

On the design of ultrafast shutters for time-resolved synchrotron experiments

Milan Gembicky* and Philip Coppens*

Chemistry Department, State University of New York at Buffalo, Buffalo, NY 14260-3000, USA.
E-mail: gembicky@buffalo.edu, coppens@buffalo.edu

A comprehensive treatment of the limitations and possibilities for single-pulse selection in synchrotron operating modes with ~ 150 ns bunch separation, as occurs in the standard operating mode at the Advanced Photon Source, is presented. It is shown that the strength of available materials and allowable kinetic energy build-up limit single-bunch selection for this separation to sample sizes of ~ 100 μm , and that for minimization of kinetic energy build-up it is preferable to increase the r.p.m. within physically acceptable limits rather than increase the disc radius to obtain a desirable peripheral speed. A slight modification of the equal-bunch spacing standard fill patterns is proposed that allows use of samples as large as 500 μm . The corresponding peripheral speed of the chopper wheel is ~ 600 m s^{-1} , which is within the limits of high-strength titanium alloys. For smaller samples, peripheral speeds are proportionally lower. Versatility can be achieved with interchangeable chopper wheels and the use of different orientations of the rotation axis relative to the X-ray beam, which opens the possibility of larger, rather than one-of-a-kind, production runs.

Keywords: time-resolved experimentation; ultrafast pulse selector; synchrotron science; synchrotron fill pattern.

1. Introduction

The development of fast and accurate shutters for the isolation of short X-ray pulses or pulse trains at third-generation synchrotron sources is of crucial importance in time-resolved scattering and diffraction experiments, which have received considerable attention in the past decade (see, for example, Helliwell & Rentzepis, 1997; McPherson *et al.*, 2000; Moffat, 2001; Techert *et al.*, 2001; Coppens & Novozhilova, 2002; Wulff *et al.*, 2002; Schotte *et al.*, 2002, 2003; Rajagopal *et al.*, 2004; Anderson *et al.*, 2004; Yasuda *et al.*, 2004; Coppens *et al.*, 2005). The developments are based on earlier pioneering work performed in the 1980s in which the rotating slot design was analyzed (Mills, 1989; LeGrand *et al.*, 1989). More recently, several alternatives to mechanical choppers, including designs based on a rotating mirror (Kosciesza & Bartunik, 1999), and by diffraction by acoustic surface waves (Tucoulou *et al.*, 1998), and a rotating silicon crystal (McPherson *et al.*, 2002), were analyzed. However, unlike mechanical shutters, not all these designs are applicable to pink- or white-beam experiments.

In a preceding paper (Gembicky *et al.*, 2005) we have described a fast and accurate mechanical shutter for sub-microsecond resolution experiments, capable of either selecting the single- or the super-bunch of the 'hybrid' SOM-1

operating mode of the APS synchrotron source. A major advantage of the shutter is its moderate cost compared with comparable equipment currently available. It is also highly flexible, as the slotted disc can be replaced readily, and capable of high repeat rates of the transmitted X-ray pulses. However, the shutter does not have sufficient resolution for use in the more common operating modes in which the electron bunches are closer to each other and equally spaced around the storage ring. Such a capability is highly desirable, as it eliminates the need for special operating modes that may have disadvantages for other experiments being conducted at the facility. Single-pulse selection under standard operating conditions can dramatically increase the beam time usable for time-resolved experiments.

As no comprehensive treatment is available in the literature, we discuss here the considerations involved in single-bunch selection during synchrotron operating modes with ~ 150 ns bunch separation, as is the case in the 24-bunch mode of the Advanced Photon Source (APS). The results depend on the sample size when specimens smaller than the shutter aperture are used with an essentially non-converging X-ray beam. In practice, the limitations on sample size can be eliminated by a slight modification of the filling pattern of the synchrotron ring with minimal impact on other users, as discussed further below.

