DELIVERABLE REPORT

WP9 Research on Time-resolved ultrafast probes on nanosystems

D9.4

Setup for pump-probe small and wide angle scattering experiments

Expected date

M42

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Emmanuel Stratakis (FORTH)

DELIVERABLE DETAILS

DELIVERABLE ID	DELIVERABLE TITLE
D9.4	Setup for pump-probe small and wide angle scattering experiments

DELIVERABLE DESCRIPTION

TUG develops a pump-probe setup for small and wide angle scattering experiments. The scope is to follow the structural evolution of ultra-fast chemical/catalytic reactions. This activity will prepare future implementation of FEL experiments.

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NATURE

P - Prototype

DISSEMINATION LEVEL

☒ P - Public
☐ PP - Restricted to other programme participants & EC: (Specify)
☐ RE - Restricted to a group (Specify)
☐ CO - Confidential, only for members of the consortium

REPORT DETAILS

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Executive Summary

With the drastic need for renewable energy sources, materials with solar harvesting capabilities receive increasingly attention for technological and scientific purposes. In such energy-conversion materials, the principal interaction between light and matter occurs on the nanometer level – a regime in which most properties of bulk-matter do not hold. A comprehensive understanding of these phenomena, particularly the structural material response within the first pico- and nanoseconds after the absorption of light, is critical for the design of novel nanomaterials with enhanced energy conversion efficiency. Deliverable 9.4 aims at building a time-resolved optical-pump X-Ray-probe setup, capable of studying the picosecond lattice dynamics of nanomaterials. This is achieved by installing an ultrafast high-repetition-rate laser at the AustroSAXS beamline of the Elettra synchrotron, where X-Ray scattering / diffraction patterns are taken before, during and after optical excitation. Pilot-experiments were successful, paving the way for accessibility by future NFFA-Europe users.

1. Concept

The concept of this deliverable is to facilitate the pulsed nature of the Elettra storage ring by synchronizing a laboratory laser source to the synchrotron bunch-structure. In this context, we defined three major objectives of this task: i) synchronization of beamline-electronics to the synchrotron-frequency, ii) construction and implementation of laser-related infrastructure on-site and iii) spatial and temporal overlap of laser and X-Ray pulse. In this report, the following paragraphs of this section will introduce the setup’s boundary conditions, predefined by the synchrotron/beamline infrastructure. The specific technical details of the implementation will be presented in section 2, whereas section 3 include results of the first proof-of-principle experiment. In the final section 4, we will give an outlook on planned improvements and further conclude the potential impact for the NFFA-Europe user-community.

1.1 Infrastructure Boundary Conditions

This deliverable is to be implemented at the TUG host-facility: the Austrian SAXS beamline at the Elettra synchrotron in Trieste. The beamline is positioned at the trajectory of a wiggler insertion device, delivering approx. 10^{12} photons s^{-1} mm^{-2} at 5.6, 8 and 16 keV. As X-Ray radiation is generated by deflection of electron-buckets within the wiggler, the temporal width of the radiation pulse is predefined by two factors: i) the electron-bucket length (approx. 90 ps at 400mA ring current) and ii) the diversion of the electron path though the wiggler compared to the direct trajectory (approx. 15 ps). The resulting X-Ray pulse width at sample can therefore be estimated to be approx. 92 ps (std.dev), which is feasible for picosecond resolved experiments.

As electrons are distributed in “bunches” within in the storage ring, one has to consider their temporal sequence, so the “bunch-structure”. In case of the Elettra storage ring, the cavities responsible for temporal ring-stability operate at 500 MHz (2 ns) – the ring radio frequency (RF). Given by the ring-size, one therefore finds a total of 432 “buckets” each spaced 2 ns apart, resulting in a ring repetition rate (RR) of approx. 1.16 MHz (864 ns). Each of these buckets may (or may not) be filled with electrons, resulting in the previously termed "bunch-structure". While Elettra may operate at several different bunch configurations, the most feasible operation mode for the purpose of time-resolved experiments is the "hybrid mode" (see Figure 1a), consisting of: i) a continuous filling mode and ii) a single electron bunch with approx. three times higher current in a 160 ns camshaft - the "dark-gap". The advantage of this operation mode is that the radiation of the single electron bunch can be isolated easily for time-resolved experiments, while the continuous filling regime delivers an overall high average flux for all other beamlines on the ring.

