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Abstract
Recent developments have allowed x-ray diffraction techniques at
synchrotron radiation facilities to be employed with a temporal resolution of
around 1 ps. These developments are the availability of firstly, high
brightness third-generation x-ray sources, secondly, passively mode-locked
lasers with 100 fs temporal duration and thirdly, averaging streak cameras
with sub-picosecond temporal resolution. In this paper, we discuss how
these novel devices are combined in time-resolved x-ray diffraction
experiments at synchrotron radiation facilities.

Keywords: time-resolved x-ray diffraction, synchrotron radiation, streak
camera, ultrafast laser pulses, phase transitions

1. Introduction

The use of time-resolved x-ray diffraction for studies of laser-induced ultra-fast phenomena occurring in crystals has been of recent interest [1–6]. The focus has been on studies of ultra-fast disordering of the crystal structure [1–4] and of acoustic phonons [5, 6]. Time-resolved x-ray diffraction has a long history dating back to the 1940s when x-ray tubes were used together with detectors yielding a temporal resolution of milliseconds. Today, we are going towards sub-picosecond temporal resolution. This is due to two developments. Detectors with temporal resolution nine orders of magnitude higher are available. We have access to sources with brightness nine orders of magnitude higher. These dramatic improvements are what allow us to directly follow atomic motion in real time on a time scale of approximately 1 ps. These developments have been made in parallel by separate research communities, resulting in two distinct technologies. These two technologies are firstly laser-based table-top sources and secondly particle accelerators (e.g. storage rings for electrons).

Laser-produced plasmas have, for many years, been used as sources of x-rays. The development of intense laser pulses of short duration has provided short-pulse x-ray radiation with high photon energies. The first hard x-ray source (1.6 keV) for which a temporal resolution of less than 1 ps has been measured was demonstrated by Rischel *et al* [1]. This pump–probe station has sufficiently short pulse duration to directly probe dynamics of structures without relying on detectors to provide the resolution. Since the laser pulse which generates the plasma may be used to photo-activate the sample, synchronization of pump and probe is straightforward.

Synchrotron radiation readily provides pulsed x-ray radiation. However, the pulse duration is relatively long. As a typical example, the pulse duration at the ESRF storage ring in Grenoble is approximately 100 ps. Such pulses are now routinely isolated for pump–probe experiments. In order to study the rapid dynamics of matter, a rapid detector may be employed (such as a streak camera) or new methodologies must be developed (see the contribution by Catravas *et al* in this issue). A description of ultra-fast laser sources can be found in the paper by Rousse *et al* in this issue. In the present paper instrumentation and experiments combining state-of-the-art femtosecond lasers with synchrotron radiation are described, starting with a historical background.

1.1. The historical background to combined laser synchrotron radiation experiments

Already in the 1980s work in which lasers and synchrotrons were used in combination was being done. These measurements were time resolved, but did not take advantage of the pulsed nature of the radiation. The temporal resolution is obtained from the detector whereas the synchrotron radiation is used as continuous wave (CW) background illumination [7].

Pioneering work combining lasers and synchrotron radiation was started at LURE, Paris in the early 1990s [8]. Atomic radiative lifetimes and ionization cross-sections were measured using VUV radiation from the synchrotron in combination with a laser with pulses duration of a few tens of picoseconds. In these experiments the synchrotron was used as a pump to excite atoms to a high-lying state and the laser was used as an ionizing probe.

With the advent of third-generation synchrotron radiation sources in combination with 100 fs lasers utilizing passive mode-locking the concepts of pump–probe experiments could be applied to time-resolved x-ray diffraction with a temporal resolution down to 2 ps. Lasers have been synchronized to storage rings with accuracy better than 5 ps. However, this relatively large jitter does not influence the temporal resolution in the pump–probe experiment.

1.2. Instrumentation

This section describes the instrumentation that is necessary for experiments that use visible radiation to initiate an event and x-ray radiation from an electron storage ring in order to probe the atomic structure of the sample as function of time following the event.

