

Phase-locked, time-delayed harmonic pulses for high spectral resolution in the extreme ultraviolet

M. Bellini

*Istituto Nazionale di Ottica Applicata, European Laboratory for Nonlinear Spectroscopy and
Istituto Nazionale di Fisica della Materia, Largo E. Fermi 6, 50125, Florence, Italy*

S. Cavalieri, C. Corsi, and M. Materazzi

*Istituto Nazionale di Fisica della Materia, Sezione di Firenze, Florence, Italy, and Dipartimento di Fisica and
European Laboratory for Nonlinear Spectroscopy, Università di Firenze, Largo E. Fermi 2, 50125 Florence, Italy*

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We present experiments in the time and frequency domains aimed at confirming the measured mutual phase coherence of time-delayed, collinear harmonic pulses. We show that pairs of phase-locked harmonic pulses of medium order can be generated for peak intensities up to $\sim 1.5 \cdot 10^{14}$ W/cm² in xenon, demonstrating the possibility of performing high-resolution spectroscopy in the extreme ultraviolet with Ramsey-like techniques.

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High-order harmonic generation is a promising new tool for the production of short-wavelength coherent radiation, and, because of its unique features, it has the potential to find a wide range of applications, from atomic and molecular spectroscopy to plasma and surface studies, in the extreme ultraviolet (XUV) and soft-x-ray regions.¹ One of the main barriers to its more-widespread use is the extremely broad bandwidth associated with the short duration of the harmonic pulses: Short and intense laser pump pulses are indeed necessary for efficient generation of harmonics.

It was recently demonstrated that a technique (which we term time-delay spectroscopy, or TDS) that relies on pairs of time-delayed and phase-locked ultrashort pulses can overcome the limitations associated with the broad single-pulse spectrum and can reach a spectral resolution that is limited only by the length and stability of the delay line.²⁻⁴ Use of TDS has also been suggested for the study of high-lying bound atomic states and autoionizing levels by means of one-photon transitions: In such cases harmonics are good candidates to provide the short-wavelength source required.⁵ The application of the TDS technique to harmonic pulses is, however, not straightforward, mainly because the Michelson interferometer that is used to create the time-delayed pulses cannot be built to work in the XUV owing to a lack of good mirrors and beam splitters.

Recent experiments⁶ have demonstrated that the process of harmonic generation does not scramble the phase of the XUV radiation and that two phase-locked pump pulses can indeed produce two phase-locked harmonic pulses. In those experiments, however, the two harmonic pulses were generated in two spatially separated zones, whereas in the case of TDS the pulses need to be collinear to focus onto the same spot in the region of interaction with the sample. Also, the two time-delayed laser pulses have to interact with the same atoms to produce harmonics, and the second pulse of the pair might generate harmonics in a less

efficient way or with significant phase disturbances caused by the presence of free electrons. As a result, the possibility of using the harmonic pulses that are produced for TDS would be seriously compromised.

To check the phase locking, we set up an experiment in which we could observe optical interference between the two collinear harmonic pulses generated by phase-locked laser pulses. To analyze our data, we first need to express the temporally integrated signal $I(\omega_s, \tau)$, which is observable at the exit slit of a spectrometer centered at ω_s , as a function of the delay τ . We start by considering the harmonic field of order n as the sum of two identical and temporally separated pulses (i.e., we do not consider the simple interference of the two driving laser pulses) of central frequency $n\omega_L$ and temporal envelope $E_0(t)$:

$$E(t) = [E_0(t) + E_0(t + \tau)\exp(-in\omega_L\tau)\exp(-in\omega_L t)]. \quad (1)$$

If $F(\omega - \omega_s)$ is the transmission function of the filter that we assume to be symmetric and much narrower than the single-pulse spectrum $|\tilde{E}_0|^2$, we obtain

$$I(\omega_s, \tau) \propto |\tilde{E}_0(\omega_s - n\omega_L)|^2 [1 + \tilde{F}(\tau)\cos(\omega_s\tau)], \quad (2)$$

where $\tilde{F}(\tau)$ is the Fourier transform of $F(\omega)$ normalized to $\tilde{F}(\tau = 0) = 1$. \tilde{F} is simply seen to correspond to the contrast of the resultant interference fringes.