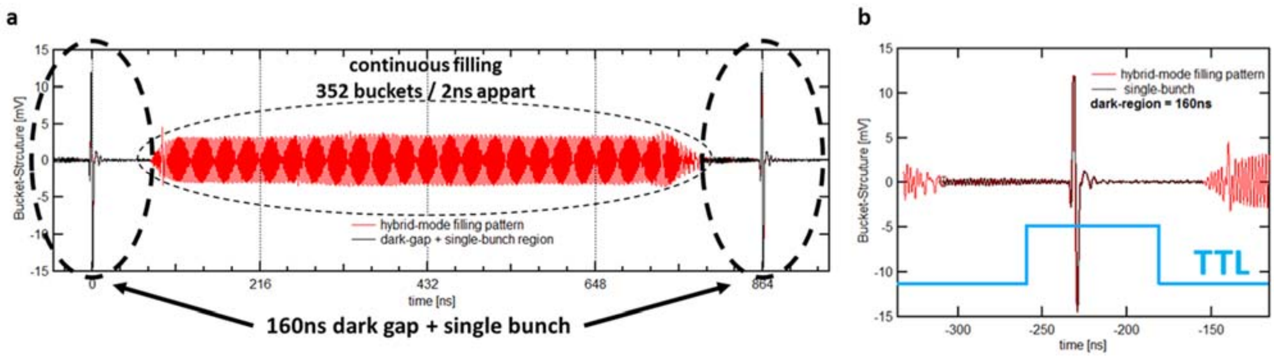

Figure 1: a) Hybrid filling mode of the Elettra storage ring, showing a "continuous filling" regime of approx. 700 ns and a single electron bunch within the 160 ns camshaft – the "dark gap". b) Detailed view of the single-bunch in the "dark gap", which is used as X-Ray probe for the time-resolved experiments.

Considering these ring- and beamline-specific restraints, we derive a list of specifications, predefined by the available infrastructure (see Table 1). As seen, the overall X-Ray flux from a single X-Ray pulse is in the order of 10^3 photons on the sample. In order to still achieve feasible counting statistics in the experiment with timely manner, experiments must be performed at repetition rates in the order of 100-500 kHz resulting in an overall X-Ray flux at 10^9 photons s^{-1} mm^{-2}. Consequently, we chose to install a high-power/high-repetition rate Nd:YAG laser (at 1030, 515 and 343 nm), delivering 240 fs pulses with up to 40 µJ at 500 kHz repetition rate.

Table 1: Setup-specifications predefined by the Elettra storage ring and the SAXS beamline X-Ray optics.

Setup Parameter	Value
X-Ray pulse length (std.dev.)	92 ps
storage ring RF (= sinusoidal timebase)	500 MHz
max. probe repetition rate	1.16 MHz
max. dark-gap length	160 ns
max. usable flux @ sample	7 x 10^9 photons s^{-1} mm^{-2}
max. flux / single-pulse	6 x 10^3 photons mm^{-2}

2. Technical Implementation

2.1 Temporal Synchronization

In order to perform single-bunch experiments at the Elettra facility, the necessary electronic equipment must be synchronized to the timebase of the storage ring, i.e. the RF signal. In this case, two major components are to be synchronized: i) the X-Ray detector and ii) the laser cavity. An overview of the underlying timing-scheme is shown in Figure 2.

