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Towards high-resolution spectroscopy in the XUV with phase-locked harmonic pulses

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Abstract

We report the results of measurements aimed to check the phase lock of time-delayed, collinear, harmonic pulses and to verify the possibility of performing Ramsey-like spectroscopy in the XUV. We demonstrate that for harmonics of medium order and for the peak intensities available with our laser system, the generation of collinear phase-locked harmonic pulses is indeed possible and that such pulses can be used to achieve high-resolution spectroscopy in the short-wavelength region.

1. INTRODUCTION

The nonlinear process of high-order harmonic generation (HHG) in noble gases is a promising new tool for the production of coherent short-wavelength radiation with the potential to find a wide range of applications, from atomic and molecular spectroscopy to plasma and surfaces studies, in the XUV and soft X-rays (for a recent review, see Salières *et al.*, 1999*b*).

Though some spectroscopic applications of the new harmonic sources have already appeared, one of the main limitations to a more widespread use is the extremely broad bandwidth associated with the short duration of the harmonic pulses. Short pump pulses are indeed necessary to extend the plateau, the region of the spectrum where harmonics can be generated in an efficient way; even the common 100-fs laser systems have an associated bandwidth of the order of several THz, and prevent any possible spectroscopic application unless special techniques are adopted.

A technique (which we may call time-delay spectroscopy, or TDS) relying on pairs of time-delayed and phase-locked ultrashort pulses has been recently demonstrated to overcome the limitations connected with the broad single-pulse spectrum and to reach a spectral resolution which is only limited by the length and stability of the delay line (Haberle *et al.*, 1995, Fourkas *et al.*, 1989, Bellini *et al.*, 1997).

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The frequency spectrum associated with two identical time-delayed pulses has a broad single-pulse spectral envelope but is sinusoidally modulated with a period that is the inverse of the time delay τ. It is the maximum time delay between the two pulses that, as in Fourier-transform spectroscopy, sets the instrumental spectral resolution.

Looking at the same scheme in the temporal domain, one can think of the first pulse of the sequence as inducing a dipole moment oscillating at the frequency fixed by the ground and the selected excited states of the investigated atom; after the first pulse has gone, the polarization keeps oscillating at its eigen-frequencies and can vanish or be further excited depending on the phase delay of the second pulse. Similar to Ramsey spectroscopy, the time delay between the pulses increases the effective interrogation time and improves the spectral resolution. If two or more levels are excited by the broad bandwidth associated to the short pulses, beatings show up in the total excited population and the spectrum of the involved levels can be recovered by a simple Fourier transformation.

An alternative and interesting way to look at the phenomenon from the temporal point of view is to consider the absorbing transition of the atomic or molecular system under study as a narrow spectral filter for the broadband incoming light. Two short, time-delayed pulses that would no longer show optical interference due to their temporal separation may thus be made to overlap again thanks to the spreading of their temporal profile introduced by the spectral filtering. If the two light pulses are phase locked, interference effects show up while scanning the delay and last as

long as the time separation is compensated by the pulse spreading. The width of the spectral filter (or the linewidth of the investigated transition) sets the maximum time delay for the existence of interference fringes. While in the case of an instrumental spectral filter (like a slit in the focal plane of the monochromator) we are dealing with optical interference fringes that can be observed with a photodetector, in the case of an absorbing transition we speak of quantum interference of the matter; in such a case the interference fringes must be measured in some of the atomic observables, such as the population of the levels.

Time-delay spectroscopy has also been suggested for the study of high-lying bound atomic states or autoionizing levels by means of one-photon transitions: In such cases harmonics are good candidates to provide the short-wavelength source required (Cavalieri & Eramo, 1998). The application of the TDS technique to harmonic pulses is, however, not straightforward, mainly because the Michelson interferometer used to create the time-delayed pulses cannot be built to work in the XUV due to the lack of good mirrors and beamsplitters.

Recent experiments (Bellini *et al.*, 1998) have demonstrated that the process of harmonic generation does not randomly scramble the phases of the XUV radiation and that two phase-locked pump pulses, obtained by a Michelson interferometer, can indeed produce two phase-locked harmonic pulses. In those experiments, however, the two harmonic pulses were generated in two spatially separated zones while, in the case of TDS, the pulses need to be collinear in order to focus into the same spot in the region of interaction with the sample. This also means that the two time-delayed laser pulses have to interact with the same atoms to produce harmonics. Harmonic generation always implies a certain degree of medium ionization and the second pulse of the pair may generate harmonics in a less efficient way or with significant phase disturbances due to the presence of free electrons. As a result, the possibility of using the produced harmonic pulses for TDS would be seriously compromised.

