Single-shot timing measurement of extreme-ultraviolet free-electron laser pulses

Theophilos Maltezopoulos\textsuperscript{1,4}, Stefan Cunovic\textsuperscript{2}, Marek Wieland\textsuperscript{3}, Martin Beye\textsuperscript{3}, Armin Azima\textsuperscript{1}, Harald Redlin\textsuperscript{1}, Maria Krikunova\textsuperscript{3}, Roland Kalms\textsuperscript{3}, Ulrike Frühling\textsuperscript{1}, Filip Budzyn\textsuperscript{3}, Wilfried Wurth\textsuperscript{3}, Alexander Föhlisch\textsuperscript{3} and Markus Drescher\textsuperscript{3}

\textsuperscript{1} HASYLAB at DESY, Notkestrasse 85, 22607 Hamburg, Germany
\textsuperscript{2} Department of Physics, Bielefeld University, Universitätsstrasse 25, 33615 Bielefeld, Germany
\textsuperscript{3} Institut für Experimentalphysik, University of Hamburg, Luruper Chaussee 149, 22761 Hamburg, Germany

E-mail: theophilos.maltezopoulos@desy.de

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Abstract. Arrival time fluctuations of extreme-ultraviolet (EUV) pulses from the free-electron laser in Hamburg (FLASH) are measured single-pulse resolved at the experimental end-station. To this end, they are non-collinearly superimposed in space and time with visible femtosecond laser pulses on a GaAs substrate. The EUV irradiation induces changes of the reflectivity for the visible pulse. The temporal delay between the two light pulses is directly encoded in the spatial position of the reflectivity change which is captured with a CCD camera. For each single shot, the relative EUV/visible arrival-time can be measured with about 40 fs rms accuracy. The method constitutes a novel route for an improvement of future pump–probe experiments at short-wavelength free-electron lasers (FELs) by a pulse-wise correction with simultaneously measured arrival times of individual EUV pulses.
1. Introduction

Free-electron lasers (FELs) are exceptionally powerful pulsed light sources at extreme wavelengths. The free-electron laser in Hamburg (FLASH) [1] at the Deutsches Elektronen Synchrotron (DESY), is the first of its kind operating in the extreme-ultraviolet (EUV) range, delivering femtosecond (fs) pulses at photon energies of 90 eV [2] and higher. Among other exciting prospects, FELs facilitate a novel class of pump–probe experiments at short wavelengths with pulse intensities orders of magnitude higher than other sources can provide [3]. While the EUV pulse duration is in the range of 20–30 fs [1], the timing with respect to external light pulses deserves further attention to fully exploit the potential for dynamics studies. Short-wavelength FELs are currently operated in the so-called self-amplified spontaneous emission (SASE) mode [4], where a relativistic electron bunch passes through a long undulator magnet structure. At FLASH, the length of the leading spike inside the electron bunch is typically 120 fs full width at half maximum (FWHM) [1]. The electron acceleration process introduces a pulse-to-pulse variation of the arrival time of the electron beam at the undulator entrance 5. Together with the statistical nature of SASE and the several tens of metres long beamlines, this leads to an unpredictable temporal jitter of the FEL pulses at the experimental end-stations. This inevitable jitter currently puts constraints on the temporal resolution of pump–probe experiments that correlate EUV and external laser pulses.

The temporal resolution will be significantly improved, if the random delay fluctuations are measured for individual pulses and the data from a simultaneously operated pump–probe experiment are sorted accordingly. We note that the arrival time of the electron bunch before its injection into the undulator has already been measured using an electro-optical sampling (EOS) technique [5, 6]. This way, x-ray diffraction data have been successfully arranged by using the EOS data in order to study structural dynamics in bismuth [7]. A photon-based measurement of the FELs delay fluctuations at the experimental end-station will include, additionally, the statistical nature of the SASE process together with any jitter sources connected with the long EUV beam transport behind the undulator. One approach for the temporal characterization of EUV pulses at a FLASH end-station is based on a EUV/visible cross-correlation in the gas phase, where the kinetic energy of FEL generated photo-electrons is shifted to spectral sidebands by a synchronized near-infrared laser field [8, 9]. Single pulse delay measurements, however, have so far been prevented by space-charge induced perturbations occurring at high FEL intensities.