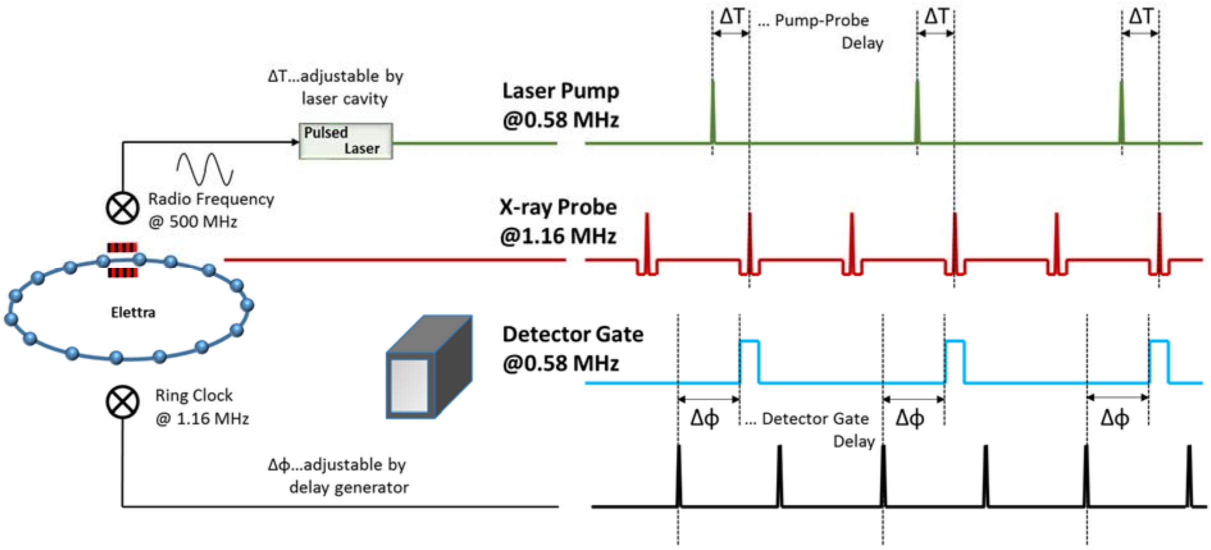

Figure 2: Timing-scheme used to synchronize the laser cavity as well as the detector gate to the timebase of the storage-ring.

In regard of i), the X-Ray detetor operates in a "gated" configuration: an electronic TTL pulse temporarly activates the sensing area of the detector such that all incident X-Rays outside of this pulse are neglected. If timed correctly, the detector can be activated exclusively during the "dark-gap", such that only photons from the single-bunch are counted (see Figure 1b). We achieve synchronization of the detector-gate via an in-house made ring-clock (delivering an electronic pulse every ring repetition – see black trace in Figure 2) followed by an electronic delay generator (generating the detctor gate – see blue trace in Figure 2). Hence, by scanning the detector-gate delay over the ring-clock phase, the single-bunch in the dark gap can be isolated.

In regard of ii), the oscillator cavity of the laser was tuned to operate at 1/6 of the RF, so at 83.33 MHz. Additionally, the two cavity mirrors were placed on motorized stages, which allows rapid fine-adjusment of the oscillator frequnecy. By analogue phase comparison of the RF/6 signal to the actual oscillator frequency, the precise phase offset can be measured and therby used as input for a PID controlled feedback loop. This scheme is implemented in a commerical timing module, which is installed on site, resulting in a laser-to-RF jitter of 0.4 ps. The RF/6 signal can further be phase-shifted within the timing module, effectively allowing tuneability of the laser-pulse delay (see green trace in Figure 2).

2.2 Construction of Laser Infrastructure

In order to accommodate the acquired femtosecond laser, we had to adjust and expand our existing beamline infrastructure. In detail, this included three major milestones: i) construction of an "offline" laser hutch (to allow useability of the laser independtly from beamline operation), ii) construction of a beam-transfer line (porting the laser-beam to the X-Ray hutch) and iii) construction of the pump-probe sample stage (where both beams are spatialy overlapped on the sample). A graphic overview of the final infrastructure can be seen in Figure 3.

