Timing modes at the MAX IV storage rings

Background

The design of the MAX IV storage rings aims to achieve ultralow emittance at low cost. The MAX IV laboratory will provide a 3 GeV storage ring with an emittance down to 0.2 nmrad for hard x rays and a 1.5 GeV storage ring for soft x rays. The laboratory has promised to deliver this source during the spring of 2016. The design specifies uniform bunch filling patterns in the storage rings, and employs passive harmonic cavities to damp beam instabilities and lengthen the bunches.

The FEMTOMAX short-pulse beam line will deliver a 100 fs x-ray pulse of spontaneous radiation from an undulator or wiggler with a100 Hz repetition frequency. Other timing experiments have so far not been considered for MAX IV. A variety of techniques for pump-probe measurements, for multi-particle coincidence measurements or for time-resolved detection are routinely used in laser laboratories, at free-electron lasers or at synchrotron light sources which offer users storage-ring based timing modes that provide x-ray pulses with up to 800 ns pulse-to-pulse intervals.

A workshop was held in March, 2014 in order to investigate the interest of the user community for timing experiments at MAX IV. The workshop was informal and was not announced to the broader community but it attracted 50 scientists. The workshop generated a set of user-oriented beam parameters, and initiated a dialogue between users and accelerator physicists. Based on this dialog we have put together a brief scientific case, a catalog of users who would carry out experiments using timing modes which are not possible at MAX IV within the present design.

I. Brief scientific case for timing modes at MAX IV

Time-resolved laser pump/X-ray probe techniques have proven to be versatile tools for investigating ultrafast dynamics in gases, solids and liquid solutions. The time resolution is generally limited by the x-ray pulse duration, which at synchrotrons ranges from 50-200 ps, and for lasers is usually on the 10 femtosecond level.

Ultrafast spectroscopy provides detailed insights on the mechanisms of chemical reactions. In a pump–probe experiment the pump pulse triggers a chemical reaction and the probe pulse monitors the ongoing reaction at a fixed time delay. By changing the time delay between pump and probe pulses, the development of the photoinduced reaction is mapped out. A complete description of chemical dynamics also requires spatial resolution on the atomic scale. Since many fundamental reactions in chemistry and biology take place in a solvent environment, structural techniques which can be implemented in a variety of sample environments are desirable.

Temporal resolution in the picosecond/femtosecond domain can be obtained with atomic-scale spatial resolution using ultrafast time-resolved x-ray absorption spectroscopy (XAS**)** . Here the probe pulse consists of an x-ray pulse, while the reaction is triggered by a laser pulse. The x-ray probe yields information on the oxidation state of an atom of interest and on the unoccupied valence states via x-ray

absorption near-edge structure spectroscopy. Information on the local molecular structure around the probed atom is obtained via extended x-ray absorption fine structure **(EXAFS).**

Protein conformational changes are believed to participate in the function and regulation of photosynthetic reaction centers. X-ray diffraction studies of crystals of the Rhodobacter sphaeroides reaction center (RCsph) at room temperature showed substantial movement of QB upon its reduction. A concerted movement of the H subunit was also observed in crystals of the same reaction center after prolonged illumination. Time-resolved Laue diffraction provides a direct approach for observing protein conformational changes in real time at room temperature. This method has captured light induced electron density changes in myoglobin: carbon monoxide complexes and photoactive yellow protein. Time-resolved Laue diffraction can be used to probe light-driven conformational changes in such crystals.

Research on luminescence processes in wide-gap semiconductors benefits uniquely from the ability to detect fluorescence in a time-resolved fashion. When a material contains multiple substances (luminescence centers), their fluorescence spectrum may contain contributions from several centers, and from several different types of processes. An effective approach in such cases is to observe the light emission dynamics by carrying out fluorescence lifetime measurements. The time required for the substance excited by the pulsed light to return to its ground state is measured in the *sub-nanosecond* to *millisecond* region.

Fundamental research on atoms and on ions is important for understanding a variety of processes in plasmas, in astrophysics and in industrial processes. Studying the electronic properties of positively charged ions is difficult using traditional methods where ion beams or trapped ions are photoionized due to the low sample densities. An alternative method using synchrotron radiation for single-photon multi-ionisation of a neutral atom, but detecting all of the electrons using multi-coincidence techniques provides the same information on a sample which is easily handled. This method has two advantages: the huge intensity generated and the accurate configuration of the initial states of ions which can be easily calculated.

