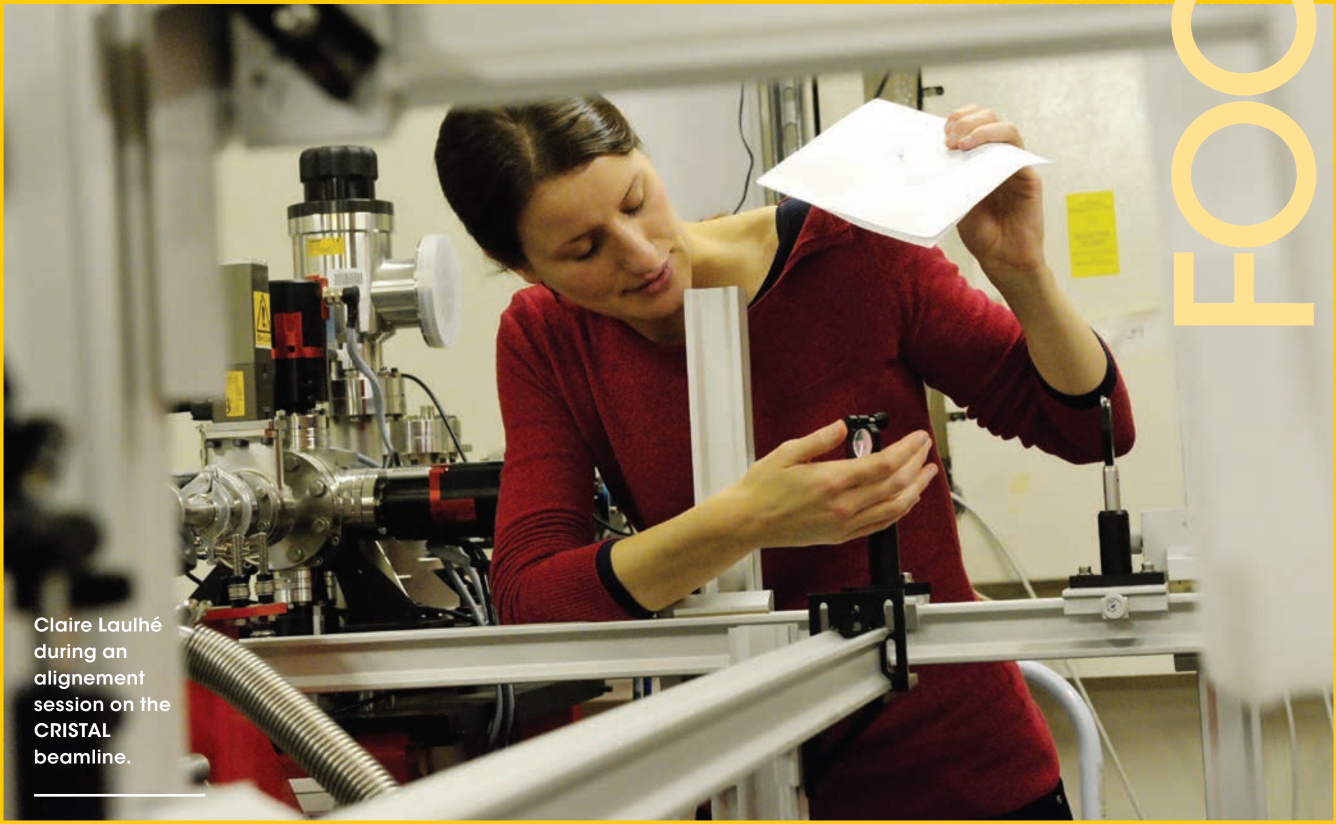
SOLEIL, using light as an instrument



Claire Laulhé during an alignement session on the CRISTAL beamline.

2015 will be the International Year of Light, a topic at the core of the science carried out at SOLEIL, as in the synchrotron facility, light is produced, shaped, and used for all its various properties. Let's take a look at a few examples giving an overview of the research taking place at SOLEIL.