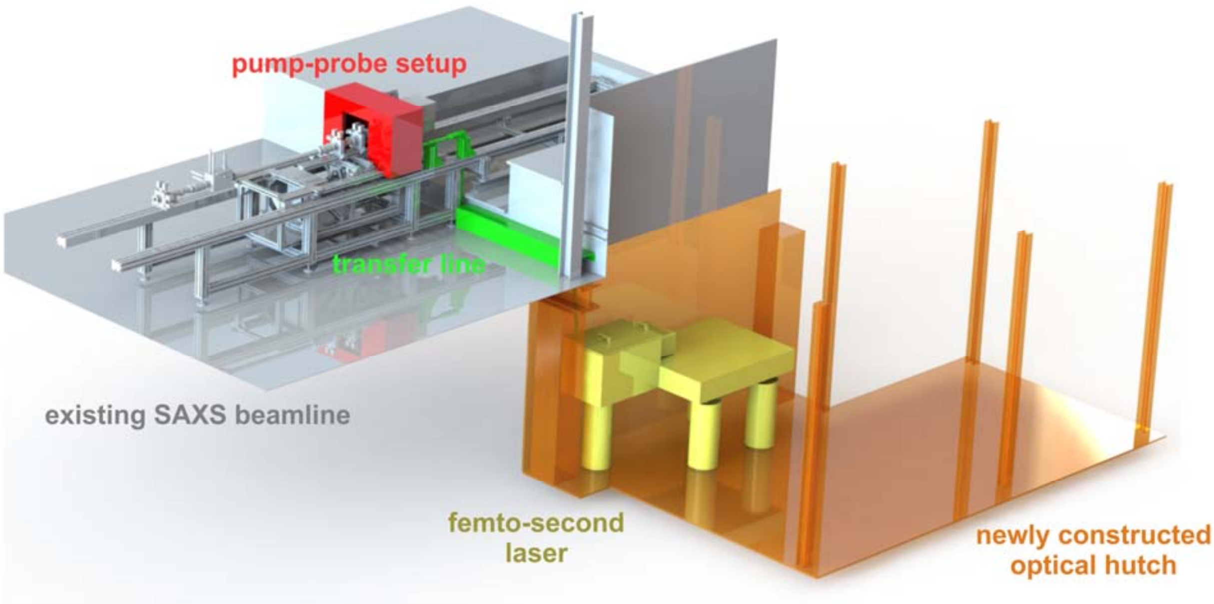

Figure 3: Overview of the beamline modifications and expansions that were made in order to accommodate the acquired femto-second laser (grey items existed prior to NFFA-Europe, coloured items were added/expanded in the framework of this project).

In addtion, all designs and constructions had to meet X-Ray radiation and laser safety standards of the Elettra research facility, which included implementation of a multilevel laser intetrlock-system (see Figure 4). The PLC based interlock-system was designed, implemented and tested in-house with the principle objective to provide the highgest possible safety-standard for users with little to no laser-experience. E.g. open-beam experiments at the X-Ray beamline are not possible by any means, in order to outrule any health hazard of beamline and visiting personel.

Figure 4: Photograph of the laser-head enclosure in the final laser hutch with a detailed view of the custom made PLC laser safety interlock-system.

2.3 Spatial & Temporal Beam Overlap

As the trajectory of the X-Ray beam is not adjustable, the laser beam direction and focal point must be tuned in order to achieve spatial overlap of both beams. We hence constructed a modular optical enclosure that can be placed directly on the beamline-frame. This module contains all necessary optical elements necessary for laser-beam positioning and focusing (see Figure 5), including: a beam switch to control sample exposure to the laser beam, a motorized focusing system, a periscope to port the laser beam above the sample and a two-axis motorized mirror above the sample. Finally, the reflected laser-beam is captured inside a beam dump. For pump-probe experiments on substrates, the sample is positioned on a grazing-incidence (GI) goniometer system that allows independent 3-axis positioning and 2-axis rotation.