1.2.1. Temporal properties of synchrotron radiation.
The radiation from a synchrotron radiation facility is pulsed. The pulsed nature of the radiation has only recently been exploited. As an example, it can be mentioned that pulses of radiation from the Swedish synchrotron radiation facility MAX II are emitted with a repetition rate of 500 MHz and the pulse duration is 150 ps. The repetition rate is given by the electron bunch structure, which in turn is defined by the radio frequency, which is used to give energy to the electrons in order to compensate for losses due to the synchrotron radiation. The possible fill positions (in the case of MAX II every 2 ns) are called buckets. One can decrease the repetition rate by filling fewer buckets. The limit is given by the repetition rate when only one bucket is filled (the ring-revolution frequency). At a small ring like MAX II, this lowest repetition rate is 3 MHz, whereas at the European Synchrotron Radiation Facility (ESRF) in Grenoble, it is 355 kHz.

1.2.2. The choice of laser.
There are a few factors to consider. As can be seen in the subsection above, x-rays can be obtained at a very high repetition rate. The spot size of the x-rays is given by the quality of the x-ray optics in the beamline. A typical number for a well designed beamline with 1:1 imaging is a focal spot with an area of a few times 10^{-4} cm^2. The next thing to consider is the wavelength range and maximum fluence (energy per unit area) necessary for the experiment. In every pump–probe experiment it is desirable to pump a volume that is large relative to the probed region. The pumped region should have an area of about 10^{-3} cm^2. Also for experiments in which the laser wavelength is not of fundamental importance, one may, in order to ensure overlap in depth inside the target, choose the wavelength of the laser such that a sufficiently high transmission is obtained. In general, the intensity of the pump varies exponentially with the depth and in each experiment one must consider how much variation can be tolerated. Typically commercial pulsed fixed frequency lasers have an average power of 10 W, whereas tunable lasers are often driven by fixed frequency lasers with an efficiency of 10–20%. If a fluence in the range 0.1–1 W cm^{-2} is desired, a repetition rate in these experiments should be approximately 1 kHz. An increase of the repetition rate would not yield a larger number of detected photons on average unless the imaging conditions for the x-rays are changed. The large mismatch of the repetition rates means that only a small fraction (10^{-3}–10^{-6}) of the x-ray photons provide useful information about the time shortly after the pumping of the sample.

1.2.3. Laser synchronization.
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, as described in many electronics textbooks. Locking the laser oscillator to the rf gives the fine tuning. The laser amplifier can then be synchronized to a given sub-harmonic of the ring-revolution frequency.

1.2.4. Changing the timing.
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 taken 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 rf phase-shifter in the laser synchronization control loop. A temporal resolution of a few picoseconds and a range of a few nanoseconds can be achieved.

1.2.5. The choice of detector.
When it comes to choosing the detector, three main concerns should be taken into account, namely the temporal resolution, quantum efficiency and area of the detector. The only type of detector that can be used to obtain temporal resolution in the picosecond regime is a streak camera. Owing to the low number of photons in every x-ray pulse and the low quantum efficiency of the detector, averaging over multiple pulses is necessary. The quantum efficiency depends on the photocathode material, the thickness of the cathode and the x-ray wavelength. Henke et al [9] have described how to calculate quantum efficiencies from relevant parameters. We note that typical numbers are 0.1% around 10 keV and 10% around 100 eV. Generally a streak camera has a slit for an active area. This gives
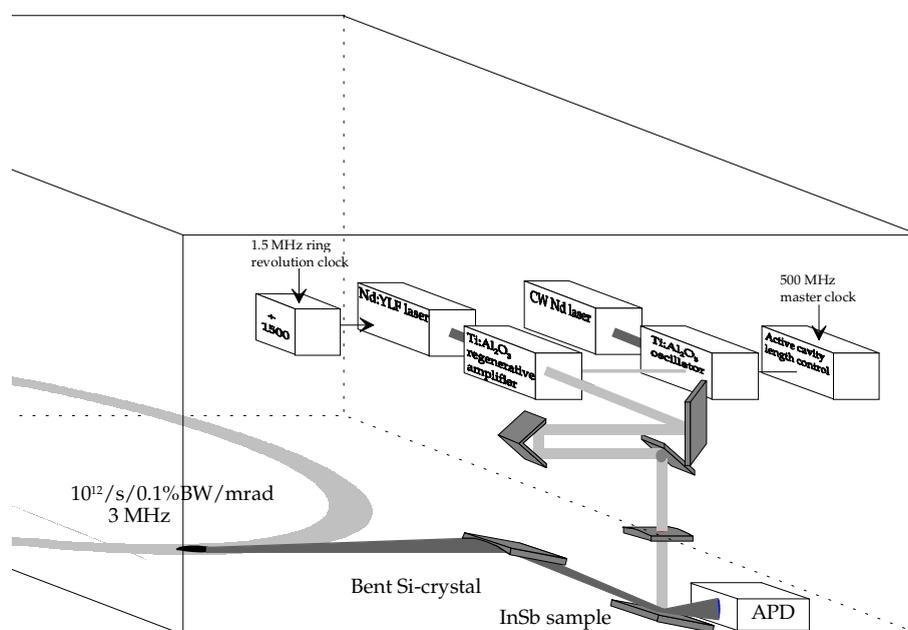
Figure 1. An experimental set-up for x-ray diffraction experiments combining laser and synchrotron radiation.

