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Extreme Ultraviolet Fourier-Transform Spectroscopy with High Order Harmonics


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We demonstrate a new scheme for extreme ultraviolet (xuv) Fourier-transform spectroscopy based on the generation of two phase-locked high-harmonic beams. It allows us to measure for the first time interferograms at wavelengths as short as 90 nm, and open the perspective of performing high-resolution Fourier-transform absorption spectroscopy in the xuv. Our measurements also demonstrate that a precise control of the relative phase of harmonic pulses can be obtained with an accuracy on an attosecond time scale, of importance for future xuv pump-xuv probe attosecond spectroscopy.

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Fourier-transform spectroscopy (FTS) is a widely used technique in the infrared, visible, and ultraviolet ranges [1,2]. This is due to both its wavelength accuracy and its extremely high resolution, only limited by the length and stability of the delay line, that can surpass that of the best grating spectrometers. Unfortunately few attempts have been made to extend this technique to the extreme ultraviolet (xuv) range [3], because many difficulties arise in this demanding spectral domain. First, the beam to characterize has to be split in two replicas. Since xuv beam splitters did not exist until recently (good optical elements with small absorption are still a challenge) [4,5], wave front division of a spatially coherent beam was proposed [6,7]. Second, the delay between the two replicas must be precisely controlled and varied on the time scale of the xuv optical period, i.e., on an attosecond time scale; the stability and resolution of the delay line must be on a nanometer length scale, which is challenging in a lab environment. Finally, the beams should be combined in order that they interfere, which raises the problem of the combining element. The signal oscillating as a function of the delay [or optical path difference (OPD)] is a first-order autocorrelation (or interferogram) of the incoming beam. Its Fourier transform gives the power spectrum of the radiation. The scaling of both optical and mechanical tolerances with the wavelength, together with the strong absorption in the xuv range, are thus the main reasons for the absence of FTS data at wavelengths below 130 nm.

An elegant way to produce replicas (first requirement) is to generate two mutually coherent xuv beams, without need of splitting. This has recently become possible thanks to the generation of high harmonics (HHG) of intense laser pulses [8]. Two space-separated, usually undelayed, phase-locked harmonic pulses can be generated from two replicas of the laser pulse focused at two different locations in a rare gas jet [9]. In this scheme, the two beams combine in the far field by natural diffraction and produce an interference pattern similar to the Young’s double slit fringes. Similarly, two time-separated, spatially superposed, phase-locked harmonic pulses are generated from two time-delayed laser pulses focused at the same location [10,11]. This scheme allows frequency-domain interferometry. Both techniques have been applied to plasma diagnostic using xuv interferometry [10,12]. The time-separation method has been used to perform Ramsey-fringe spectroscopy [13,14] or to generate a frequency comb [15,16]. However, none of the two methods in their initial form is adapted to FTS.

Spectral measurement of harmonics has been reported in a scheme related to space separation, but under assumption of spatial coherence [17]. In the time-separation scheme, the intrinsic nonlinearity of HHG does not allow overlapping the two pulses in time without strongly distorting them. In contrast, the combination of a space and a variable time separation makes FTS possible. In a first step, by sampling the contrast of the far-field fringes at different delays, one has access to the coherence time [18,19]. Now, by continuously monitoring the variation of the fringe pattern as a function of the delay, one should measure the harmonic first-order autocorrelation. One therefore accesses to both the absolute wavelength and the line shape of the xuv light.

In this Letter, we demonstrate xuv Fourier-transform spectroscopy of high-order harmonics in the space- and time-separation scheme. Our interferometric setup does not rely on any xuv optics, but on an infrared Michelson-type setup, and would allow performing broadband absorption spectroscopy of a target gas. To our knowledge, we measure for the first time interferograms at wavelengths as short as 90 nm, corresponding to harmonic 9. We study in detail the influence of the separation between the two laser foci, and observe a transition from a first-order autocorrelation of the harmonic beam to a high-order autocorrelation...
of the laser beam. Measurements show good agreement 
with simulations of interferometric traces including both
the single-atom response and the beam propagation.

