Frequency-Domain Interferometry in the XUV with High-Order Harmonics

P. Salières, L. Le Déroff, T. Auguste, P. Monot, P. d'Oliveira, D. Campo, J.-F. Hergott, H. Merdji, and B. Carré
CEA/DSM/DRECAM, Service des Photons, Atomes, et Molécules, Centre d'Etudes de Saclay, 91191 Gif-sur-Yvette, France
(Received 17 August 1999)

We demonstrate that frequency-domain interferometry can be performed in the extreme ultraviolet range using high-order harmonics. We first show that two phase-locked harmonic sources delayed in time can be generated in the same medium despite ionization. This gives insight into the dynamics of the generation and ionization processes. We then apply the technique to the study of the temporal evolution of an ultrashort laser-produced plasma at the femtosecond time scale.

PACS numbers: 42.65.Ky, 32.80.Rm

Recently, high-order harmonic generation (HHG) has attracted considerable interest, not only as an intriguing and spectacular phenomenon, but also as a potential source presenting unique properties in the extreme ultraviolet (XUV) range. The two main characteristics of this radiation are the ultrashort pulse duration and the good coherence that are both unprecedented in this spectral region. The short pulse duration (down to a few tens of femtosecond) has been characterized [1–3] and is already used in atomic and molecular spectroscopy [4]. On the other hand, a number of experiments in the last few years have shown that good coherence properties could be obtained in some generating conditions [5–8] but no applications of this coherence have been performed yet. XUV interferometry, developed so far with x-ray lasers [9–12], would benefit a lot from the HHG properties. First, the tunability of the radiation allows one to adapt the wavelength to the probed medium, for example, far from resonances. In plasmas, different electron densities can be probed by changing the harmonic order, leading to a precise density mapping. Note that beside this coarse tunability, a fine adjustment of the wavelength can be obtained by generating the harmonics with a chirped laser [13] or with a laser mixed with an optical parametric amplifier [14]. Second, the ultrashort harmonic pulse duration is well adapted to the study of ultrafast processes, and could prevent, for example, the blurring of the fringes due to fast evolution of the density profile of a plasma close to the critical surface, as is the case when using x-ray lasers [9]. Finally, HHG is a tabletop XUV source, naturally synchronized with the generating laser at the same repetition rate (10 Hz to 1 kHz) allowing systematic experiments. In this work, we perform frequency-domain interferometry with high-order harmonics. We first demonstrate that the technique can be transposed to harmonics, and then apply it to probe the electron density of a laser-produced plasma in a high density gas jet with a femtosecond temporal resolution.

Frequency-domain interferometry is now a widely used technique in the infrared for femtosecond time-resolved studies in solid-state and plasma physics [15–17]. It can be described as the temporal analog of the Young two-slit experiment [18]. In the latter, two *spatially separated* phase-locked sources lead to interferences in the far field after the beams have diffracted. In the former, two *temporally separated* phase-locked pulses interfere in the spectral domain after dispersion on a grating. The physical analogy between diffraction and dispersion results in similar fringe patterns. It has been shown recently that two phase-locked high-order harmonic sources *separated in space* could be created by splitting a laser beam into two beams focused at different locations in a gas jet [19]. The problem of creating two phase-locked harmonic sources *delayed in time* by focusing an intense double pulse laser beam at the same location in a gas jet is even more challenging and generally considered as hopeless. Indeed, HHG is highly connected to the ionization of the medium, and it is widely accepted that an efficient harmonic generation implies a high degree of ionization at the end of the (first) laser pulse. This means that the second laser pulse interacts with a partially—if not entirely—ionized medium, and consequently the HHG efficiency should be strongly reduced (due to depletion of the medium, defocusing of the laser beam, phase mismatch induced by free electrons, etc.). Moreover, the dephasing induced by the free electron dispersion, which affects strongly the spatial coherence [7,8], can result in a loss of mutual coherence between the two harmonic beams and to a blurring of the fringes. In this Letter, we demonstrate that two phase-locked harmonic sources in time can be generated efficiently, leading to highly contrasted spectral fringe patterns. The incidence of the ionization on the spectra gives insight into the generation process (efficiency vs ionization, chirp).

