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Published in:
Journal of Synchrotron Radiation

DOI:
10.1107/S1600577514019286

2014

Journal of
Synchrotron Radiation

ISSN 1600-5775

Received 28 August 2014
Accepted 28 August 2014

Diffraction-limited storage rings – a window to the science of tomorrow

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This article summarizes the contributions in this special issue on Diffraction-Limited Storage Rings. It analyses the progress in accelerator technology enabling a significant increase in brightness and coherent fraction of the X-ray light provided by storage rings. With MAX IV and Sirius there are two facilities under construction that already exploit these advantages. Several other projects are in the design stage and these will probably enhance the performance further. To translate the progress in light source quality into new science requires similar progress in aspects such as optics, beamline technology, detectors and data analysis. The quality of new science will be limited by the weakest component in this value chain. Breakthroughs can be expected in high-resolution imaging, microscopy and spectroscopy. These techniques are relevant for many fields of science; for example, for the fundamental understanding of the properties of correlated electron materials, the development and characterization of materials for data and energy storage, environmental applications and bio-medicine.

Keywords: diffraction-limited storage rings; new science opportunities.

1. Diffraction-limited storage rings: why and how?

Synchrotron scientists have been always longing for brighter sources. Today, advances in accelerator technology open a new window of opportunities (Tavares *et al.*, 2014; Liu *et al.*, 2014). Accelerator physicists have long known that one way of providing higher brightness and coherence in the X-ray beams is to decrease the bending angle θ_d in each of the dipole bending magnets, and to allow tighter focusing by multipole magnets between the dipole bend magnets (Einfeld *et al.*, 2014). In fact, the horizontal emittance ε_0, which determines the average spectral brightness $B_{avg}(\lambda)$ and the coherent fraction $f_{coh}(\lambda)$, scales inversely with the third power of the number of bending magnets N_d: $\varepsilon_0 \sim N_d^{-3}$ (Hettel, 2014). Until recently it was unclear, however, how this strong dependence could be exploited without significant cost increase due to the need to fabricate and install many hundreds of magnets and the need for a very large building (circumference $\geq$1 km). This challenge was met by drastically decreasing the magnet gaps (Johansson *et al.*, 2014), and by innovative vacuum technology relying entirely on non-evaporable getter (NEG) pumps for pumping vacuum systems of a few centimetres diameter and several hundred metres length (Al-Dmour *et al.*, 2014). Today two rings are being constructed that use the multi-bend achromat approach to achieve a diffraction-limited storage ring in the medium-energy X-ray range: MAX IV in Sweden (Tavares *et al.*, 2014) and Sirius (Liu *et al.*, 2014) in Brazil. MAX IV will open to users in 2016, Sirius soon thereafter. Many existing facilities are working on upgrades of their present machines based on these concepts, and entirely new machines are under consideration. MAX IV and Sirius aim for emittances of $\varepsilon_0 \approx 3 \times 10^2$ pm rad; planned future machines will push for a few 10^1 pm rad or even lower.

Higher brightness of the source will be advantageous for almost any experiment. This is the case if, for example, a small spot needs to be illuminated such as in high-resolution RIXS (Schmitt *et al.*, 2014) or EXAFS (Frenkel & van Bokhoven, 2014), a high-pressure experiment has to be conducted in a tiny diamond anvil cell (DAC) (McMahon, 2014) or a (spectro-)microscopy experiment is performed (Thibault *et al.*, 2014; Rotenberg & Bostwick, 2014; de Jonge *et al.*, 2014; Hitchcock & Toney, 2014). The amount of light that perfect optics can focus into a diffraction-limited spot is given by the coherent flux provided by the source (Siewert *et al.*, 2014; Yabashi *et al.*, 2014; Susini *et al.*, 2014; Schroer & Falkenberg, 2014; de Jonge *et al.*, 2014). The most obvious benefits are for the new class of imaging experiments using coherent illumination of a sample to reconstruct information beyond the size of the X-ray focus (Thibault *et al.*, 2014; de Jonge *et al.*, 2014) as well as for X-ray photon correlation spectroscopy (Shpyrko, 2014), where a correlation function depends on two scattering events separated by a certain delay time.

