JITTER REDUCED PUMP-PROBE EXPERIMENTS

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Abstract

For two-colour pump-probe experiments carried out at the free electron laser FLASH at DESY, the XUV FEL laser pulses must be synchronized with femtosecond precision to optical laser pulses (120 fs, 20 μJ Ti:Sapphire). An electro-optical (EO) sampling diagnostic measures the arrival time jitter of the infrared pump-probe laser pulse with respect to the electron bunch of the FEL. Here, the electron arrival time is spatially encoded into the laser pulse profile and read out by an intensified camera. In this paper we report about the improvement of the temporal resolution of pump-probe experiments on gaseous targets using the arrival time data acquired by the EO-diagnostic.

INTRODUCTION

Fast physical processes in the fs time domain can be observed using pump-probe experiments. Two ultrashort pulses are temporally and spatially overlapped in some target. The first pulse induces a reaction and the second pulse probes the changes of the target material induced by the first pulse. To scan the time evolution of the reaction, one introduces a variable time delay \( \Delta T_{PP} \) between both pulses. The infrastructure to perform two-colour pump-probe experiments was built up at the FLASH facility. One pulse in the near infrared range is generated by an amplified Ti:Sa oscillator at a wavelength of 800 nm. The Ti:Sa pulse is amplified up to 20 μJ by an optical parametric amplifier. Its pulse duration at the experimental site is 120 fs FWHM. The focal intensity is in the range of a few TW cm\(^{-2}\). FLASH generates the second ultra-short pulse of high intense (≈ 10\(^9\) W) XUV light in the spectral range from 13.8 nm to 40 nm. The XUV pulses have a pulse duration of about 30 fs. The pulse pattern of both sources consist of pulse trains with up to 30 pulses with a 1 MHz repetition rate within the burst and a burst repetition rate of 5 Hz.

Since the sources of the two laser pulses are independent from each other, they have to be synchronized. The synchronization is jitter afflicted. The temporal jitter worsens the temporal resolution of pump-probe experiments to about five times the resolution determined by the pulse duration of the optical laser. One source of this temporal jitter is the energy fluctuation of the electron bunch, which becomes an arrival-time fluctuation after the electron bunch compressors. The second source of timing jitter is due to the imperfect synchronization of the Ti:Sa laser oscillator with respect to the reference RF line. Both jitter add to about 250 fs RMS measured during one hour.

To increase the temporal resolution, one can precisely measure the jitter pulse-by-pulse and sort the experimental data according to the measured arrival-time. This is achieved by a diagnostic called Timing by Electro-Optical sampling (TEO). Utilizing this technique, the temporal resolution is no longer dominated by the jitter. It is dominated only by the precision of the timing measurement, which can be made substantially better than the width of the temporal jitter.

EXPERIMENTAL SETUP OF TEO

The TEO diagnostic was set up measure the precise relative arrival-time \( \Delta T_{el} \) of the electron bunch and the IR laser pulse in the accelerator tunnel.

![Figure 1: Overview of the principle setup of the TEO experiment. The relative arrival time between FEL pulse and IR pulse is measured inside the accelerator tunnel using the electron bunches instead of the XUV pulses.](image-url)

It uses the electro-optical effect with the spatial encoding technique to map time into space and produces a signal, from which the relative arrival-time between laser pulse and electron bunch can be extracted (fig. 2). The electron bunch and the IR pulse are arriving simultaneously at an electro-optical GaAs crystal. Here, the longitudinal electron bunch profile is encoded inside the polarization of the transverse laser profile and transformed into an intensity modulation by a polarizer. The transversal position of the signal is depending on the relative arrival-time of electron bunch and optical laser pulse at the EO crystal because of an angle of incidence of 45° of the optical laser to the EO crystal surface. See [3] for more details. Since the used EO...
technique detects primarily the arrival of the electron bunch peak, which produces the FEL SASE pulse, the measured electron bunch arrival-time is in good approximation equal to the relative arrival time of both photon pulses at the experiment. Thus, by measuring the electron arrival-time the photon arrival-time can be predicted within the error bars of the method (≈ 90-120 fs RMS). A similar diagnostic on an incoherent light source at SLAC [2] has shown comparable experimental error bars of ≈ 60 fs RMS of the photon pulse arrival-time.

![TEO signal](image)

**Figure 2:** TEO signal with GaP crystal, 180 μm thickness, non-wedged, crystal edge distance from e^- -beam about 1.5 mm. The arrival-time of the electron bunch is indicated with highest precision by the rising edge, which has a 10%-90% rise time of 190 fs. The inflexion point on this edge is used to read off the arrival-time.

The IR laser system is located in a laser laboratory at the end of the FEL linear accelerator in close vicinity to the pump-probe experiments. To transport the laser pulse Fourier limited to the location of TEO in the accelerator tunnel, which is located 160 m away from the laser laboratory (see fig. 1). The laser pulse transport system is the most complex part of the TEO experiment, because the dispersion of 160 m bulk glass fibre has to be recompressed accurately. Additionally the fibre length is modulated by thermal expansion and micro-phonics, which has to be compensated by active path length stabilization. A detailed description of the dispersion compensation can be found in [3].