5 The Technical Design Report of the European XFEL, http://xfel.desy.de/tdr.
Space charge effects are completely avoided in a photon-in/photon-out cross-correlation set-up: two laser pulses are temporally and spatially superimposed on a surface where the reflectivity for the probe pulse is changed by the pump pulse. Using this technique, femtosecond-resolving pump–probe measurements on surfaces have been demonstrated [10]. Picosecond pulse duration has been measured in [11]. The temporal shape of a mid-infrared FEL pulse was sampled in [12], and atomic displacements of an optically pumped InSb sample were probed with x-ray pulses in [13]. With the high intensities delivered by FEL sources, short-wavelength pulses can now take over the role of efficiently pumping a surface: an EUV-induced reflectivity change for visible light was recently observed on a GaAs sample [14]. The EUV pulse is mainly absorbed by atomic photo-ionization of inner-shell electrons [14]. Auger decay and auto-ionization convert the initial inner shell excitation into valence excitations within a few femtoseconds [14]. These valence excitations are responsible for the change in optical reflectivity [14]. From experiments with excitations in the visible it is known that the Drude model alone is insufficient to describe optical excitations in GaAs below the damage threshold, but models including screening of ionic potentials, many body effects and modification of the band-structure must be considered [15, 16].

We utilized this reflectivity change in a specific geometry that facilitates mapping of EUV/visible delay fluctuations at FLASH on a single-shot basis. EUV pulses (wavelength $\lambda = 28$ nm and pulse duration $\tau = 20–30$ fs) are orthogonally crossed with visible pulses ($\lambda = 400$ nm and $\tau \approx 130$ fs) on a GaAs sample in an ultra-high vacuum environment ($p \approx 10^{-7}$ mbar). The visible light reflected from the semiconductor surface is imaged onto a two-dimensional (2D) detector. The EUV pulse reduces the reflectivity of GaAs for this wavelength with the onset of the reflectivity change directly mapping the delay between the EUV and the visible pulse.

2. Experimental set-up

At the FLASH facility, a near-infrared laser system ($\lambda = 800$ nm, $\tau \approx 130$ fs and pulse energy $E \approx 15 \mu$J) is electronically synchronized to the EUV beam based on the 1.3 GHz master clock of the facility. EUV and near-infrared pulses therefore share the same temporal pulse pattern, which consists of $n_{\text{micro}} = 1–30$ micro-pulses separated by 1–10 $\mu$s from each other forming a macro-pulse at a repetition rate of 5 Hz. In the present work, $n_{\text{micro}} = 1$ was used in order to comply with the limited acquisition rate of the CCD camera used for imaging. The 800 nm radiation pulses were frequency-doubled to 400 nm using a BBO crystal. A simulation with SLNO\(^6\) did not reveal any significant pulse lengthening upon this frequency conversion. In order to avoid saturation and damage of the imaging CCD, we attenuated the visible beam by approximately three orders of magnitude, to a pulse energy of $\approx 10$ nJ on the substrate surface. The experiment was mounted at the monochromator beamline PG2 of FLASH [17, 18]. Using the 200 lines mm\(^{-1}\) grating in zeroth-order, the beamline has a calculated transmission of about 47% for 28 nm FEL radiation.

A scheme of the experimental geometry is shown in figure 1. The unfocused visible laser pulse illuminates the entire area of the polished GaAs sample ($3 \times 3$ mm\(^2\)) from the bottom. The surface normal is oriented at an angle of 22.5° with respect to the vertical axis, such that

