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Direct measurement of the pulse duration and frequency chirp of seeded XUV free electron laser pulses

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Abstract
We report on a direct time-domain measurement of the temporal properties of a seeded free-electron laser pulse in the extreme ultraviolet spectral range. Utilizing the oscillating electromagnetic field of terahertz radiation, a single-shot THz streak-camera was applied for measuring the duration as well as spectral phase of the generated intense XUV pulses. The experiment was conducted at FLASH, the free electron laser user facility at DESY in Hamburg, Germany. In contrast to indirect methods, this approach directly resolves and visualizes the frequency chirp of a seeded free-electron laser (FEL) pulse. The reported diagnostic capability is a prerequisite to tailor amplitude, phase and frequency distributions of FEL beams on demand. In particular, it opens up a new window of opportunities for advanced coherent spectroscopic studies making use of the high degree of temporal coherence expected from a seeded FEL pulse.

1. Introduction

Free-electron lasers (FELs) are unequalled in providing tuneable, intense, ultrashort light pulses in the soft- and hard-x-ray regime. The self-amplified spontaneous emission (SASE) principle, underlying most FEL implementations, guarantees a high spatial coherence of the beam. Its longitudinal coherence, however, is known to be rather limited [1, 2]. This important property is now being improved using FEL seeding techniques, where a seeding wave at the target wavelength [3] or an integer fraction of it [4, 5] imprints a well-behaved and reproducible time evolution on the phase of the amplified electro-magnetic field. This opens the door for a novel class of experiments investigating the response of matter on the mutual action of synchronized light fields in a previously unexplored spectral regime. A first such coherent-control type experiment [6] has demonstrated to steer photoelectrons formed in a nonlinear ionization event by adjusting the phase between the EUV pulse and its simultaneously generated 2nd harmonic. The full exploitation of these opportunities calls for a precise measurement of the temporal pulse properties, including it is temporal phase evolution. Existing temporal pulse property methods for FEL pulses in the soft and hard x-ray regime adapting measurement principles developed for optical fs-laser pulses, like the autocorrelation [7] or SPIDER [8] technique. Recently, the duration of pulses from the seeded FEL FERMI has been determined using nonlinear cross-correlation with an infrared laser pulse [9] and a chirp has been indirectly inferred by successively following the change in the FEL pulse duration, when modifying the chirp of the seed beam [10]. In the work presented here, we aimed at a direct measurement of the temporal properties of a seeded FEL pulse by employing the THz streak camera principle [11], which provides information on both pulse duration as well as spectral chirp [12]. Importantly, the single-shot capability of THz-
streaking allows to assess the reproducibility of these pulse parameters. While this method is not able to achieve a complete single-shot field reconstruction (as potentially being provided by the SPIDER method), it can deliver the pulse’s duration and linear chirp, if more than only one time-of-flight spectrometer is used \[13\]. Furthermore, as the XUV pulse structure is not affected by the gas target or streaking electrical THz field, this method provides a non-invasive and fully parasitic temporal diagnostic allowing for a re-use of the undisturbed and characterized FEL pulse for downstream experiments.

In the last section we will explicitly compare the seeded FEL pulse measurement results obtained from the THz streaking method with an electron bunch related measurement method, which uses a transverse deflecting rf-structure (temporal deflecting structure, TDS, \[14\]), to observe the energy modulation imprint of the seeding process in the electron bunch directly.

### 2. Transient THz light field streaking of photo electrons

To directly measure both, pulse duration and chirp of individual XUV pulses, this diagnostic utilizes the energetic streaking of photo electrons in a strong electro-magnetic field at zero-phase. This technique has been developed in the field of attosecond science \[15\] and was adapted to be used on ionizing light sources with pulses in the femtosecond to picosecond regime \[11\]. The underlying principle is to map the temporal intensity and phase profile of the XUV pulse onto an electron yield distribution over kinetic energies. It also has conceptual similarity to other ‘zero-phase’ techniques for analyzing the structure of an electron bunch of a linear electron accelerator as discussed in \[16\] or \[17\]. In order to comply with the expected pulse duration of the seeded FEL in the range of several tens to hundreds of femtoseconds, the frequency of the adjacent field is chosen to be in the Terahertz regime. The THz streaking diagnostic has been explained in detail elsewhere \[12\], so here we focus on the salient points for its application at a seeded FEL. Figure 1 depicts the experimental setup. The THz pulse is transported via metallic mirrors and is focused with a plano-konvex lens (Zeonex polymer 440r, Viaoptics) of 75 mm focal length to a spot size of 1.2 mm FWHM width and 1.9 mm FWHM height. The XUV pulse is collinearly guided through 2 mm apertures in the THz lens and the metallic mirror (see figure 1). The XUV pulse ionizes an argon gas target, which is located 5 mm in front of the entrance of a time-of-flight spectrometer. The beam diameter of the XUV pulse is reduced to 0.5 mm diameter with a pinhole aperture. Using a temporal delay stage and a set of temporal and spatial diagnostics like an electro-optical crystal, a fluorescent screen, a pyro detector and a fast XUV diode, the spatio-temporal overlap between the THz pulse and the XUV pulse is established.
3. Discussion of the involved light pulses

