Opportunities and challenges using short-pulse X-ray sources.

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Abstract. Free-electron lasers will change the way we carry out time-resolved X-ray experiments. At present date, we use laser-produced plasma sources or synchrotron radiation. Laser-produced plasma sources have short pulses, but unfortunately large pulse-to-pulse fluctuations and large divergence. Synchrotron radiation from third generation source provide collimated and stable beams, but unfortunately long pulses. This means that either the time-resolution is limited to 100 ps or rather complex set-ups involving slicing or streak cameras are needed. Hard X-ray free-electron lasers will combine the best properties of present-day sources and increase the number of photons by many orders of magnitude. Already today, a precursor to the free-electron lasers has been built at Stanford Linear Accelerator Centre (SLAC). The Sub-Picosecond Photon Source (SPPS) has already shown the opportunities and challenges of using short-pulse X-ray sources.

1. Background
Since the discovery of X-rays, the average brilliance of X-ray sources has been increasing by more than 3 orders of magnitude every 10 years. New machines for producing X-ray radiation have been created enabling new kinds of measurements. Time-resolved X-ray studies require instruments with intense and very short X-ray pulses. Time-resolved X-ray diffraction is a rapidly growing research field in spite of the limitations of present-day sources. Non-thermal melting of semiconductor [1-3], dynamics of acoustic and optical phonons [4-8] as well as ferroelectric phase-transitions [9-10] and nanostructured materials have been investigated [11-12]. Probing chemical reactions, with diffraction
or X-ray absorption spectroscopy [13-14] and the study of the dynamics of photo-excited biomolecules [15-16] are examples of time-resolved X-ray techniques applied outside the physical sciences. New methods in structural determination will also require bright, short-pulse sources [17].

The next quantum leap in source technology will be taken when the hard X-ray SASE-FEL (Self-amplified spontaneous emission free electron laser) at SLAC Stanford becomes operational in 2009 [18]. The first SASE FEL where saturation was reached in the visible part of the spectrum was LEUTL [19] at APS (USA). The TTF (Tesla Test Facility, Hamburg) has pushed the SASE principle further [20] and performed experiments using the radiation around 100 nm [21]. This machine has since been rebuilt into VUV-FEL which is a user facility operating at 30 nm. The most advanced development is today pushed by the construction of LCLS (Stanford USA) [18] and European X-FEL (Hamburg, Germany) [22] which both aim at creating an X-ray laser-like source. There are plans to extend the Japanese soft X-ray-source [23] which is under construction to the hard X-ray range. These machines will require extreme electron beam characteristics regarding energy, emittance, peak current and stability.

The Sub-Picosecond Photon Source (SPPS) [24, 25] is a short-pulse X-ray source in the fine-focus test beam (FFTB) branch of the SLAC Linac, 80 fs electrons generate short incoherent X-ray pulses in an undulator. In a highly collimated beam (0.01 mrad) $10^7$ photons in a few percent bandwidth reaches the X-ray hutch.

In order to focus R&D efforts and to bring in key know-how from user groups world-wide the SPPS collaboration was launched at Stanford Linear Accelerator Center (SLAC). A collaboration team consisting of partners from Europe and the US has been formed. R&D on synchronisation, bunch-compression and bunch diagnostics has been carried out [26] in synergy with scientific studies on non-thermal melting [27]. Due to the fact that LCLS will occupy the same physical space as SPPS the latter facility will most likely close early 2006. In the timeframe up to the commissioning of the LCLS and also in the coming years due to the limited beamtime available, further sources inspired by SPPS could play an important role.

2. Timing issues

2.1. Synchronisation between a short-pulse laser and the accelerator

In order to synchronize a short-pulse laser to the electron bunch structure it is necessary to control the optical length of the cavity. This can be done by mounting the end mirror on a piezo-electric crystal. The key is to realize that the repetition rate of a mode-locked laser is defined by its cavity length. The ability to change the cavity length by controlling the piezo crystal turns this mode-locked laser into a voltage-controlled oscillator. Synchronization can then be performed by a phase-locked loop, described in many electronics textbooks. Locking the laser oscillator to the rf-frequency gives the fine-tuning. It should be noted that synchronization is made to the rf used to accelerate the electrons and not to the electrons them selves.

2.2. Changing the timing between the laser and the accelerator

The most obvious way of changing the timing between the pump and the probe is to use an optical delay line for the visible pump. This is used for all visible pump-probe experiments and gives a very high temporal resolution limited only by the time for light to propagate the distance of the spatial resolution of the stage. A more convenient way in these often remote controlled experiments is to use a radio-frequency phase-shifter in the laser synchronisation control loop. The phase stability of commercial devices is typically a degree, but better performance can be achieved.

