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Manipulating intense XUV coherent light in the temporal domain

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Abstract

In this article, we demonstrate the generation of four phase-locked harmonic pulses separated in time using frequency-domain interferometry. The spectra present a high sensitivity to a change of the relative phase on an attosecond time scale. The spectral resolution and the control of this relative phase could be used to perform high resolution measurements.

Keywords: Attosecond; High harmonics generation; Phase-locking; XUV interferometry

1. INTRODUCTION

Studies of high harmonic generation (HHG) have become a very active domain in high-field physics. Together with above threshold ionization, this spectacular nonlinear process is exemplary of the atom–high-field interaction in the tunneling regime, defined by the Keldysh parameter $\gamma < 1$. Besides its fundamental interest, HHG has demonstrated unique properties as a XUV source. Microjoule energies per pulse in the range 80–30 nm to a few nanojoules at shorter wavelengths have been obtained (Hergott *et al.*, 2002; Takahashi *et al.*, 2002). The ultrashort pulse duration of $\sim$10 fs to subfemtoseconds, together with the high spatial coherence and wave front quality, lead to high peak brightness. Now available terawatt laser sources with kilohertz repetition rates make possible high average power and brightness that actually compare with those of third-generation synchrotron sources in the 10–100 eV range. This opens considerable perspectives for the dynamical studies at an unprecedented subfemtosecond time scale in a variety of fields, from atomic and molecular physics, chemistry, and biology to solid-state or plasma physics (Larsson *et al.*, 1995; Gisselbrecht *et al.*, 1999; Descamps *et al.*, 2000; Quéré *et al.*, 2000; Merdji *et al.*, 2000*a*, 2000*b*; Hergott *et al.*, 2001).

High-order harmonic radiation presents unique properties of intrinsic spatial and spectral coherence allowing the transposition of many infrared techniques to the XUV range. Interferometry is a widely used technique offering a variety of possibilities as a tool for probing but also manipulating and controlling ultrafast phenomena. The demonstration of the generation of *two* temporally separated phase-locked harmonic pulses has been recently reported (Salières *et al.*, 1999; Hergott *et al.*, 2003). This XUV frequency-domain interferometer has been applied to the study of the temporal evolution of a dense laser plasma at the femtosecond time scale. Recently Ramsey-like spectroscopy has been demonstrated using the coherence properties of high order harmonic generation (Cavalieri *et al.*, 2002). If you now generate *N* temporally separated phase-locked XUV pulses separated in time you will observe in the spectral domain fringes that are becoming narrower and brighter (scaling as N^2). This technique can be applied to perform high resolution spectroscopy in the XUV range. The control of the phase on these time scales offers a powerful tool to perform Fourier transform spectroscopy with an unprecedented resolution. Another exciting application could be to use multiple time-delayed beams to perform Ramsey-like spectroscopy in the XUV range. Frequency-domain interferometry is a widely used technique offering a variety of possibilities as a tool for probing but also manipulating and controlling ultrafast phenomena, for instance, in solid-state and plasma physics. The extension of the technique considering two temporally separated harmonic pulses to multiple phase-locked harmonic pulses will lead to highly contrasted spectral fringe patterns and an increased phase sensitivity. In this article we will describe the extension to four phase-locked XUV pulses and will show the extreme sensitivity of this method to a change of the relative phase on an attosecond time scale.

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2. EXPERIMENTAL RESULTS AND DISCUSSION

The experiment has been performed on the LUCA laser facility in Saclay. This Ti:Sa system delivers 800-nm pulses with 50-fs pulse duration at 20 Hz with an energy up to 100 mJ. We studied the 11th harmonic generated in xenon at an estimated intensity of 4×10^{13} W/cm^2. The four phase-locked harmonic pulses have been generated using four IR laser pulses separated in time as shown in Figure 1. The first double laser pulse is created by using the group velocity difference on the two axes of a birefringent plate rotated at 45° from the laser polarization. A polarizer placed after the plate projects both components on the same axis. The calibrated thickness of the plate fixes the time delay τ between the two pulses at 120 fs. This couple of laser pulses is sent into a Michelson interferometer where two replicas (C_1 and C_2) are produced with the same time delay τ between the pulses. The length of the first arm of the interferometer is controlled by a stepping motor, whereas the second one is moved by a piezoelectric translation with nanometric precision. It is then possible to control very precisely the optical paths of (and thus the time delay ΔT between) the two laser pulse couples C_1 and C_2. These four IR laser pulses (500 μJ each) are focused in a pulsed xenon jet where they generate four harmonic pulses that are sent into a spectrometer. The spectral fringe pattern obtained after dispersion by the grating is recorded on microchannel plates mounted in the focal plane of the spectrometer at grazing incidence (12°) in order to increase the resolution to 0.1 Å. The signal level is high enough to perform a single shot of the power spectrum of the four pulses.

