Cross-correlation measurement of femtosecond noncollinear high-order harmonics

A. Bouhal, R. Evans, G. Grillon, and A. Mysyrowicz

Laboratoire d’Optique Appliquée, Ecole National Supérieure des Techniques Avancées–Polytechnique,
Chemin de la Hunière, 91125 Palaiseau, France

P. Breger and P. Agostini

Service des Photons, Atomes et Molecules, Centre d’Etudes de Saclay, 91191 Gif Sur Yvette, France

R. C. Constantinescu and H. G. Muller

Stichting voore Fundamental Onderzoek der Materie (FOM), Nederlandse Organisatie voor Wetenschappelyk Onderzoek (NWO), Institute for Atomic and Molecular Physics, Kruislaan 407, 1098 SJ Amsterdam, The Netherlands

D. von der Linde

Department of Physics, Institut für Laser and Plasmaphysik, Postfach 103764, D4300, Essen, Germany

Received March 11, 1996; revised manuscript received August 21, 1996

The durations of extreme-ultraviolet femtosecond pulses (harmonics 21 and 27 of a Ti:sapphire femtosecond laser) have been measured by a cross-correlation technique. Photoelectron energy spectra resulting from ionization of helium atoms by one harmonic photon in the presence of the infrared laser field show sidebands that provide a cross-correlation signal from which the durations of the harmonic pulses can be obtained. The measurements yield a mean value of 130 fs for both harmonics. The accuracy of the method depends on the actual intensity dependence of the sidebands. © 1997 Optical Society of America [0740-3224(97)01204-6]

PACS number(s): 32.80.Hd, 32.80.Wr, 42.65.Ky, 06.60.Jn.

1. INTRODUCTION

Noble-gas atoms subjected to an intense infrared (IR) laser field radiate odd harmonics of this field up to high orders. From a microscopic standpoint, the dynamics of such a process is now well known to be intimately related to the classical motion of an electron in an electromagnetic field. For instance, the cutoff energy $IP + 3.2U_p$ (where $IP$ is the atomic ionization potential) is well predicted by the maximum classical kinetic energy of the electron at the position of its parent ion, $3.2U_p$, where $U_p = I/4\omega^2$ is the average oscillating kinetic energy of a free electron in a field of intensity $I$ and frequency $\omega$. Quantum mechanically, the delay between ionization and recollision translates into a phase shift between the induced harmonic polarization and the fundamental. The angular distribution and the temporal evolution of the harmonics result from both these phases and the propagation properties. Whereas the angular distributions have been investigated in detail, the wavelength range and the short duration of the harmonics make time measurements rather difficult because no direct technique is available.

Ionization of atoms by one high-order harmonic (HOH) photon and one fundamental photon, a two-color above-threshold ionization process, offers one of the few experimental methods for determining the temporal characteristics of a HOH in the subpicosecond domain. The corresponding photoelectron energy spectra contain peaks whose amplitudes are, in first approximation, proportional to the product of both beam intensities and therefore supply a cross-correlation signal from which the HOH duration can be obtained, provided that the fundamental signal is known. Practically, the correlation function is measured in a pump–probe arrangement in which the extreme ultraviolet (XUV) pulse pumps into the continuum an electron population that is subsequently probed by the fundamental pulse. The IR pulse is sometimes called the dressing pulse because the sidebands can be considered to result from transitions to a continuum dressed by the IR field. From the point of view of the pump–probe measurement two-color above-threshold ionization is ideally suited to the measurement of ultrashort XUV pulses because the continuum’s intermediate state has no lifetime that could limit the temporal resolution. The two-color above-threshold ionization is also interesting in itself and will potentially permit coherent control of the ionization process.

