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Extreme ultraviolet interferometry measurements with high-order harmonics

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We demonstrate that high-order harmonics generated by short, intense laser pulses in gases provide an interesting radiation source for extreme ultraviolet interferometry, since they are tunable, coherent, of short pulse duration, and simple to manipulate. Harmonics from the 9th to the 15th are used to measure the thickness of an aluminum layer. The 11th harmonic is used to determine the spatial distribution of the electron density of a plasma produced by a 300-ps laser. Electronic densities higher than $2 \times 10^{19}$ electrons/cm$^3$ are measured. © 2000 Optical Society of America

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High-order harmonic generation in gases has been studied extensively during the past few years, motivated by the potential of this extreme ultraviolet (XUV) radiation to become a useful source for applications.\textsuperscript{1–4} Much effort has been devoted to improving the harmonic-generation conversion efficiency\textsuperscript{2–7} as well as characterizing the radiation properties spatially\textsuperscript{8,9} and temporally.\textsuperscript{10,11} Recently we showed\textsuperscript{8,9} that two spatially separated sources of high-harmonic radiation created by the same laser pulse are phase locked\textsuperscript{12} and interfere when they are superposed in the far field. This interesting property was used to investigate the temporal coherence of high-order harmonic emission.\textsuperscript{13} Our aim in this Letter is to show that phase locking also allows us to perform interferometry in the XUV range to probe thin solid films and dense plasmas.

Interferometry has been demonstrated in the soft-x-ray wavelength range by use of x-ray lasers.\textsuperscript{14,15} High-order harmonic generation adds to the technique the major improvements of tunability, ultrashort (subpicosecond) pulse duration, compactness, and simplicity. For some experiments, it is very useful to adjust the wavelength far from an absorption line or band. The spectrum of the harmonic source allows us to obtain interference patterns at different wavelengths corresponding to odd harmonics of the fundamental radiation. High-order harmonic sources can emit during a few tens of femtoseconds. This property offers the unique possibility of studying very fast mechanisms that occur, for example, in ultrashort laser–plasma experiments. Finally, the possibility of creating two phase-locked independent harmonic sources allows us to work with a simple optical setup in which splitting and alignment are done with light beams in the near infrared and not in the XUV.

In this Letter we report what is to our knowledge the first interferometric measurements in the XUV by use of high-order harmonics generated in a gas jet as a source of coherent radiation. The experimental setup is quite similar to that used in previous harmonic temporal coherence studies.\textsuperscript{13} We use a terawatt Ti:sapphire laser, operating at 10 Hz and delivering 200-mJ 110-fs pulses in a 9-nm bandwidth centered around 790 nm. The infrared laser pulse is split into two identical pulses in a setup resembling a Michelson interferometer (Fig. 1). In one arm, a mirror is slightly tilted so that focusing of the laser beams results in two spatially separated foci. At the output of the setup, we reduce the aperture of the beam from 25 to 11 mm with an iris located just before the 500-mm focal-length lens that focuses the beams into a pulsed krypton gas jet. The experiment is performed with energy of typically 0.5 mJ in each infrared pulse after the aperture, corresponding to a total energy of 6.5 mJ before the first beam splitter. In these conditions, the peak intensity in each focus is estimated to a few times $10^{14}$ W/cm$^2$. A normal-incidence spherical grating (1200 lines/mm, $f$ = 500 mm) is used to select, with the help of a slit, a given harmonic order and to image, with a magnification of 1, the gas-jet plane. From the image plane, the two harmonic beams diverge and overlap in the far field, resulting in an interference pattern. This pattern is observed with microchannel plates (MCP’s) coupled to a phosphor screen and captured with a 16-bit CCD camera through a camera objective. The distance between the image plane and the MCP is fixed at 0.9 m.

The far-field fringe pattern can be observed with single-shot acquisition and exhibits good contrast, better than 30% over a large part of the harmonic beam. Using the knife-edge technique in the image focal...
plane, we find a 130-μm separation between the two images of harmonic sources, as is confirmed by the fringe spacing, and measure the diameter of each of the two foci of the 13th harmonic to be roughly 25 μm (FWHM). The divergence of the harmonic beams is deduced from the spot size on the MCP detector and found to be equal to approximately 4 mrad.