The experiment makes use of a variety of different light pulses, which all originate from the same optical femtosecond laser pulse. It is created in a 10 Hz flash-lamp pumped, chirped-pulse-amplification titanium-sapphire laser system (HIDRA, Coherent) with 30 mJ pulse energy at 800 nm central wavelength and a Fourier limited pulse of 35 fs FWHM duration. The Titanium sapphire laser is optically synchronized to the master oscillator of the FEL with a temporal precision of 13 fs rms [18]. The amplified output pulses are split into two parts, one is used for the seeding of the FEL process and the second for the THz streak camera. The output pulse of the laser system is temporally uncompressed. To reduce unwanted nonlinear effects during the pulse transport the compression of each of the two parts is performed in front and close to any nonlinear frequency conversion component. For seeding a pulse with 5 mJ pulse energy is frequency tripled to 266 nm wavelength and a pulse energy of 500 µJ. The pulse duration of the optical seed pulse has been measured using an optical single-shot line cross-correlator between the fundamental and the frequency-tripled pulse to \( t_{\text{laser}} = (140 \pm 10) \) fs rms. A spectral measurement of the seed pulse provides a spectral width of 1.35 nm, which corresponds with a Fourier limited pulse duration of 34 fs rms. Thus, the seed pulse was affected by an initial chirp as a result of the tripling process. The nature of this chirp is currently unknown and will be subject to further measurements. The frequency tripled pulse is focused into a dedicated section of the linear accelerator of the FEL, where the amplification process is initiated by means of external seeding [19]. In the so-called high-gain harmonic generation mode of operation, a sinusoidal modulation of the energy of an ultra-relativistic electron beam is introduced by overlapping it with a GW-level optical seed laser pulse with 266 nm wavelength inside an undulator magnet. Arrangements of four dipole magnets generate longitudinal dispersion and convert the energy modulation into a current density modulation. The harmonic content of the current spikes allow the coherent emission of radiation at an integer fraction of the initial seed wavelength in a subsequent undulator. In contrast to FEL radiation pulses which are initiated by the spontaneous synchrotron radiation in the so-called SASE mode, the seeded FEL pulses exhibit a high degree of temporal coherence [19]. In this experiment, the 8th harmonic of the 266 nm seed laser radiation was generated.

The THz pulse with a central frequency of \((0.5 \pm 0.1)\) THz and a maximum field strength of \((80 \pm 10)\) kV cm\(^{-1}\) in the focus was generated via optical rectification [20] using a nonlinear crystal (LiNbO\(_3\)). The field strength information about the streaking THz field is extracted from the result of a THz streaking temporal delay scan as will be discussed in the next section. The optical driver pulse energy in front of the LiNbO\(_3\)-crystal was 1.5 mJ with a pulse duration chirped to 800 fs FWHM. The beam profile of the optical pulse is cylindrically shaped and reduced in size by using a spherical and a cylindrical Galilei telescope (1:4 and 1:2) before passing the grating, which induces the pulse front tilt. Behind the LiNbO\(_3\)-crystal a gold coated parabolic mirror \((f = 150 \text{ mm})\) collimates the THz radiation. The THz transport is performed with gold and silver coated mirrors and the path length in air is limited to 0.6 m to minimize absorption. A \( f = 75 \text{ mm plastic lens (Zeonex 480R) with a 2 mm diameter hole}\) focused the THz radiation into the interaction area (figure 1) with a spot size of 1.5 and 2.2 mm FWHM. With a pulse duration of 1200 fs FWHM a THz pulse energy of 0.42 µJ is obtained. Including energy losses due to Fresnel reflection, absorption of water in air and clipping in the vacuum entrance window, the conversion efficiency is estimated to be 0.05%. The reason for this relatively low conversion efficiency (see [21] for example) is attributed to a too low available energy of the optical driver laser during that experimental campaign, which prevented saturation of the THz conversion process.