The feasibility of such a measurement of HOH durations in the subpicosecond range was demonstrated by Schins et al. The drawback of the setup described in that paper was that only the falling edge of the cross-correlation function could be sampled over a (small) num-
number of absorbed or emitted IR photons, Coulomb-corrected Volkov wave function. In the case approximation can be further improved by use of a tron wave function is described by a Volkov state. This free-electron wave function is the essence of the Keldysh quantum-mechanical treatment. The amplitude of a given sideband requires a full context of laser-assisted auger decay, later independently by Glover et al., and in the research reported here. In spite of many similarities, these last two setups differ in two aspects: in the optical paths of the harmonics and the dressing beams and in the elimination of the fundamental remaining in the pump beam after harmonic generation. The advantages and drawbacks of both methods are discussed later in this paper.

The principle of the two-color experiment involves using a high-frequency, weak-intensity beam to lift the electron energy into the continuum and a high-intensity, low-frequency beam to induce free–free transitions involving the absorption or emission of one or more photons after the initial ionization. In the electron energy spectrum such free–free transitions result in sidebands on both sides of the ionization peak, separated by the (low-frequency) photon energy. The energy of the electrons exposed to both frequencies is given (in atomic units) by

$$E_{q,n} = \omega_{XUV} \pm n \omega_{IR} - IP - U_p,$$ (1)

where $\omega_{XUV} = q \omega_{IR}$ (harmonic of order $q$) and $\omega_{IR}$ are the photon energies, $n$ is an integer that accounts for the number of absorbed or emitted IR photons, $IP$ is the ionization-potential of the target atom, and $U_p$ is the ponderomotive energy in the IR field.

According to lowest-order perturbation theory, the population of the $n$th sideband, $I_{sb}^{(n)}$, is proportional to the product of the intensity of the pump pulse, $I_{XUV}$, and of the probe pulse raised to the power $n$, $I_{IR}^n$. The lowest-order perturbation approximation, however, breaks down rapidly inasmuch as the free–free transitions are easily saturated. In the limit of high intensities for the IR beam or of high energy for the XUV photon a classical approximation is useful for calculating the envelope of the sidebands’ spectrum. However, to evaluate the amplitude of a given sideband requires a full quantum-mechanical treatment.

Neglecting the influence of the Coulomb field on the free-electron wave function is the essence of the Keldysh–Faisal–Reiss (KFR) approximation in which the electron wave function is described by a Volkov state. This approximation can be further improved by use of a Coulomb-corrected Volkov wave function. In the case of linearly polarized beams, for an electron ejected at an angle $\theta$ with respect to the polarization direction the transition probability can be expressed as $p_n^2 (1 + p_n^2)^4 J_n^2(x, y)$, where $p_n = \frac{\sqrt{2E_n}}{\omega_{IR}}$ is the electron’s final momentum and $J_n(x, y)$ is the generalized Bessel function, defined by

$$J_n(x, y) = \sum_{m=-\infty}^{+\infty} J_{n-2m}(x)J_m(y).$$ (2)

The arguments of $J_n(x, y)$ are $x(I_{IR}, n) = p_n \sqrt{I_{IR}/\omega_{IR}^2} \cos \theta$, where $\sqrt{I_{IR}/\omega_{IR}^2}$ is the amplitude of the oscillating motion of the photoelectron in the field and $y(I_{IR}) = U_p/2\omega_{IR}$. When the probe pulse is delayed by $\tau$ with respect to the harmonic pulse, the total population of the $n$th sideband is proportional to the convolution of the two pulses:

$$I_{sb}^{(n)}(\tau) \propto \int_{-\infty}^{+\infty} I_{IR}(\tau - \tau) I_{XUV}(\tau) d\tau.$$ (3)

The linearity of $I_{sb}^{(n)}$ on $I_{XUV}$ is difficult to check experimentally because the intensity of the harmonics can be controlled only indirectly, through the IR intensity. However, this intensity is much too low to induce nonlinear effects, and linearity will be assumed in the deconvolution. The function $f$ results from integrating the probability with respect to $\theta$:

$$f(I_{IR}) \propto p_n^2 [(1 + p_n^2)]^4 \times \int_0^\pi \sin \theta \cos^2 \theta J_n^2(x(I_{IR}), y(I_{IR})) d\theta$$ (4)

and goes asymptotically to the perturbative law $(I_{IR})^n$ in the limit of small arguments of the Bessel functions. From relation (3) it can be seen that measurement of $I_{sb}^{(n)}$ as a function of the time delay $\tau$ yields the temporal shape of the XUV pulse, $I_{XUV}(\tau)$, through a deconvolution operation, assuming that the IR pulse $I_{IR}(\tau)$ is known.