In our experimental setup, shown in Fig. 1, an intense
infrared (IR) pulse from the 810 nm, 80 mJ, 60 fs LUCA
laser is sent in a Michelson-type interferometer that pro-
duces two time-delayed IR pulses. By slightly misaligning
one arm, the two pulses can be focused at different loca-
tions (separated by distance \(d\)) in a 90 Torr xenon jet,
generating two phase-locked harmonic sources. The laser
intensity in each focus reaches \(10^{14}\) W/cm\(^2\), and the waist
is 100 \(\mu m\). We then filter out the strong fundamental
radiation, either with a low-resolution grating monochro-
mator for observing one single harmonic, or with an alu-
inium filter (1000 \(\AA\)) for transmitting the full spectrum
of the laser beam. Measurements show good agreement 
with simulations of interferometric traces including both
the single-atom response and the beam propagation.

A crucial question for realizing the above FTS scheme
is: how separated must the two laser foci be in order to
generate two “independent” harmonic sources? We first
show in Fig. 2(a) the trace obtained for two overlapping
laser foci. The 9th harmonic signal shows pronounced
oscillations at the laser optical period. In contrast to the
FTS scheme, these oscillations are due to the modulation
of the total radiated harmonic energy. Indeed, two IR laser
beams interfere in the generating medium, which modu-
lates the total laser intensity and consequently the har-
monic production. Trace 2(a) is thus a high-order
autocorrelation of the IR laser pulse [20]:

\[
S_q^p(\tau) = \int_{-\infty}^{\infty} |E_q(t) + E_q(t-\tau)|^2 dt,
\]

where \(E_q\) is the laser electric field, and \(p\) the effective order of
nonlinearity. Since we are not in a perturbative regime, \(p\)
is much smaller than the harmonic order \(q\) [21]. Frequency
spectrum of trace 2(a) (FT) is plotted in Fig. 2(b). It ex-
hibits a peak at the laser frequency and rapidly decreasing
harmonic peaks, corresponding to the decomposition of a
quasiperiodic function into its discrete frequency com-
ponents. Note that there is no detectable signal at the 9th
harmonic. In order to measure the first-order autocorrela-
tion of the harmonic beam, we now separate the two laser
foci by slightly tilting one mirror in the interferometer. The

![FIG. 1 (color online). Experimental setup for the Fourier-
transform spectroscopy of high harmonics.](image)

![FIG. 2. (a) Experimental trace obtained for the 9th harmonic
generated by two collinear laser pulses \((d = 0)\). (b) Fourier
transform of the trace.](image)
influence of this separation is shown in Fig. 3. For partially separated laser beams \((d = 90 \, \mu \text{m})\) is of the order of the laser waist), the trace is still strongly modulated at the laser period, but shows the onset of oscillations at the 9th harmonic [Figs. 3(a) and 3(b)]. At intermediate separation \((d = 135 \, \mu \text{m})\), the trace exhibits pronounced harmonic oscillations over almost the full observation range. Finally, at maximum separation \((d = 300 \, \mu \text{m})\), the trace frequency modulation is strongly reduced [Figs. 3(e) and 3(f)]; the period of the far-field fringes is very small, which means that signal is reduced and more sensitive to vibrations. In all these traces, we performed a very fine sampling to demonstrate the high accuracy and stability of our setup: a harmonic period (300 as) is sampled with 10 points (1 point every 33 as). The clear oscillations in the zooms of Fig. 3 demonstrate the accurate control of the relative phase of the harmonic pulses on an attosecond time scale. This will be crucial for performing attosecond xuv pump-xuv probe spectroscopy [22]. Note that, due to the absence of axial symmetry in the two-foci geometry, a full 3D simulation is required in principle. On the other hand, the accurate calculation of the interferometric traces requires a fine sampling of the delay between the two laser pulses. To limit the calculation time, we thus use a 2D model neglecting the spatial dimension perpendicular to the plane of the two laser beams, as unrelated to the interference effect. The Fourier transforms of the simulated traces are plotted in Figs. 4(b), 4(d), and 4(f). They show qualitatively the same evolution with increasing source separation as the experimental curves. The trace simulated for maximum beam separation of 300 \(\mu \text{m}\) was compared to the first-order autocorrelation of the harmonic beam generated by a single laser beam, and is indeed exactly the same, so that its Fourier transform shown in Fig. 4(f) is really the power spectrum of each harmonic beam, as would be obtained with a conventional (high-resolution) spectrometer. The inset in Fig. 4 shows the good agreement between the experimental and simulated H9 spectra of Figs. 4(e) and 4(f).