4. THz streaking scan

In figure 2 the temporal delay scan between the XUV and the THz pulse is presented. For each delay time, kinetic electron energy spectra are recorded and plotted vertically in false color encoding the electron signal amplitude normalized to the overall maximum. The scan was recorded in time steps of \( \Delta t = 50 \) fs with \( n = 10 \) spectra measured at each step. In the figure, all single-shot spectra are plotted such that for each spectrum of delay time \( t_i \) a new interpolated delay time \( t_j \) according \( t_j = t_i + n \cdot \Delta t \) is assigned, with \( j = 1 \ldots n \). This plotting technique preserves the single shot characteristics such as shot-to-shot fluctuations of the pulse amplitudes and the relative pulse arrival time. The red trend line connects the centers of mass of each spectrum. It represents the time-dependent energy gain \( W(t) \) and is further used to derive the streaking THz field \( E_{\text{THz}}(t) \). The scan was limited to the central part of the THz vector potential since we extract the relevant information to calculate the XUV pulse information only from spectra taken at the streaking points \( O_n \) and from unstreaked spectra of the region \( R_n \) as will be further explained. According to [12], the energy modulation \( \Delta W(t) \) is connected with the streaking THz field by the relation \( \Delta W(t) = \frac{\hbar}{m_e} A_{\text{THz}}(t) \), with \( W_0 \) as the kinetic energy of the unstreaked electrons, \( m_e \) the electron mass and \( A_{\text{THz}}(t) = \int_r E_{\text{THz}}(t) dt \) the vector potential of the streaking THz field. It should be
emphasized that the quality of a THz streaking scan strongly depends on the technical abilities to keep any source of spatio-temporal jitter of the THz and XUV pulse as well as fluctuations of the energy and of the spectral center-wavelength of the XUV pulse as small as possible. Since the THz scan is measured using a seeded FEL pulse, the relative pulse arrival time jitter of THz and XUV pulse is expected to be small as will be discussed later [22]. \( R_0 \), \( R_+ \) and \( R_- \) denote regions of similar kinematics: in the region of \( R_0 \) the temporal overlap is not yet established and spectra are not streaked, thus, representing the unperturbed spectrum of the ionized 3p-argon valence electrons. In this area, one can study the shot-to-shot variation of the pure FEL pulse, which is mandatory to further evaluate the average XUV pulse properties from the streaked spectra. Region \( R_+ \) reaches from 0.3 to 0.6 ps. In this delay time interval the slope of the energy—gain curve is positive and almost constant as is the case at \( R_- \). ranging from 0.9 to 1.2 ps, where the slope is negative. In the two inflexion points of the energy—gain curve the slope has an extremum providing highest streaking strength. Those two positions are further denoted as positive and negative operating points \( O_+ \). A linear fit of the streaking trace at these two points reveal streaking speeds \( s_+ \) and \( s_- \) with \(+16.8 \text{ meV fs}^{-1}\) and \(-23.0 \text{ meV fs}^{-1}\), respectively.

5. Determination of XUV pulse duration and chirp

Under the assumption that the pulse may by subject to a frequency chirp of no more than linear order, it is possible to extract pulse duration and chirp from the spectral broadenings \( \sigma_\chi \) at both streaking points together with the original width \( \sigma_0 \) in region \( R_0 \) [12]. Assuming a Gaussian temporal shape, the electric field of a XUV pulse with chirp parameter \( c \) and rms pulse duration \( \tau_{\text{XUV}} \) is given by

\[
E_{\text{XUV}}(t) = E_{\text{XUV}}^0 \, e^{-\left((t-t_\text{f})^2 + c(t-t_\text{f})^3\right)}.
\]