2. EXPERIMENT

The experimental setup is shown in Fig. 1. The output of a self-mode-locked Ti:sapphire laser is stretched to 400 ps and amplified in two stages. After recompression one obtains 60-mJ pulses of 160 fs at 800 nm (IR) with a repetition rate of 10 Hz. The temporal profile of the amplified laser pulses is best fitted with a squared hyperbolic secant function. The Michelson interferometer divides the incoming pulse into two. One of the beams provides the (IR) dressing field. The second generates the high-order harmonics. It was reported by Peatross et al. that harmonics can be efficiently generated by a beam with an annular profile.

To create an annular beam we block the central part (8 mm) of the pump beam in one arm of the Michelson interferometer. At the focus the spatial intensity pattern is practically the same as that of a Gaussian beam, except for small diffraction rings. The energy in this beam is controlled by an aperture and adjusted to a few millijoules, corresponding to intensities of $\sim 10^{14}$ W cm$^{-2}$. The high-order harmonics are generated by the central part and propagate in the direction of the fundamental beam. As the beam emerges from the focus the annular shape reappears. The remaining ring of the fundamental beam is then blocked by a 4-mm aperture placed 90 cm after the first jet, while the harmonic beam is allowed to pass.

In the second arm of the interferometer we therefore placed a variable-diameter pinhole. We were thus able to control to a certain extent the energy and the geometry
of the probe beam that propagates collinearly with the generated harmonic beam. Typical intensities needed to saturate the continuum–continuum transitions are of the order of $10^{11} - 10^{12}$ W cm$^{-2}$ and were easily achieved with pulse energies of a few microjoules. The length of the second arm could be changed in steps of 1 $\mu$m by use of a motorized stage. Both beams were actually focused by the 1-m focal-length lens into an argon-gas jet (1 mm thick, $1.1 \times 10^{-1}$ Torr, synchronized with the laser), albeit with different $f$-numbers (50 and 300, respectively) owing to the different apertures placed in the Michelson arms. The position of the focus with respect to the jet is not known with precision. The focusing lens is simply adjusted for maximum harmonic signal. Theory predicts drastic variations of both the spatial and the temporal profiles of the harmonic when this position is varied. Glover et al. have already reported such variations. A systematic study is in progress.

A major requirement of the cross-correlation method is to ensure good spatial overlap between the two pulses. In the experiment described by Glover et al., the harmonic and the dressing beams propagate along different pathways and are focused by different optics in the interaction region. In our setup we use a Michelson interferometer, which allows for the optical delay between the beam that is generating the harmonics and the dressing beam. After the interferometer the two beams are superposed in space, follow the same path, and are focused by the same spherical mirror. This guarantees an automatic overlap of the beams at focus. The drawback is that control of the harmonic and IR spot sizes is achieved through variation of the apertures that also control the intensities. The solution of independent paths and optics chosen by Glover et al., on the other hand, allows for independent adjustment of the foci at the expense of a more delicate alignment.

A second crucial feature is the separation of the harmonics from the IR radiation remaining in this beam after the harmonics were generated. To eliminate the fundamental beam after harmonic generation Glover et al. make use of an aluminum filter. This solution, though, has a number of drawbacks: The filter itself is a fragile element that can easily be damaged by the IR pulse; furthermore, it absorbs more than half of the XUV photons. As mentioned above, we use an annular profile for the IR beam that generates the harmonics. This permits filtering out of the unwanted fundamental by a simple aperture that lets through the harmonic beam. The advantage of this method is obvious. Its limitations come from the refraction of the IR by a too-strong free-electron density. The intensity of the IR pulse and the gas density must be therefore adjusted to prevent the electron density in the jet from being too high. This condition is required anyway for harmonic generation that saturates when the medium gets strongly ionized, because of both depletion of the neutral population and the defocusing of the IR beam by the free electrons. It is easy to detect if the fundamental beam does get refracted because then sidebands appear in the spectrum, independently of the delay in the interferometer. Eventually, if too much intensity is present on axis, the multilayer mirror can be damaged.

