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Measuring the temporal coherence of a high harmonic generation setup employing a Fourier transform spectrometer for the VUV/XUV

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Abstract

In this experiment we used an 800 nm laser to generate high-order harmonics in a gas cell filled with Argon. Of those photons, a harmonic with 42 eV was selected by using a time-preserving grating monochromator. Employing a modified Mach-Zehnder type Fourier transform spectrometer for the VUV/XUV it was possible to measure the temporal coherence of the selected photons to about 6 fs. We demonstrated that not only could this kind of measurement be performed with a Fourier transform spectrometer, but also with some spatial resolution without modifying the XUV source or the spectrometer.

Keywords

Fourier transform spectrometer
HHG
Temporal coherence
Spatial coherence
Soft X-ray
XUV

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1. Introduction

The measurement of temporal coherence of visible or infrared radiation can be done employing equipment like a Michelson interferometer [1]. In a Michelson interferometer the beam is split in two components, where one of them is delayed with respect to the other by changing the path length in one of the interferometer arms. Both beams are then recombined and the resulting interference pattern is recorded as a function of path length difference. By scanning the delay and observing the changes of the interference fringes, one can measure the field autocorrelation of the radiation [2]. From that one can get information about the temporal coherence as well as the spectrum of the radiation. Temporal coherence is seen in the presence or absence of interference between the two overlapping beams while spectral information can be retrieved through a Fourier transformation of the recorded interference fringes as function of path length difference [2]. Although the principle is the same for shorter wavelengths in the Vacuum Ultraviolet (VUV) or Extreme Ultraviolet (XUV) regime, the implementation is much more difficult. Key reasons for this are that VUV/XUV radiation is strongly absorbed by all kinds of materials, even air, making it necessary to work in vacuum and to use only reflective optics as well as special radiation detectors. Furthermore, one has to keep the angle of incidence large enough to achieve total external reflection, since reflectivity at normal incidence is very poor at these photon energies [3]. This also implies that the number of reflections should be kept at a minimum. Additionally, the requirements on the surface quality of the optics increase with decreasing wavelength, and so do the demands on mechanical stability and positioning precision of movable parts.

Although the demands on a setup rise with the photon energy, the temporal coherence of XUV pulses has been measured. This was done e.g. for Free Electron Lasers (FELs) pulses with schemes employing wave front dividing beam splitters [4,5] or for collisionally pumped soft-x-ray lasers using multilayer intensity dividing beam splitters [6].

To measure the temporal coherence of XUV radiation produced in a High Harmonic Generation (HHG) process [7], one can use dedicated setups as done in Refs. [8,9]. In these setups the driving laser beam is split prior to the HHG generation in a semitransparent intensity-dividing beam splitter. These two phase-locked beams are then used to generate two separate XUV sources in the same gas very close to each other. Hence, the XUV radiation from both sources is also phase-locked and the coherence properties can be analyzed by overlapping the two XUV beams. However, this method cannot be used easily at

existing sources and beamlines due to limitations in laser power and accessibility of the setups.

The development of wave front dividing beam splitting methods made a recent development of Fourier transform spectroscopy into the VUV and XUV region possible [10,11]. This allows Fourier transform spectrometers to be used in the VUV and XUV range to analyze the so-called visibility of the interference pattern in the same way as can be done for visible and infrared sources. Thus, the measurement of the temporal coherence of a source, without the need of modifying it is possible.

A Fourier transform spectrometer, although capable of measuring the coherence length, is however not capable of measuring the pulse length. A proper measurement of the pulse length is a topic of its own and is discussed for optical wavelengths in detail e.g. in Ref. [12]. The major difference is that one needs to measure not the field autocorrelation of the signal but the intensity autocorrelation which requires a second order process. Such a second order process requires high light intensity and adds additional complexity which makes the measurement of the pulse length unachievable with a conventional Fourier transform spectrometer.