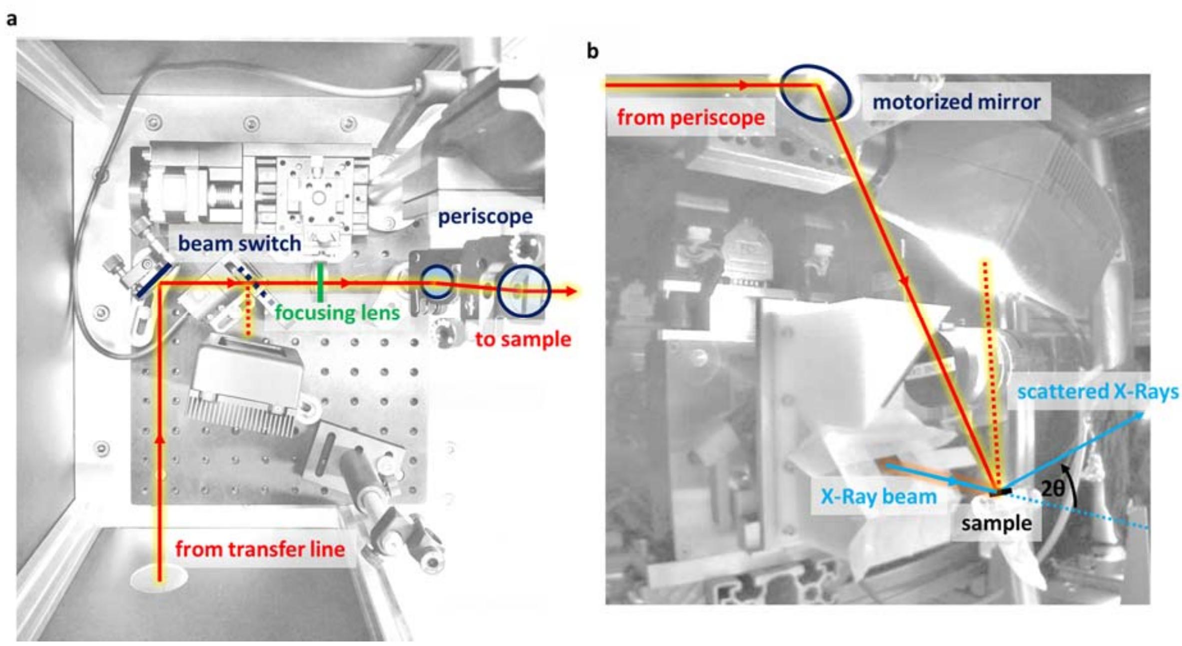

Figure 5: Laser path inside the optical enclosure module installable directly on the beamline frame. a) View of the optical components capturing and focusing the laser beam. b) Direct view of the sample stage, where X-Ray and laser beam are overlapped.

Regarding the temporal overlap, one faces the general issue of locking the device delays to an "absolute" time – an unaltered storage-ring signal that is not subject to any device-specific behaviour. Here, we use the measured ring current (see Figure 1a), in detail the single-pulse peak (see Figure 1b) as reference signal. In a first step, we determine the detector-gate delay (see section 2.1) and lock it to the reference signal. In regard of the laser, we use the diode signal from the laser amplifier as laser specific timing signal. Hence, in the second step (assuming successful spatial overlap of X-Ray and laser beam) the laser delay is scanned over one entire pump-repetition period in order to determine exact temporal overlap. Once this overlap is known, comparison of the ring-current to the laser amplifier signal yields an exact (±0.6 ps) timing basis such that the exact laser delay can be adjust straightforward by simple arithmetic means.

3. Results

The first pilot experiment was performed on an epitaxial grown Ga/Al/InAs superlattice. Here the laser was tuned to the fundamental wavelength of 1030 nm. At this energy, GaAs and AlAs are quasi transparent while InAs has a 1/e penetration length of only 235 nm. Hence, the IR laser pulse is absorbed exclusively in the InAs layers such that they act as immediate phonon sources.