Expressions (1) and (2) are valid when the two pump laser pulses are separated such that they no longer directly interfere. Of course, when the two laser pulses overlap in time, the dependence of $I(\omega_s, \tau)$ on the delay is given simply by the turning off and on of the pump intensity with the period of the laser field; the intermediate region between this situation and that of well-separated laser pulses shows a behavior

that is similar to the analogous situation of multi-photon TDS.^{7,8}

From expression (2) it can be seen that two short, time-delayed pulses that would not normally show interference owing to their temporal separation may be made to overlap again as a result of the broadening of their temporal profile introduced by spectral filtering. The width of the spectral filter sets the maximum time delay for the existence of interference fringes. Note that, in the absence of such a filtering effect, interference fringes would last only for delays of no more than the coherence time of the pulses, of the order of a few tens of femtoseconds.

In the case of identical harmonic pulses there should be no degradation of the fringe contrast other than that caused by the limited spectral resolution of the monochromator and given by expression (2). If, however, one of the two harmonic pulses is degraded, for example, by passing through a partially depleted medium, the fringe contrast should show a much faster decay with the time delay. The same effect is obtained not only in the case of a simple intensity imbalance between the two pulses but also for a phase disturbance in one of them.

We performed experiments in two configurations: In the first one we selected a narrow wavelength interval and observed the temporal interference fringes as the time delay between the harmonic pulses was scanned. Note that, by observing optical interferences at time delays much longer than the pulse separation, we have already performed a sort of simple TDS on the exit slit of the monochromator, making it possible, for example, to measure directly its spectral width and shape with a spectral resolution much higher than that given by the single-pulse limit.

In the second set of measurements we observed spectra that correspond to sequences of harmonic pulses at different time delays to confirm our results from the spectral point of view also. Although a similar experiment was recently reported by Salières *et al.*,⁹ for harmonic orders higher than those studied here they used fixed delays to record the spectra, and no analysis in the temporal domain was thus possible.

Our experimental setup is composed of a Michelson interferometer used to provide the temporal delay between the pump laser pulses, a vacuum chamber for the interaction between the laser pulses and the pulsed gas jet, and a vacuum normal-incidence monochromator for spectral selection of the harmonic orders produced. The Ti:sapphire amplified laser system provides 100-fs pulses centered about 800 nm and with a 1-kHz repetition rate. The monochromator is equipped with a spherical, 600-line/mm diffraction grating that allows for a spectral resolution of $\Delta\lambda_{\text{FWHM}} = 0.056$ nm when the exit slit is narrowed to ~ 10 μm . We used a 200-mm focal-length lens to focus the laser pulses to peak intensities up to $\sim 1.5 \times 10^{14}$ W/cm² below the exit nozzle of a pulsed valve in the interaction chamber. Xenon was used at a backing pressure of 1–1.5 bars, with an estimated density in the interaction region (~ 0.8 -mm long) of 4×10^{17} cm⁻³. In these conditions, on the basis of Ammosov–Delone–Krainov theory, medium ioniza-

tion is complete only on the laser beam axis, but it may quickly drop to a few percent, depending on the actual position of the atoms with respect to the laser focus and to the beam axis.

Harmonics are observed downstream from the exit slit of the monochromator by means of a phosphor tetraphenyl buthadiene screen and a photomultiplier. The signals are then processed by a digital oscilloscope and stored by a computer, which also controls the monochromator wavelength and the delay line of the interferometer. We observed interference fringes by scanning either the time delay between the pulses or the monochromator's wavelength. Rapid scans of the time delay are performed with a stepper motor, whereas we make shorter and smoother scans by applying a triangular wave voltage to a piezoelectric transducer, in both cases by moving one arm of the interferometer.