In the first part of the experiment, we probe a free-standing aluminum step filter, using harmonics of orders 9 to 15 (14–24 eV). Aluminum is chosen for its relatively good transmission in the XUV range and being of the refractive index is quite different from unity allowing its absorp-

**Fig. 1.** Experimental setup.

**Fig. 2.** Interference pattern with the 13th harmonic through an aluminum step filter (two-shot integration). The envelope of the spatial distribution of the harmonic beam is removed by use of a Fourier-transform technique.

imation coefficient is 1 order of magnitude higher in this spectral region. The delay between the plasma-producing laser pulse and the harmonic pulses at the target is held constant at 1.2 ns.

The optimum probe wavelength for studying this plasma is found to be that of the 11th harmonic. Single-shot interference patterns are presented in Fig. 3, together with the approximate positions of the aluminum target. In Fig. 3(a), the target is not irradiated, and the fringe pattern presents good fringe visibility. Figures 3(b) and 3(c) show results obtained with an irradiated target but with slightly different target–probe distances; the probe beam is closer to the target in Fig. 3(c). Compared with that in Fig. 3(a), the noise level in Figs. 3(b) and 3(c) increases slightly owing to self-emission, and the fringes are tilted. To obtain an estimate of the electron density of the...
plasma, we can approximate the observed fringe shift $N$ (in fringe units) at wavelength $\lambda$:

$$N = \int_0^d \frac{1 - n_{\text{ref}}}{\lambda} \, dx \approx \frac{d}{2\lambda N_{\text{cr}}} \left(1 - N_{\text{cr}}/N_e\right)^{1/2}.$$

In Eq. (1), $n_{\text{ref}} = (1 - N_{\text{cr}}/N_e)^{1/2}$ is the plasma’s refractive index and $d$ is the distance that is traversed by the probe beam through the plasma. We assume that the plasma is uniform along the probe path and that the electron density $N_e$ is much smaller than the critical density $N_{\text{cr}} = 1.1 \times 10^{21} \lambda^{-2}$ (in is in micrometers and $N_{\text{cr}}$ is in inverse cubic centimeters). Refraction effects are neglected. We estimate the distance $d$ experimentally to the order of 0.1 mm by moving the plasma-producing laser beam until the fringe tilt disappears. In Fig. 3(b), only the fringes in the top part of the image are tilted. Those that are far from the target are not perturbed, which allows us to determine the fringe shift that is due to the effect of the plasma and, consequently, the electron density. Isodensity lines corresponding to electron densities of $0.5 \times 10^{20}$, $1 \times 10^{20}$, and $2 \times 10^{20}$ electrons/cm$^3$ (from bottom to top) are indicated by the dashed curves in Fig. 3(b). In Fig. 3(c), the entire probe beam is perturbed by the plasma, which results in a tilt of the whole fringe pattern. This result indicates a quite homogeneous density gradient. Our analysis allows us to give only an upper limit to the electron densities, assuming that the bottom edge of the image is not influenced much by the plasma. Fringe shifts as high as 0.8$\lambda$ are observed close to the target surface, indicating densities of at least $2.5 \times 10^{20}$ electrons/cm$^3$. A simulation of the plasma expansion in our experimental conditions has been performed\cite{17} that confirms the absolute values of the electron density as well as the shape of the electron-density gradient obtained from Figs. 3(b) and 3(c).

In conclusion, we have demonstrated the possibility of performing interferometry in the XUV range with higher-order harmonics. We have applied this technique to probe thin solid aluminum films and laser-produced plasmas. The higher-order harmonic source combines a tunable short wavelength and an ultrashort pulse duration, which makes it possible to freeze in time the electron density of an expanding dense plasma. Consequently, this experiment opens new perspectives for the characterization of dense plasmas created in different regimes, from nanosecond to subpicosecond time scales.

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