September 29th 2014, scientists are watching for the first time the interaction Terahertz signal of the Slicing project.

Synchrotron radiation begins to have quite a rather long history. It all started in the early 1960s, when it was used as part of the very first spectroscopic measurements, and it continued with a considerable contribution to structural matter determination and the use of light diffraction as a standard method to study ordered matter. Nowadays, synchrotron studies are once again becoming increasingly popular due to a growing need for coherence and temporal structure now available thanks to significant machine improvements on the one hand, and advances in beam control and optics performance on the other.

SOLEIL: a multipurpose light source

As we will be celebrating 2015 as the International Year of Light, it appeared interesting to show how SOLEIL already benefits from these advances within a few select areas such as direct and indirect imaging, ultra-high spectroscopy and time-resolved measurements. Let's start with machines: since it first started operating in 2006, SOLEIL's storage ring has undergone continual upgrades to meet the growing needs and challenges of each beamline. In this sense, SOLEIL can be seen as a multipurpose light source. Users can chose among several filling pattern options provided in Top-up injection,

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NANOSCOPIUM

Nanofocusing at NANOSCOPIUM

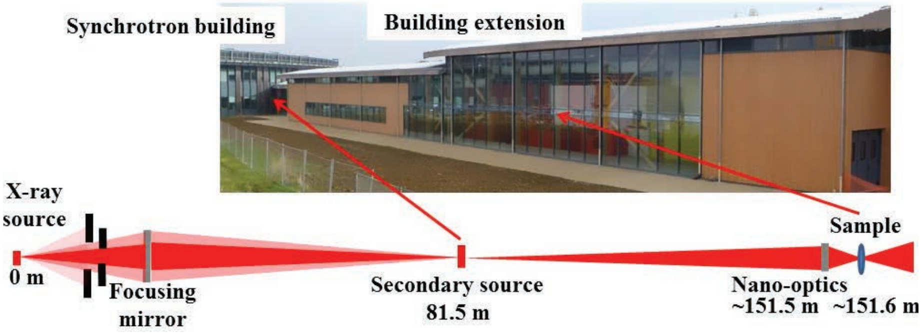


Figure 1. Optical scheme of nano-focalization at NANOSCOPIUM. The demagnification is determined by the ratio of the SS-nano-optic and nano-optic-sample distances.

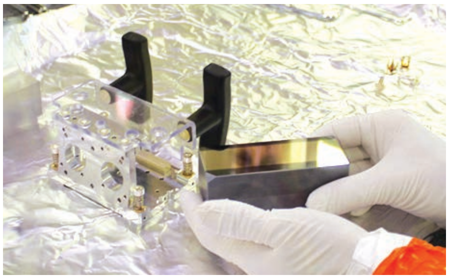


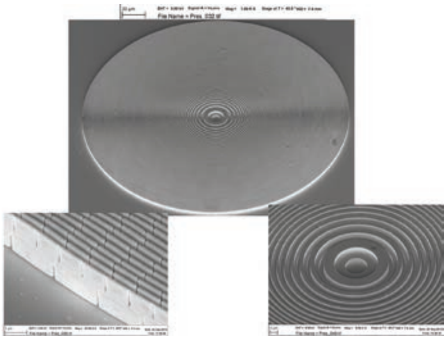
Figure 2a (top). Ultra-high quality nanofocusing (Kirkpatrick-Baez, KB) mirror. Figure 2b (low). Nanofocusing Fresnel Zone plate[2].

High societal impact research areas, such as biology, earth- and environmental sciences or bio-nanotechnology are seeking information down to nanometers scale on highly heterogeneous systems often in natural/in operando conditions. Scanning hard X-ray nano-imaging is getting increasing interest in such research providing morphological and elemental information together with high penetrating power particularly well suited for *in situ* studies. Recently X-ray microscopy witnessed tremendous development driven by this strong scientific interest.