In Fig. 1(a) we show how the fifth-harmonic signal versus time delay, near $\tau = 0$, reflects the modulation of the two-pulse fundamental field. In Figs. 1(b)–1(d) we show the fifth-, seventh-, and ninth-harmonic signals obtained by scanning of the time delay on a small scale in the region of well-separated laser pulses. Stretching of the duration of the harmonic pulses owing to the spectral filtering of the monochromator is evident, as these pulses keep interfering even for such large temporal separations.

In Fig. 2, values for the measured fringe contrast of different harmonics are shown on a larger

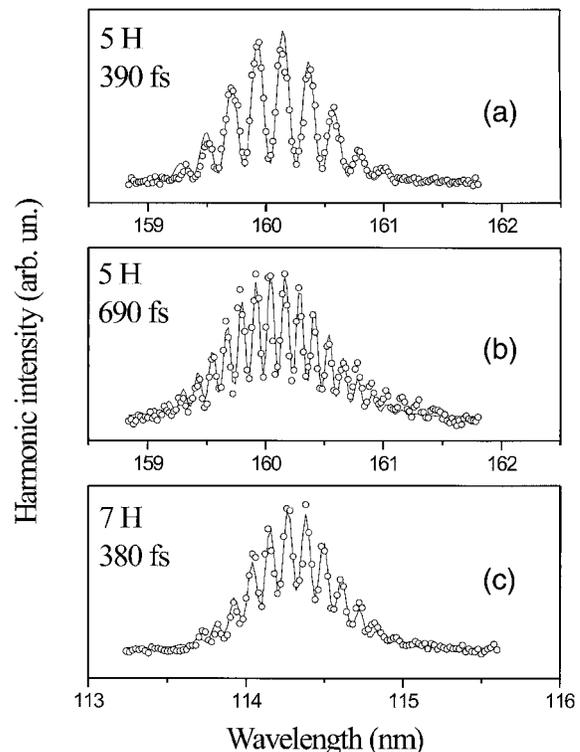


Fig. 1. Experimental measurements of the two-pulse, time-integrated intensity $I(\tau, \omega_s \approx \omega_{\text{max}})$ versus time delay τ for (a) the fifth harmonic at $\tau \approx 0$, (b) the fifth harmonic at $\tau \approx 750$ fs, (c) the seventh harmonic at $\tau \approx 750$ fs, (d) the ninth harmonic at $\tau \approx 400$ fs.

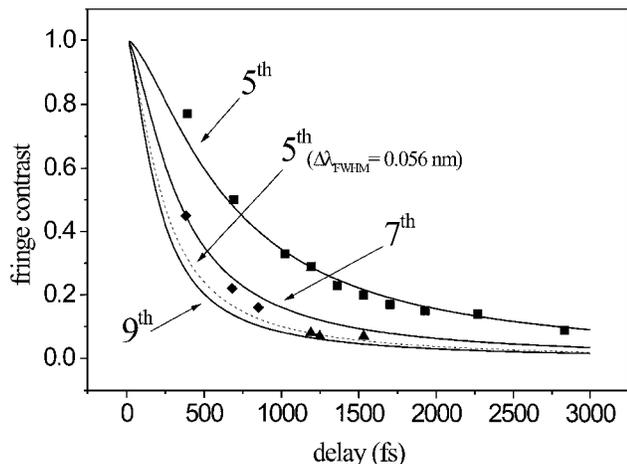


Fig. 2. Experimental fringe contrast for the fifth (squares), the seventh (diamonds), and the ninth (triangles) harmonics. Solid curves, best overall fit to the data, corresponding to $\Delta\lambda_{\text{FWHM}} = 0.02$ nm. Dashed curve, fringe contrast for the fifth harmonic, corresponding to the manufacturer's value of $\Delta\lambda_{\text{FWHM}} = 0.056$ nm.