2. EXPERIMENTAL

In order to demonstrate the capability of using a Fourier transform spectrometer (FTS) to measure the temporal coherence of short XUV pulses, the HELIOS (*High Energy Laser Induced Overtone Source*) HHG light source at Uppsala University [S. Plogmaker et al. (in manuscript)], and an in-house developed Mach-Zehnder type interferometer for the XUV radiation [11] were used. Briefly, HELIOS is an HHG setup which uses an amplified commercial Ti:sapphire based laser system (Coherent Inc.) with a center wavelength of 800 nm and a pulse duration of 35 fs. The repetition rate of the system is 5 kHz resulting in a total output power of 12.5 W. The laser is focused into a gas cell filled with Argon or Neon to generate XUV radiation. The generated radiation is inherently coherent [13] and the upper limit of its pulse duration is set by the pulse duration of the driving laser. The energies that are generated at HELIOS today are in the order of 20 eV to 70 eV and, since the radiation is generated in noble gases, the harmonics are limited to the odd harmonics of the energy of the driving laser photons.

Since several harmonics are generated in the HHG process at the same time a monochromator is used to select a single harmonic. A traditional monochromator design would prolong the pulse due to the path length difference introduced by every single groove of the grating. To keep this prolongation small, HELIOS uses a monochromator design employing a grating in a so called off-plane mount [14,15]. The HELIOS light source will be discussed in further detail in a forthcoming publication [S. Plogmaker et al. (in manuscript)].

The spectrometer used in this experiment is a FTS of modified Mach-Zehnder type [11]. It is equipped with a large-aperture comb-like wave-front-dividing beam splitter, made from a super polished single crystal silicon mirror. An identically slotted mirror is used as a beam mixer. The FTS was earlier tested at the I3 beamline [16] at the Max IV laboratory, Lund, Sweden, both in direct [17] and indirect detection of the beam, and has been shown to work at least up to 55 eV photon energy.

FIG. 1: Schematic drawing of the HELIOS source with the Fourier transform spectrometer attached behind the monochromator.

In the current experiment, Argon was used in the gas cell and the FTS was mounted directly behind the monochromator allowing measurements of the temporal coherence of a single harmonic of the HHG radiation. A schematic drawing of the setup used in the experiment is shown in FIG. 1. A typical image of the XUV radiation on the detector of the FTS can be seen

FIG. 2: FTS detector image of the 27th harmonic. The path length difference is $1.35\ \mu\text{m}$ (corresponding to $45.1\ \text{fs}$) for a) while it is zero μm for b). Hence, interference fringes are visible in b) but not in a). See text for details about the interference pattern.

in FIG. 2. Note that the path length difference in FIG. 2 a) is larger than the temporal coherence and hence no interference pattern can be seen. The fringes visible in FIG. 2 a) are static and do not change when the path length difference between the arms of the spectrometer is varied. This intensity pattern has its origin rather in the light passing first the beam splitter and then the beam mixer which both act as multi slits. The fringe pattern caused by the interference at zero path length difference can be seen in FIG. 2 b). Additionally, in both FIG. 2 a) and b), one can see a grid like structure which probably originates from a meshed aluminum foil in the monochromator that is used to block the infrared driving laser while transmitting the XUV radiation.

3. Results

125 The temporal coherence can be determined by measuring the visibility of the interference
 126 fringes when changing the delay between the two beam paths in the spectrometer. The
 127 visibility V_b is defined as [18]:

$$V_b = \frac{I_{max} - I_{min}}{I_{max} + I_{min}} \quad (1)$$

128 where I_{max} and I_{min} are the intensities of constructive and destructive interference of the
 129 observed fringes. The temporal coherence τ_c of the radiation can be calculated from the
 130 visibility as [18]

$$\tau_c = \int_{-\infty}^{\infty} |\gamma(\tau)|^2 d\tau \quad (2)$$

131 where $\gamma(\tau)$ is the complex degree of coherence which can be expressed as

$$\gamma(\tau) = \frac{V(\tau)}{V(0)}$$

132 and τ is the traveling-time difference for the light in the two arms of the spectrometer. V
 133 denotes the visibility V_b , but with a constant background subtracted. Note that the
 134 subtraction of background is necessary due to noise in the signal. This noise leads to an
 135 offset of the visibility V_b when it increases the intensity of an interference maximum I_{max} but
 136 decreases the intensity of an interference minimum I_{min} leading to an overestimation of the
 137 visibility V_b . When referring to the visibility from here on we refer to V and not to V_b (unless
 138 explicitly stated differently).

