Ultrafast laser pump X-ray probe experiments by means of asynchronous sampling

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Abstract. A high time resolution in the picosecond range is required for the time-domain investigation of phonon dynamics in crystalline systems. Following a recently developed scheme in the visible spectrum, this resolution can be achieved by a method called asynchronous optical X-ray sampling (ASOXS). A pulsed femtosecond laser with high repetition rate is synchronized to the electron bunches in a storage ring. A slight frequency detuning changes the mutual delay continuously, resulting in a time-domain X-ray sampling of the laser-excited system. At the synchrotron radiation source ANKA a machine mode with low momentum compaction factor $\alpha_c$ is available, which delivers ultra-short X-ray pulses in the picosecond range.

1. Introduction
Phonon dynamics can be studied by time-domain methods, such as femtosecond spectroscopy or time-resolved X-ray scattering [1]. Resolving non-equilibrium motion of the crystal lattice requires tools to resolve few picoseconds. For X-ray scattering as lattice-sensitive probe several sources of radiation are available. Laser-induced plasma sources emit X-ray pulses with a sub-picosecond duration, that allows to resolve lattice motion [2, 3]. Synchrotrons also emit pulsed radiation, which, however, is limited to pulse durations of about 100 ps [4]. By aid of a streak camera this time resolution can be reduced to a few picoseconds [5]. A second approach to reduce the pulse length at a synchrotron is a special mode with quasi-isochronous orbits for electrons of different energy. This "low-$\alpha_c$" mode results in a modification of phase space such that the pulses are reduced in longitudinal extension.

This mode is being employed in order to produce coherent THz radiation at the synchrotron radiation source ANKA (Karlsruhe Institute of Technology). We exploit the ability to use these short pulses to perform pump-probe experiments with a femtosecond oscillator as pump and micro-focused X-ray pulses as probe for lattice excitation in crystals. The use of the high pulse repetition rate of the synchrotron together with the asynchronous sampling allows for a sampling of a full delay span with high sensitivity.
2. Technical setup
The goal is to excite coherent lattice motion by ultrashort laser pulses and to probe the change in atomic distances by X-ray scattering or alternatively X-ray absorption spectroscopy. The ANKA ring uses a 500 MHz bunch clock, which means that pulse-to-pulse distance of the X-ray emission is 2 ns. Modes of lower frequency can be used, which implies that only selected buckets are filled with electron bunches. In the present setup the 500 MHz rate is used by synchronizing a femtosecond oscillator (Gigajet, Gigaoptics) of the same frequency to the X-ray emission. The output is up to 1 W of laser power at 800 nm. The laser can be synchronized by feeding the electronic signal from a strip-line electrode in the ring to a phase-lock loop.

Instead of using the direct synchronization, which fixes the delay between laser and X-ray pulses at the sample, a relatively low difference frequency is synthesized upon the feedback signal [6]. This difference frequency induces a continuous sweep of the delay between the two sources from 0 to 2 ns. The scheme is depicted in figure 1. At one instant in time both laser and X-ray pulses coincide on the sample. For the next pulse pair the laser arrives slightly earlier. This delay change will go on until a later laser pulse (N+1) overtakes the X-ray pulse (N). The real time span (drift time) is given by the inverse difference frequency. For example a difference frequency of 25 kHz leads to a drift time of 40 µs. Thus the pump-probe delay between 0 and 2 ns is mapped on a drift time from 0 to 40 µs. Consequently a reasonable fast X-ray detector is able to resolve picosecond dynamics while itself having only a nanosecond time resolution. We are using an avalanche photo-diode (Cyberstar, FMB Oxford) together with a multichannel scaler for time-resolved pulse recording (Nanoharp, Picoquant). This combination allows for a 4 ns resolution in drift time, which relates to a 200 fs nominal time resolution. This is shorter than the currently available X-ray pulse length. Synchronisation has been proven to work better that 1 ps jitter (fwhm, full width at half maximum) [7, 9].

The laser pulse energy is very low due to the high repetition rate. This limits the studies to mostly linear excitation regime. The important quantity is the excitation density in the sample, which is given by the laser fluence. In order to raise this fluence to a level near 1 mJ/cm² for visible lattice motion, we need to focus both beams strongly. This can be accomplished at the SUL-X microfocusing beamline at ANKA [8]. A Kirkpatrick-Baez (KB) silicon mirror pair

Figure 1. ASOXS scheme.

Figure 2. Bunch length as function of bunch current for both 1.6 GeV and 1.3 GeV electron energy. The RF voltage was set to 1 MV, the synchrotron frequency was 8 resp. 5.6 kHz for both energies [11].
produces a focal spot at the sample of about 150 \( \mu \text{m} \), which can be further reduced by a slit at an intermediate focus position of the primary mirrors. A silicon double monochromator delivers monochromatic X-rays with a 2 eV bandwidth. We are using a 35 \( \mu \text{m} \) focus size for both laser and X-ray beams. The laser beam can be steered by a motorized lens for optimal spatial overlap.