spatial resolution in one dimension only and only one or a few Bragg spots can be measured. A method which makes it possible to use a streak camera to measure the full temporal history for a two-dimensional image has been described in [10]. This would allow recording thousands of spots in a Laue experiment. The idea would be to remove the slit and use a large photocathode as the active area. As the voltage across the sweep plates of the streak camera is ramped, every Laue spot would be elongated due to the temporal history of that particular spot.

For experiments for which the 100 ps temporal resolution of the synchrotron pulse is adequate, a CCD camera with a near 100% quantum efficiency can be employed. Other detectors with larger areas, such as microchannel plates, can also be considered.

1.2.6. Synchronization of the detector. Just as with the laser, so the synchronization of the detector should be implemented on two different time scales. The obvious synchronization is for experiments in which the detector itself is giving the picosecond temporal resolution. For these experiments the detector should be synchronized to the laser with a higher accuracy than the desired temporal resolution. In order to increase the signal-to-noise ratio, averaging over multiple pulses can be performed. However, in order to use this approach, the temporal jitter associated with triggering the streak camera must be less than the required temporal resolution. This can be achieved by triggering the sweep voltage to the deflection plates of the streak camera with a laser-triggered photoconductive switch [11, 12]. An x-ray streak camera, which is capable of operating at a high (1 kHz) repetition rate with high temporal resolution (500 fs) has been developed at the ESRF. A similar system has been described by Larsson *et al* [13]. The photoconductive switch uses the fact that the conductivity of a semiconductor can be changed rapidly by exciting electrons across the band gap.
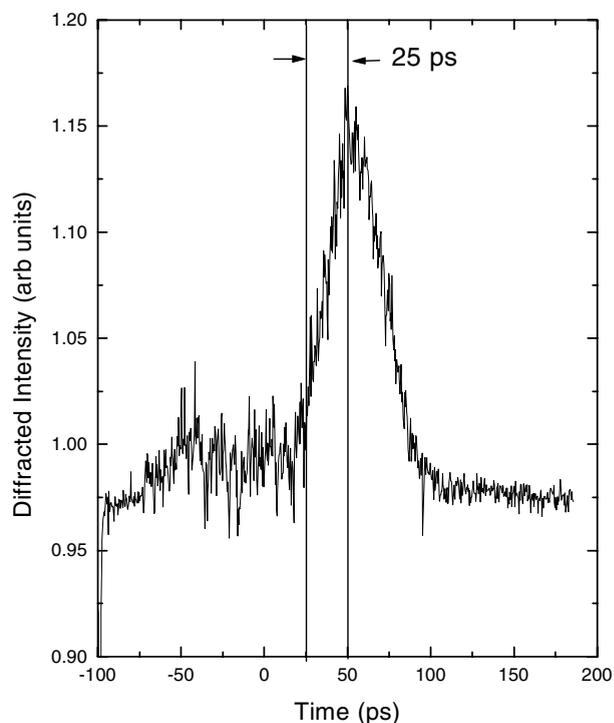
Figure 2. Laser-induced strain causes an increase of the Bragg diffracted intensity on the negative side of the rocking curve.