Table 1

Characteristics of shutters used at APS for operation under SOM-1 conditions.

| | Shutter type | Opening size (mm) | Diameter (mm) | Number of slots | Duty cycle† | Repetition rate (Hz) | TWT‡ (μs) | Peripheral velocity (m s ⁻¹) | r.p.m. |
|-------------------------|---------------|-------------------|---------------|-----------------|-------------|----------------------|-----------|--|--------|
| McPherson <i>et al.</i> | Perpendicular | 0.5 | 50.8 | 2§ | 1/102 | 2662 | 2.45 | 212 | 79867 |
| Jülich¶ | Parallel | 0.9 | 193.6 | 1 | 1/395 | 896.6 | 1.89 | 545 | 54309 |
| Buffalo-1 | Parallel | 0.35 | 140 | 45 | 1/12 | 22629 | 3.28 | 221 | 30172 |

† Defined as the sub-harmonic of the orbit frequency. ‡ Total window time, *i.e.* exposure time for a sample of the size of the slot opening. Exposure times are proportionally shorter for smaller samples. § Single symmetrical channel allowing transmission in both directions. ¶ From Wulff *et al.* (2002).

In experiments in which a sample larger than the beam is used, the size of the transmitted beam becomes the determining dimension.

2. Desirable features

The aim is to produce a versatile instrument with maximal flexibility to adjust to the requirements of a particular set of experiments. The following criteria should be met:

- (i) Separation of a single bunch during the standard operation modes at APS and storage rings with similar bunch-to-bunch separations of ~150 ns.
- (ii) Ability to accommodate a sample size of at least 100 μm in the vertical direction.
- (iii) Minimization of jitter, machining and alignment errors which can negatively affect the size and the shape of the transmitted beam.
- (iv) Operation with pink and white beams when used in conjunction with a heat-load shutter.
- (v) Reasonable price and low maintenance requirements.
- (vi) Portability from beamline to beamline.

3. Shutter design

An important distinction is made between shutters with rotation axes aligned perpendicular and parallel to the X-ray beam. The two geometries are compared in Fig. 1. To achieve the short opening time the perpendicular geometry must be selected, as originally proposed by LeGrand *et al.* (1989) and applied in both the shutter developed by McPherson (McPherson *et al.*, 2000; Mills, 1989) and the ‘Jülich’ shutter (Wulff *et al.*, 1997), whereas the parallel geometry is preferred if a high repeat rate is required (Comsa *et al.*, 1981; Möller & Zimmerer, 1987; Mills, 1989; Coppens *et al.*, 2005). Table 1 compares the Jülich shutter, as installed at the European Synchrotron Radiation facility (ESRF), and the McPherson shutter with the Buffalo-1 shutter (Gembicky *et al.*, 2005), which operates in the parallel geometry. The calculations assume a fully illuminated sample of the same size of the shutter opening, giving a triangular profile of the diffracted beams. While this is not the best geometry for an experiment, it presents an upper limit for the time resolution that can be achieved when samples smaller than the beam are employed. If the sample is smaller than the slot opening, it effectively acts as a slit and the profile becomes trapezoidal, with a plateau

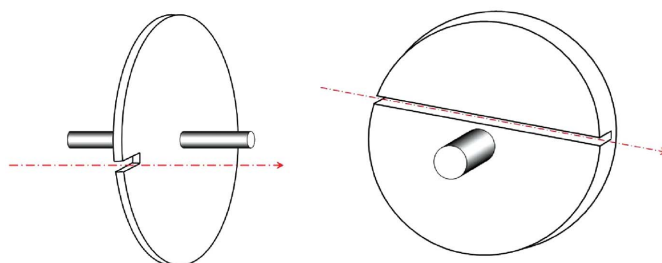


Figure 1 The parallel and perpendicular designs of X-ray shutters (after LeGrand *et al.*, 1989). For simplicity only one slot/channel is shown. Most shutters will have more than one opening, depending on the application. The channel on the perpendicular shutter on the right can pass through the center rather than being offset. This arrangement will double the frequency as the X-ray beam can then pass in both directions.