While diffraction-limited storage rings (DLSRs) provide high average brightness, $B_{avg}(\lambda) \approx 10^{22}$ photons s^{-1} mm^{-2} mrad^{-2} (0.1% bandwidth)$^{-1}$, they cannot compete with freeelectron lasers (FELs) as regards the peak brightness $B_{\mathrm{peak}}(\lambda)$ required for ultra-fast time resolution or single-shot experiments. This complementarity would make it attractive to locate a DLSR and a FEL on the same site. In that case a large number of scientific experiments could be conducted simultaneously on many beamlines at the DLSR, while specialized experiments are scheduled for the FEL, at which only one or a few experiments can be conducted at a given time.

Exploitation of the full potential of a DLSR requires near-perfect optics (Siewert *et al.*, 2014; Yabashi *et al.*, 2014; Susini *et al.*, 2014; Schroer & Falkenberg, 2014), dedicated beamlines and sample environments (McMahon, 2014; Susini *et al.*, 2014), and specialized detectors (Denes & Schmitt, 2014). Together they will produce huge data rates ($\sim$10 GB s^{-1}) and data volumes ($\sim$10 TB per experiment) requiring dedicated infrastructure and specialized software that also allows non-expert synchrotron users to extract the relevant information within a realistic time.

2. Accelerators: achievements and challenges

The performance of synchrotron radiation sources has increased tremendously in terms of brightness during the last decades. The rate of brightness gain even beats the fast improvement rate of semiconductors (Moore's law) by a factor of two. As a consequence, new science areas have been and are being opened up and our scientific knowledge is continuously deepened and widened. Such scientific expansion rests on technology pillars. We have seen how the storage ring technology has improved, along with the technology of insertion devices (IDs), beamlines, detectors and computing facilities. It is a prerequisite that all technologies are matched and it is often difficult to identify which area is driving the development of others.

The introduction of DLSRs and FELs paves the way for reaching unprecedented performance in terms of temporal and spatial resolution at accelerator-based X-ray sources. But these technologies are just gate-openers; in order to fully exploit them, the full technology chain from source to detector must be developed.

The basic idea behind DLSRs is to radically reduce the horizontal emittance of the storage ring. This is achieved by building the ring from a large number of focusing cells (Hettel, 2014; Einfeld *et al.*, 2014). Each cell contains a bending magnet as well as higher-order multipole magnets necessary for focusing the electron beam. This exploits the fact that the horizontal emittance is inversely proportional to the number of cells cubed. Such a lattice is called a multi-bend achromat magnet lattice. The price to pay for this scheme is a drastic reduction of the dispersion in the ring, which requires powerful sextupole magnets compensating for a large negative natural chromaticity. These strong sextupoles are not trivial to build and moreover decrease the dynamic aperture of the ring (Tavares *et al.*, 2014), making it more difficult to operate and commission it (Borland *et al.*, 2014; Nagaoka & Bane, 2014).

One crucial point in this context is miniaturization of magnets. A way to keep the cost under control when increasing the number of cells is to make the individual magnets, and in particular their gaps, smaller. This also eases the problem of making strong magnet lenses (quadrupoles, sextupoles, octupoles) (Hettel, 2014), since the number of Ampère-turns can be kept low and room-temperature technology can be used without saturation of the magnet poles (Johansson *et al.*, 2014). Reduction of the magnet gaps requires a decreased diameter of the vacuum system, which will lower the conductance of the vacuum chambers. Lumped pumping by ion-getter pumps is therefore often replaced with linear pumping through the introduction of fully NEG-coated vacuum chambers (Al-Dmour *et al.*, 2014).

The reduced dynamic aperture is still an issue. However, our knowledge base regarding the operation and computer modelling of third-generation storage rings has reduced the safety margins needed (Hettel, 2014; Borland *et al.*, 2014; Nagaoka & Bane, 2014). In the case of an extreme DLSR, we can now even sacrifice electron beam lifetime (Nagaoka & Bane, 2014; Liu *et al.*, 2014; Tavares *et al.*, 2014) (to some extent) by relying on frequent top-up injection.

Smaller magnets imply lower mechanical tolerances and higher demands on mechanical stability (Hettel, 2014). Integrated magnet blocks housing several magnets offer a high degree of structural rigidity, system integration and precise alignment, where the advantages of modern CNC-machining are fully exploited (Johansson *et al.*, 2014).