For the voltages required for a streak camera ($\simeq 1000$ V) approximately 0.1 mJ of energy per pulse is needed in order to achieve a good performance.

An alternative approach is to drive the plates directly with the rf from the laser. This is known as synchroscanning. Synchroscanning is often used for machine diagnostics, but then the driving frequency is derived from the rf cavities. This method has the advantage of not requiring energy from the
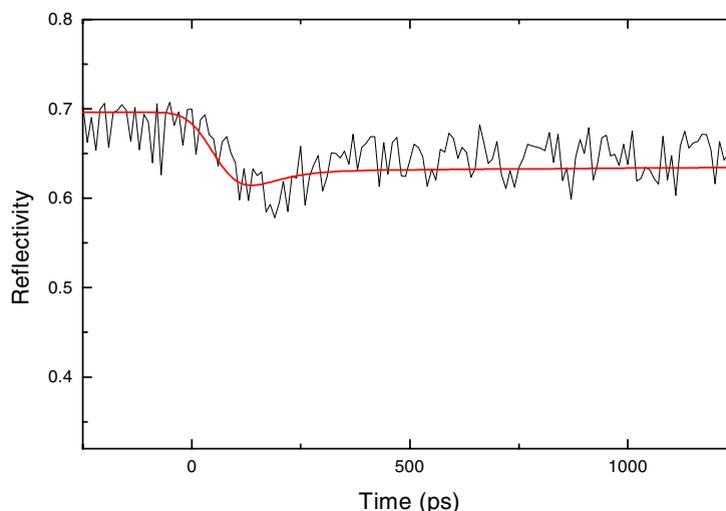
Figure 3. The x-ray reflectivity of a crystal of InSb as a function of time. The crystal is oriented to be on the compression side of the rocking curve. Experimental data recorded by a photodiode and a simulation assuming the strain profile given in [15] are shown.

pump laser to be split off for triggering. This is a concept that would allow work to be performed at higher repetition rates.

There is also a need to gate the detector on a longer timescale to reduce the background. This means that the detector must be gated and synchronized to the laser so that only the x-ray pulses arriving during or shortly following the laser pulse are detected.

Another approach has been taken by Wulff *et al* [14]. A mechanical chopper with an opening time of 280 ns is used to reduce the pulse repetition rate of the x-rays to 900 Hz in order to match the laser repetition rate without the use of electronic gating of the detector.

2. Experiments

We now focus our attention on some of the new experiments that have been made possible with this technology. For example, conformational changes in the nanosecond time domain were studied at the ESRF, using Laue diffraction from a rapidly evolving sample of myoglobin [15]. The time resolution in this experiment was limited by the duration of a single pulse of x-rays from the storage ring. With improved resolution in the range of 1 ps, x-ray diffraction techniques could be applied to the study of ultra-fast processes such as laser-induced phase transitions and transient excited modes in solids.

Since this issue is dedicated to ultra-fast phenomena, we restrict ourselves to work in which the temporal resolution is higher than what is given by the duration of the electron bunch. We particularly discuss recent work involving the detection of strain waves propagating into the material. A set-up for such studies is shown in figure 1.

It has been shown that diffraction experiments provide the means to selectively study the various longitudinal acoustic (LA) phonon modes that make up the strain wave. The mechanism for this is described below. In the absence of coherent phonons, the Bragg condition is fulfilled if the difference of the k-vectors of the incident and diffracted x-ray beams corresponds to a reciprocal lattice vector. However, in

the presence of a phonon, the Bragg condition is fulfilled when the difference of the k-vectors equal the sum of the reciprocal lattice vector and the phonon wavevector. A different way of expressing this is by saying that, when the diffraction efficiency at a given angle close to the Bragg peak of the unperturbed crystal is studied, a range of phonon frequencies is interrogated. The range of phonon frequencies is defined by the angular spread and by the bandwidth of the system. The first direct observation of these coherent phonons was performed by Lindenberg *et al* [6].