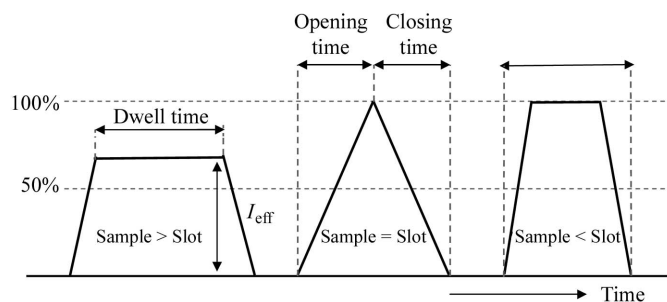


Figure 2 Effective transmission function *versus* time for three different cases: sample size larger than slot opening (parallel geometry only) (left); sample equal to the slot opening (middle); sample size smaller than slot opening (right). If an auxiliary slit smaller than the sample is used, the slit size replaces the sample size in determining the profile of the secondary beam. In the perpendicular geometry the central figure applies even for large sample sizes. A parallel beam is assumed. The two figures on the left are as discussed earlier in terms of slit size for the perpendicular geometry (Mills, 1989).

when the sample is fully illuminated, as illustrated in Fig. 2 for both the parallel and the perpendicular designs.

For a single slot used in the parallel geometry with a continuous source, the exposure of the sample begins when the leading edge of the chopper projects on the sample edge when looking along the beam direction. It ends when the trailing edge passes the opposite boundary of the sample, as illustrated in Fig. 3. This implies that the shutter opening has to travel twice the cross-section length of the sample. For a channel-type shutter with perpendicular geometry this factor of two does not occur, as the back edge of the shutter moves at

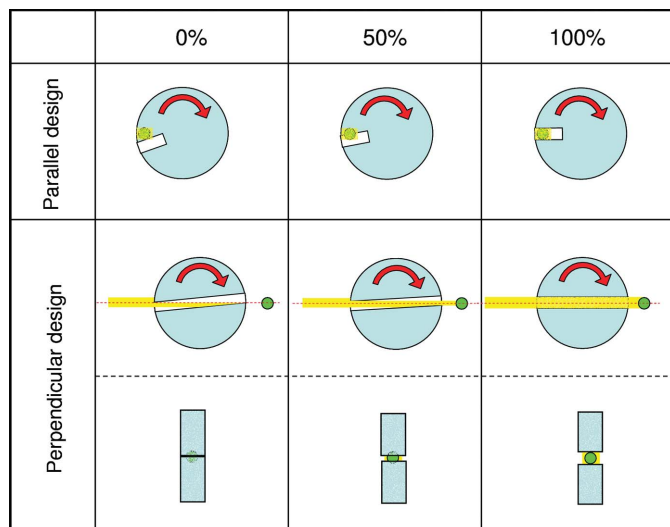


Figure 3 Schematic showing shutter opening for the parallel and perpendicular shutter designs for a sample of size equal to that of the slit opening. Closing of the shutter is identical except for a reverse of the order. The beam is drawn in yellow. In the parallel geometry the beam is perpendicular to the plane of the figure. For the perpendicular geometry both the views perpendicular and parallel to the beam are shown. Note that in the perpendicular design the peripheral speed is half of that of the parallel design for a given opening time, as both the entrance and the exit apertures of the tunnel truncate the beam. In the parallel design the projection of the chopper slit scans through the sample, whereas in the perpendicular design the slit opening remains centered on the sample.

equal speed and closes the beam when the projection of the trailing edge of the front opening has traversed the sample. Thus, the perpendicular geometry has the advantage that the opening of the channel through which the X-ray beam travels and its exit both act as shutters truncating the beam, leading to a factor of two improvement in time resolution relative to the parallel geometry. If the perpendicular geometry is implemented with the beam being cut by a straight-edged slot rather than by both sides of a channel, the factor of two is lost. For a pulsed source the transmission function is multiplied by the time structure of the pulse (or pulses) admitted by the window. The absence of a plateau in the transmission function in the case where the sample size is equal to the slot opening, represented by the middle figure of Fig. 2, implies that this geometry should be avoided, as the diffraction intensity from a pulsed source becomes sensitive to small jitters in the chopper wheel (Mills, 1989).