139 The way the temporal coherence was defined in equ. (2) is the same as used for measuring
 140 the longitudinal coherence of FLASH by Schlotter et al. [4]. This definition was introduced by
 141 Manel 1959 and its importance was exemplified in [19].

142 This definition bases the temporal coherence on an integral over the squared complex
 143 degree of coherence $\gamma(\tau)$. That way, one gets rather independent of the shape of the
 144 frequency spectrum, the temporal shape of the pulse or its chirp. This is an advantage
 145 compared to using peak-shape dependent parameters as full width at half maximum
 146 (FWHM), especially since one can see in FIG. 4 that the peak shape of the visibility seems to
 147 be more complex than a Gaussian peak.

FIG. 3: Intensity of a single pixel out of the interference pattern of the 27th harmonic (~42 eV) in gray plotted against the pulse delay. A zoomed cutout of a) is displayed in b), showing how the interference pattern consists of single interference fringes. Black dots denote measured points, gray and white modulations of the background show the slices used to gain I_{max} and I_{min} . The blue line/bars in a)/b) show the upper envelope used as I_{max} while the red line/bars show the lower envelope used as I_{min} .

148 To measure the temporal coherence of the XUV radiation behind the monochromator (see
 149 FIG. 1) the intensities of the interference patterns were recorded as a function of path length
 150 difference by recording the full 2D detector image while scanning the path length difference
 151 in the spectrometer. In this way, the intensity progression of every pixel of the detector
 152 image was obtained. Such an intensity progression for a single representative pixel is shown

in figure FIG. 3. The successive intensity values of a single pixel were divided in slices of ten measurements as can be seen in FIG. 3 b). One slice contains slightly more than one period of the interference fringes. The highest and lowest value within such a slice was assigned to $I_{\max}$ and $I_{\min}$ respectively. That way, it was possible to calculate a visibility curve for V_b by means of equ. (1) for every single pixel on the detector. Due to the slicing, the density of successive values of the visibility was reduced by a factor of ten compared to the intensity measurement. This results in one visibility value per 42 nm scanned path length difference (corresponding to a delay of 0.14 fs). To obtain the visibility of a whole detector region, all single-pixel visibilities in a specific region were averaged leading to a clear visibility distribution although the interference pattern on the detector looks quite complicated.

Due to assigning the maximum and minimum value of ten measurements to $I_{\max}$ and $I_{\min}$ the visibility V_b of a single pixel also contains the noise of the measurement. Since the interference picture contains about 16000 pixels this noise adds up to a constant background which can be subtracted. By subtracting this background one gets a background free visibility V as discussed in the beginning of this chapter.

FIG. 4: Measured visibility of the fringes of the 27th harmonic (42 eV) in red measured on the whole detector image as shown in FIG. 2 plotted against the path length difference of the spectrometer. A linear background has been subtracted. The black line represents a fit consisting of one Gauss peak plotted in light gray symmetrically surrounded by two smaller ones per side in dark gray.

FIG. 4 shows this visibility V for the whole detector region plotted against the path length difference in the spectrometer. The visibility curve can be fitted with good agreement by a main Gaussian, which is symmetrically surrounded by two smaller Gauss peaks per side. The parameters for the fit can be found in

Table 1 together with the temporal coherence determined using equ. (2). The origin of this structure is not known but could come from a pulse shape that is more complex than that of a Gaussian.

Note that the visibility is unlikely to reach one, even for absolutely coherent radiation. This has its reason in the nature of the beam splitter and beam mixer in the FTS. Due to the multi slit character of the beam splitter, the beams hitting the beam mixer will have a diffraction pattern. This means that the intensity from each arm of the spectrometer does not necessarily need to be equal for all parts of the detector. This is due to the fact that this ratio depends on whether rather diffraction maxima or minima of the beam splitter are blocked by non-reflective/non-transitive parts of the beam mixer.