Without using an ultra-fast detector, it is possible to implicitly determine whether coherent phonons are excited. The temporal structure of this phenomenon differs from that of pure heating. If a crystal is rotated to an angle at which the reflectivity is within an order of magnitude of that at the peak of the rocking curve and subsequently heated, the reflectivity would go monotonically to the value for the new lattice spacing of the heated expanded crystal. We show experimental curves of the time-resolved reflectivity at a given point of the rocking curve of the crystal. As can be seen in figures 2 and 3, an overshoot or undershoot in the x-ray reflectivity occurs. This is the signature of the shockwave propagating through the wafer. This shockwave can be thought of as being built up from phonon oscillations. In figure 3 the absolute reflectivity is decreased as the lattice spacing is changed due to the propagation strain pulse. The overall features of figure 3 can be explained in the following qualitative way. The crystal has been rotated so that the angle is not at the peak, but on the low angle side of the rocking curve. The shape of the propagating strain is known [16]. As the shockwave enters the crystal there is a thin layer of compressed material, a sharp edge and then expanded material. The thickness of the expanded material will be larger than the amount of compressed material and thus dominate. Hence, the reflectivity will fall rapidly. When the shockwave has passed through the material, the lattice has been heated and thermal expansion will remain until the heat is transported away by diffusion. This occurs on a 100 ns timescale.

The data shown in figure 2 were acquired using a streak camera in order to observe the same phenomenon on a more
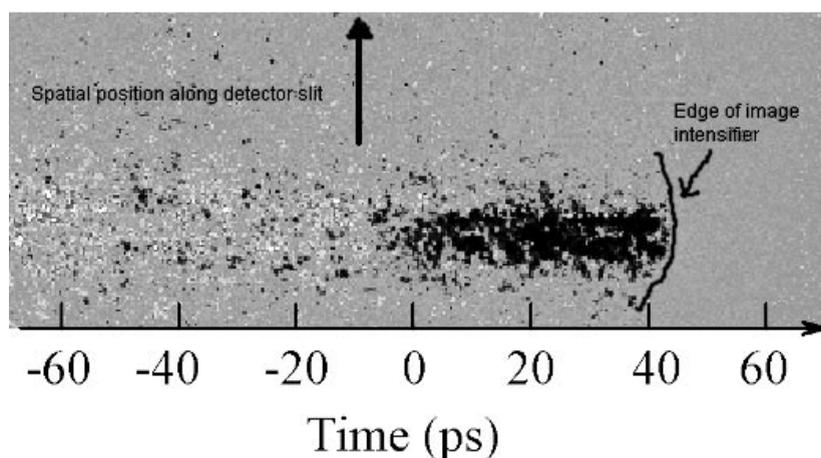
Figure 4. A streak camera image showing a disordering of the structure of InSb.

rapid timescale. The crystal was rotated so that the angle is not at the peak, but on the high angle side of the rocking curve. In this study low frequency modes close to the peak of the rocking curve were studied. From the temporal shape and the rise in reflectivity, shown in figure 2, we have been able to estimate the peak strain in GaAs irradiated by a fluence of $100 \, \text{mJ} \, \text{cm}^{-2}$. This fluence is 20% below the fluence that would cause damage under our experimental conditions. We find the strain to be about 60% of the strain that would be induced by lattice expansion due to a rise in temperature up to the melting temperature.

Another field that has generated some interest is the possibility of looking for phase transformations in semiconductors. The study of the properties of semiconductors following irradiation with ultra-short-pulse lasers has been studied for nearly two decades [17, 18]. One motivation for this work has been the determination of the mechanism of ultra-fast disordering (melting) and other potential phase transitions. Pump–probe techniques have generally been employed. On ultra-fast timescales (<1 ps), these studies have involved time-resolved reflectivity and second-harmonic generation of optical light pulses. Theoretical studies have suggested that excitation of about 10% of the valence electrons in crystals with a diamond structure would give rise to melting even though the lattice temperature remains below the temperature for melting [19]. The signature of non-thermal melting would be the timescale for melting, that should be less than 1 ps, whereas transfer of energy to the lattice via the electrons would require several picoseconds. By probing laser-irradiated material using ultra-fast time-resolved x-ray diffraction, it should be possible to directly determine the mechanism for ultra-fast disordering. However, the lack both of ultra-short-pulse x-ray sources and of ultra-fast x-ray detectors has so far prevented such studies.