We have studied harmonics 11th to 23rd generated in argon by the 10 TW, 60 fs, Ti:sapphire UHI10 laser in Saclay. In this part of the experiment, less than 10 mJ were focused by an $f/110$ lens into the pulsed gas jet. The double laser pulse was 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. Both birefringent plate and polarizer were placed before the laser compressor in order to avoid self-phase modulation by the intense compressed beam. We used two birefringent plates of calibrated thicknesses to produce 120 and 450 fs delays, respectively. Even though fixed delays are obtained, the advantage of this setup compared to the Michelson is that it is highly stable so that integration over thousands of shots can be performed without blurring the fringes. Indeed, a dephasing between the two laser beams of only half a fundamental period *divided by the harmonic order* is enough to shift the harmonic fringes out of phase. The observation of the harmonic spectral fringes raises a technical problem: the period in the wavelength space is given by $\Delta\lambda = \lambda^2/c\Delta t$, where Δt is the time delay between the two harmonic pulses. It decreases rapidly with the harmonic order, implying that a high resolution spectrometer is needed to resolve the fringes. For achieving the best resolution, the exit slit of our XUV spectrometer was closed to only 16 μm, corresponding to a resolution better than 0.1 Å. The data are averaged over 40 shots with an allowed dispersion of 7% in laser energy.

The experimental spectra of harmonics 11, 15, 19, and 23 generated by two phase-locked laser pulses delayed by 120 fs and focused at 2×10^{14} W/cm^2 are shown in Fig. 1. Regular contrasted fringes are measured for all orders, with a reduced contrast when the order increases: from 90% for harmonic 11 to about 25% for the 23rd. The period of the fringes decreases quadratically with the order, as expected from the above formula. For the 23rd harmonic, $\Delta\lambda = 0.34$ Å approaches the resolution limit. For all orders, the higher energy part of the spectrum does not seem to be modulated. This part broadens as the order increases, to almost half of the spectrum for harmonic 23.

We investigated the influence of the laser intensity on the fringe pattern. This is shown in Fig. 2 for the 15th harmonic in the otherwise same conditions as Fig. 1b. An evolution similar to that observed when increasing the order is obtained when increasing the intensity from 2×10^{14} to 3.5×10^{14} and 5×10^{14} W/cm^2. The blue part of the spectrum gets more intense and does not exhibit any modulation, whereas the red part is still modulated but with a reduced contrast: only 25% at 5×10^{14} W/cm^2 vs 65% at 2×10^{14} W/cm^2. This results in a strong asymmetry in the spectra. At even higher intensity, the fringes completely disappear while the spectrum becomes very broad. This trend is also observed for the other harmonics, very clear for the 19th but less dramatic for the 11th. Note that the gas density plays an important role: the above spectra have been measured for a backing pressure of 600 Torr (the pressure in the jet is roughly a factor of 10 lower). For a pressure of 150 Torr, the variation with intensity is similar but less dramatic.

The fringe pattern also depends on the position of the laser focus relative to the gas jet. When the laser focus is moved from 3 cm before the medium (condition of Fig. 1a) to the center of the jet as in Fig. 3, the fringe contrast is reduced: for the 11th harmonic, it drops to 70% (Fig. 3a). Finally, when the time delay Δt is increased to 450 fs, the period of the fringes decreases as expected by more than a factor of 3, as shown in Fig. 3b. The contrast is reduced but the intensity is slightly higher (3×10^{14} W/cm^2). Note that for this low-order harmonic at relatively low intensity, the envelope of the fringes remains roughly the same when changing the delay. We have checked that this envelope is very close to the spectrum of the 11th harmonic generated by a *single* laser pulse at the same intensity.

In order to understand the experimental trends, we have performed numerical simulations in conditions close to the experimental ones. We use the harmonic dipole calculated from the strong field approximation approach [20] and a 3D propagation code accounting for all the consequences of ionization in the jet (defocusing, depletion, phase mismatch) [21]. Simulated profiles are shown in Figs. 3c and 3d, corresponding to the conditions of, respectively, Figs. 3a and 3b. The envelope of the spectra as well as the period and the contrast of the fringes are