The entries in Table 1 for the Jülich shutter are for the instruments currently installed at 14-ID at APS and 9-ID at ESRF with a wheel diameter of 194 mm. In all cases, for a given slit size the opening time depends only on the peripheral velocity. If the synchrotron is used as a pulsed source, the duty cycle is defined by the sub-harmonic of the orbit frequency that is used, rather than by a combination of the actual width and number of the openings in the shutter wheel. The duty cycles given in Table 1 are obtained in this way.

The slot opening size of the Buffalo-1 shutter can be varied by changing the chopper disc; a 350 μm size is used in the example in Table 1. Owing to the larger number of slots, the

Table 2 Characteristics of parallel- and perpendicular-geometry shutters.

For a certain peripheral speed the r.p.m. is inversely proportional to the wheel radius. Bold entries represent acceptable solutions in terms of pulse-to-pulse separation and peripheral speed.

| | Required TWT (ns)† | Sample size (μm) | Parallel axis peripheral speed (m s ⁻¹) | Perpendicular axis peripheral speed (m s ⁻¹) |
|----------|--------------------|------------------|---|--|
| SOM-1 | 2400 | 500 | 417 | 208 |
| 24 bunch | 204 | 250 | 2451 | 1225 |
| 24 bunch | 204 | 200 | 1961 | 980 |
| 24 bunch | 204 | 100 | 980 | 490 |

† Total window time defined as the time needed for the leading edge of the opening to traverse the sample + the time for the trailing edge to traverse the sample for the limiting case of a sample the size of the slot opening.

duty cycle in the Buffalo-1 chopper in the parallel mode is higher than for the other instruments, even at lower r.p.m. values, the repetition rate and therefore the exposure time being proportional to the number of slots. This is of importance for narrow-bandwidth techniques, such as ultrafast X-ray spectroscopy and imaging, and synchrotron Mössbauer inelastic spectroscopy.

Table 2 compares the possibilities for single-bunch separation in the 24-bunch APS mode for the parallel- and perpendicular-spindle geometries. Total window time is assumed to be two-thirds of the maximal pulse separation time. The calculations do not take into account the effect of possible timing, machining and alignment errors. However, these can be minimized at reasonable cost. For example, the commercially available linear amplifier/controller (Motion Control Systems LA 2000-62) combined with the Buffalo-1 shutter with its precision bearing has a jitter of only a few nanoseconds, sufficient for single-pulse selection.

Limitations are imposed by the maximum strain in the material (Hearn, 1997). They can be expressed as a function of the speed at the periphery of the chopper disc.

The stress σ in a uniform flat disc depends on Poisson's ratio ν and the density of the disc material ρ , and is given by

$$\sigma = (1/8)(3 + \nu)\rho\omega^2 R_o^2, \quad (1)$$

where R_o is the disc radius and ω is the angular velocity. This equation is equivalent to that given by LeGrand *et al.* (1989), apart from the dependence on Poisson's ratio of the material, which was omitted in the earlier expression.

The most important measure of the feasibility of the shutter design is the peripheral speed as it cannot exceed the physical limitations of the material. Substituting the peripheral speed $v_p = \omega R_o$, we obtain

$$\sigma = (1/8)(3 + \nu)\rho v_p^2. \quad (1a)$$

With a value of ν equal to ~ 0.3 , valid for a typical material such as stainless steel, we obtain approximately

$$v_{p,\max} = [\sim 2.4(\sigma_{\max}/\rho)]^{1/2}. \quad (2)$$

For the perpendicular design of the ultrafast shutter with the pulses defined by the channel, high-quality titanium can be chosen as the disc material. It is both light ($\rho = 4500 \text{ kg m}^{-3}$)

and strong ($\sigma_{\max} > 1000$ MPa). This gives, with expression (2), $v_{p,\max} \simeq 730$ m s⁻¹. Therefore, with the materials presently available, one-bunch selection in standard operating modes is limited to a 100–150 μm sample size. Larger samples can be accommodated by a modest modification of the standard operating mode, as discussed further below.