In molecules the multicoincident detection of several ion fragments has been used with multibunch synchrotron radiation for many years for photoionization where a fast electron is utilized as a start signal for time-of-flight measurements. Knowledge of the molecular orbital where the electron originates provides detailed information on the localization, the character and even on the coherence of the ionization process that correlates to the fragmentation process. There are some methods that involve fast keV extraction pulses that can provide an independent timing signal, but these reduce the resolution and efficiency of the spectrometers severely and prohibit the use of imaging spectrometers.

Since the nuclear dynamics of many-body systems such as molecules or clusters can be mapped more or less directly onto the electronic final state this is an attractive research. Pulses synchrotrons are much longer than the typical times of most molecular vibrations. Isomerization processes or excitation of high-lying vibrational states can be induced by a laser pulse and then be probed with a synchrotron pulse (fragmentation plus momentum spectroscopy): By doing so, the rearrangement

processes respectively the inner nuclear dynamics can be probed time-resolved as a function of the laser-synchrotron-delay. Combining pump and probe pulses with momentum spectroscopy represents the ultimate tool to unveil the inner dynamics of atoms and molecules. Even with single-photon ionization this combination represents the softest way to see dynamics in atoms and molecules but with laser synchronization the time coordinates can be obtained. These multi coincidence experiments are not simple (high flux, stability and full control for the two different pulses are needed) but highly desirable. They will play an important role in future investigations of many particle dynamics and time resolved spectroscopy of chemical reactions.

II. User case:

While there is no established timing user community at MAX Lab today a large number of Nordic scientists are doing experiments at other facilities that provide timing pulses. For soft x ray spectroscopy, imaging and coincidences BESSY and SOLEIL provide a variety of timing modes for users. The ESRF and the APS have several beam lines where pulsed lasers are synchronized to the pulse structure of the storage ring for time-resolved diffraction and EXAFS studies.

Another part of the community has been using pump-probe methods using a variety of different techniques both at synchrotron radiation laboratories and at laser laboratories (mainly where high harmonic generation sources are available in the XUV range). Several users are presently utilizing free-electron lasers but the difficulty in obtaining beam time and the need for more thorough experimental investigations makes storage-ring studies using either laser activation or gated detectors not only attractive, but necessary in order to obtain a complete picture of dynamical processes in large molecules.

Without providing a detailed description of the scientific scope of the users it is now clear that there is interest from the atomic and molecular physics community to study fundamental processes in multiply-charged systems, from physical chemists working on energy storage and transformation materials, from structural biologists who investigate the coupling between structure and light-induced function, materials scientists who study magnetic transitions in two-dimensional systems and a variety of other areas. The primary storage ring needs cover both the VUV, soft x-ray and hard x-ray ranges.

Most of the applications do not specify exact energy ranges or polarization needs, but many of the imaging methods (molecular spectroscopy) and methods requiring either a mechanical or optical shutter request a very small spot size for technical reasons. Several of the imaging methods rely upon a small source in order to obtain optimal resolution.

Criteria for timing experiments specified by the user community:

1. Coincidence and time-of-flight experiments require a 10-100 kHz x-ray pulse at the experiment. Pulse durations of up to 500 ps are acceptable.

RMS bunch length (ps)	Up to 500
Nr of photons/s (within 1% ΔE/E)	10^7
Rep rate at experiment	3 MHz
Interval between neighboring pulses (ns)	±150 or more for synchronization

2. Short x-ray pulse durations (5-50 ps) for electron time-of-flight measurements using the ARTOF electron spectrometer. Requires a trigger from an external source.

RMS bunch length (ps)	10 (optimal) – 200 (OK)
Nr of photons/s (within 1% ΔE/E)	10^7
Rep rate MHz	0.5-3 MHz
Interval between neighboring pulses (ns)	100

3. Pump-probe schemes requiring synchronization and longer time intervals, and adjustable pulse lengths. The time resolution is generally determined by the photon pulse duration.

RMS bunch length (ps)	100
Nr of photons/s (within 1% ΔE/E)	10^{14}
Rep rate MHz	Up to 6 MHz
Interval between neighboring pulses (ns)	±130 for synchronization

4. VUV-XUV pulses with intervals of 100-1000 ns for luminescence experiments on wide-gap crystals. Longer intervals (up to milliseconds) might be interesting if the pulse intensity is large enough.

There are a number of parameters that vary from experiment to experiment and it is hard to specify precisely what the optimal or minimal parameters for pulse duration, repetition rate, photons per pulse, photon energy, focus, etc. It is clear that the majority of timing users are interested in either short pulses or in longer time intervals between pulses with 200-500 ps durations. If an accelerator solution could be found that can deliver single pulses with 10-100 microsecond intervals at specific beam lines while other beam lines on the same storage ring receive essentially multibunch flux that this would be ideal.