The spatially resolved detector image of the FTS makes it possible to measure the temporal coherence not only for the whole beam at once but also spatially resolved. This was done for seven subareas at different places of the detector as shown in FIG. 5. This analysis shows that the visibility as well as temporal coherence differs at different areas of the beam. As one can see, the visibility of the subareas is in all cases higher than the one on the full detector. This behavior has its origin in the fact that the average intensity in the subareas is higher than the average intensity on the whole detector image. This means that the dark parts of the detector image have a rather low visibility of fringes, most due to the much lower signal to noise ratio in them.

It is important to note that the numbers shown in

Table 1 should not be seen as quantitative values describing the exact behavior of the visibility in the single subareas of the detector regions. They are rather meant to show a more qualitative picture of the visibility changes in FIG. 5. The fit parameters might vary quite drastically when repeating the measurement although the actual visibility curve does look quite similar. This behavior might have its origin in the large number of free parameters that are used to describe the visibility curve.

The temporal coherences reported here are about half of the ones measured by Bellini et al. [8] for long electron trajectories in the HHG process. Furthermore, we did not detect a

200 component of the radiation with much larger coherence length as expected in the middle of
201 the HHG radiation cone originating from short electron trajectories in the HHG process [8].
202 This might be due to uncertainties in the alignment of the spectrometer relative to the XUV
203 beam. On account of this, the amount of radiation originating from electrons on short
204 trajectories might have been reduced so much that their signal was drowned in the noise of
205 the detector.

FIG. 5: a) Detector image of the 27th harmonic (42 eV). Green rectangles denote the areas, which were analyzed separately to extract their visibilities. b) Visibilities measured for the full detector image as well as the subareas from a) plotted in dark red against the path length difference of the spectrometer. A linear background was subtracted and each spectrum was vertically offsetted by 0.075 from the previous one for visual clarity. Light gray curves showing the visibility of the full detector image are plotted to ease the comparison between the subareas. The temporal coherences for each subarea are noted to the left.

Table 1: Fit parameters obtained from fitting the visibility of the whole detector image of the 27th harmonic (~42 eV) by a main Gaussian peak symmetrically surrounded by two smaller ones. Note that the positions of the side peaks are at $x_0 + x_s$ and $x_0 - x_s$. The width σ denotes the standard deviation.

areas	temporal coherence [fs]	main peak			1 st side peak			2 nd side peak		
		amp. A [10 ⁻²]	pos. x_0 [fs]	width σ [fs]	amp. A [10 ⁻²]	pos. x_s [fs]	width σ [fs]	amp. A [10 ⁻²]	pos. x_s [fs]	width σ [fs]
full image	5.5	7.6	0.0	2.7	0.85	8.5	1.6	0.65	14	8.0
1	6.6	9.0	-0.033	3.3	1.1	7.1	2.1	0.75	17	5.8
2	4.9	12.6	-0.070	2.6	1.7	8.3	1.6	0.75	18	8.2
3	5.2	12.8	-0.049	2.6	3.1	8.5	1.6	1.6	18	3.9
4	7.7	11.1	0.054	3.6	2.5	11	3.0	0.82	15	5.7
5	5.4	12.1	0.30	2.8	1.9	9.5	1.7	0.92	18	5.5
6	5.2	12.7	0.31	2.7	2.0	8.9	2.1	0.71	21	6.1
7	6.4	11.3	0.31	3.4	0.72	8.4	1.8	0.88	18	5.3

4. Conclusion

We have shown in the example of the 27th harmonic of HHG radiation (~42 eV) that a Fourier transform spectrometer for the XUV can be used to directly gain knowledge about the temporal coherence of HHG pulses. In the future, one could ensure that the whole XUV beam is imaged onto the middle of the detector of the Fourier transform spectrometer by developing a new, more rigorous alignment process. This should allow distinguishing radiation with different coherence times at different parts in the XUV beam which was not possible with the current beam alignment. Nevertheless, we have noticed a spatial variation of the temporal coherence within the beam but on smaller scales than expected.

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