Another interesting property of many DLSRs is that the dipole magnet fields are reduced as a consequence of their increased number (Einfeld *et al.*, 2014; Johansson *et al.*, 2014; Liu *et al.*, 2014; Tavares *et al.*, 2014). Since the ring emittance is defined by the electron-optic properties of the dipoles, the relative impact of IDs on the emittance, which still generally have strong magnet fields, is increased. The eigen-emittance of especially short-period undulators is negligible compared with the ring emittance, so these IDs act as emittance-damping items. For the DLSRs already under construction, the horizontal emittance of the ring is roughly halved when fully equipped with IDs (Tavares *et al.*, 2014; Liu *et al.*, 2014).

As the beam size is reduced, collective electron beam effects become more pronounced. One class of collective instabilities is due to the higher electron density (Nagaoka & Bane, 2014). In some cases the electron bunches are stretched to reduce this effect (Tavares *et al.*, 2014). This, however, increases the temporal length of the X-ray pulses, thus compromising time-resolved studies. To counteract such effects, concepts to provide ultra-short X-ray pulses even from DLSRs are being developed (Huang *et al.*, 2014).

One exception among the collective effects is the Touschek instability where electrons are lost due to electron transverse kinetic energy being transferred into longitudinal energy deviations. This may bring the electrons above the bucket height of the RF system, with a reduced beam lifetime as a consequence (Nagaoka & Bane, 2014; Tavares *et al.*, 2014; Liu *et al.*, 2014). However, if the beam emittance is reduced, the transverse oscillation energy is reduced as well. This might bring us into the situation that the beam lifetime is increased in spite of the increased electron density (Tavares *et al.*, 2014; Liu *et al.*, 2014).

Today, we see a number of (almost) DLSRs being built or planned. MAX IV in Sweden (Tavares *et al.*, 2014) and Sirius in Brazil (Liu *et al.*, 2014) are the fore-runners, but large facilities such as ESRF, APS and SPring-8 follow closely. Several national sources are also investigating the possibility of introducing the multi-bend achromat scheme.

3. New science enabled by brighter sources

In order to allow experiments to profit from the immense progress being realised in storage rings, it is necessary to match the parameters of the electron beam, which is the light source, to the sample or the detector. This requires three steps: (i) matching source size and divergence, (ii) transporting the photon beam while preserving brightness and coherence, and (iii) diagnostics, which allow achieving and maintaining optimum performance over hours to days.

3.1. Matching source and sample

Today's third-generation synchrotron radiation sources have an extreme asymmetry in the geometrical source size (σ_x, σ_y) (Hettel, 2014). The horizontal beam size σ_x is dependent on the emittance of the accelerator $\varepsilon_{0,x}$ which is typically of the order of a few nm rad (Einfeld *et al.*, 2014). The vertical source size σ_y is determined by the coupling to the horizontal emittance: $\varepsilon_{0,y} = \kappa\varepsilon_{0,x}$, with κ the coupling parameter. Such coupling comes from skew components of the magnetic field, alignment errors, scattering processes, *etc*. In modern accelerators it can be made very small: $\kappa \simeq 10^{-2}$–10^{-3}. As a result, the source is geometrically very asymmetric, having the form of an ellipse that is much wider than it is high. While this is a good source for illuminating the horizontal entrance slit of a spectrometer, which was a major application of early synchrotron radiation experiments, it is not well suited for the many imaging or diffraction experiments performed at third-generation synchrotrons. For today's imaging experiments in which typically uniform illumination of the sample is required and the scattered (or reflected, transmitted) light has to be recorded by a two-dimensional detector, a round or square source would be more adequate: $\sigma_x \simeq \sigma_y$.

The new DLSRs will reduce the horizontal emittance by an order of magnitude or more (Tavares *et al.*, 2014; Liu *et al.*, 2014; Hettel, 2014). This allows relaxing the coupling parameter κ and getting closer to a symmetrical source. Present projects still aim for couplings in the 10^{-2} range, thus maximizing the brightness. The ultimate dream for almost any imaging and diffraction experiment is a storage ring with an emittance so low that one can operate it at full coupling ($\kappa = 1$) and still achieve a source that is at the diffraction limit $\varepsilon_r(\lambda) \approx \lambda/(2\pi)$ well into the hard X-ray range. This is the ambition of future projects, which target emittances of the order of 10 pm rad.