In order to validate our measurements, we have performed systematic simulations of the interferometric traces in the experimental conditions [23]. The distribution of nonlinear atomic dipole in the total driving field (sum of the two fields separately focussed) is calculated in the strong-field approximation (SFA) [24]. The macroscopic xuv field is obtained from the propagation equations, taking into account linear and nonlinear atomic dispersion, ionization of the medium [under Ammosov-Delone-Krainov theory [25]], and electronic dispersion. Note that, due to the absence of axial symmetry in the two-foci geometry, a full 3D simulation is required in principle.

![FIG. 3. Experimental traces of the 9th harmonic (left panels) and corresponding zooms of the first two laser periods (right panels), for different values of the laser foci separation \(d\): (a), (b) \(d = 90 \, \mu \text{m}\), (c), (d) \(d = 135 \, \mu \text{m}\), (e), (f) \(d = 300 \, \mu \text{m}\).](image)

![FIG. 4. (a), (c), (e) Fourier transforms of the experimental traces presented in Fig. 3. (b), (d), (f) Fourier transforms of the corresponding simulated traces. The inset shows enlarged H9 spectra of (e) (white circles) and (f) (black dots).](image)
situation arises at the intermediate separation of \(d = 135 \mu m\) [Figs. 4(c) and 4(d)], with the appearance of sidebands at even harmonic frequencies 8 and 10; these sidebands result from a modulation, at the laser period, of the harmonic electric field. This is an intermediate situation between Eqs. (1) and (2) with a mixing between first-order autocorrelation of the harmonic beam and high-order autocorrelation of the laser beam.

By setting the grating in the zeroth order and using an aluminum filter, our setup allows performing a broadband spectral measurement. The interferogram then exhibits a beating pattern with the periodicity of a half laser cycle. The corresponding spectrum contains the contribution of the experimentally selected harmonic order (H11 to H17). By placing a target gas jet in the image plane of the toroidal mirror (see Fig. 1), our setup has the potential for Fourier-transform absorption spectroscopy in the gas phase. The close enough locations of the harmonic beams ensure that they probe the same target. Today, structure and energetics of atoms and molecules [26,27], ions, and reactive species (radicals) [28] are primarily investigated in high-resolution xuv spectroscopy, both on narrow band lasers and on incoherent broadband sources such as synchrotron radiation. Moreover, time-resolved spectroscopy should become a key tool for studying dynamics in photochemistry: after pump pulse triggers the reaction, transient intermediate is probed in absorption. In such pump/probe scheme, femtosecond resolution requires broadband and coherent xuv pulses, the spectral resolution being appropriately adapted. To that goal, the proposed FTS scheme using ultrashort harmonics should be particularly suitable.

The large number of photons [\(>10^{11}\) per harmonic pulse [29]] and short pulse duration [a few 10 fs [30]] as compared to the picosecond regime [31] allow to probe highly dilute transient species. Moreover, the xuv probe pulse is naturally synchronized with the generating laser beam that can provide the pump pulse. In our experiment, the resolving power was limited to 2 \(\times\) 10\(^5\) both by the piezotranslation range (90 \(\mu m\)) and by the long acquisition time at 20 Hz laser rep rate. With the currently available 1 kHz lasers, and using external indexation of the position allowing an OPD of 1 cm [6], a resolving power of 2 \(\times\) 10\(^5\) at 90 nm could be achieved.

In conclusion, we have demonstrated the feasibility of xuv Fourier-transform spectroscopy based on the generation of two phase-locked harmonic beams. This scheme does not rely on xuv optics and can cover a large spectral range (many 10 eV) with a high accuracy. Thanks to the intense ultrashort pulses, it has the potential for time-resolved spectroscopy on short-lived transient species with a pump-probe setup. We therefore believe that such a system will be an important tool for the development of xuv high-resolution Fourier-transform spectroscopic applications.

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[23] In the simulation, a 90 fs laser pulse is focused 3.5 mm after a 3 mm xenon jet. The other parameters are indicated in the text.