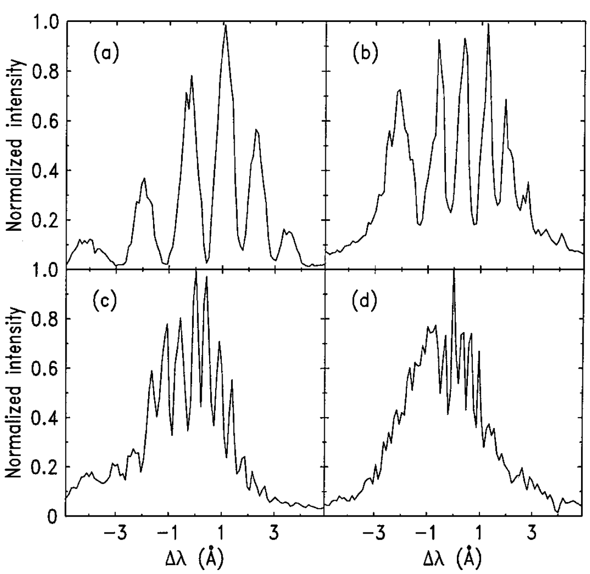

FIG. 1. Experimental spectra of harmonics 11 (a), 15 (b), 19 (c), and 23 (d) generated by two laser pulses delayed by 120 fs and focused at 2×10^{14} W/cm^2.

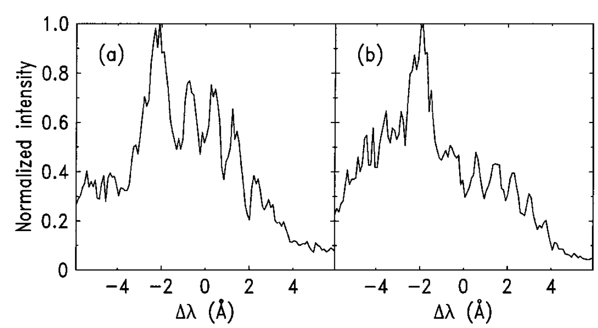

FIG. 2. Experimental spectra of harmonic 15 generated with a delay of 120 fs: (a) at 3.5×10^{14} W/cm^2; (b) at 5×10^{14} W/cm^2.

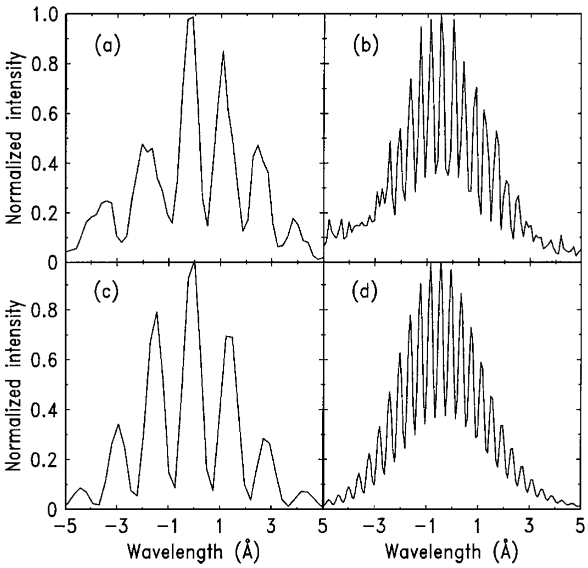

FIG. 3. Experimental [(a) and (b)] and theoretical [(c) and (d)] spectra of harmonic 11. (a) and (c): Generated at 2×10^{14} W/cm^2 with a delay of 120 fs. (b) and (d): Obtained at 3×10^{14} W/cm^2 with a delay of 450 fs.

remarkably reproduced. We can thus interpret the reduced contrast of Fig. 3b: the higher intensity induces more ionization which in turn results in a stronger defocusing of the laser beam. The intensity of the second harmonic beam is then reduced by more than a factor of 4. By studying the contrast as a function of the laser intensity, we get direct information on the ionization of the medium that can be compared to the generation efficiency, providing a test of the HHG theories. The reduced contrast observed in Fig. 1 when increasing the order reflects the fact that higher order harmonics are generated efficiently later in time, at higher intensity, and are thus more affected by ionization.

Using our numerical analysis, we are also able to interpret the spectral asymmetry observed for high harmonics. It happens to be particularly interesting because it gives indication on a possible harmonic chirp. Indeed, ionization introduces a temporal asymmetry in the problem: the leading edge of the first laser pulse is less affected than its trailing edge and than both edges of the second pulse. This results in a larger asymmetry between the leading than between the trailing edges. If there is a negative chirp of the harmonic emission, the contrast on the blue side of the spectrum (associated with the leading edges) should be more affected than on the red side. The observed behavior is thus consistent with a negative harmonic chirp, in agreement with recent studies [22–24]. Note that the relative influence of the ionization and of the chirp can be varied by playing with the relative intensity of the two laser pulses. By slightly rotating the birefringent plate, we were able to increase or decrease the contrast on either side of the spectrum depending on which pulse is more intense.