In the pilot experiment, the laser-beam was focused to a spot size of 300 µm (FWHM) at the sample (with approx. 2 mm depth of focus), whereas the X-Ray beam was cut to 200 x 200 µm (H x V – FWHM). Spatial overlap was determined by monitoring the Bragg-peak position of the sample while scanning the motorized laser mirror (see Figure 5). Here, the convoluted beam-width was found to be 525 x 407 µm (H x V – FWHM) – elongated along the X-Ray trajectory due to the sample-incidence angle of approx. 12°. Considering a pulse power of 6µJ at a repetition rate of 1.16 MHz / 3 = 386 kHz, we impose an energy density of approx. 4 mJ/cm^2 on the sample.

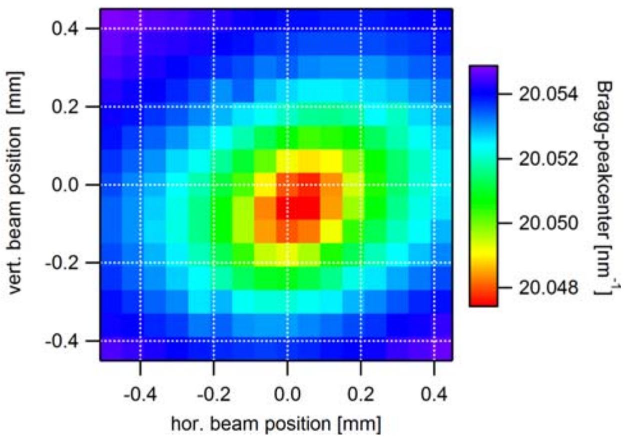

Figure 6: Spatial overlap of X-Ray and laser beam by monitoring the Bragg-peak position of the sample.

A temporal scan over the full pump-probe delay range (3 x 864 = 2592 ns) shows a clear change in the statistical moments of the Bragg reflection (see Fig. 7a). Comparison of single acquisitions 200 ps before and after the pump show a clear broadening and slight suppression of the Bragg peak, a typical observation for heat-induced strain/lattice expansion (see Figure 7b).

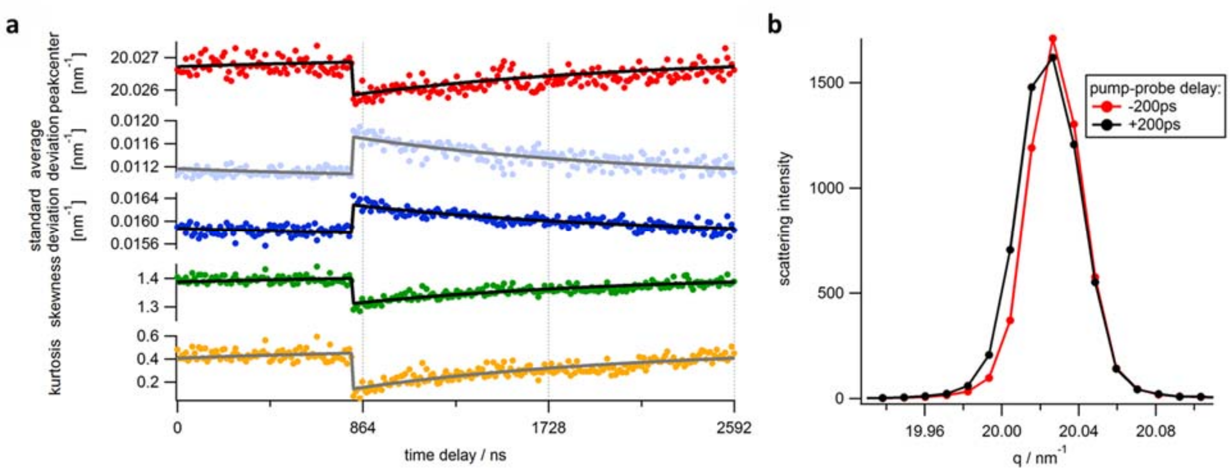

Figure 7: a) Statistical moments calculated from the sample's Bragg reflection over a full pump-probe period. b) Scattering intensity of the Bragg reflection 200 ps before and after the laser pump.