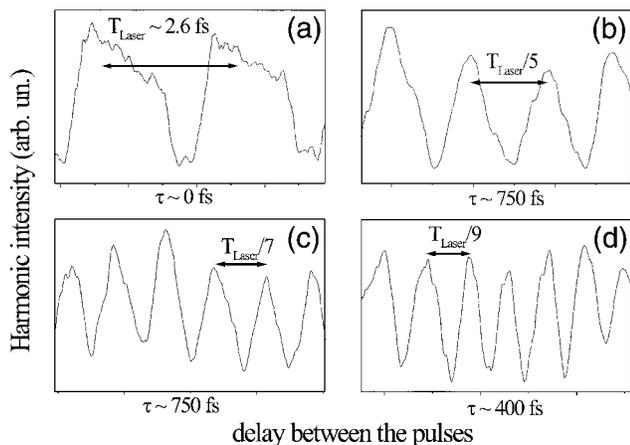


Fig. 3. Two-pulse spectra of the fifth harmonic at (a) $\tau \approx 390$ fs and (b) $\tau \approx 690$ fs. The spectrum of the seventh harmonic at $\tau \approx 380$ fs is shown in (c). The solid curves come from expression (4), with $F(\tau)$ derived from the fit presented in Fig. 2.

scale of time delay. We find that the best overall agreement for $F(\tau)$ (solid curves) is achieved with a corresponding width of the spectral filter of $\Delta\lambda_{\text{FWHM}} = 0.02$ nm. Note that this value is smaller than that found by the monochromator manufacturer ($\Delta\lambda_{\text{FWHM}} = 0.056$ nm) by use of a xenon discharge and was independently measured and confirmed by us by measuring the spectrum of a mercury lamp: The corresponding $F(\tau)$ curve for the fifth harmonic is shown by a dashed curve in Fig. 2.

Our data clearly show that there is no degradation of fringe visibility except that given by the monochromator resolution itself. The apparently surprising result of a fringe contrast even better than that given by the instrumental limit is probably due to the different conditions of measurement of the spectra: In the case of harmonics, the beam illuminates only a portion of the grating surface, whereas in the case of the mer-

cury lamp or xenon discharge, the whole grating is uniformly illuminated. Inasmuch as the resolution power has a negligible limit, the less divergent harmonic field can actually give that better resolution that is not affected by the aberrations on the whole surface of the grating.

In Figs. 3(a)–3(c) we show modulated two-pulse spectra that correspond to the fifth [Figs. 3(a) and 3(b)] to the seventh [Fig. 3(c)] harmonics at different time delays between the pump pulses. It can be seen that, according to expectations, the two-pulse spectra exhibit the broad envelope of the single pulse, with a superimposed sinusoidal modulation showing fringes with a period $\delta\lambda = \lambda^2/c\tau$. The solid curves are fits to expression (2), assuming a Gaussian single-pulse spectrum and the $F(\tau)$ found above. When the fringe period approaches the spectral resolution of the monochromator, the fringe contrast starts to decrease as a result of the blurring of adjacent minima and maxima that finally washes out the whole fringe pattern.

In conclusion, our measurements are in agreement with the simple expression (2) that takes into account only the limit given by the finite resolution of the monochromator. We have found that there is no degradation of the fringe contrast caused by the generation process itself in our experimental conditions. We conclude that, in the regime of medium-order harmonics and for laser intensities as great as 1.5×10^{14} W/cm², it is indeed possible to generate collinear pairs of phase-locked extreme-ultraviolet radiation pulses. These measurements represent a first step toward the realization of time-delay spectroscopy in the XUV: Replacing the monochromator slit with the narrow resonance of an atomic system will permit study of the spectral characteristics of TDS with an unprecedented resolution for this wavelength region.

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