In this paper, we report the results of some measurements aimed to check the phase lock of time-delayed, collinear, harmonic pulses and, ultimately, to verify the possibility of performing time-delay spectroscopy in the XUV. We demonstrate that, at least for harmonics of medium order and for peak intensities of the order of 10^{14} W/cm^2 in xenon, the generation of collinear phase-locked harmonic pulses is indeed possible and that such pulses can be used to achieve high-resolution spectroscopy in the short-wavelength region.

Experiments have been performed in two different configurations: In the first one, we observe the spectra corresponding to sequences of harmonic pulses with different time delays. A similar experiment has been recently reported by Salières *et al.* (1999*a*) for harmonic orders higher than those studied here but, in that case, fixed delays had been used to record the spectra. In the second set of measurements, we select a narrow wavelength interval and we observe the temporal interference fringes as the time delay between the harmonic pulses is scanned. It should be noted that, by observing optical interferences at time delays longer than the pulse separation, we are already performing a sort of simple TDS on the exit slit of the monochromator, making it possible, for example, to directly observe its spectral width and shape.

2. EXPERIMENT

The experimental setup is sketched in Figure 1 and is composed of a stable, motorized Michelson interferometer used to provide the temporal delay between the pump laser pulses,

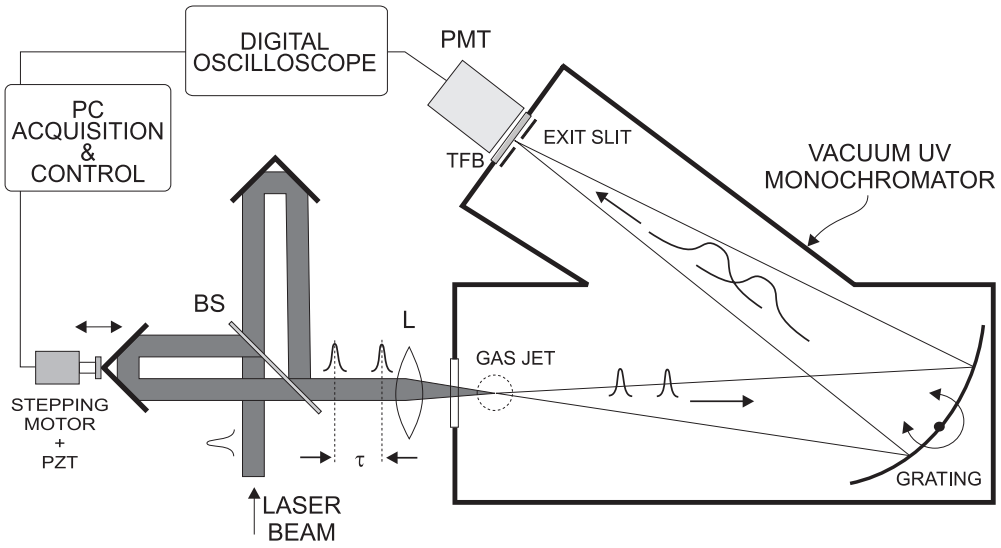

Fig. 1. Experimental setup for the test of the phase lock between two collinear harmonic pulses. A 50% beamsplitter (BS) splits the laser pulses from the laser and the lens (L) focuses them in the gas jet with a variable relative delay τ. The harmonic pulses are selected and temporally stretched by the grating and are then detected by a phosphor screen (TFB) and a photomultiplier (PMT).

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a vacuum chamber equipped with an electromagnetic valve for the interaction between the laser pulses and the pulsed gas jet, and a vacuum, 1-m, normal incidence monochromator for the spectral selection of the different harmonic orders produced. The Ti:Sa amplified laser system provides 100-fs pulses centered around 800 nm and with a 1-kHz repetition rate. Due to the limitations connected to the valve and to the pumping system used to evacuate the monochromator, the rate of gas injection in the chamber is generally kept to a lower value, around 50 Hz.

The monochromator is equipped with a spherical, 600 lines/mm diffraction grating that allows for a spectral resolution of about 0.056 nm (FWHM) when the exit slit is closed to about 10 μm. A 200-mm focal length lens has been used to focus the laser pulses to peak intensities up to 1.4×10^{14} W/cm^2 below the exit nozzle of the pulsed valve in the interaction chamber. Xenon, thanks to its high polarizability that results in a high yield of harmonic photons, has been used for the experiments at a backing pressure of 1–1.5 bar.

Harmonics are observed downstream the exit slit of the monochromator by means of a phosphor (TFB) screen and a photomultiplier. The signals are then processed by a digital oscilloscope and stored by a computer which also controls the grating angle and the delay line of the interferometer.