When the two arms of the Michelson are slightly misaligned, the two couples are not spatially superposed and we observe two identical spectral fringe patterns corresponding to the interference between the two pulses of each couple, with a fringe period fixed by the same time separation τ (Fig. 2a). In this case the observed fringe patterns are independent of the time delay ΔT between C_1 and C_2. A realignment of the Michelson allows a spatial superposition of the harmonic couples. A dramatic change of the fringe pattern is observed (as shown in Fig. 2b), which gives evidence for the generation of four phase-locked harmonic pulses separated in time. The delay ΔT between C_1 and C_2 was chosen to be 2τ, corresponding to an equal separation of the four pulses of τ. Due to this regular time separation, an effective fringe narrowing is obtained (factor 2 with respect to the 2×2 sources case). By slightly varying the delay to $\Delta T = 2\tau + T_q/2$ ($T_q/2$ is the harmonic half period, ~120 as for the 11th harmonic, corresponding to a mirror displacement of 18 nm), we observe a clear change of the fringe pattern that becomes more regular with a fringe period twice as small as in Figure 2a (Fig. 2c).

In Figure 3, we show the corresponding spectral profiles. In the case of a single couple, the modulation of the harmonic spectrum follows $\cos^2(\omega\tau/2)$. When the two couples interfere, an additional modulation comes into play in $\cos^2(\omega\Delta T/2)$. When $\Delta T \sim 2\tau$, the same periodicity as in the case of a single couple is observed, with a significantly narrowed central peak surrounded by satellite structures (Fig. 3a). When $\Delta T \sim 2\tau + T_q/2$, the periodicity seems to decrease to half the former value, with regular peaks of equal intensity (Fig. 3b). In both cases, there is a good agreement with the results of the fit using the theoretical

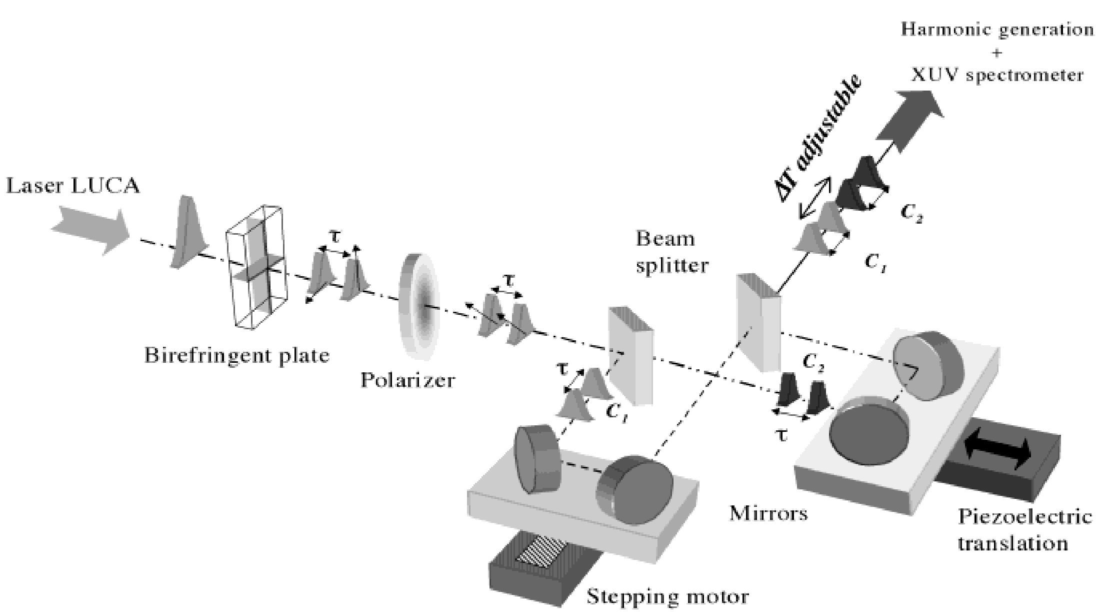

Fig. 1. Diagram of the setup used for the generation of four laser pulses delayed in time.