From a quantum-mechanical calculation [12, 23] one finds that the streaked spectrum preserves the Gaussian shape and has an rms width of \( \sigma_{\text{XUV}} = \sqrt{\sigma_0^2 + \tau_{\text{XUV}}^2 (s_+^2 + 4cs_-)} \), with equal streaking speeds \( s_+ = s_- = s \). The sign in the index corresponds to the previously introduced regions in figure 2. In this work we introduce one more degree of freedom to account for for different streaking speeds \( s_+ \neq s_- \) in the two operating points, thus obtaining \( \sigma_{\text{XUV}} = \sqrt{\sigma_0^2 + \tau_{\text{XUV}}^2 (s_+^2 + 4cs_-)} \). To extract pulse duration and chirp of each individual XUV pulse requires measuring \( \sigma_0 \) simultaneously with the streaked widths \( \sigma_{\text{XUV}} \) and solve the two equations for pulse duration and chirp. One obtains

\[
\tau_{\text{XUV}} = \sqrt{\frac{(\sigma_{\text{XUV}}^2 - \sigma_0^2) s_+ + (\sigma_{\text{XUV}}^2 - \sigma_0^2) s_-}{(s_+ + s_-) s_+ s_-}} \tag{1}
\]

\[
c = \frac{\left(\sigma_{\text{XUV}}^2 - \sigma_0^2\right) s_+^2 - \left(\sigma_{\text{XUV}}^2 - \sigma_0^2\right) s_-^2}{4s_+ s_- (s_+ + s_-) \tau_{\text{XUV}}} \tag{2}
\]

with the definition \( \sigma_{\text{XUV}}^2 = \sqrt{\sigma_0^2 - \sigma_{\text{XUV}}^2} \) for the widths of the deconvoluted streaked spectra. Because in this THz streaking experiment only a single time-of-flight detector was used, the average values \( \sigma_{\text{XUV}}, \sigma_0 \), and thus the averages of \( \sigma_{\text{XUV}}, c \) and \( \tau_{\text{XUV}} \) were acquired successively. The error of the average values for pulse duration and chirp is thus dominated by the shot-to-shot fluctuations of the FEL pulse. However, as this experiment was performed on a seeded FEL, these fluctuations are expected to be smaller than in the case of an unseeded SASE FEL. This expectation is underlined by a closer inspection of streaking data: in figure 3 (left column) a series of single pulse spectra acquired at the two streaking points and in region \( R_0 \) are presented. The center column displays individual spectra of arbitrarily chosen XUV pulses as waterfall plots. All amplitudes are normalized to
the maximum amplitude found in the series of the non-streaked spectra. The right column shows the histogram of the spectral widths of a subset of spectra, which are used to calculate the average temporal pulse properties.

Due to shot-to-shot fluctuations of the temporal and spatial overlap of the seeding pulse and the electron bunch in the undulator magnet, the seeding process performed with fluctuating efficiency leading to fluctuations of the XUV pulse energy and thus of the amplitude of the kinetic photo-electron spectra. By selecting only spectra with amplitude higher than 10% of the highest amplitude of a measurement series, only spectra of good signal-to-noise ratio were used for the evaluation. The total number of selected spectra used in one series varied between 100 and 300 individuals. The temporal resolution for the provided pulse duration measurement is limited by the smaller streaking speed, hence $s_{\text{res}} = 0 \pm 16.6$ fs. A detailed analysis of the influence of THz-wave front deformations in the focus shows that the spectra are also subject to broadening due to the Guoy-phase shift, which has been discussed in the literature\[12\]. With a gas jet of 1.2 mm diameter FWHM and a THz Rayleigh-range of 4.7 mm one calculates a broadening of 25 fs rms. To correct this effect, one has to deconvolute the spectral widths $\sigma_i$ according to $\sigma_i^{\text{real}} = \sqrt{\sigma_i^{2,\text{meas}} - s_{\text{Guoy}}^2}$. The corresponding Guoy-phase shift corrected average values for the streaked spectra widths and standard deviations as well as the derived temporal pulse properties are presented in table 1. The negative sign of the chirp factor means in this context an anomalous chirp in the sense that higher frequencies come first. The standard deviations of the derived values $\tau_{\text{XUV,rms}}$ and chirp $c$ are calculated from the Gaussian law of error propagation.

From the center of mass variation of the $\sigma_i$ single-pulse spectra, we retrieve a pulse-to-pulse arrival-time jitter between THz and FEL pulse of 17 fs rms. The main source are length fluctuations along the two about 60 m long optical beam paths of the seed pulse and the NIR pulse generating the THz radiation.

