

Report of the Work Group III: Measurement of Temporal Properties

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Number of participating discussion partners: 13

(Massimo Altarelli, Alan Fisher, Alexander Föhlich, Markus Grabrysch, Gerhard Ingold, Michael Meyer, Serguei Molodtsov, Klaus Sokolowski-Tinten, Ryszard Sobierajski, Sven Toilekis, Marek Wieland, Helmut Zacharias, Beata Ziaja)

Parameters:

The following parameters should be assessed for a characterization of the pulses of XFEL as well as for the user experiments:

- i) pulse duration and shape,
- ii) arrival time of the x-ray pulses with respect to the pulses of an external optical laser,
- iii) temporal coherence,
- iv) spectral distribution.

Requirements:

For the pulse duration two modes of operation are possible: a short pulse mode with pulses of a few femtoseconds or even below, and a long pulse mode with durations of about 100 fs. Characterization of the overall average values as well as of the individual pulses (single-shot) is required, since it determines directly the temporal resolution of all two-color pump-probe experiments. The shape of the pulses should be measured for each individual shot, since the particular shape (spikes) is an essential parameter for the quantitative analysis of non-linear processes.

The arrival time has to be known with a precision better than 10fs, ideally better than the pulse duration of the optical laser or the FEL whichever is shorter. It is expected that in the beginning such an optical laser will have pulse durations of about 20 fs. Later few-cycle pulses or even attosecond pulses may be envisioned.

Depending on the wavelength generated the coherence time is expected to be in the range from 0.2 fs (at 12 keV) to 1.4 fs (at 250 eV).

The spectral distribution should be measured for each individual pulse. Similar to the pulse shape, the spectral distribution has to be taken into account for non-linear resonant processes.

Devices:

Arrival time monitor:

Several methods have been identified which can serve as arrival time monitors. Besides the optical/x-ray methods (a, b, c), which directly monitor the x-ray pulse, also methods, which observe the electron bunch (d, e), were discussed.

(a) X-ray induced changes in the electronic structure of solid-state materials. Here, changes in both the reflectivity and in the transmission have been discussed.

The reflectivity changes should work similarly as in the XUV range, where the feasibility of the method has been demonstrated (C. Gahl et al., *Nature Photonics* 2, 168 (2008)). At present the temporal resolution is limited by the duration of the optical pulse.

K. Sokolowski-Tinten presented the transmission version, where the active material (in the specific case considered: diamond) is placed inside an optical interferometer. Then only phase changes in the optical beam induced by the traversing x-ray pulse have to be monitored. He calculated that only about 1.5% of the x-ray pulse is sufficient to induce a notable phase shift. Also, the optical beam requires only very low intensity. Therefore, this method shows the capability to be used online with an experiment, as a diagnostic tool to provide user information. The temporal resolution is estimated to be in the 50 fs regime, for relative measurements possibly 20 to 30 fs. Definite numbers for the resolution can, however, not be given at present due to a lack of information about the basic physical process.

Both methods are capable to be used as a single-shot 1-D monitor.

(b) Side-band generation.

This method successfully implemented in the XUV (P. Radcliffe et al., *Appl. Phys. Lett.* 90, 131108 (2007) and *NIM A* 583, 516 (2007)) may also be useful in the hard X-ray regime. It requires however the temporal overlap of X-ray and optical pulses and the knowledge of the pulse shapes. The technical limitations are seen in the regions below 10 fs.

For these few femtosecond pulses other physical phenomena ("streaking") become important. The application of the streaking method for the determination of the arrival time will be tested at the LCLS (see proposals R. Kienberger and A. Cavalieri)

(c) Structural phase transitions.

Optically induced structural phase transitions in materials which contain light atoms and show optical phonon frequencies in the 6 to 10 THz are seen as promising approach. Although at present no dedicated experiment has been performed, the group felt that one should monitor the development in this area. In a recent experiment, the almost complete disappearance of a superlattice Bragg reflection within 200 fs has been measured due to a bisection of the unit cell in a photoinduced metal-to-insulator phase transition (P. Beaud et al., *PRL* 103, 155702 (2009)). Phonon driven structural phase transitions involving light atoms should yield much shorter time scales in the order of 30-50 fs.

This method would also allow to be used as an on-line user monitor, because the development of new Bragg peaks from the phase transition requires only weak x-ray intensities. A beam-splitter will be needed. The temporal resolution, however, is seen more at the upper end of the pulse duration of XFEL.

