0 [Eq. (5)]. For simplicity, we restrict ourselves to the case where the kinetic energy U_d that the electron acquires from the deflecting field is bigger than the initial kinetic energy U_{k_0} ($U_{k_0} = h\nu_{\mathrm{XUV}} - I_p = \frac{1}{2} m_e V_0^2$). The condition $U_d \gg U_{k_0}$ ensures that the electron deflection is mainly due to the low-frequency field. Since $U_d \sim U_p$ this is equivalent to $U_p \gg U_{k_0}$.

If the initial angular velocity distribution is isotropic, the uncertainty on the total energy of an electron released at a given time is $\Delta U \sim \sqrt{U_{k_0} U_d}$. This uncertainty determines the resolution of the streak camera. This isotropic case leads to the maximum uncertainty on the electron energy and gives a lower bound for the resolution (this is the case considered in the following). For the specific case where the single-photon transition occurs from an s state with a linearly polarized XUV pulse, this uncertainty is reduced to $\Delta U \sim \sqrt{U_{k_0} U_d}/5$, when the low-frequency field is polarized perpendicularly to the XUV field. The resolution limit τ_{res} of the streak camera is the time needed to increase the electron drift energy U_d by $\Delta U[U_d(t) - U_d(t+\tau_{\mathrm{res}}) = \Delta U]$. For an electron kinetic-energy distribution determined by the phase ωt_0 of a linearly polarized deflecting field, the drift energy changes from zero to the maximum value $U_{d\,\max}$ in a quarter of an optical period of the deflecting field $T_0/4$. With $U_{d\,\max} = \alpha U_{k_0}$, the typical time needed for the energy to change by ΔU is $\tau_{\mathrm{res}} = T_0/2\pi\sqrt{\alpha}$. Combining this value of τ_{res} with $U_{d\,\max} = \frac{1}{2} e^2 E_0^2/m\omega^2$, $T_0 = 2\pi/\omega$, and I, the intensity of the low-frequency field, leads to

$$\tau_{\mathrm{res}}\ (\mathrm{fs}) = 1.22\sqrt{\frac{U_{k_0}\ (\mathrm{eV})}{I\ (\mathrm{TW/cm^2})}}.$$

The initial kinetic energy can be reduced down to the bandwidth of the XUV pulse $\delta U = \hbar\,\delta\omega$ by using an atom whose ionization potential matches the wavelength of the ionizing pulse. The vicinity of the ionization threshold is usually a region where the photoionization cross section changes rapidly. This restricts the bandwidth of the pulses that can be measured since the cross section must be constant over the entire bandwidth of the XUV pulse. However, this restriction is not too severe. For helium, for instance, the cross section changes only by a factor of ~ 2 when the energy of the XUV photon changes from 25 to 35 eV [46]. When the uncertainty on the initial electron kinetic energy is reduced down to the bandwidth of the XUV pulse, we obtain

$$\tau_{\mathrm{res}}\ (\mathrm{fs}) = 1.22\sqrt{\frac{\delta U\ (\mathrm{eV})}{I\ (\mathrm{TW/cm^2})}}.$$

As outlined in Sec. IV B, the uncertainty on the initial kinetic energy can also be reduced to $\delta U = \hbar\,\delta\omega$ by using a linearly polarized deflecting field and a detector with spatial and temporal resolution. This is correct provided that electrons released in the deflecting field without initial kinetic energy ($U_{k_0} = 0$) are deflected in the direction of the polarization as they are in the classical model. The angular spread associated with tunnel ionization in a low-frequency field was considered by Delone and Krainov [42]. For the experimental case considered below, it leads to an angular spread of $\sim 2°$ in the direction of deflection and is not a limitation.

Improving the resolution for the measurement of a short pulse of a given bandwidth requires an increase in the intensity of the low-frequency field. However, its amplitude is restricted since the deflecting field must not ionize the atoms by itself. The maximal amplitude of the deflecting field that allows us to neglect the ionization that it induces depends on many experimental parameters (volume of gas irradiated, pressure of the gas, and duration of the deflecting field). In many cases, an ionization rate $W_{\mathrm{ADK}} < 1\ \mathrm{s}^{-1}$ gives a very conservative estimate of the maximum field. According to the ADK tunnel ionization rate [21], the condition $W_{\mathrm{ADK}} < 1\ \mathrm{s}^{-1}$ is fulfilled provided that E_0 (V/m) $< 1.6\times 10^8 I_p^{3/2}$ (eV). This fixes the resolution limit of the streak camera:

$$\tau_{\mathrm{res}}\ (\mathrm{fs}) = 20\sqrt{\frac{\delta U\ (\mathrm{eV})}{I_p^3\ (\mathrm{eV})}}.$$

In a realistic experiment, the intense low-frequency field provided, for instance, by a CO_2 laser ($\lambda \sim 10\ \mu$m), can have an amplitude of $E_0 = 2\times10^{10}$ V/m ($\sim 5\times10^{13}$ W/cm^2) [22] when helium is the atomic photocathode ($I_p \sim 24.6$ eV). When the XUV photons have a mean energy of 50 eV, the ponderomotive energy (500 eV) is larger than the initial kinetic energy of the electron $U_{k_0} \sim 25$ eV. The photoionization cross section of helium around 50 eV is $\sigma \sim 2\times10^{-18}$ cm^2 [46] and therefore 10^6 XUV photons propagating in 20 mTorr of helium for 1 mm release approximately 100 photoelectrons. A 2-ps-long deflecting field propagating in the same medium with a maximum amplitude of 2×10^{10} V/m and a waist of 200 μm releases no electron. Under these conditions the direct measurement resolution is $\tau_{\rm res} \sim 0.8$ fs. Assuming an ~ 5 eV bandwidth for the XUV pulse (corresponding to a bandwidth limited duration of ~ 300 attoseconds), the resolution can be reduced down to ~ 400 attoseconds in the isotropic case and to 180 attoseconds for the specific case where the linearly polarized deflecting field is polarized perpendicularly to the XUV pulse.

V. CONCLUSION

In this work we presented two techniques for measuring the duration of ultrashort XUV pulses. The first, based on control of the ellipticity of the fundamental pulse that creates the high harmonics, has a resolution equal to one optical period of the fundamental. The second is the optical analog of the streak camera and has subfemtosecond resolution.

Both techniques can be reversed to create ultrashort pulses. The time-dependent polarization technique was already presented as a candidate for generating attosecond light pulses. The subfemtosecond streak camera technique can also be adapted to generate ultrashort pulses of electrons with well-defined characteristics. For instance, a circularly polarized CO_2 deflecting field deflects electrons in a 2π angle in 33 fs. If the electrons are initially released in less than 33 fs, the rapid change of the direction of the electron velocity can be used to select a portion of the electron bunch and therefore obtain an ultrashort electron pulse. This electron pulse created at a well-defined phase of the deflecting field could be accelerated by a second phase related deflecting field.

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