For the first series of measurements, fixed delays have been selected between the incoming pulses, and the corresponding spectra have been recorded by scanning the monochromator grating angle while recording the photon flux beyond the exit slit. For the second set of measurements, the grating angle has been kept fixed, and the exit slit has been used to select a narrow wavelength region close to the maximum of the single-pulse spectrum. The temporal interference fringes have been observed by scanning the time delay between the pulses. Rapid scans are performed by means of a stepping motor, while shorter and smoother scans are done by applying a triangular wave voltage to a PZT, both moving one arm of the interferometer.

3. RESULTS AND DISCUSSION

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. The comparison with experimental results then requires the expression of the temporally integrated signal at the exit slit of the spectrometer in order to take into account the instrumental spectral resolution.

Let us consider the harmonic field of order n as the sum of two 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 $E_0(t)$:

$$E(t) = (E_0(t) + E_0(t+\tau)e^{-in\omega_L\tau})e^{-in\omega_L t}. \quad (1)$$

This expression with two identical pulse fields is valid if one assumes that the generation of the second pulse is not perturbed by the generation of the first one. The corresponding frequency spectrum is

$$\tilde{E}(\omega - n\omega_L) = \tilde{E}_0(\omega - n\omega_L)(1 + e^{-i\omega\tau}). \quad (2)$$

The measured quantity is the temporally integrated signal at the exit slit of the spectrometer centered at ω_s:

$$I(\omega_s, \tau) \propto \int_{-\infty}^{\infty} |\tilde{E}_0(\omega - n\omega_L)|^2 F(\omega - \omega_s)(1 + \cos(\omega\tau))d\omega, \quad (3)$$

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

$$I(\omega_s, \tau) \propto |\tilde{E}_0(\omega_s - n\omega_L)|^2(1 + \tilde{F}(\tau)\cos(\omega_s\tau)) \quad (4)$$

being $\tilde{F}(\tau)$, the Fourier transform of $F(\omega)$ normalized to have $\tilde{F}(\tau = 0) = 1$. It is evident that $\tilde{F}$ corresponds to the fringe contrast defined as $(I_{max} - I_{min})/(I_{max} + I_{min})$.

The above expressions are valid in the case that the two pump laser pulses are so separated that they no longer interfere. When the laser pulses are overlapped in time, the de-

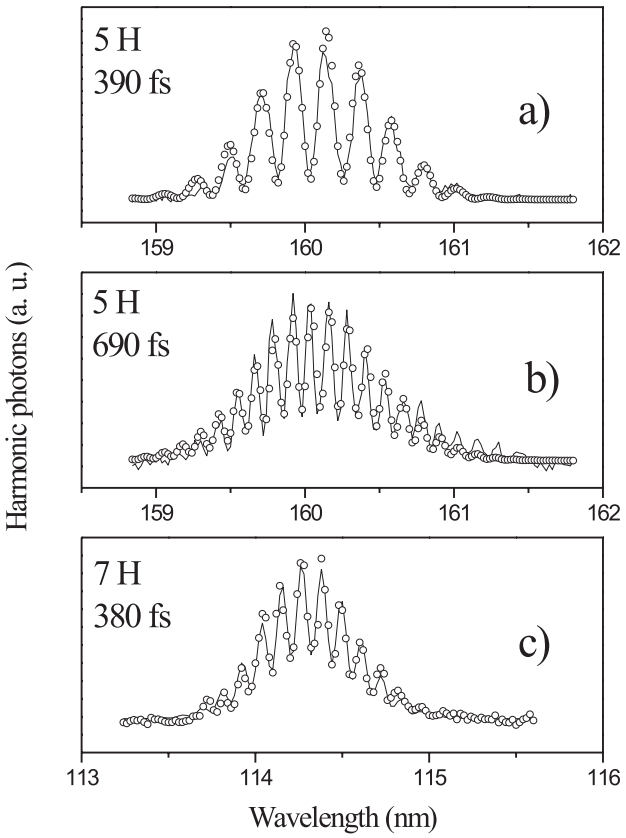

Fig. 2. Two-pulse spectra of the fifth harmonic at $\tau \approx 390$ fs (a) and $\tau \approx 690$ fs (b). The spectrum of the seventh harmonic at $\tau \approx 380$ fs is shown in (c). The solid lines are fits with expression (4) where the width of $F(\omega)$ has been fixed to 0.056 nm.