Matching the source (electron beam) to the sample or detector will further be simplified by new options in IDs. The lower horizontal emittance poses less stringent requirements on the horizontal field profile of undulators (Hettel, 2014). Future undulators will have close to fourfold symmetry at very low gap sizes, of $\sim$3 mm. This allows stronger peak fields and more periods for a given undulator length L. The coherent fraction $f_{\rm coh}$ increases with the number of periods $N_{\rm ID}$ squared, $f_{\rm coh} \propto N_{\rm ID}^2$, in the limit of negligible electron energy and angle spread. This alone will increase the coherent flux by up to a factor of three to four. All of this will provide higher spectral flux (photons per unit bandwidth).

3.2. Transporting source brightness onto the sample

Brightness is a conserved quantity; the best that an optical system can do is to maintain the brightness provided by the source. This has led to the proverb 'The best optics is no optics'. In reality, optics will almost always be needed to collect, monochromatize and focus X-ray beams. Very significant work and creativity will be required to develop, fabricate and install brightness- and coherence-preserving optics up to wavelengths of the order of 1 Å. Several relevant technologies have been identified and first promising results are presented in this issue for polishing of optics (Siewert *et al.*, 2014; Yabashi *et al.*, 2014; Susini *et al.*, 2014), coating with single or optimized multilayers (Siewert *et al.*, 2014; Susini *et al.*, 2014), focusing (Siewert *et al.*, 2014; Yabashi *et al.*, 2014; Schroer & Falkenberg, 2014), as well as filters and diagnostics (Yabashi *et al.*, 2014). The individual elements will then need to be positioned and moved with respect to the source and the sample with unprecedented accuracy, posing new engineering challenges (Siewert *et al.*, 2014; Yabashi *et al.*, 2014; Susini *et al.*, 2014). Last but not least, the community will need to develop proper beam simulation tools from source to detector, which take coherence and fabrication errors into account and allow global optimization and testing of new optical concepts (Siewert *et al.*, 2014; Yabashi *et al.*, 2014; Susini *et al.*, 2014; de Jonge *et al.*, 2014).

3.3. Diagnostics

Active control of the electron beam through diagnostic information provided by the X-ray beam will be mandatory for operating the storage ring at its performance limit. These diagnostics should include position and angular information in both planes and possibly even time domain feedback through, for example, measurements of coherent synchrotron radiation emitted in the infrared spectrum.

Finally an optimized detector is needed to record the scattered (or absorbed, emitted) signal from the sample. Maybe this is an area where in recent years progress has been greatest, but potential is still largest (Denes & Schmitt, 2014; Shpyrko, 2014; de Jonge *et al.*, 2014). Continuing to develop and install pixelated detectors with high efficiency, low noise and large collection angle is likely to be much more cost effective than upgrading the accelerator to higher current for increasing flux. Apart from the cost argument, this is the only way to mitigate radiation damage, which already today is the limiting factor for many experiments in the fields of polymer science, soft matter and biology (Shpyrko, 2014; de Jonge *et al.*, 2014; Hitchcock & Toney, 2014).

In general, in the future it will be more important than ever to optimize the entire value chain from the source to the detector. Cutting-edge experiments will only succeed if each single component is optimized. This requires thinking out of the box and necessitates discussions and collaborations across the fields of accelerator physics, X-ray optics, sample preparation, detectors, data storage and analysis. This special issue is an example of the challenges ahead but also of the ongoing discussions between the experts.

4. New science enabled by brighter sources

Light sources are a tool to see the world around us and storage rings are nothing but light sources for the X-ray range. The significant improvement provided by the DLSRs under construction and in the design stage will enlighten our view of the world and allow science which is not possible, or not even thinkable, today. Several articles in this issue attempt to describe why improved light sources are needed and what they will allow (McMahon, 2014; Thibault *et al.*, 2014; Frenkel & van Bokhoven, 2014; Rotenberg & Bostwick, 2014; Schmitt *et al.*, 2014; Shpyrko, 2014; de Jonge *et al.*, 2014; Hitchcock & Toney, 2014). However, predictions are difficult.