To summarize, we have shown that it is possible to generate two phase-locked harmonic sources in time, and to observe spectral fringe patterns. Very high contrasts can be obtained, which are degraded when ionization of the medium becomes important. The distortion of the fringe pattern as a function of the generating conditions gives insight into the underlying physics of harmonic generation (chirp, efficiency, ionization). This result is interesting from a fundamental point of view but also, it opens the possibility of performing XUV frequency-domain interferometry. We now demonstrate it in a plasma experiment where we probe the electronic density created in a high density gas jet by an intense laser pulse.

The experimental setup is shown in Fig. 4. The two 11th harmonic pulses are generated in a xenon gas jet with a 300 fs delay and refocused without magnification with a toroidal mirror into a helium jet. After analysis by an XUV flat-field spectrometer, the fringes are detected on microchannel plates (MCP) coupled to a phosphor screen and a charge-coupled device (CCD) camera. To achieve the best spectral resolution, the MCP were tilted to a grazing incidence of 8°. The focal spot diameter measured for the 11th harmonic by the knife edge technique was 157 μm (at $1/e$). The plasma was created by focusing about 160 mJ of the UHI10 laser with a $f/3.5$ off-axis parabolic mirror in the high pressure helium jet. A gas density of 2.4×10^{19} cm^{-3} was measured by IR interferometry. The overall size of the plasma measured by IR shadowgraphy was 270 μm. The plasma was probed at 45° from the propagation axis of the TW laser. Note that harmonic 11 is below the first excited state of helium (21 eV), thus preventing absorption of the harmonic by the neutral gas surrounding the plasma.

We have recorded the harmonic fringe pattern as a function of the delay between the TW pulse generating the plasma and the harmonic probes (zero delay for pump in between the two probes). When the plasma is generated before (positive delays) or in between the two pulses, the fringes are shifted. Since the harmonic beams are smaller than the plasma, the shift is homogeneous in the spatial dimension. In Fig. 5a, we show the spatially integrated

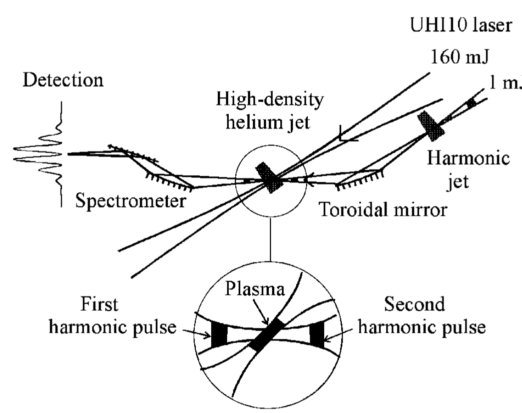

FIG. 4. Experimental setup for the spectral interferometry with harmonics.

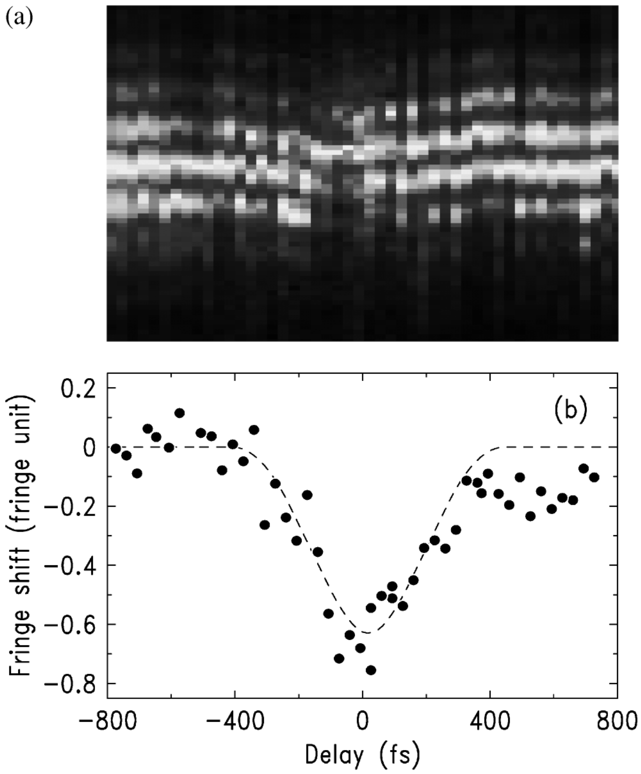

FIG. 5. Variation of (a) the spatially integrated fringe pattern and (b) the average fringe shift, with the delay between the plasma generating laser beam and the 11th harmonic beams.