3. Implications of the low-\( \alpha_c \) mode
Pulse length and emittance are a function of several parameters of the synchrotron optics. First of all, \( \alpha_c \) determines the ratio of longitudinal pulse compression. This results in the shortest pulses at lower electron energy. Best results are obtained at 1.3 and 1.6 GeV. Changes in the \( \alpha_c \) parameter can be expressed in terms of the synchrotron frequency (an easily measurable quantity), a particular mode excitation mode of the electron bunches [10]. Typical values are 30 kHz in normal operation mode down to several kHz for strongly compacted beams.

A second parameter is the charge per bunch. Higher charges lead to stronger repulsion and instabilities and thus to bunch lengthening. Two measurements of pulse length are given in figure 2. A comparison has been made between the pulse length (rms, root mean square) at 1.3 and 1.6 GeV at variable bunch charge [11]. The data has been measured for the visible-light emission with a streak camera. These curves have a characteristic shape for each synchrotron frequency. At the lowest bunch charge the pulse length reaches a constant independent of charge. At higher charges at a diagonal line in the diagram the pulse length would increase with a power law. At that point the electron bunches experience microbunch instabilities which leads to a blow up of phase space. For THz production these microbunch instabilities are beneficial as coherent emission is amplified by strong gradients in charge. In the X-ray regime, in contrast, smallest pulse lengths are wanted, which limits the usable charge and thus the obtainable X-ray flux. While operation at 1.6 GeV on one hand shows a larger minimum bunch length, the microbunch instabilities here start a higher charge, thus a higher flux is achievable. Together with the critical energy of the permanent-magnet wiggler at SUL-X decreasing from 2.4 keV to 1.6 keV from 1.6 to 1.3 GeV this amounts for a substantial flux change. Furthermore the accelerating voltage (\( V_{RF} \)) in the radiofrequency cavities changes the bunch length proportional to \( \sqrt{V_{RF}} \). Thus a factor of about 20% can be gained by going from the usual 1 MV voltage to 1.6 MV.

4. Discussion of settings and results
A typical ASOXS setup at the beamline SUL-X therefore uses a 1.6 GeV electron beam with as many as possible filled buckets in the ring to reduce the individual bunch charge at given ring current and to fill the ASOXS pattern (see figure 1) as regularly as possible. Typically 4 trains of 30 buckets each are filled. The synchrotron frequency is reduced to 6-8 kHz by tuning the magnet optics. Then the RF is raised to 1.6 MV. The lifetime of a 30 mA beam is about 2 hours immediately after fill and increases to 14 hours below 20 mA. The X-ray energy is chosen between 4.5 and 6 keV as an optimum between resolution in scattering, X-ray flux and achievable momentum transfer.

With a laser power of 1 W the fluence on the sample is limited to about 0.2 mJ/cm\(^2\). At a laser absorption depth of 0.6 \( \mu \text{m} \) for GaAs a temperature rise of 0.9 K is achievable for each pulse [9]. The accumulation effect by the high repetition rate is nevertheless much more prominent with \( \Delta T > 100 \) K. Thus the change in scattering intensity will be quite small and a high accuracy is needed for the detection of lattice dynamics. Figure 3 shows several traces of changes of reflection intensity near the symmetric (400) peak of undoped GaAs. At the expansion side intensity increases transiently by some % after laser impact (delay 0). This can be followed by oscillations which represent the lattice movement due to the excitation of coherent phonons. As these phonons are long-wavelength longitudinal acoustic phonons their frequency depends linearly on the momentum transfer relative to the reciprocal lattice point.
Thus a change $\Delta \theta$ relative to the Bragg angle position produces traces that bear the signature of different oscillation frequencies $[12, 1]$.

![Figure 3](image_url)

**Figure 3.** Laser-induced difference intensity close to the GaAs (400) reflection for two difference angles, $\Delta \theta$ (photon frequency) 0.02 degrees (11.7 GHz), shifted upwards for clarity and 0.005 degrees (2.9 GHz).

In Figure 3 a sinusoidal curve with the frequency given by the dispersion relation is displayed along with the intensity traces for different $\Delta \theta$. The acquisition time per trace was 80 seconds. It can be clearly seen that for increasing $\Delta \theta$ the slope of intensity changes at delay zero becomes steeper in agreement with the increasing frequency. The rise time is about 9 ps (root-mean-square) for 0.02 degrees and limited by the phonon propagation $[9]$. Partially the oscillations are resolved. There are several reasons for the low visibility of the coherent phonon signature. First, the X-ray beam has a finite divergence due to the KB optics (0.004 degrees for the slit setting), which limits the accuracy of definition of $\Delta \theta$. Secondly the focus size for both laser and X-ray beams is similar so that a gradient of excitation fluence is detected. Thirdly the X-ray penetration depth is slightly higher than the laser penetration depth. Therefore the signal will not be a pure oscillatory change of intensity but also detect the propagation of the strain front into the material. The signature of this process is a monotonous increase and relaxation of intensity at the expansion side of the Bragg reflection $[13]$. Non-statistical signal fluctuations might be seen at negative delays due to laser intensity variation due to the feedback loop.

In conclusion we have shown that an asynchronous pump-probe scheme can be utilized for the ANKA low-$\alpha_c$ mode with reduced pulse length, despite the high repetition rate and low pulse flux. Extremely small lattice motion can be detected. A gain in sensitivity is expected for an undulator source beamline as well as an improved detection scheme with spatially resolving detectors.

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