Certain to profit from enhanced brightness and coherence are the science communities interested in materials that derive their properties from phenomena happening on a range of different length scales. Examples of these are found in energy materials (Frenkel & van Bokhoven, 2014; Hitchcock & Toney, 2014), biological materials (de Jonge *et al.*, 2014) and correlated electron systems (Rotenberg & Bostwick, 2014). In all these materials the increased brightness will enable better focusing of the X-ray beam, thus allowing for X-ray fluorescence, NEXAFS, diffraction, *etc.* on a sub-micrometre or possibly nanometre scale. One may thus investigate the activity of catalytic nanoparticles resting on substrates (Frenkel & van Bokhoven, 2014), chemical reactions and diffusion in batteries and fuel cell membranes (Hitchcock & Toney, 2014), trace element distributions in biological cells and tissue (de Jonge *et al.*, 2014), and novel materials for electronic applications (Rotenberg & Bostwick, 2014).

The availability of partially or fully coherent X-ray sources has enabled novel imaging and diffraction techniques (Thibault *et al.*, 2014; Hitchcock & Toney, 2014). We may be seeing a paradigm change. In the past, incoherent beams revealed the structure of periodic crystals through conventional diffraction. In the future, the structure of non-periodic objects will be studied over a wide range of sizes and composition; they can be illuminated with coherent X-rays of sufficient intensity to enable extracting spatial information by numerical phase retrieval. Examples for which this is relevant are the multi-scale materials mentioned above, as well as amorphous materials, or matter close to phase transitions, where the deviation from perfect order becomes relevant and determines the properties.

When considering focusing of the X-ray beam, two different classes of experiments should be distinguished. In some cases the size of the X-ray focus directly determines the spatial resolution obtainable in the experiment. Here the increased coherence from the source directly translates to an increase of intensity, which a perfect optical element can provide. Different optical elements have been conceived and tested and provide resolutions down below 10 nm already (Yabashi *et al.*, 2014). The new DLSRs will provide orders of magnitude more intensity. They will also challenge the makers of focusing optics because any increase in source quality will expose any existing weakness in the optical element (Siewert *et al.*, 2014).

In other cases the focused X-ray beam is more a tool than a goal. Examples are experiments at high pressure (McMahon, 2014) or inelastic scattering (Schmitt *et al.*, 2014). In the former, the smaller X-ray beam allows samples to be illuminated in a smaller diamond anvil cell, which can reach higher pressure and thus enable entering previously uncharted scientific territory. In the latter, the X-ray spot defines the entrance spot for a secondary spectrometer collecting the X-rays emitted by the sample and dispersing them according to their energy loss. Here, any reduction of focus size directly translates into better energy resolution and thus into increased sensitivity for lower-energy excitations in the sample, associated with, for example, charge, orbital and spin order, and superconductivity.

Recent imaging techniques utilizing coherent illumination such as ptychography (Thibault *et al.*, 2014) have the capability of efficiently bridging the gap between nanometre and micrometre length scales. Although they aim for the best possible spatial resolution, they do not necessarily push for the smallest possible X-ray focus. Instead, they utilize a coherent beam with moderate focus size in the tens or hundreds of nanometres range. In ptychography, a spatial resolution below the focus diameter is obtained by numerical reconstruction of the diffracted phase information resulting from overlapping beam spots during scanning of the sample. Ptychography, in combination with high source brightness and improved quality of optics and detectors, will likely be the tool of choice for microscopy on 'real world' samples over a wide range of length scales. It provides quantitative electron densities, and photon-energy-dependent ptychography across an absorption edge enables determination of the local elemental composition underlying the electron density distribution. However, it is not suitable for detecting trace elements. Here, X-ray fluorescence (XRF) is the technique of choice, and one can envision combining ptychography with simultaneous XRF for microscopy of the matrix in which the trace elements are embedded (de Jonge *et al.*, 2014).