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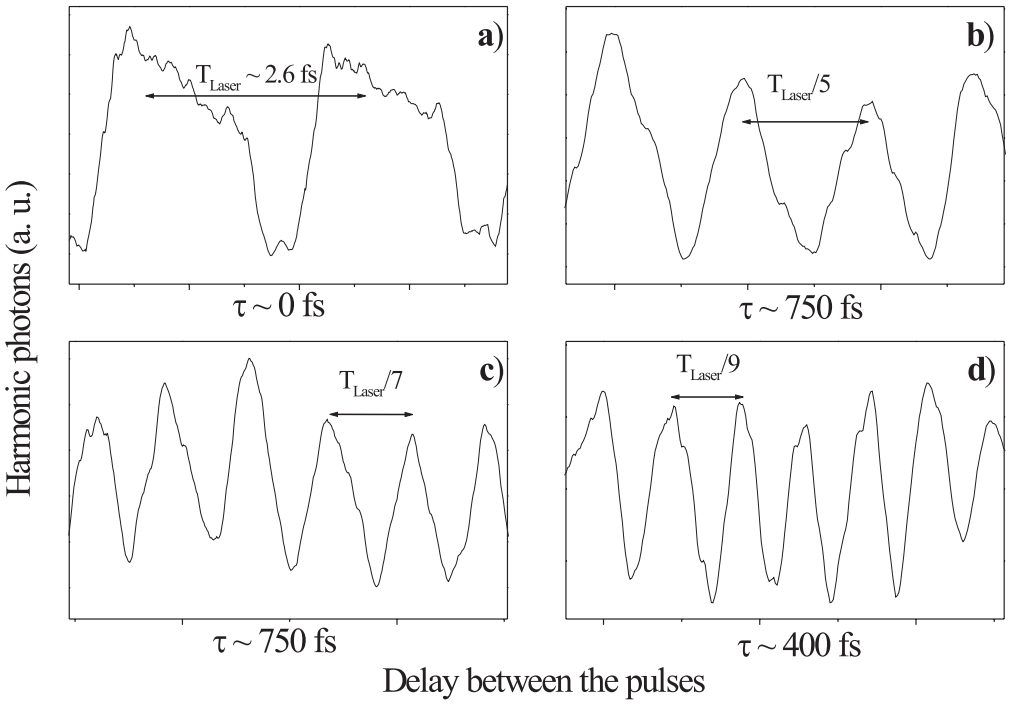

Fig. 3. Experimental measurements of the two-pulse, time-integrated intensity $I(\tau, \omega_s \approx \omega_{max})$ for: (a) fifth harmonic at $\tau \approx 0$, (b) fifth harmonic at $\tau \approx 750$ fs, (c) seventh harmonic at $\tau \approx 750$ fs, (d) ninth harmonic at $\tau \approx 400$ fs.

pendence from the delay is simply given by the turning off and on of the pump intensity with the period of the laser field. The intermediate region from overlapped laser pulses to the well-separated case behaves similarly to the analogue case of multiphoton time delay spectroscopy (Blanchet *et al.*, 1997; Cavalieri *et al.*, 2000).

If one of the two harmonic pulses is degraded, for example, by passing through a partially depleted medium, then the fringe contrast should show a much faster decay with the time delay. Notice that the same effect is obtained not only in the case of a simple intensity unbalance between the two pulses but also in the case of a phase disturbance on one of them.

In Figure 2 we show modulated two-pulse spectra corresponding to the fifth (2a and 2b) and to the seventh harmonics (2c) at different time delays between the pump pulses. It can be seen that, according to expectations, the two-pulse spectra present the broad envelope of the single pulses with a superimposed sinusoidal modulation showing fringes with a period $\delta\lambda = \lambda^2/c\tau$. The solid lines are fits with expression (4), assuming Gaussian pulses and a Gaussian $F(\omega)$ with the value of $\delta\lambda_{FWHM} = 0.056$ nm as given by the monochromator manufacturer. When the fringe period approaches the spectral resolution of the monochromator, the fringe contrast starts to decrease due to the blurring of adjacent minima and maxima that finally washes out the whole fringe pattern.

Our measurements are in agreement with the simple expression (4) that only takes into account the limit given by the finite resolution of the monochromator. We conclude that there is no degradation of the fringe contrast due to the generation process itself in our experimental conditions.

In Figure 3a we show how the fifth harmonic signal versus time delay around $\tau = 0$ reflects the modulation of the two-pulse fundamental field. In Figures 3b, 3c, and 3d, we show the fifth, seventh, and ninth harmonic signals obtained by scanning the time delay on a small scale in the region of well-separated pulses. The stretching of the duration of the harmonic pulses due to the spectral filtering of the monochromator is evident because such pulses keep interfering even for large temporal separations.

4. CONCLUSIONS

We have found that, in the regime of medium-order harmonics and for laser intensities below 1.5×10^{14} W/cm^2, it is possible to generate collinear pairs of phase-locked XUV radiation pulses. We have observed interferences between the time-delayed harmonic pulses both in the spectral and in the time domain; these measurements represent the first step towards the realization of time-delay spectroscopy in the XUV.

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