<table>
<thead>
<tr>
<th>( \sigma_0 )</th>
<th>( \sigma_+ )</th>
<th>( \sigma_- )</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.28</td>
<td>0.96</td>
<td>1.61</td>
</tr>
<tr>
<td>0.06</td>
<td>0.13</td>
<td>0.25</td>
</tr>
<tr>
<td>21</td>
<td>13.5</td>
<td>15.5</td>
</tr>
</tbody>
</table>

**Table 1.** Measured rms widths $\sigma_0$, $\sigma_+$, $\sigma_-$ and retrieved pulse properties $\tau_{\text{XUV,rms}}$ and chirp $c$.

<table>
<thead>
<tr>
<th>( \tau_{\text{XUV,rms}} ) (fs)</th>
<th>$c$ (THz ps$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>58</td>
<td>$-1940$</td>
</tr>
<tr>
<td>7.5</td>
<td>$800$</td>
</tr>
<tr>
<td>13</td>
<td>41</td>
</tr>
</tbody>
</table>

**Figure 3.** Single-shot measurement of THz streaked seeded XUV FEL pulses.
6. Comparison with TDS measurements

The FLASH setup allows to temporally resolve the energy spectrum of the electron beam after it has generated the FEL radiation \[17\]. In a TDS, an \(x-z\)-correlation is introduced to the electron beam that allows mapping the longitudinal phase-space into the \(x-y\) coordinate system. The increase of the local energy spread for the seeded region inside the electron pulse compared to the unseeded electron pulse was used to determine the FEL pulse duration. A direct comparison of the FEL pulse duration measured by TDS and THz streaking is shown in figure 4. With only one time-of-flight detector, it is not possible to measure the widths at the positive and negative streaking slopes simultaneously. Thus, for single-shot comparison with TDS any chirp effect on the streaked widths was ignored and only by using the single shot widths from the positive streaking point \(O_+\) we calculated the XUV rms pulse duration by

\[
\tau_{\text{XUV}} = \frac{\sigma_{\text{decon,Gouy}}}{s_+} = \frac{\sqrt{\sigma_{\text{decon,meas}}^2 - \sigma_0^2}}{s_+} = (s_+ \tau_{\text{Gouy}})^2
\]

with \(\sigma_0\) the average width of the unstreaked spectra. While the statistical fluctuations obscured significant correlations on the single-shot level, the average pulse durations of 54 and 57 fs rms of the two methods match within the standard deviation.

It is of high interest, how the pulse duration of the optical seed pulse and the final seeded FEL pulse depend on each other. In \[24\], a relation of \(\tau_{\text{XUV}} = 0.94 \times H^{1/2} \times \tau_{\text{laser}}\) with the harmonic order \(H\) has been proposed, resulting in \(\tau_{\text{XUV}} = 0.47 \times \tau_{\text{laser}} = (66 \pm 5)\) fs for the 8th harmonic, which agrees with our measured pulse durations within the error bars.

7. Summary and conclusion

We have measured the pulse duration as well as the frequency chirp of a seeded XUV free-electron-laser using the THz streaking method. The obtained pulse duration matches well with an independent TDS based measurement of the duration of the energetic modulation of the electron bunch. The spectral components of the FEL XUV pulse were found to have a temporal chirp. In \[24\] it is predicted that, due to the high degree of involved nonlinearity, the HGHG seeding process is very sensitive to temporal phase variations of the optical seed pulse. Such transfer of temporal phases from the seed to the amplified FEL beam has also been experimentally confirmed at FERMI \[10\]. A detailed quantitative investigation of this transfer mechanism, however, is beyond the scope of this work and will be the subject of a further study. Our simulations of the frequency-tripling process generating the seed beam are compatible with a chirp of positive or negative sign, depending on the exact settings of the laser compressor, the phase-matching conditions in the tripling crystals, and the residual dispersion introduced by the optical materials in the beam path. In the present experiment, the resulting XUV-chirp of the seeded FEL pulse was found to exhibit a negative sign. Yet, with currently built phase diagnostics for the seed beam being implemented, we expect to be able to realize and measure chirps of the seed pulse of both signs and variable magnitude. With a powerful metrology for the spectro-temporal properties of individual FEL pulses now at hand, we will aim at a full control over the pulse shape and longitudinal coherence properties of seeded FEL pulses.

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Figure 4. Pulse durations from TDS and THz streaking evaluated at positive streaking point \(O_+\).
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