In scanning X-ray imaging the sample is scanned in the intensive nano-beam created by a high quality X-ray optic. Next to the geometrical demagnification of the X-ray source, a physical limitation of beam focusing is due to the diffraction of X-rays. This "diffraction limited" beam-size, which is proportional to the X-ray wavelength, determines the ultimate spatial resolution of the optic. This is below of the diffraction limited resolution of visible light microscopes.

Fabrication of ultra-high quality optics is crucial for nano-focusing and comes with methodological and fabrication challenges. These are successfully tackled by latest nanofabrication technologies. Recent X-ray focusing devices, such as Fresnel Zone plates consisting of hundreds of concentric rings with decreases widths, or ultra-precisely figured elliptical mirror-pairs, or new class of x-ray optics as multilayer Laue lenses (MLL), provide near-diffraction-limited focusing. As such modern x-ray microscopy reaches a few tens of nanometers resolution and most recent results report even sub-10 nm focusing opening unprecedented research possibilities.

The NANOSCOPIUM[1] 155 m-long beamline is dedicated to 2D/3D scanning hard X-ray nano-imaging aiming to reach down to 30 nm spatial resolution. The large 60-70 m distance of the state of the art nano-focusing optics from the secondary source (SS) ensures high ~0.0015 demagnification (Fig. 1) necessary for obtaining nano-sized beams (i.e. 10 μm SS size is demagnified to 10 nm at the sample position).
The outstanding optical quality of the elliptically figured mirror-pair (Fig. 2a) (JTEC, Japan) is crucial for focusing down to 50-100 nm. The measured elliptical mirror shape agrees within ≤0.5 nm, almost atomic scale!, over the 100 mm mirror length with the theoretical shape. Beam-sizes down to 30 nm will be obtained by dedicated FZP's (Fig. 2b) developed by the group of C. David[2] (PSI, Villigen, Switzerland).



With these nano-optics NANOSCOPIUM will offer state of the art imaging with stable and high intensity nano-beams, which can be tailored in the 30-500 nm size-range according to the experimental needs. Moreover, fast continuous scanning[3] makes multi-scale imaging and tomography feasible during a typical user experiment. This, together with the combination of complementary techniques (X-ray fluorescence, absorption, phase contrast and dark field) provides complete quantitative information about the sample structure, composition and chemistry for cutting edge sciences.

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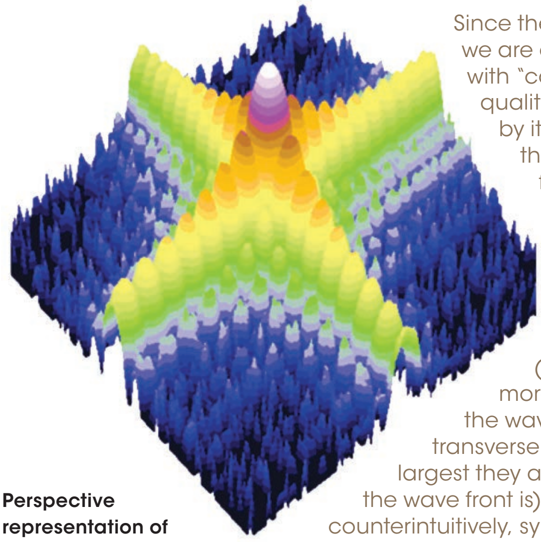
References:
[1] **A. Somogyi et al.**
Instruments and Methods, 885104 (2013).
[2] **I. Mohacsi et al.**
Journal of Synchrotron Radiation, 21, 497–5 (2014).
[3] **K. Medjoubi et al.**
Journal of Synchrotron Radiation, 20(2), 293 (2013).

CRISTAL

Extreme coherence



Perspective representation of the X-ray coherent diffraction pattern obtained with 5x5 µm² slits at the CRISTAL beamline. Intensities are in log scale. The existence of fringes demonstrates the coherence of the beam.

