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An optical chopper for generation of short X-ray pulses to allow in-house time-resolved photocrystallography

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As part of a project to implement in-house time-resolved diffraction of short-lifetime species, a fast shutter with a custom-designed enclosure has been installed. The device is suitable for generation of X-ray pulses with time lengths down to about 5 µs. The design does not require major modifications to commercially available diffractometer setups. Significant airflow, generated by the rotating chopper disc, which interferes with temperature control at the sample, has been eliminated by a compact enclosure, which does not interfere with sample monitoring and conditioning devices. It allows for full rotation of the diffractometer circles.

1. Introduction

Studies of light-induced dynamics at the atomic level are crucial for understanding the behaviour of functional materials and their potential application (Gorfman, 2014; Cailleau *et al.*, 2010; Coppens, 2011). Such studies are possible with time-resolved photocrystallographic techniques, in which a well defined crystal is pumped with a laser pulse, quickly followed by a synchronized X-ray pulse. This approach has been successfully applied to fast photo-induced processes in model systems of transition metals (Coppens, 2011; Makal *et al.*, 2012), as well as macromolecules (Ren *et al.*, 1999; Schotte *et al.*, 2012). However, such studies are typically performed at synchrotron sources, at which the generation of X-ray probes is achieved by isolation of single pulses from the synchrotron beam. In the case of monochromatic experiments, pulses are selected with high-speed shutters (Gembicky *et al.*, 2007; Gembicky & Coppens, 2007). When the more intense polychromatic (pink Laue) radiation is applied, a combination of high-speed choppers and heat-load shutters is employed to isolate single X-ray pulses (Graber *et al.*, 2011; Cammarata *et al.*, 2009; Wulff *et al.*, 2002).

With the continuing increase of intensity of in-house X-ray sources (Sakabe *et al.*, 2013; Hemberg *et al.*, 2003; Otendal *et al.*, 2008), and the parallel increase of sensitivity and reduced read-out time of X-ray area detectors (Fertey *et al.*, 2013; Broennimann *et al.*, 2006; Basolo *et al.*, 2005), it is timely to explore the possibility of performing similar time-resolved studies in-house. To accomplish this, X-ray pulses must be generated from a continuous X-ray beam. This can be achieved with an optical chopper. Gembicky & Coppens (2007) have discussed various chopper designs. They show that the setting with the rotation axis perpendicular to the X-ray beam provides pulses shorter by a factor of two at the same RPM (revolutions per minute) than those achievable with a parallel setting. However, the space requirements of the perpendicular arrangement prevent its use for in-house experiments at commercially supplied X-ray sources.

2. Location of the chopper

It would be preferable to install the chopper before the collimator, just after the X-ray source. However, in the in-house experimental arrangement the X-ray beam traverses from the source through one or more shutters, a monochromator or multilayer optics, and a collimator. All of these components are carefully aligned such as to obtain the maximal intensity and the optimal profile of the X-ray primary beam at the sample position. As it is not advisable to make significant changes to the diffractometer design by placing the optical chopper between any of these components, the chopper was mounted after the collimator. This avoids the need to alter the primary beam path and preserves the possibility to easily remove the chopper from the beam and thus allow other crystallographic experiments. However, tests show that the rapidly rotating chopper generates a significant airflow, affecting the direction of the cryostream used for sample cooling (Fig. 1) and thereby disturbing the temperature stability at the sample. The commercially supplied enclosure (Scitec Instruments Ltd; http://www.scitec.uk.com) is reasonably effective in

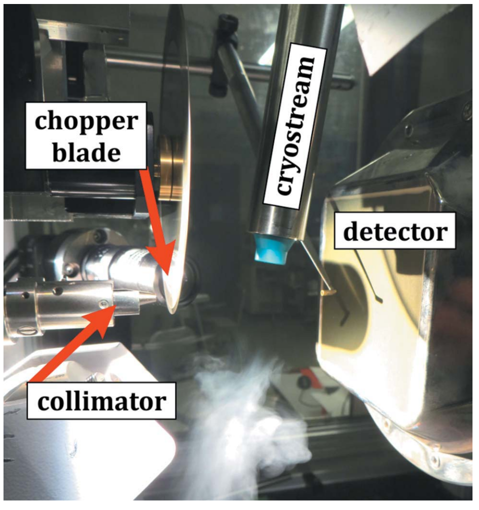

Figure 1
Optical chopper mounted in front of the collimator rotating at the chopping frequency of 50 kHz. Significant disturbance of the cryostream is clearly visible.

this respect, but its dimensions limit the oscillation range of the diffractometer circles. We therefore designed a compact enclosure with Kapton windows, the latter to further reduce the chopper-generated airstream at the sample crystal. The enclosure does not interfere with any of the three-circle goniometer axes, thus allowing the sample to be freely oriented and moved during the experiment.