2.3. Extracting temporal information in the experiment

The most straight-forward way of extracting temporal information is in a classic pump-probe experiment. If there is a significant jitter between the laser and X-ray pulses as discussed above, each shot needs to be accompanied with an absolute timing reference. It may even be that if the jitter is
random and spans a larger time than what is to be investigated, no active time scans are needed. It is only necessary to sort the data according to the relative time between laser and X-rays as measured by the absolute timing monitor.

A way of capturing the full temporal history in a single shot was proposed by Neutze and Hajdu [28] and later demonstrated by Synnergren et al. [29]. The experimental configuration as implemented at SPPS [27] is shown in Figure 1.

Since the laser arrives at an angle relative to the X-rays they will not interact with the sample at the same time over the interaction area. There will be a sweep with a speed which is determined by the relative angle of the two beams. The sweep duration is given by $L/c \left( \sin X - \sin V \right)$ where $V$ is the angle between the propagation vector of the laser and the surface normal and $X$ is corresponding angle of the X-rays. $L$ is the length over which the laser and X-rays have a uniform beam profile and $c$ is the speed of light. In addition, many reflections where the corresponding Q-vector is at an angle to the surface normal can be found by simply rotating the sample. The geometrical construction illustrating this can also be found in Figure 1. The reconstruction is shown for the case when the incident X-ray beam is grazing the surface of the crystal. This was the case in our measurement where the incidence angle of 0.4 degrees also helped matching the penetration depth of the laser and probe depth of the X-rays. By rotating the crystal and thereby the Q-vector, the intersection with the circle defined by the $k_s$ vector is found, which is when the Laue formulation of Braggs Law is fulfilled.

Crossed-beam topography is a technique which brings many advantages. Other than the fact that pulse-to-pulse fluctuations are negated in averaging experiments, it also allows the capture of data in a single shot which relaxes the need for a timing monitor unless the excitation time (often refereed to as

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**Figure 1.** Experimental set-up for studying non-thermal melting of InSb, using the cross-beam technique. $Q$ is the lattice vector, $k$ the k-vector of the incident X-ray beam and $k_s$ denotes the k-vector of the scattered X-ray beam. In the geometry used, several reflections could be reached while keeping the laser and X-rays aligned. Later times (increasing $t$) means that the X-rays are later with respect to the laser.
time zero) must be known. Single-shot measurement will be applicable to many samples when high-power free-electron lasers are the source.

The drawbacks are that the requirement of the laser beam profile uniformity is high and that an imaging detector is needed.

In some studies it may be desirable to have a larger temporal window than what practically can be achieved by using the flight time of electro-magnetic radiation over the interaction region. This was addressed in [29], where a grating arrangement was used to provide a 200 ps temporal window. The time delay at different spatial locations along the crystal’s surface was introduced by reflecting the laser pulse from a diffraction grating prior to it being imaged onto the sample. As such the reflected wave front became tilted relative to its direction of propagation, with the result that different rays traversed different optical path lengths before arriving at the semiconductor’s surface.

2.4. Measuring timing between the laser and the accelerator

The most straightforward way of measuring the relative timing between a laser and X-rays from an accelerator is using an X-ray sensitive streakcamera [30, 31]. By using a gold-cathode, a relatively high sensitivity for radiation with a wavelength <400 nm can be obtained. The absolute timing can be measured at the interaction point and it is also possible to use a streakcamera as an online jitter monitor.

Another type of online jitter monitor is to use an optical cross-correlator. Such a device can be used in a scanning [32] or single-shot [33] geometry.

Recent studies at SPPS [26] has shown that the electro-optic effect which occurs in a Pockels active crystal when the electrical field originating from the relativistic bunch is a powerful synchronization tool. By sending a part of the laser-pulse through the EO-crystal and analyzing the polarization change of the light, the relative timing of the laser and X-rays can be deduced. A major advantage is that it does not require extraction of light from the accelerator. It is possible to use a pulse from the oscillator which is sent out one roundtrip before the pulse which is amplified and used for experiments.

A very direct timing method, which is not practical to implement as an online monitor is the non-thermal melting of InSb discussed below. Although the complexity is too high to use it as an online monitor, the method can be used to evaluate all other timing techniques. In the top of Fig. 2 we see the total interaction area by deliberately making the laser arrive too early. Below is when the laser is timed and the jitter causes the edge to move around.