\(^6\) SNLO nonlinear optics code available from Smith A V, Sandia National Laboratories, Albuquerque, NM 87185-1423.
the reflected light is imaged with an f/4.5 lens system onto a CCD camera under a 45° angle. The GaAs sample is mounted on a manipulator, which allows for adjusting the reflected beam. In order to prevent un-reproducible modifications of the surface properties like, for example, engineered thick oxides by the wafer manufacturer, or other contaminations, 800 nm of GaAs was grown on an undoped GaAs(001) wafer by molecular beam epitaxy. The native oxide thickness of GaAs is only of the order of a few nanometres [19]. The horizontally propagating focused FEL pulse is almost completely absorbed in the sample. For the data presented in this paper, the typical EUV pulse energy was 60 ± 20 µJ rms, which was attenuated to 10 ± 4 µJ rms with a gas absorber behind the undulator. Together with the beamline transmission of about 47% this leads to a pulse energy of 4.7 ± 1.9 µJ rms at the experiment. The FEL spot size in the interaction region was about $70 \times 200 \, \mu m^2$ FWHM, which corresponds to a footprint of $70 \times 520 \, \mu m^2$ in the projection on the GaAs substrate. Thus, the resultant fluence was about $13 \pm 5 \, mJ \, cm^{-2}$ rms. At this fluence, measurements over several hours could be performed at the same location on the GaAs sample. We found, however, that higher average FEL fluence inhibits long-term measurements and leaves permanent damage on the GaAs surface after a few minutes, requiring the selection of a new location on the sample. On the other hand, a lower average fluence leads to very weak contrast in the images, the above range of the EUV fluence must be selected for the method to work properly.

Figure 1 depicts the region of the EUV-induced reflectivity change for the visible pulse on the GaAs sample and how it is imaged onto the CCD. This geometrical assembly directly maps time- into space-information. The correlation between the space coordinate $s$ (see figure 1) and
time coordinate \( t \) can be deduced from geometrical considerations to:

\[
s = \frac{ct}{1 + \tan \varphi},
\]

where \( c \) is the vacuum speed of light, \( \varphi \) the angle of the GaAs sample, and \( t \) a relative time delay between the visible and FEL laser pulse. In the present case of \( \varphi = 22.5^\circ \), one obtains \( \Delta t/\Delta s = 100 \text{ fs}/21.2 \text{ \( \mu \)m}. \)

Spatial overlap between the two pulses is easily adjusted, because the EUV radiation induces a visible fluorescence on the GaAs substrate which can be monitored with the imaging system. Temporal pre-alignment is accomplished with \( \pm 10 \text{ ps} \) accuracy using a high-bandwidth copper photocathode. It can be placed into the interaction region with a manipulator and it is read out by a 6 GHz amplifier and oscilloscope.

### 3. Experimental results

A single-shot image acquired at temporal and spatial overlap is presented in figure 2(a), where the space-coordinate \( s \) is already converted into a time coordinate \( t \). The area where the FEL pulse has excited the GaAs substrate prior to the arrival of the visible pulse wavefront appears darker, with the boundary of reflectivity change being clearly discernible. To obtain such an image from the raw CCD data, image processing was carried out. It included a background subtraction in order to minimize interference fringes from scattering objects in the visible beam path and a filtering in order to reduce inhomogeneous illumination of the field of observation. An average of the signal of pixels corresponding to the same time bin within the region-of-interest (ROI) marked in figure 2(a) is shown in (b). The slope width of the rising edge of about
130 fs is in good accordance with the visible pulse duration. The inflection point was determined with a Gaussian fitting applied to the binned-data in figure 2(b). In order to obtain a measure of the fit quality, we performed fits of the same image with different starting parameters. With this procedure applied to many images, we obtained an error for the determination of the inflection point of about 40 fs rms. This inflection point will further be used as the time-marker for the EUV/visible delay.

The visible laser pulses can be temporally shifted relative to the FEL pulses with an optical delay line. A selection of 17 single-shot images from a delay scan over 2.3 ps is assembled in figure 3. The nominal delay stage setting is marked at the bottom of the images and the time axis \( t \) is shown on the left. Upon progression of the delay line to larger times, the FEL pulse arrives earlier. Thus, the boundary of reflectivity change moves downwards (larger \( t \)). We delayed the visible laser over more than 10 ps, but only within this 2.3 ps window could the region of reflectivity change be observed. This is in excellent agreement with the expected time window \( t_w \) of our set-up, which can be calculated from the FEL beam height \( h_{\text{FEL}} = 200 \mu \text{m} \) to:

\[
t_w = \frac{h_{\text{FEL}}(1 + \tan\varphi)}{c \tan\varphi} = 2.3 \text{ ps}.
\]

Note that these are single-shot measurements, where an arbitrary delay due to the FEL jitter is superimposed to the delay scan, which is clearly visible in the pictures. The appearance of the dark pulse tails differs from shot to shot, reflecting fluctuations of the FELs spatial profile.