3. The chopper design

Efficient chopping of the continuous X-ray beam requires a slotted blade rotating at a very high speed. For this purpose the Scitec Instruments 310CD optical chopper was selected (Scitec Instruments Ltd). With a 445-slot blade, it can chop the X-ray beam at a rotation rate of 16 000 RPM, giving a maximal chopping frequency of almost 120 kHz. The blade, about 0.25 mm thick, is made out of stainless steel (type: 304; temper: hard; chemically blackened: black zinc passivate), which perfectly absorbs the X-ray beam. The 445-slot blade supplied has slots of about 0.2–0.3 mm wide, which matches the size of our X-ray beam (Bruker AXS TXS rotating anode, Helios multilayer optics) (Fig. 2), and at the maximal RPM produces pulses of 4.17 µs length. We note here that slower processes can readily be investigated with lower rotation rates and differently designed chopper blades, as presented in our previous synchrotron work (Fullagar *et al.*, 2000).

The enclosure design consists of two parts, the main scaffolding and the cover. A technical drawing of the scaffolding is shown in Fig. 3. Both parts of the design were machined from brass. The choice of material is dictated by its excellent X-ray beam attenuation properties and ease of machining. Three cover joints were attached on the edge of the main body using an eutectic solder at appropriate positions. The cover consists of a 1/16 inch thick (1 inch = 25.4 mm)

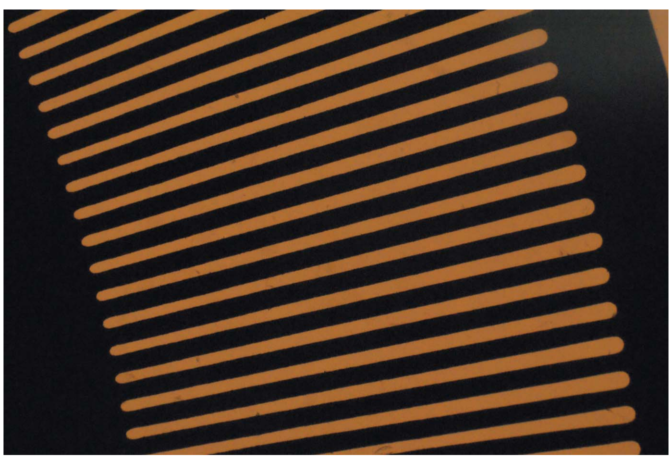

Figure 2
Microscope photograph of part of the 445-slot chopper blade.

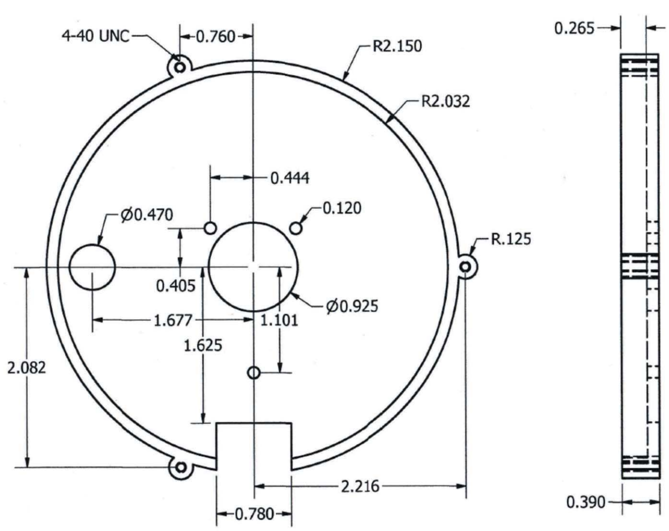

Figure 3
Technical drawing of the chopper enclosure main scaffolding. Left panel – front view; right panel – side view; dimensions in inches.