Another important consideration for chopper design is the kinetic energy stored in a rotating disc. For a rotating disc of uniform thickness, the kinetic energy can be expressed as a function of the angular or peripheral speed,

$$E_k = (1/4)\pi h \rho R_o^4 \omega^2, \quad (3)$$

where h is the chopper disc thickness. Substituting for $\omega = v_p/R_o$, we obtain

$$E_k = (1/4)\pi h \rho R_o^2 v_p^2. \quad (3a)$$

It follows that for a given peripheral speed the chopper disc radius must be kept as small as possible to avoid kinetic energy build-up. Safety reasons dictate that such energy build-up should be avoided. It is therefore preferable to increase the r.p.m. within physically acceptable limits, rather than increase the disc radius to obtain the desired peripheral speed.

4. Alleviation of sample size limitations by minor modification of the standard synchrotron filling pattern

A drawback of the hybrid operation modes is that experiments that are dependent on single-photon counting are adversely affected when the X-rays are concentrated in a short part of the ring orbit flight-time. However, it is important to realise that the presence of a superbunch carrying most of the ring current, and concentrated in 495 ns (SOM-1 at APS), is not a requirement for single-bunch selection. It is fully adequate to have a single bunch separated from its neighbors by, for example, 306, 462 or 602 ns (*i.e.* two, three or four times 1/24 of the period of the APS orbit flight time), while the rest of the ring is filled with equally spaced bunches. If the two bunches on each side of the single bunch to be selected are omitted in a modified standard mode with 22 bunches (21 + 1 separated bunch) (Fig. 4), the slot size can be increased and the sample size limitation is reduced by a factor of two (Table 3). In general, a $(24 - 2n)$ ring filling will allow an n -fold increase in sample size. As for many experiments, 50–200 μm samples are desirable, and single-bunch selection with the current chopper design in the perpendicular mode of operation is entirely feasible.

Further detail is given in Table 3. In the (19 + 1) mode with perpendicular geometry of the shutter design, limitations in sample size become much less severe as acceptable linear velocities can be used with larger samples.

5. Concluding remarks

We have presented a comprehensive treatment of the limitations and possibilities for single-pulse selection in standard operating modes of APS and light sources with comparable bunch separations in frequently-used operating modes. They

Table 3

Shutter operation with the (21 + 1) and (19 + 1) modified standard ring patterns.

Bold entries indicate acceptable solutions in terms of pulse-to-pulse separation and peripheral speed.

| | TWT (ns) | Sample (mm) | Parallel geometry peripheral speed (m s ⁻¹) | Perpendicular geometry peripheral speed (m s ⁻¹) |
|--------|----------|-------------|---|--|
| 21 + 1 | 408 | 0.5 | 2451 | 1225 |
| 21 + 1 | 408 | 0.25 | 1225 | 613 |
| 21 + 1 | 408 | 0.1 | 490 | 245 |
| 19 + 1 | 816 | 0.5 | 1225 | 613 |
| 19 + 1 | 816 | 0.25 | 613 | 306 |
| 19 + 1 | 816 | 0.1 | 245 | 123 |

Modified Standard Operation I:
22 bunch (21+1)



Modified Standard Operation II:
20 bunch (19+1)

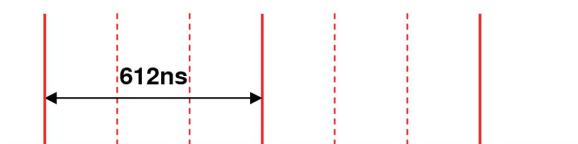


Figure 4

Schematic showing two modified standard operating modes.

include SPring-8 (2006) and many of the other synchrotron light sources currently under construction.

Practical considerations limit single-bunch selection to sample sizes of ~ 100 μm for ~ 150 ns bunch separation when the chopper is operating in the perpendicular mode, and half that for parallel operation. For samples larger than the beam, the chopper aperture becomes the determining factor and the same limitations apply. However, a slight modification of the standard fill pattern of the storage ring allows use of samples as large as 500 μm with a peripheral speed of the chopper wheel of ~ 600 m s⁻¹, which is within the limits of high-strength titanium alloys. Although even this small modification will require the consent of the broader synchrotron community, the proposed modified standard modes have significant advantages over common special operating modes for experiments that depend on single-photon counting.

The kinetic energy stored in the chopper can be minimized by reducing the diameter of the chopper wheel within physically acceptable limits and increasing its rotational velocity to compensate for the concomitant reduction in peripheral speed.