Since the advent of the laser, we are accustomed to deal with "coherent light", a high quality light characterized by its spectral purity and the regularity of its wave front (planar or spherical). Physicists characterize these properties by two lengths, the longitudinal coherence length (the largest it is, the more "monochromatic" the wave is), and the two transverse coherence ones (the largest they are, the more perfect the wave front is). Rather counterintuitively, synchrotron light is not so coherent, especially in the X-ray range. Let's see why.

3rd generation sources are based on undulators, in which bunches of billions of electrons (a few cm long, *i.e.* a few tens of picosecond at the speed of light) undulate a hundred times. One electron of this bunch generates a nice plane wave of a hundred of oscillations (which is ~100 nm in the hard X-ray regime). Because each electron emits light **independently**, synchrotron light consists in a pulse of a few tens of picosecond, with a longitudinal coherence length of ~100 nm. The transverse lengths depend on the transverse sizes of the bunch: the smaller it is, the larger they are. In practical they are about a few tens of microns. Because the beam size is larger than that (~100 µm), the beam is **partially coherent**. So, how to use the coherence properties of such a beam?

The first trick is to use a monochromator, which increases the spectral quality of the beam, and thus the longitudinal coherence length, up to a micron. The second one is to place an aperture of about 10x10 µm² close to the sample in order to select the **coherent part** of the beam. This way a coherent beam is obtained, as shown by the diffraction pattern measured after the aperture, made of a pair of slits. Unfortunately, this is done at the expense of the intensity: a factor 10 000 is lost in this process. Still, it allows one to perform diffraction, that we call "coherent diffraction". On CRISTAL, we have developped coherent diffraction to study nanocrystals or multiphase compounds.

How to get more coherence? The first idea is to decrease the transverse size of the bunch by decreasing the so-called emittance of the electron beam till its minimum: the **diffraction limit**. This will be done in some synchrotron light centers, which will then become "Diffraction Limited Synchrotron Rings" (DLSR). The second way is to increase the size of the undulator in order to reach the self-amplified stimulated emission (SASE), in which the electrons of the bunch emit coherently! This is the principle of the **X-ray free electron lasers**, which have started to work ten years ago, but are not yet operating in these nominal conditions. Extreme coherence is knocking at the door!

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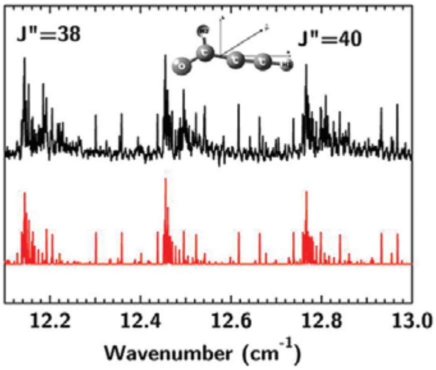
designed to meet their expectations in terms of high brilliance, time structure and short experiments. The beamline capacity has also increased by using canted undulators in single straight sections. Through the high reliability and stability of its photon beam, the SOLEIL synchrotron allows for versatile modes of operation that are tailored to meet the needs of each user. One of SOLEIL's unique features is also its ability to accept a multibunch current up to 500 mA and store more than 20 mA in a single bunch for time-resolved experiments. In addition to the mode where every single possible bunch (416 in total) is filled, the operating modes of SOLEIL's storage ring allow for a strong emphasis on the temporal structure exploitation. The hybrid mode consists of a series of 312 bunches of electrons with a total current reaching 425 mA over three quarters of the ring circumference, supplemented by an additional bunch with 5 mA in the middle of the last quarter. Two weeks per year are devoted to the so-called «eight-bunch mode» with a 90 mA total current, each separated by 148 ns and with a 25 ps RMS bunch length, while another two weeks are dedicated to the single-bunch mode with 16 mA current and 1.18 µs time interval.