One field that has profited immensely from progress in storage-ring-based light sources and has in turn pushed such sources to ever higher ambitions is macromolecular crystallography (MX). DLSRs will boost this synergy even further. MX is not covered by a separate article in this issue, but the benefits are obvious. Today some of the most challenging questions in structural biology involve proteins that do not crystallize in the size and quality needed for experiments at existing third-generation sources. The increased source brightness of DLSRs allows the beam to be focused to smaller diameters while retaining the low divergence necessary for diffraction from crystals with large unit cells. Optics developments will enable adaptive illumination of odd-shaped crystals (needles, flakes) or even sub-structures of these in order to reduce mosaicity or eliminate salt crystals. In case the crystals are too small for the generation of a full data set, serial crystallography with data collection at room temperature can be exploited as applied at FELs (Stellato *et al.*, 2014). Using, for example, a high-viscosity liquid jet (lipid cubic phase or similar carrier material), fresh crystallites of (sub-)micrometre size may continuously be delivered into the X-ray beam. Alternatively, a collection of crystallites may be dispersed on a thin substrate or on a growth template (in solution) and each one is illuminated by the focused beam. At a FEL, each crystallite that is hit by an X-ray pulse is destroyed. During the shot each crystallite presents itself in one particular orientation, which may make it difficult to index and process the diffraction pattern. By contrast, at a DLSR more data can be collected by monitoring and even correcting the radiation damage over time using a fast-frame-rate pixel array detector (Denes & Schmitt, 2014). One may even try to collect, for each crystal on the substrate, reflections over a small oscillation angle ('fine phi-slicing'), which makes indexing easier and determination of the integrated intensities of the reflections more accurate. Sorting thousands of such diffraction patterns by their crystallite orientation and then summing them up can lead to data sets with sufficient information to solve the structure.

While FELs carry the motto 'Diffract before you destroy', at DLSRs one may rephrase it as 'Diffract while you destroy'. We note here that radiation damage is expected to be reduced in a tiny crystal because a substantial part of photoelectrons can simply escape (Sanishvili *et al.*, 2011). This would work to the advantage of fine phi-slicing at a DLSR. Staying at room temperature, crystallites generally exhibit less mosaicity and defects than at cryogenic temperatures, which results in low-background diffraction patterns with sharp reflections. As a result, sparse data can be analysed.

In the above considerations for MX, we implicitly assumed the use of a Si-crystal monochromator having an energy bandwidth of a few 10^{-4}. Even at a DLSR, the diffraction pattern from a single micro- or nano-crystal illuminated by such a highly monochromatic beam may be too weak. For some applications one may therefore consider use of a larger-bandwidth (a few 10^{-2}) beam as provided by a multilayer monochromator.

DLSRs will also be used for four-dimensional imaging (Thibault *et al.*, 2014; Shpyrko, 2014; de Jonge *et al.*, 2014; Hitchcock & Toney, 2014). Their increased brightness allows routine acquisition of three-dimensional tomograms with a time resolution of milliseconds or below for relevant sample sizes (millimetres or tens of millimetres) and resolutions $\Delta x <$ 1 µm. If processes are studied that can be triggered repeatedly in a pump–probe set-up by a fast external stimulus like a laser pulse or a magnetic field, the time resolution can be pushed to the length of the electron bunch in the storage ring (10^1–10^2 ps). However, in the ultra-fast time domain the DLSRs will reach a limit and certainly need to be complemented by FELs. The gain in brightness at DLSRs comes from decreasing both the horizontal (σ_x) and the vertical beam size (σ_y). This increases the electron density in the bunch, inducing collective effects, which in turn cause beam instabilities or loss of electrons (Tavares *et al.*, 2014; Nagaoka & Bane, 2014). To keep these effects within acceptable limits, present projects deliberately stretch the electron bunches in the longitudinal direction by using low-frequency RF and high-harmonic cavities (Tavares *et al.*, 2014). Unless novel ideas are developed and implemented in regular user operation (Huang *et al.*, 2014), DLSRs will have longer bunches than present third-generation rings. For example, the MAX IV ring will have a pulse duration of $\Delta t \simeq 400$ ps, while typical third-generation sources today have $\Delta t \simeq 70$ ps. In any case, storage rings can, and should, not compete with linear accelerators when it comes to the ultimate time resolution. The sub-fs scale that is being targeted by FELs today is certainly out of reach for a storage ring.

5. Conclusion

As shown in the papers collected in this special issue, accelerator physics has taken the lead in bringing synchrotron X-ray sources closer to their diffraction limit. Using new concepts in magnet design, vacuum technology and an improved understanding of beam dynamics, light sources of an unprecedented quality are and will be built. The users have embraced this challenge and now design instrumentation to exploit this increased performance as well as experiments demanding it. In our common effort to make the invisible secrets of nature visible we have again come one step further. We all look forward to what will be found.

The authors thank the many colleagues in the world-wide network of synchrotron radiation sources for sharing their unpublished results and for fruitful discussions. Progress in our field depends on open exchange of ideas and information.

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