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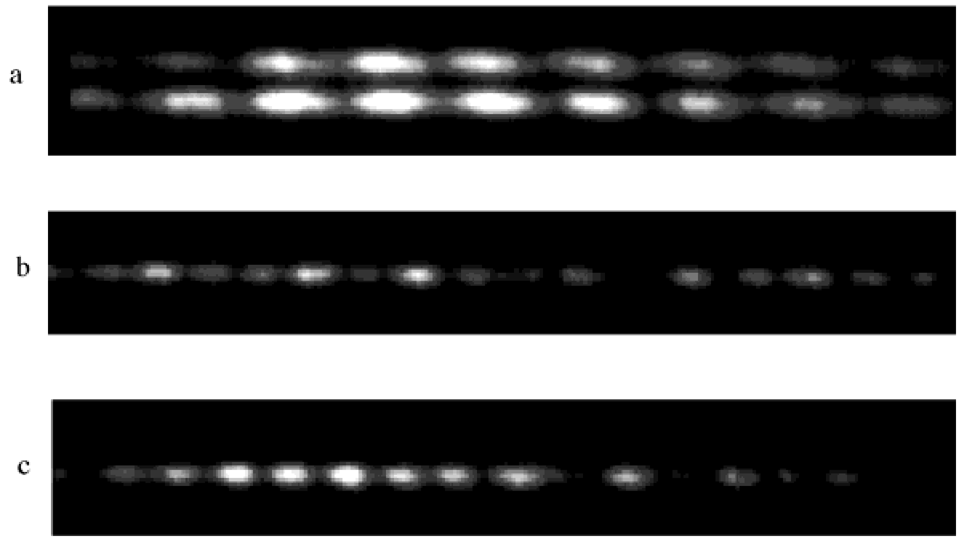

Fig. 2. Fringe patterns for the 11th harmonic generated in xenon by two laser pulse couples separated by $\tau = 120$ fs. a: The Michelson is misaligned. b, c: The Michelson is aligned and the delay ΔT between the two couples is set, respectively, to 2τ and $2\tau + T_q/2$.

formula of interference between four phase-locked pulses with the corresponding delays (dashed lines). This demonstrates the high sensitivity of our experimental setup to a change of the relative phase on an attosecond time scale: A phase shift between the two couples of a half harmonic period (~120 as) results in a strong modification of the whole fringe pattern, whereas for the simple two-pulse case (variation of τ), there is only a general shift of the whole fringe pattern in the envelope, which is much more difficult to diagnose unambiguously due to the experimental fluctuations.

3. CONCLUSIONS

We have performed the generation of four phase-locked harmonic pulses separated in time. The spectral profiles obtained from frequency-domain interferometry present a high sensitivity to a change of the relative phase on an attosecond time scale, indicating that such a control is achieved on the harmonic emission. Interesting perspectives arise from the application of this technique. The sensitivity of this technique could be useful for, for example, the precise diagnostic of dense plasmas. Moreover, pump probe experiments with two spatially separated harmonic couples (see Fig. 2a) would benefit from an absolute reference pattern or the possibility to probe large plasmas with relative density measurements.

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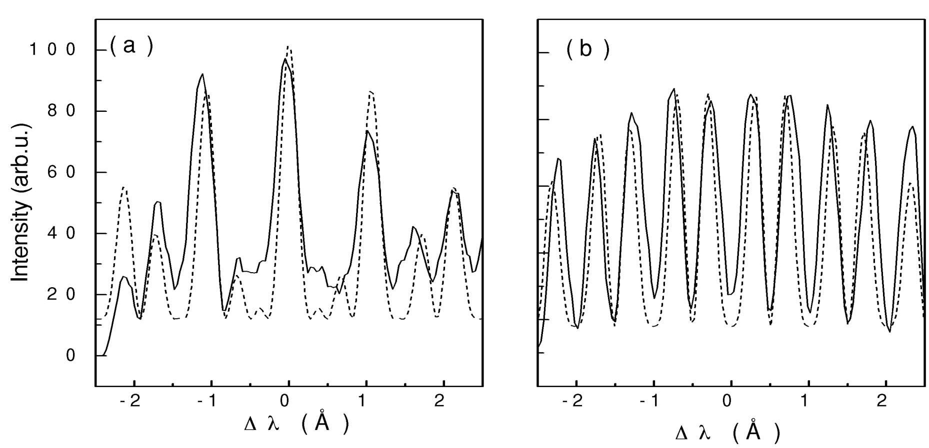

Fig. 3. Spectral profiles (solid lines) obtained from the interference of two couples (H11) delayed by about 2τ (a) and $2\tau + T_q/2$ (b). Dashed lines indicate the fits from the theoretical formula. a: The spectral profile obtained in the case of a single couple.

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