One major issue concerns the possibility that the measurement itself perturbs harmonic generation. This could happen if the probe beam significantly changed the ionization in the harmonic jet. We keep the intensity of the probe beam low enough to avoid this: Assuming a diffraction-limited spot size ($f$-number $\approx 400$) and an energy of 50 $\mu$J/pulse yields an intensity of $3.2 \times 10^{11}$ W cm$^{-2}$. The ratio of the two fields is $\sim 30$, and the intensity can therefore be changed by at most 6% when the two pulses interfere. Far from saturation, in the tunnel ionization regime a change of 6% in the field
changes the ionization rate by several orders of magnitude. However, the intensity in practice is close to saturation, and a change of a few percent is of no consequence to the rate and the electron density, except for a small shift in time.

The group-velocity dispersion in the harmonic gas jet itself is another subject of concern, inasmuch as the electron density gets close to neutral density during the course of the pulse. It is well known that both the harmonic and the probe pulses are affected by propagating through a medium with a time-dependent refractive index. This phenomenon is by itself a subject of study that can be addressed through the use of the cross-correlation method. We simulated the propagation of the fundamental beam in the jet by a finite-difference method and observed no significant change in the temporal profile for the experimental parameters. The helium target gas, though, is much more robust against ionization, and the dressing beam is totally ineffective in that respect. The harmonic pulse does produce free electrons, but their number is limited to fewer than 100 per shot (to avoid space charge), which corresponds to a low density that cannot disperse the pulses.

As Fig. 1 shows, the harmonic and the dressing beams must cross the interaction region before being focused by the multilayer mirror. Even unfocused, the XUV beam can then ionize the target gas, whereas the dressing beam will not have enough intensity to create sidebands, thus deteriorating the signal-to-background ratio. Therefore a 1-mm-diameter wire movable from outside is used to block the central part of the incoming beams, shadowing the sensitivity zone of the spectrometer. One adjusts the positioning of the wire by monitoring the electron signal and verifying that it is generated only by the beam reflected from the multilayer mirror. This ensures that only reflected beams with a good spatial overlap are actually producing a signal. Both of these beams are focused into a (second) pulsed gas jet of the target gas (helium) by a concave multilayer mirror (radius 70 mm) inside the electron spectrometer. Two different mirrors were used, one coated for 30 eV, the other for 40 eV. Both coatings consisted of alternating layers of silicon and molybdenum. Both mirrors have a reflectivity of \( \frac{1}{2} \) within a bandwidth of \( \pm 6 \) eV. The mirror focus is positioned \( \pm 2 \) mm past the spectrometer’s sensitivity zone. This distance provides a large enough interaction volumes and appropriate dressing intensity. The magnetic-bottle, time-of-flight electron spectrometer has been described elsewhere. Helium is chosen as a target gas because its ground state has no fine structure that would complicate the interpretation of the energy spectra.

3. RESULTS

Figures 2(a) and 2(b) show two spectra taken with the 30-eV mirror, and two Figs. 2(c) and 2(d) show two spectra taken with the 40-eV mirror. We recorded the spectra in Figs. 2(a) and 2(c) with only the harmonic beam present by blocking the other beam in the interferometer, whereas we took the spectra in Figs. 2(b) and 2(d) by using both the harmonic and the IR beams. The presence of the sidebands is clearly visible. In Figs. 2(c) and 2(d) an electron retardation potential of +15 V was used to eliminate the electrons resulting from the ionization by the 23rd and 25th harmonics that are actually more reflected than harmonic 27 within the bandwidth of the 40-eV mirror. No retardation was used for Figs. 2(a) and 2(b); hence three harmonics are observed in the case of the 30-eV mirror.