3. Non-thermal melting of InSb

When a semiconductor is excited by intense femtosecond laser pulses, a dense electron-hole plasma is formed. Stampfli and Bennemann calculated that the changes in the inter-atomic potential that occurs under these conditions may lead to disordering of the crystalline lattice on a time scale faster than the thermal equilibrium time [34, 35]. This is sometimes referred to as non-thermal melting. Already 20 years ago it was found that the optical properties of the surface change from that of a solid to that of the liquid in a few 100 fs [36]. In a pioneering study by Rousse et al.[1] the lattice disordering was directly measured to atomic pathways have not been possible to deduce in this and subsequent studies using laser-produced plasma sources.
Using the cross-beam technique together with the SPPS we have been able to study non-thermal melting with higher temporal resolution and signal-to-noise ratio compared to what has been possible before. We have also studied two different Bragg-reflections. We observe two distinct characteristics of the disordering process. The intensity drop is non-exponential and follows Gaussian temporal profile $I(t) = e^{-Q^2\omega^2t^3/3}$ from the time of excitation and for a few 100 fs until the intensity was small. The ratio between the disordering time of the 111 and the 220 reflections are $(\sqrt{8}/3)$ [26], implying an inverse linear dependence in $Q$. Both of these observations are accounted for in a numerical description using the Debye-Waller formalism which yields the diffracted intensity as function of the mean displacement, which relates the time-dependent decrease in scattered intensity to a time-dependent rms displacement: $I(Q, t) = e^{-Q^2\omega^2t^3/3}$. This behaviour is consistent with an intuitive model. Before the laser excitation the ensemble of atoms are vibrating with random phase in the atomic potential. As the laser excites the sample, the inter-atomic bonds are severed and the atoms move according to their initial velocities so that the ensemble of atoms moves in the direction and with the velocity they had when the laser excited the sample. The thermal rms velocity is 2.5 Å/ps. The extracted velocity from our data is 2.3 Å/ps. There are uncertainties in the determination and it is also possible that atoms lose or gain speed in the modified potential landscape which results after laser excitation. The change in speed may also be different in different directions. This is under further investigation.

4. New source initiatives

Discussions about creating a free-electron lasers operating in the UV, VUV and X-ray region are currently ongoing in many laboratories world-wide. As an example, a soft X-ray FEL is envisioned as a part of the MAX IV project. So far funding has only been secured for a test facility operating at 90 nm. During the last year, the direction of development has slightly switched and the focus for several of the new sources is leaning towards seeding schemes (the amplification process is defined by an initial well defined light beam) and harmonic generation (the seed laser wavelength is multiplied). These principles have already been tested in the 80ies for example at Super-ACO in Paris [37] and at MAX I [38] but are now implemented on dedicated short-pulse accelerators. As a branchline to the free-electron laser a femtosecond incoherent X-ray beamline is being designed. The aim is to provide a source which delivers the photon numbers shown in the figure below The graph below requires an upgrade of the existing 500 MeV Linac to 900 MeV and the installation of a 5 T 50-pole wiggler. Note that even at 9 keV, the photon numbers rival those of the SPPS. The brilliance of SPPS is much higher as the divergence is orders of magnitude smaller. Bunch compression will be installed and sub-ps pulses produced. The aim is to go down to 100 fs. A 10 Hz, TW laser will be synchronized to the installation and a station for pump-probe experiments developed. Due to the relatively small size of the MAX Laboratory (<50 m) it is likely that the same laser front end will be used as a photocathode laser and as pump-probe laser.

Figure 3. Non-thermal melting of InSb. The elongated diffraction spot is due to the extreme asymmetry. The top curve is before laser irradiation. The central one is during laser excitation. The bottom streak is after irradiation.
Figure 4. The number of photons per pulse as function of the photon energy. The calculation was carried out using the SPECTRA [38]. The following parameters were used: Electron energy = 900 MeV, Electron emittance 1.7 e-9 mrad mm$^2$, Electron energy spread dE/E 5e-4, Wiggler field strength = 5 T, Wiggler period = 6 cm, Wiggler length = 3 m = 50 periods, Bunch current 1 nC, Slit 5 x 1 mRad. The wiggler data is based on the MAX II superconducting wigglers assuming a smaller gap and an extension to twice the length.

Figure 5. A view of the MAX-Laboratory housing the MAX I, MAX II and MAX III electron storage rings and the 500 MeV LINAC. A 90 nm FEL and a hard X-ray station are planned inside the MAX II ring.
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