In order to determine the exact temporal resolution of the experiment and hence of the setup, the laser delay was scanned three times with 5 ps resolution over pump-probe edge. An error-function fit of the peak's skewness-parameter (see Figure 8a) yields a temporal X-Ray pulse width of 99 ps (std.dev.), in agreement with the theoretically expected value (93 ps – std.dev.). On a side-note: the total measurement time for all three scans was approx. 70 minutes.

By evaluating the average peak-center as a function of the laser delay, more insight on the sample's thermal response can be gained (see Figure 8b). From the relative peak shift, we estimate a transient temperature increase of approx. 45°C. Furthermore, phonon-propagation simulations of the sample's superlattice structure based on the used laser fluence yield outstanding agreement with the experimental data (see black trace in Figure 8b)

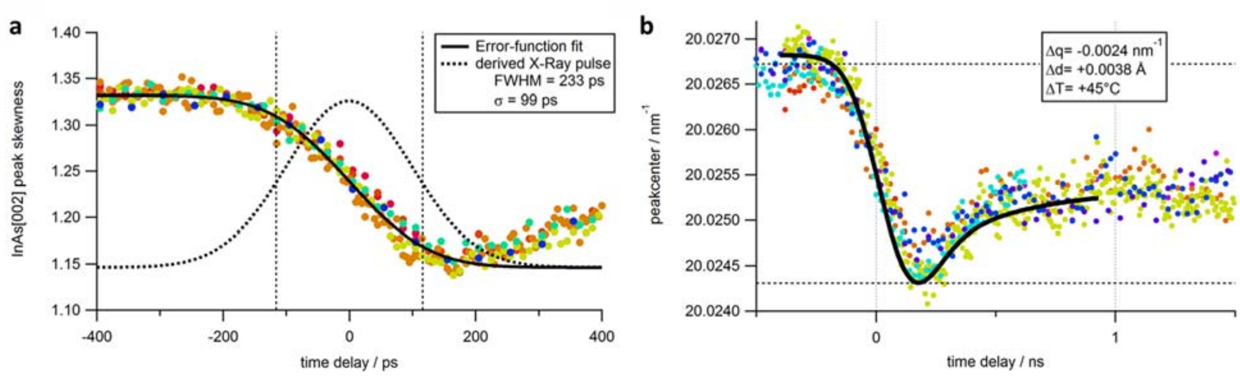

Figure 8: a) Fine-scan over the pump-probe edge to determine the precise time-resolution given by the temporal X-Ray pulse width. b) Experimental results of the transient thermal response of the sample compared to corresponding phonon-propagation simulations (black trace).

4. Conclusions and perspectives

A transient X-Ray scattering, so an optical-pump X-Ray-probe, setup was succesfully implemented at the Austrian SAXS beamline of the Elettra synchrotron. Even though the laser delivery was delayed and therefore fully installed in May/June 2018, the setup has extensively been tested, it is fully operable and it is readily available to NFFA-Europe users. Pilot experiments determine the temporal time-resolution of the beamline at approx. 100 ps (std.dev.), confirming feasibility of the implemented setup to study the transient behaviour of nanostructures. In particular, the flexible layout of the "online" is also compatible laser-transmission geometries, required for liquid samples. Future efforts will be devoted to the installation of a laser-feedback system to accelerate and simplify user operation. In summary, the built workstation offers the unique possibility to conduct picosecond-resolved pump-probe experiments in the hard-X-Ray regime, which allows atomic-scale tracking of the structural response of condensed matter systems. On the long term the up-grade program for ELETTRA 2.0 forseen 2024/2025 will allow provisions for the fast pump probe experiments with resolution times down in the ps-time regime, which will ensure the sustainability of the set-up.