As Fig. 3 shows, the probe pulse has a FWHM duration of \( 160 \pm 10 \) fs. However, linear dispersion through the Michelson beam splitter, the lens, and the chamber entrance window results in an increase in the duration of what \( \sim 30 \) fs.

The experimental cross correlations are displayed in Fig. 4. They are fitted by a convolution between a squared hyperbolic secant function raised to a certain power (FWHM 190 fs, corresponding to the temporal profile of the laser inside the spectrometer) and a Gaussian whose FWHM is adjusted for best fit. The duration of the harmonic pulse derived from this procedure depends critically on the power law that directly affects the effective duration of the fundamental pulse: The smaller the exponent, the longer the effective duration of the probe pulse and hence the shorter the harmonic pulse for a given cross correlation. For the case of Gaussian pulses an analytical relation is easily derived: The convolution full width at half-maximum (FWHM) \( \tau_H \) is related to the FWHM \( \tau_F \) of one pulse and the FWHM \( \tau_{EF} \) of the second pulse, assumed raised to the power \( a \), through

\[
\tau_H = (\tau_F^2 - \tau_F^2/\alpha)^{1/2}. \tag{5}
\]

Therefore \( \tau_H \) is an increasing function of \( \alpha \). Note, however, that as \( \alpha \) goes to the limit \( (\tau_F/\tau_C)^2 \) the uncertainty on \( \tau_F \) for a given uncertainty on \( \alpha \) becomes infinite.

In the case of the 30-eV mirror (harmonic 21) the sideband used for the cross correlation is an incoherent sum of contributions from the three harmonics (19, 21, 23) reflected by the mirror. Labeling their intensities \( h_q \), we can write the population of the sideband used as the cross-correlation signal as

\[
I_{sb}^{(n)} = \sum_{q=19}^{23} h_q P_{q,n}^3 (1 + P_{q,n}^2)^{-4} \times \int_0^\pi \sin \theta \cos^2 \theta J_n^2 (\sqrt{n} I_{IR}, \theta) \, d\theta,
\]

where \( P_{q,n} \) stands for the momentum of the photoelectron that has absorbed a photon from the harmonic of order \( q \) and absorbed \( (+) \) or emitted \( (-) \) \( n \) IR photons. The \( q \) index runs over the three harmonics reflected by the mirror (19, 21, 23), and \( n \) takes the values \( +1, -1, \) and \( -3 \) for the three terms, respectively, of the sum. \( I_{sb}^{(n)} \) is a complicated analytical function of intensity, but it can easily be calculated numerically. One can use relation (6) to determine the dressing intensity by fitting the calculated sideband intensity to the experiment. The best fit is obtained for \( 2 \times 10^{-5} \) atomic units (7 \( \times 10^{11} \) W cm \(^{-2} \)), in reasonable agreement with the experimental estimate. The KFR theory appears to work rather well in this case, even though the photoelectron’s initial energy is not very high (8 eV for harmonic 21).
the IR intensity is quite moderate. The theoretical intensity dependence of $I_{sb}(n)$ near $2 \times 10^{-5}$ is well fitted by a power law of index 0.72. For the convolution procedure the analytical form in relation (6) is replaced in the calculation by this fit. Using the KFR exponent yields a value of 130 fs for the duration of harmonic 21, whereas using the lowest-order perturbation exponent yields a value of 150 fs. As we have already pointed out, this is a direct consequence of Eq. (5). The KFR and the linear least-squares fits are shown in Fig. 4.

For the 40-eV mirror (harmonic 27) we have attempted also to monitor the higher-order sidebands, possibly to take advantage of their higher effective indices (and hence the shorter effective probe durations). To obtain the overall best fit between the KFR and the experimental relative amplitudes of the sidebands one must use an intensity of $2.5 \times 10^{-5}$ atomic units, again in agreement with direct estimates. Experimentally, the first sideband scales as $I_{sb}^{0.7}$ (Fig. 5) and the intensity that will yield this slope from the KFR expression is $\sim 1.5 \times 10^{-5}$ atomic units. Using this exponent yields a duration of 130 fs, comparable with that of harmonic 21.