It has been the ambition of many groups for a number of years to verify this effect by a clean x-ray experiment. The definitive proof of non-thermal melting was given in a study by Rousse *et al* [20], but indications had been seen in other work [2–4]. Here we discuss laser-induced melting of InSb measured at the ID9 beamline using the ESRF ultra-fast x-ray streak camera.
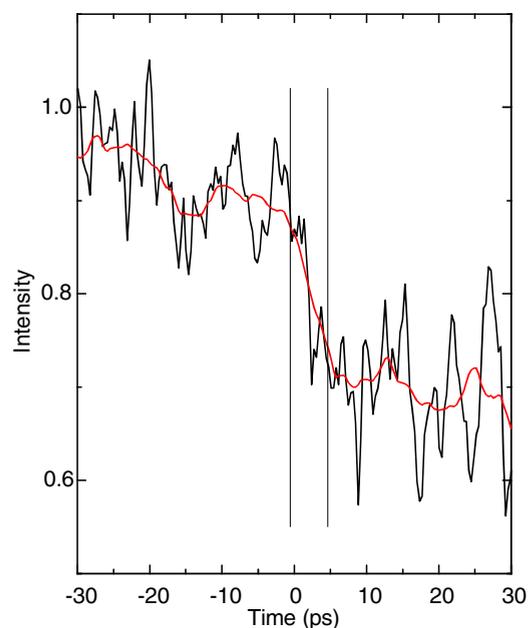
Figure 5. The lineout of figure 4, showing the relative reflectivity as a function of time. The laser interacts with the sample at time $t = 0$. A rapid decrease in intensity of a Bragg spot is found when the top layer is molten.

The results shown here can be used as a straightforward way of producing rapid structures in the time profile of the x-ray radiation. Such rapid structures can be used for the development of ultra-fast detectors and methodology.

In a measurement of the peak reflectivity (at the peak of the rocking curve) of InSb, we observe a rapid drop of the reflectivity. In the raw data this appears to occur in about 1.5 ps. However, there is a difficulty in extracting timescales if the signal-to-noise ratio (SNR) is too low. In order to improve the SNR, the data were smoothed and an upper time limit for the drop in reflectivity was determined to be 6 ps. The physics behind this rapid drop in reflectivity is not well understood, but the drop in reflectivity of 15% is consistent with the absorption that occurs if a molten layer with the depth of the laser penetration depth is formed at the surface. It should be noted that it is not possible to explain the rapid drop in

intensity by invoking the strain/phonon model which provides excellent agreement with experiments for lower fluences. At fluences of about 5 mJ cm^{-2}, coherent phonons are seen. The results shown below were obtained at fluences in the range 30–100 mJ cm^{-2} incident on the sample. The temporal resolution of the streak camera was mainly limited by the static focusing and by the temporal jitter. At the time of the measurement, the temporal jitter was approximately 1 ps and the effect of static focusing was 1.5 ps. This can be seen by observing the signal from laser-generated UV radiation with 100 fs temporal resolution that was measured simultaneously with the x-ray radiation. The image in figure 4 is derived from two images of the phosphor of the streak camera. The image is built up by streaks from approximately 10^6 photons and the noise is associated with noise in the back end of the streak camera. One image was recorded with the laser present and a second was recorded without the laser. The image shown has been obtained by dividing one of the two images by the other pixel by pixel. The intensity across the central part of the image is shown as a line graph in figure 5.

3. Outlook

Over the next few years we expect to see a steady increase in the number of laser systems at synchrotron radiation facilities worldwide. We foresee applications not only in physics but also in chemistry and biology. The x-ray brilliance of available sources has been demonstrated to be sufficient to follow simple photochemical reactions in solution, thereby opening a new window on the role of the solvent in mediating caging and relaxation processes. A number of experiments facilitating the study of the most rapid structural rearrangements within specific biological systems which can be triggered by light will also emerge. The work discussed in this paper should be seen in the context of new and even more powerful x-ray sources. When fourth-generation sources become available, a new generation of pump–probe experiments using x-ray radiation will emerge.

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