Currently the diagnostic can deliver the arrival time for one arbitrary bunch of each bunch train.

**PUMP-PROBE EXPERIMENT**

The performance of the TEO diagnostic can be tested utilizing sideband generation. [6]. For that, the FEL is focussed inside a low pressure Xe gas target of 10^{-6} mbar to ionize the gas atoms. The photo electrons are collected by a magnetic bottle type time of flight spectrometer [5]. The IR pulse from the amplified Ti:Sa laser is focussed into the gas collinearly with the XUV beam of the FEL. Once the two laser pulses overlap in space and time, the electric field of the IR laser pulse interacts with the photo electrons and modulates their kinetic energy by a multiple of hωIR = ± 1.55 eV. Thus, additional sidebands appear in the photo electron spectrum. The intensity and number of sidebands depend on the strength of the modulating electric field of the IR laser pulse, which interacts with the photo electrons. The better the overlap, the more photons are absorbed from the IR field, thus, the sideband intensity increases and sidebands of higher order become visible (fig. 3). Without any spatial jitter (i.e. changing spatial overlap due to pointing fluctuations of the FEL) the sideband intensity is an indicator of the temporal overlap of both pulses with a precision of about 50 fs, according to [6].

**Experimental parameters**

During the experiment, the FEL was operating at 13.8 nm (89.9 eV) in single-bunch mode with a peak energy of up to 50 μJ per pulse. The diameter of the XUV-FEL beam in the interaction volume was 100 ± 10 μm. The IR laser was focussed down to 50 ± 10 μm in diameter with about 5 μJ per pulse and a pulse duration of 120 fs FWHM. Some of the measured electron energy spectra are shown in fig. 3. The excited state 5p^-1 of the Xe atoms has a binding energy of 12.13 eV [4]. Its photo-electron line was observed at about 78 eV inside the time-of-flight spectrum. It splits up into two lines of the spin 1/2- and 3/2-states.

![Time of flight spectra of Xe photo electrons with varying temporal overlap between the optical and XUV laser pulse](image)

**Figure 3:** (Figure adapted from [6]) Time of flight spectra of Xe photo electrons with varying temporal overlap between the optical and XUV laser pulse.
Experimental results

In the following, only the first sideband of the photo electrons is used. The sideband amplitude is correlated with the time information set by a delay stage to scan a time interval of 3 ps around time zero point of best temporal overlap. For each 0.1 ps time step the sideband amplitude is averaged over 250 shots. The correlation is shown in fig. 4 by the outer graph. This trace is the scanned sideband correlation of the XUV and IR laser pulse.

![1st sideband with Xe - TEO reconstruction](image)

Figure 4: Final result of the bunch by bunch correlation between the sideband amplitude and the arrival time measured by TEO. The envelope of the measured data points is the reconstructed temporal cross-correlation of both pulses (dashed line). The outer line (dotted) is the averaged cross-correlation from a pump-probe scan and the inner line (gray) is the theoretically expected cross-correlation.

In a second experiment the delay stage was set to time zero. The arrival-time of each event is measured by TEO and correlated with the sideband amplitude normalized by the integral of the photo electron main line, which is proportional to the FEL energy per pulse. The spread of the time jitter of the FEL was large enough to cover the expected time interval of possible temporal overlap between both pulses. Therefore, a time scan over a large interval was not necessary. The result of the correlation is shown in fig. 4 by the data points.

The width of the scanned correlation \( \Delta T_{\text{scan}} \) (outer line) is 800 fs FWHM (\( \approx 350 \) fs RMS). This width is due to the influence of the temporal jitter much larger than the optimal width \( \Delta T_{\text{optimal}} \approx 125 \) fs FWHM for the cross-correlation of both laser pulses (inner line).

The envelope of the data points is the reconstructed sideband cross-correlation. The width of the envelope \( \Delta T_{\text{corrected}} \) is \( 280 \pm 10 \) fs FWHM. It is two times smaller than the width of the averaged signal. Thus, the signal of the sideband correlation of XUV and IR laser pulse was measured with reduced temporal jitter utilizing the TEO timing information.

Compared with the duration of the theoretical signal, the remaining temporal jitter and drift was reduced from \( \sqrt{\Delta T_{\text{scan}}^2 - \Delta T_{\text{optimal}}^2}/2.35 \approx 340 \) fs RMS to \( \sqrt{\Delta T_{\text{corrected}}^2 - \Delta T_{\text{optimal}}^2}/2.35 \approx 105 \) fs RMS. (In general the variances of two independent Gaussian distributions are added quadratically.)

The data points below the envelope are a result of spatial jitter. That is, despite a good temporal overlap for several data points, the measured sideband amplitude was small, because the spatial overlap of these shots was imperfect.

CONCLUSION

We have shown in a pump-probe experiment, that the Electro-Optical Spatial Decoding technique provides the possibility of an online non-destructive single-shot measurement of the relative arrival time between a high intensity optical laser and the FEL-XUV pulse with a resolution of better than 110 fs. This opens the door for further precise pump-probe experiments at FLASH.

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REFERENCES