At a constant delay setting, the erratic EUV/visible delay fluctuations were monitored. Such a measurement over a period of 5 min is presented in figure 4(a). Each data point corresponds to one single-shot image, like figure 2(a), with binning and fitting procedures applied in order to determine the inflection point as a time-marker. During this 5 min observation, the extreme EUV/visible delay values are 1.5 ps apart while the rms jitter

\[
\pm \sqrt{\frac{1}{N} \sum_{i=1}^{N} (\bar{x} - x_i)^2 / N}
\]
Figure 4. (a) Single-shot delay fluctuations of FLASH over 5 min. The time origin is set arbitrarily. The rms jitter is ±250 fs. (b) Histogram of (a).

amounts to ±250 fs. Similar values have been found with EOS measurements at FLASH [6]. Noticeably, EUV/visible delay values around 0 and 300 fs appear more often as is also revealed in the histogram of figure 4(a), shown in (b). Similar arrival time jumps have also been observed in previous EOS data.

Under the present conditions, about 30% of the single-shot data can be evaluated. For figure 4(a), for example, only 457 out of 1500 captured images have shown sufficient contrast and signal-to-noise ratio to be further processed. This is partly attributed to EUV pulse intensities falling below the acceptable limit, but is mainly ascribed to incomplete correction of interference patterns from scattering objects in the beam path. With an improved optical layout the latter perturbation will be significantly reduced.

With the EOS set-up available at FLASH [6], simultaneous arrival-time measurements using both techniques were possible. We found a correlation between the EUV/vis and the electron-bunch/vis arrival-time, where the latter is measured with the EOS technique. This result is shown in figure 5. The time-origins of both timing tools are set deliberately so as to eliminate any offset in the straight-line fit. The straight-line slope is 1.003 ± 0.024, thus, the calibration from geometrical considerations mentioned above is confirmed with EOS. Note that we deliberately enhanced the FEL delay variations by changing the phase of FLASH accelerator ACC1. Thus, we could test the correlation to EOS within the full temporal window of our experiment. The rms deviation between both measurements is 160 fs within 4 min of data acquisition. This deviation is an upper limit for the additional jitter from the beamline and SASE process, because it still includes the intrinsic errors of both techniques. In future pump–probe experiments, both techniques will be used for sorting the spectra. Application of this concept to an ultra-fast process-like side-band generation in a gas medium, which is known to provide sub-fs response time [20], will finally aid in identifying and quantifying jitter sources that are connected with the SASE process and the subsequent radiation transport.
4. Summary and outlook

In summary, we presented single-shot measurements of arrival time fluctuations of EUV pulses from the free-electron-laser FLASH using a purely photon-based method which encodes temporal information in a spatial coordinate. In contrast to measurements of the electron bunch timing, the technique introduced here analyzes the delay directly at the experimental end-station and takes all possible jitter sources into account. The current accuracy is estimated to be 40 fs rms. Shorter laser pulses together with a noise reduced optical set-up provide room for a further improvement of the temporal resolution towards the EUV pulse duration. The developed tool can be operated with a fraction of the fluence delivered by the FEL and needs very little energy from the visible pulse, thus not requiring sample scanning. At present, the experimental set-up is limited to a single micro-pulse per macro-pulse due to the low capture rate of the CCD used; advanced 1D line detectors should enhance the burst acquisition rates to 100 kHz and more. It is shown in [14] that the sample recovers its equilibrium state between sub-bunches at 500 kHz repetition rate. While being demonstrated here in the EUV range, we expect the technique to remain applicable for harder radiation as well. For photon energies extending into the kilo-electron-volt range, the decreasing photo absorption could be compensated by an increasing efficiency of the charge creation in the relevant topmost surface layer [14]. Therefore we expect that this timing tool can be used also in future x-ray FELs like XFEL at DESY\textsuperscript{5} or LCLS at SLAC\textsuperscript{7}. The single-shot delay measurements demonstrated here represent a way to improve the temporal resolution of pump–probe experiments with FEL and synchronized optical laser pulses. This will be achieved by sorting the pump–probe spectra pulse-by-pulse according to the measured delay.

\textsuperscript{7} The Linac Coherent Light Source (LCLS) at the Stanford Linear Accelerator Center (SLAC), \url{http://www-ssrl.slac.stanford.edu/lcls/}.
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