fringe pattern from single-shot images as a function of the pump-probe delay. A clear shift can be seen in the middle of the image, when the plasma is created in between the two harmonic pulses. A blurring of the fringes occurs for two different delays (around -400 and around 0 fs, respectively) when each harmonic probe coincides with the fast ionization front of the pump. The time varying refractive index induces a fast dephasing on this harmonic beam resulting in a blurring of the fringes. Note that the short wavelength should ensure that refraction on the sharp density gradients is negligible. The average shift is plotted in Fig. 5b. The dashed curve is the theoretical shift obtained for the relevant experimental parameters. The calculated curve fits well the experimental one for an electron density of 6×10^{19} cm^{-3} which is in quite good agreement with the expected density (4.8×10^{19} cm^{-3}) assuming fully stripped ions. The slower shift of the fringes compared to the expected very fast optical field ionization front is, in fact, due to the geometry (probe at 45°) that lowers the time resolution to about 200 fs. One notices that a nonzero phase shift exists when both harmonic pulses propagate through the plasma. This behavior was systematically observed and is not understood yet. It can be due to an increase of the plasma size due to postpulse ionization of the surrounding gas by energetic electrons. Further studies are underway to clarify this point.

In conclusion, we have performed the first time-resolved XUV interferometry experiment at the femtosecond time scale using high-order harmonics. Frequency-domain interferometry is made possible thanks to the phase locking existing between two harmonic pulses generated at different instants in the same medium despite ionization. The technique is well suited to the study of plasmas created on solid target where densities as high as 10^{23} cm^{-3} could be probed. More generally, it opens new perspectives for the study of ultrafast phenomena in ultrashort laser-matter experiments.

[1] T. E. Glover *et al.*, Phys. Rev. Lett. **76**, 2468 (1996).
[2] A. Bouhal *et al.*, J. Opt. Soc. Am. B **14**, 950 (1997).
[3] Y. Kobayashi *et al.*, Opt. Lett. **23**, 64 (1998).
[4] J. Larsson *et al.*, J. Phys. B **28**, L53 (1995).
[5] P. Salières *et al.*, Phys. Rev. Lett. **74**, 3776 (1995).
[6] M. Bellini *et al.*, Phys. Rev. Lett. **81**, 297 (1998).
[7] T. Ditmire *et al.*, Phys. Rev. Lett. **77**, 4756 (1996).
[8] L. Le Déroff *et al.*, Phys. Rev. A (to be published).
[9] L. B. Da Silva *et al.*, Phys. Rev. Lett. **74**, 3991 (1995).
[10] P. Zeitoun *et al.*, Nucl. Instrum. Methods Phys. Res., Sect. A **416**, 189 (1998).
[11] J. J. Rocca *et al.*, Opt. Lett. **24**, 420 (1999).
[12] In parallel to this work and in collaboration with the Lund group, we used harmonics in a spatial interferometry experiment: D. Descamps *et al.*, Opt. Lett. (to be published).
[13] J. Zhou *et al.*, Phys. Rev. Lett. **76**, 752 (1996).
[14] M. Gaarde *et al.*, J. Phys. B **29**, L163 (1996).
[15] E. Tokunaga *et al.*, Opt. Lett. **17**, 1131 (1992).
[16] J. P. Geindre *et al.*, Opt. Lett. **19**, 1997 (1994).
[17] S. Guizard *et al.*, J. Phys. C **8**, 1281 (1996).
[18] B. Colombeau *et al.*, Acta Phys. Pol. A **78**, 85 (1990).
[19] R. Zerne *et al.*, Phys. Rev. Lett. **79**, 1006 (1997).
[20] M. Lewenstein *et al.*, Phys. Rev. A **49**, 2117 (1994).
[21] A. L'Huillier *et al.*, Phys. Rev. A **46**, 2778 (1992).
[22] K. J. Schafer *et al.*, Phys. Rev. Lett. **78**, 638 (1997).
[23] Z. Chang *et al.*, Phys. Rev. A **58**, 30 (1998).
[24] P. Salières *et al.*, Phys. Rev. Lett. **81**, 5544 (1998).