(d) Transverse electro-optic sampling.

This proven method for electron bunches at existing FELs (e.g. A. Azima et al., *Appl. Phys. Lett.* 94, 144102 (2009)) should also work for electron bunches in the XFEL. The temporal resolution of the set-up used at FLASH was measured to less than 90fs (rms), which is mainly determined by the stability of the fiber transport of the optical laser and its pulse duration. The ultimate time resolution is therefore not known, but it is estimated be better than 10-20 fs.

(e) Phase shift in RF cavities.

This method is implemented presently at the LCLS. It measures the electron bunch arrival time relative to an external RF clock. For the precision of the method, values of 30-50 fs are discussed at the moment.

For the latter two methods one has to notice that they might show some uncertainty, because a time jitter may exist between the electron bunch and the x-ray pulse generated from this bunch.

Pulse duration:

(a) Saturated absorption is foreseen as a suitable process to measure the pulse duration. This might be achieved in a saturated K and L shell absorption in appropriate solid state material, similar to the saturated L shell absorption recently observed in Al ([B. Nagler et al., Nat. Phys. 5, 693 \(2009\)](#)).

Also, similar processes in the gas phase, as multiple ionization processes, up to the complete stripping of all electrons from the atoms, may result in a sufficiently simple nonlinear process, i.e. theoretically well characterized. Here one should watch the discussion about the Ne experiment recently performed at LCLS.

As a device a two-pulse split and delay unit (i autocorrelator), which works in the hard x-ray regime is required. Both short and long femtosecond pulses can be measured.

(b) Streaking.

Streaking is a linear method which principally should also work in the hard x-ray regime. A difficulty is seen in the timing of a far-IR pulse and the x-ray pulses. In the experiment performed in the XUV at FLASH ([U. Fr, hling et al., Nature Photonics. 3, 523 \(2009\)](#)) the IR is generated in an undulator at the end of the XUV undulator from the same electron bunch. This minimizes the jitter which is believed to be in the 5 fs range. Both pulses are then optically delayed appropriately. The present lack of ∞ mirrors in the hard x-ray regime with a broad spectral acceptance bandwidth poses some problems. Bragg reflections at angles around 45° , possible at certain wavelengths, are in this case no remedy, because the extreme narrow spectral range does not permit to measure the pulse duration of the XFEL pulses.

An alternative route might be to derive the far-IR radiation with pulse energies in the 10 μ J range from a laser source synchronized to the electron bunch by difference frequency mixing or optical rectification ([K.L. Yeh et al., Appl. Phys. Lett. 90, 171121 \(2007\)](#), [K. Reimann, Rep. Prog. Phys. 70, 1597 \(2007\)](#)) It is at present uncertain whether the timing jitter allows such measurements.

(c) Two-color cross correlation.

As for the arrival time measurements two-color cross correlation and side band generation may be a suitable tool for longer XFEL pulses. The methods rely on performing a scan of the time delay between optical and x-ray pulses, therefore only average values, but no single-shot data will be available.

(d) Additional optical undulator.

In principal a second optical undulator may generate radiation in the optical regime which then may be frequency doubled. In that case, however, one gains information about a depleted electron bunch, which may have nothing to do with the bunch generating the x-ray pulse. At present, we judge this method as not suitable to assert the x-ray pulse duration.

Temporal coherence:

Although not discussed extensively, single shot temporal coherence measurements may also be employed to learn about the pulse shape of the x-ray pulses. As presented by R. Sobierajski and noticed by Mitzner et al. ([R. Mitzner et al., Opt. Expr. 16, 19909 \(2008\)](#)) at a beamtime of FLASH, sub-pulses do cross correlate even in a linear correlation. This then yields at a certain delay time relative to zero delay a local maximum in the visibility. In a single-pulse temporal cross correlation measurement this should show up even when the sub-pulses have no fixed phase relationship from one pulse to the next.

A resolution of a few femtoseconds is foreseen, and since the measurement does not require high intensities it can be used as a monitor for user experiments. As a device a two-pulse split and delay unit is required.

Spectral distribution:

It was discussed whether the single shot spectral distribution could give information about the overall pulse duration and the temporal sub-structure. The group came to the conclusion that from the spectral distribution only a lower bound of the pulse duration can be derived, because the chirp of the x-ray pulses are not known. For some experiments this might be sufficient as a sorting criterion.