A list of participants in the two workshops which were held in spring and fall 2014 is included in Appendix 1.

III. International status of the field

There are a large number of published articles using timing modes at different storage rings. For the purpose of this application there are a few cases that are worth mentioning.

There are two current methods that allow nearly multibunch operation while providing a single-bunch pulse for a particular beam line. Both of these have been tested at other sources for a cam-shaft filling mode.

At BESSY ‘resonant pulse picking’ has been demonstrated to deliver a photon flux of up to 10^7–10^9 ph s_1/0.1%BW at purity values of 10^4–10^2 at a repetition rate of 1.25 MHz. The quasi-resonant excitation of incoherent betatron oscillations of electrons allows horizontal pulse separation at variable polarization accessible for both regular 30 ps pulses and ultrashort pulses of 2–3 ps duration. The ring was operated in a cam-shaft mode (four cam-shaft bunches) in an ion clearing gap of 200 ns. The remainder of the ring is filled with a multibunch train.

Pseudo single bunch has been demonstrated in a hybrid-filling mode, and a single bunch is ‘kicked’ onto a different closed orbit than the multibunch train. This is achieved by one or several fast kicker magnet to coherently excite one bunch, which then follows a different orbit around the ring.
A high-repetition-rate (MHz), short-pulse (<100 ns) magnet vertically kicks a cam-shaft bunch relative to the bunch train. A collimator or aperture at the beam line where the electron bunch is displaced (bumped) stops light from the normal bunches. The time spacing can (theoretically) be tuned from milliseconds to microseconds. The system has been developed and tested at the ALS and at SOLEIL.

The standard operating mode at BESSY is **Multi Bunch Hybrid Mode** where 350 RF buckets out of 400 available buckets are filled with electrons (up to 0.9 mA per electron bunch, 30 ps bunch length). In the gap a single bunch (10 mA) is injected for pump-probe experiments using the fs-slicing facility.

Multi Bunch 3+1 Hybrid Mode for femtosecond slicing operation is based upon the multibunch hybrid mode but three additional pulses, separated by 12 ns are included in the multibunch train. A total of 280 mA is stored in the multi bunch train, the single bunches carry a maximum of 5 mA each.
Single-bunch operation at BESSY provides a single bunch with 20 mA total current every 800 ns.

In the standard SOLEIL Storage Ring 430 mA operation ¾ of the ring is filled with 18 ps bunches in top-up mode. The time-resolved modes include hybrid multibunch, eight bunches (11 mA/bunch, 148 ns bunch spacing, 30 ps RMS bunch length) and single bunch modes (11 mA/bunch, 1.18 microsecond period). The electron bunch length is 18 ps RMS for bunches in the multibunch train, and 25 ps RMS for the 5 mA single bunch of the hybrid filling pattern. The low-alpha mode producing a few ps long bunch was recently initiated.

The ESRF runs 20% in uniform bunch-filling mode and 80% in various bunch-filling modes for single-pulse experiments. There are currently four modes for single pulse experiments: the 4-bunch (40 mA) and 16-bunch (90 mA) modes with equidistant bunch fillings, the hybrid mode (24x8 +1) , and the 7/8 multi-bunch mode (both 200 mA).

At the APS three different filling patterns for timing modes are available. The 24-bunch filling mode provides pulse durations of 34 ps (RMS) with 153 ns interpulse spacing. The hybrid singlet mode provides one 16 mA pulse with 50 ps (RMS) duration with a 1.59 microsecond interval to a pulse train of total length 500 ns. The 324-bunch mode has an even filling pattern with 0.3 mA per 22 ps (RMS) long bunch, with an 11.4 ns interpulse spacing.

IV. Technical design suggestions

In the session at the User's Meeting on September 30 different approaches to timing modes were discussed, and the possible challenges that arise with the MAX IV low-emittance solution that implements passive harmonic cavities were pointed out.

A few key questions that deserve further thought and effort are:

1. Is a hybrid filling mode necessary in order to implement resonant pulse picking or pseudo single bunch operation?
2. Is it possible to implement a hybrid filling mode in the 3 GeV machine considering the instabilities noted in the calculation by Galina Skripka?
3. What are the limitations on synchronization of pulsed lasers or shutters to the pulse structure of the ring? What is the smallest time interval between pulses that is required due to jitter?

The two approaches outlined above should be investigated further to determine whether they are compatible with the MAX IV low-emittance design and if they can deliver sufficiently good timing pulses for the needs of the MAX IV user community.