For the second and the third sidebands the KFR theory predicts intensity dependences that can be fitted near $2 \times 10^{-5}$ atomic units by power laws with exponents 1.4 and 2.3, respectively. Experimentally, slopes of 0.6 and 0.9 (Fig. 5), respectively, were measured, albeit with large uncertainties. This discrepancy is not clearly under-

Fig. 2. Electron energy spectra. 30-eV mirror: (a) long delay and (b) zero delay. The peak labeled sdb in (b) is the one used for the cross-correlation measurement. 40-eV mirror: (c) long delay and (d) zero delay. The numbers are the harmonic orders, the labels sdb1, sdb2, and sdb3 in (d) refer to sidebands 1, 2, and 3, respectively.

Fig. 3. Laser pulse obtained through a $\omega - 2\omega$ cross correlation (dots) fitted with a squared hyperbolic secant (curve) with a FWHM of 160 fs.
pulse, whereas the KFR indices lead to lower but still too-large values (200–220 fs). The fit that results from the experimental exponents yields a more reasonable duration of 130 ± 30 fs. The error bar reflects the uncertainty on the slopes added to the uncertainty of the fit. It is clear that knowledge of the effective nonlinear index is a crucial requirement of the method. In the absence of an accurate value it seems wise to use only the first sideband, whose intensity dependence is not far from linear and that, at any rate, can be measured with good precision.

The two harmonics appear to have the same duration within the uncertainty of the measurement. Both durations are longer than the one deduced from the previous experiment, albeit still shorter than the fundamental one. This discrepancy might result from the poorer accuracy of the first measurement. The difference could also be related, more interestingly, to coherence effects, making the harmonic pulse durations dependent on the position of the gas jet used for their generation relative to the focus (around which large phase shifts occur). Such an effect was recently observed by Glover et al. A few remarks on the cross-correlation method can readily be made: (1) The dynamic range can be increased only at the expense of creating more sidebands, which unfortunately goes with a lowering of the effective index and a decrease in the accuracy. (2) The resolution is imposed by the effective duration of the probe pulse. Knowledge of this parameter, i.e., of the power law followed by the sideband being monitored, is a prerequisite for the method, just as is the assumed time profile of the pulses. In principle it is possible to monitor higher-order sidebands to benefit from the corresponding shorter effective probe time. In practice, however, the effective slopes are smaller than the perturbative indices and result in increased effective probe durations. Furthermore, the smaller slopes increase the uncertainty of the measurement. The experiment shows that reliable information

![Fig. 4. Experimental cross correlations and fits corresponding to different intensity dependences (see text): (a) first sideband of harmonic 21; (b), (c) second and third sidebands, respectively, of harmonic 27. lopt, Lowest-order perturbation theory; exppt, experimental intensity dependence.](image)
contained in these sidebands is difficult to extract. (3) A long averaging time (typically 0.5 h with a 10-Hz repetition rate) is required for cross correlation. (4) The presence of more than one harmonic owing to imperfect multilayer mirrors complicates the analysis and adds to the uncertainty. More-selective mirrors should be soon available that will allow for some improvements.

This method, however, remains for the moment the only technique through which information about femtosecond XUV pulses can be obtained. In the future, when the harmonic beam reaches high enough brightness, the method should be replaced by autocorrelation methods with much higher temporal resolution.

In summary, continuous monitoring of the cross-correlation function between femtosecond XUV pulses (at 28 and 30 nm) and an IR pulse of 190 fs was achieved. In agreement with the theory of harmonic production, the pulse duration that results from these measurements is shorter than that of the fundamental pulse. It is not clear yet whether this duration is affected by coherence effects associated with the dipole phase shifts around the focus, as predicted by recent calculations. In spite of serious limitations, the technique of noncollinear high-order harmonics seems to be quite promising and useful.

ACKNOWLEDGMENTS

This research has been carried out with the partial support of European Economic Community Human Capital and Mobility programs ERB-PL 95-1313 and ERB-CT 95-0019. R. C. Constantinescu and H. G. Muller gratefully acknowledge support by the FOM, which is subsidized by NWO.

REFERENCES