AILES

High-resolution THz spectroscopy with coherent synchrotron radiation at SOLEIL

A new operation mode called Low-Alpha is now fully functional and available to SOLEIL's users. In this mode, the electrons emitting synchrotron radiation are densely packed in very short bunches, which allows for significant advances on beamlines performing time-resolved experiments on fast-changing phenomena. It has also been used by researchers from the AILES beamline to identify the spectral signatures of molecules in the area that lies between microwave and infrared radiation. In fact, when the wavelengths of the emitted light are similar to those of the bunches of electrons circulating in the synchrotron, this results in a significant increase in light flux. Basically, for «long» bunches in normal operating mode, each electron behaves as an independent light source and the light intensity is proportional to the number of electrons in each bunch. With short bunches, of the same order of magnitude as the emitted wavelengths, the electrons are very close to one another and their waves are in phase. In such a configuration, the light intensity of this so-called «coherent» emission is proportional to the square of the number of electrons in each bunch, resulting in a 10,000-fold gain in emission. Such phenomenon can occur for wavelengths from about 0.3 to 1 mm (250 to 750 GHz). What makes this energy range so special is that corresponds to the rotational energy levels of molecules in the gas phase, as surveyed by recent modern astronomy instruments such as Herschel and ALMA-recent projects that are seeking reference data issued by independent laboratories.

By juggling with the instruments' parameters, physicists at SOLEIL's Machine Group were able to improve the intrinsic instability of this functional mode, and through enhanced probing devices compensating for the source fluctuations, the researchers recorded the pure rotational spectrum of an organic molecule already detected in certain interstellar clouds, within a few hours of measurements over the entire range of interest. The analysis of such data provides improvements in the quantification of molecules found in very remote mediums, and this spectral range allows for the identification of many long-unsuspected organic molecules present in the interstellar medium. Each molecule has a spectral signature consisting of thousands of absorption lines, and even for molecules that have already been identified, it is still necessary to analyze their spectrum as such work could lead to the identification of new elements emerging in the middle of such a dense forest.



Comparison of the pure rotational spectrum of the propynal molecule (top insert in black) as obtained in experiments, with the simulations based on these results (in red): the molecule's true «fingerprint» used for its identification in radio astronomy.

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References:
[1] **J. Barros et al.**
Rev. Sci. Inst.84, 033102, (2013).
[2] **J. Barros et al.**
J. Mol. Spectrosc. submitted (2014).

Promising imaging techniques

State-of-the-art optics on the NANOSCOPIUM beamline allow for 2D/3D nm imaging, with a significant impact expected in biology as well as in Earth and environmental sciences, a field overflowing with heterogeneous systems, the top candidates for this type of technique. Since January 2012, the storage ring has been operating with new optics including a quadrupole triplet that creates a double low vertical beta function in one of the long straight sections, and a four-magnet chicane to accommodate two canted in-vacuum undulators with a 6.5 mrad separation angle, producing two separate beams of light. The two 5.5 mm-gap undulators radiate independently hard X-rays in NANOSCOPIUM and ANATOMIX, the two long beamlines (180 m) to be used for contrast imaging and coherent diffraction.

As coherent diffraction is extensively used on the CRISTAL beamline, it is now possible to study nanocrystals and multiphase compounds (see page 18), while the ptychography technique allows users to probe larger samples. Bragg coherent diffraction patterns are expected to have a significant impact in the field of material science as it