The repeat rate of the perpendicular shutter depends on the number of channels cut in the disc, but can be of the order of several thousand Hz, and thus match currently available

picosecond pulse-width lasers. Although there is no single design that fits all experiments, versatility can be achieved with interchangeable chopper wheels and the use of different orientations of the rotation axis relative to the X-ray beam.

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References

- Anderson, S., Srajer, V., Pahl, R., Rajagopal, R., Schotte, F., Anfinrud, P., Wulff, M. & Moffat, K. (2004). *Structure*, **12**, 1039–1045.
- Comsa, G., David, R. & Schumacher, B. J. (1981). *Rev. Sci. Instrum.* **52**, 789–796.
- Coppens, P. & Novozhilova, I. V. (2002). *Faraday Discuss.* **122**, 1–11.
- Coppens, P., Vorontsov, I., Graber, T., Gembicky, M. & Kovalevsky, A. Y. (2005). *Acta Cryst.* **A61**, 162–172.
- Gembicky, M., Oss, D., Fuchs, R. & Coppens, P. (2005). *J. Synchrotron Rad.* **12**, 665–669.
- Hearn, E. J. (1997). *Mechanics of Materials*, Vol. 2, *The Mechanics of Elastic and Plastic Deformation of Solids and Structural Materials*, 3rd ed., pp. 117–126. Oxford: Butterworth-Heinemann/Elsevier. (Online version available at <http://www.knovel.com/knovel2/Toc.jsp?BookID=434&VerticalID=0>.)
- Helliwell, J. R. & Rentzepis, P. M. (1997). *Time-Resolved Diffraction*. Oxford: Clarendon Press.
- Kosciesza, D. & Bartunik, H. D. (1999). *J. Synchrotron Rad.* **6**, 947–952.
- LeGrand, A. D., Schildkamp, W. & Blank, B. (1989). *Nucl. Instrum. Methods Phys. Res. A*, **275**, 442–446.
- McPherson, A., Lee, W.-K. & Mills, D. M. (2002). *Rev. Sci. Instrum.* **73**, 2852–2855.
- McPherson, A., Wang, J., Lee, P. L. & Mills, D. M. (2000). *J. Synchrotron Rad.* **7**, 1–4.
- Mills, D. M. (1989). *Rev. Sci. Instrum.* **60**, 2338–2341.
- Moffat, K. (2001). *Chem. Rev.* **101**, 1569–1581.
- Möller, T. & Zimmerer, G. (1987). *Phys. Scr.* **T17**, 177–185.
- Rajagopal, S., Schmidt, M., Anderson, S., Ihee, H. & Moffat, K. (2004). *Acta Cryst.* **D60**, 860–871.
- Schotte, F., Anfinrud, P. A. & Wulff, M. (2002). *Trends Opt. Photon.* **72**, 31–32.
- Schotte, F., Lim, M., Jackson, T. A., Smirnov, A. V., Soman, J., Olson, J. S., Phillips, G. N. Jr, Wulff, M. & Anfinrud, P. A. (2003). *Science*, **300**, 1944–1947.
- SPRING-8 (2006). http://www.spring8.or.jp/en/users/status/schedule/publicfolder_view, accessed 18 September 2006.
- Techert, S., Schotte, F. & Wulff, M. (2001). *Phys. Rev. Lett.* **86**, 2030–2033.
- Tucoulou, R., Roshchupkin, D. V., Mathon, O., Schelokov, I. A., Brunel, M., Ziegler, E. & Morawe, C. (1998). *J. Synchrotron Rad.* **5**, 1357–1362.
- Wulff, M., Plech, A., Eybert, L., Randler, R., Schotte, F. & Anfinrud, P. (2002). *Faraday Disc.* **122**, 13–26.
- Wulff, M., Schotte, F., Naylor, G., Bourgeois, D., Moffat, K. & Mourou, G. (1997). *Nucl. Instrum. Methods Phys. Res. A*, **398**, 69–84.
- Yasuda, N., Uekusa, H. & Ohashi, Y. (2004). *Bull. Chem. Soc. Jpn*, **77**, 933–944.