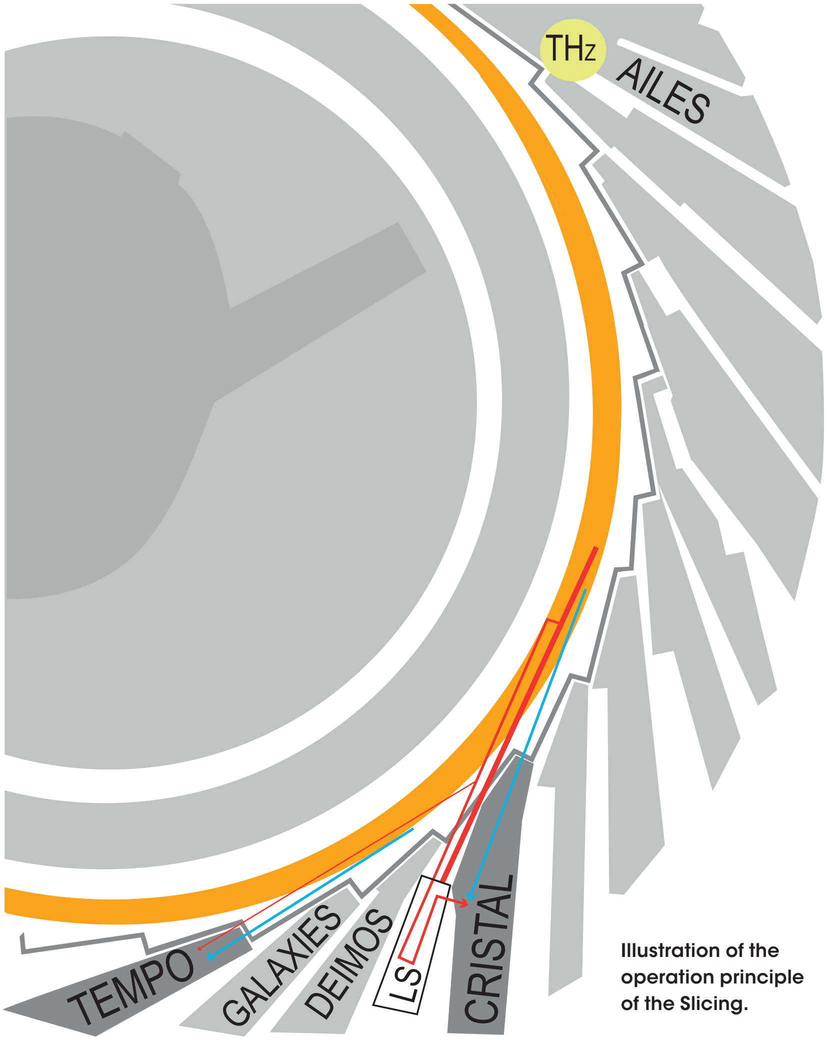
To be continued on page 20...

Generating ultra-short X-ray pulses with femtosecond visible light pulses

On September 29, 2014 the first successful observation of femtosecond slicing of an electron bunch in the storage ring of SOLEIL was made. This opens up the door for studies of ultrafast structural and electronic phenomena at SOLEIL. The femtoslicing technique relies on the interaction between a femtosecond short infrared laser pulse and one of the electron bunches circulating in the storage ring*

**See also Rayon de SOLEIL 20, p11-12.*

Figure 1. In the control room, Marie Labat and Marie-Agnès Tordeux are watching the interaction signal between the laser and the electron beam.



Laser beam and Laser System (LS) 🟥
Sliced X-ray beam 🟦
Electron beam in the storage ring 🟧
Terahertz signal 🟨

Illustration of the operation principle of the Slicing.

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allows for local constraint determination (stress, defaults etc.).

This coherence property is used quite differently on the AILES beamline, to achieve ultra-high resolution in the THz regime by means of CSR (coherent synchrotron emission) obtained when the electron beam size is of the same order of magnitude as the wavelength of the emitted photons. This has led to spectroscopic studies to obtain the spectral signature of molecules of astrophysical interest. These experiments on the AILES beamline are based on the so-called "low-alpha" mode that produces few-picosecond-long electron bunches and is also available two weeks per year (see page 19).

Probing ever-faster processes

The next new mode of operation developed at SOLEIL will be femto-slicing capabilities to several beamlines. X-ray pulses of a hundred femtoseconds will be rolled out first on the CRISTAL and TEMPO beamlines, possibly followed by DEIMOS and GALAXIES. Through these very short pulses, users will be able to probe ultra-fast dynamics (electronic and structural) processes induced in excited matter. Although the photon flux thus obtained

The femtoslicing facility at SOLEIL will be world-wide the first one providing femtosecond short X-ray pulses to several beamlines. The entire X-ray photon energy range will thus be covered at a single femtoslicing facility and femtosecond time resolution will be added to a variety of powerful X-ray techniques. Within the initial phase of the femtoslicing facility, experiments will take place at the CRISTAL and TEMPO beamlines (see figure 1). A future upgrade could provide femtosecond short X-ray pulses also at the DEIMOS and GALAXIES beamlines.

The requirement for spatial and temporal stability are stringent: scaling shows that it corresponds to pointing a laser from Paris at the Statue of Liberty in the harbour of New York – and keeping it there over the period of days, which is the typical duration of a femtoslicing experiment. To meet these requirements, significant contributions have been necessary from all of SOLEIL's divisions. To name a few of these, the laser itself had to be synchronized to SOLEIL's master RF clock; a vacuum compatible tubing had to be put in place along the transport path of the IR laser to avoid that air temperature variations or turbulences affect the laser pulse's pointing or arrival time; diagnostics had to be developed to characterize the IR beam's position, profile, energy, divergence and duration along the transport path…

All these efforts together have made it possible to reach the important milestone of observing for the first time the THz signal (see figure 1), which indicates that the electron beam and laser are spatially and temporally overlapping. Since then, a set of parameters has been optimized to further improve the overlap and to preserve it over an extended period.

The next step will be the detection of the femtosecond short X-ray pulses at the CRISTAL beamline. In preparation for the then to follow first femtoslicing experiments, scientists of the CRISTAL and TEMPO beamlines have already performed time resolved IR pump – X-ray probe experiments exploiting the currently available temporal resolution of 10 to a few tens of picoseconds. The stimulation of such out-of-equilibrium states by irradiation with ultra-short laser pulses induces electronic transitions on a timescale at which the lattice is considered to be frozen. Such excited states are inherently dynamic, involving changes over a wide range of length and time scales. Considering atomic structure, the fastest dynamics take the form of atomic vibrations with a period of the order of 100 fs. As an example figure 3 shows the time evolution of the intensity of a Bragg peak of 1T-TaS_2 during a laser driven phase transition. Femtosecond time resolution is clearly needed to get more information on the initial drop of intensity (green region).This phase transition of 1T-TaS_2 thus gives a good example why a femtosecond time resolution is needed.

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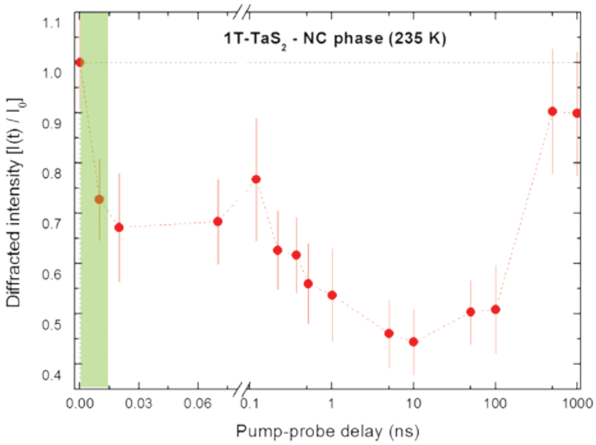

Figure 2. Intensity of a Bragg reflection as function of time after laser excitation. The intensity decrease is a direct signature of a structural transformation of the material. The first drop of intensity happens within 10 ps (region highlighted in green), which is the X-ray pulse length given temporal resolution of this measurement

does not allow for single-bunch experiments (as done on free electron lasers FEL), the unique stability of the beams is ideal for the study of reversible (repetitive) processes such as light-induced phase transition in solids and magnetization dynamics in magnetic compounds for enhanced data storage.

As is the case for other synchrotron research centers throughout the world, SOLEIL is considering upgrading to a Diffraction Limited Storage Ring (DLSR). Such an installation would include electron beam emittance near diffraction limit of the X-ray radiation, much larger coherent flux of photons and spectral brilliance 10 to 100 times greater than current third-generation synchrotron light sources. All things considered, synchrotron technology will indeed continue to serve increasingly demanding research for many years to come.