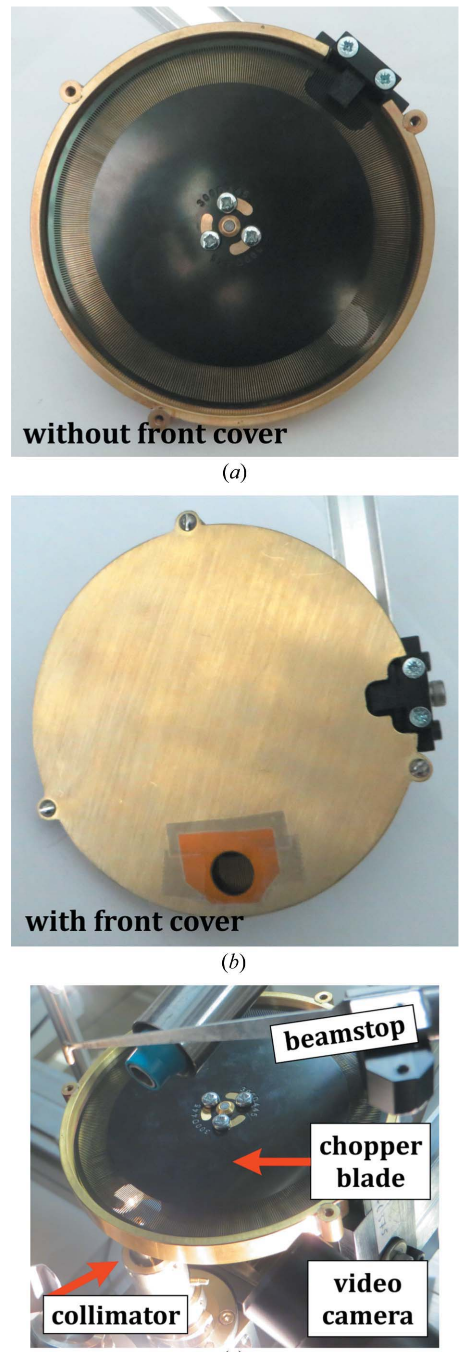

Figure 4
(*a*) Optical chopper mounted in the main enclosure scaffolding with the front cover removed. (*b*) The same as (*a*) but with the front cover installed (the X-ray window is covered with thin Kapton foil). (*c*) Chopper mounted between the collimator and the sample (tilted view; for clarity the front cover has been removed).

brass sheet (Alloy 260). Various views of the chopper are shown in Fig. 4. The front X-ray window of the chopper is covered with a thin Kapton foil to completely eliminate disturbing air flow in the vicinity of the crystal sample.

4. Testing the chopper

To examine the functioning of the chopper, an avalanche photodiode (APD; Thorlabs APD110A2) was mounted at the sample position. The chopper was set to a chopping frequency of 50 kHz, corresponding to a 20 µs duty cycle. The X-ray pulses were recorded with the APD and visualized on the oscilloscope after an accumulation time of several minutes (Fig. 5). Since the 445-slot blade has a significant jitter (∼9% of the duty cycle, according to the manufacturer), the rotor speed is not perfectly stable. Therefore, the APD readout is slightly variable and the signals are artificially broadened. As a result the true single-pulse duration of about 10 µs is broadened to about 12 µs in the diagram, as shown in Fig. 5. The image demonstrates that time-resolved diffraction experiments with microsecond time resolution are feasible at in-house sources. The pump laser pulses are synchronized with the generated X-ray pulses using delay circuit electronics. A full description of the working setup will be presented in a subsequent paper.

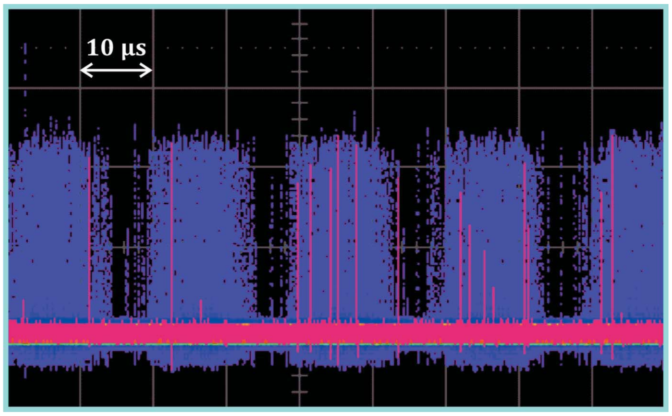

Figure 5
Oscilloscope-recorded APD signal of the X-ray beam being chopped with the frequency of 50 kHz. Single pink lines show the instantaneous APD readouts; the purple colour denotes accumulated signal.

To study the enclosure-generated background an APEXII CCD detector was positioned at 50 mm behind the sample position. 30 s single frames (see Fig. 6) were recorded with (*a*) no chopper, (*b*) the rotating chopper (50 kHz) with no enclosure, (*c*) the rotating chopper (50 kHz) with only the main scaffolding mounted and (*d*) the rotating chopper (50 kHz) with full enclosure. Is it evident that the rotating chopper lowers the average intensity of the primary beam because of the partial slot opening, but also produces powder rings originating from the rotating stainless steel blade, while the main scaffolding has a minor effect on the background. Once the front cover is mounted, an additional feature appears on the detector near the beamstop shadow. It is attributed to scattering from the front window edges. The position of the background scattering is very stable. This allows masking of the powder ring regions, as done in high-pressure crystallography (Lee *et al.*, 2014). This is of importance for the accurate recording of weak high-angle reflections, which are crucial for enhancing the quality of structure determination of transient species.

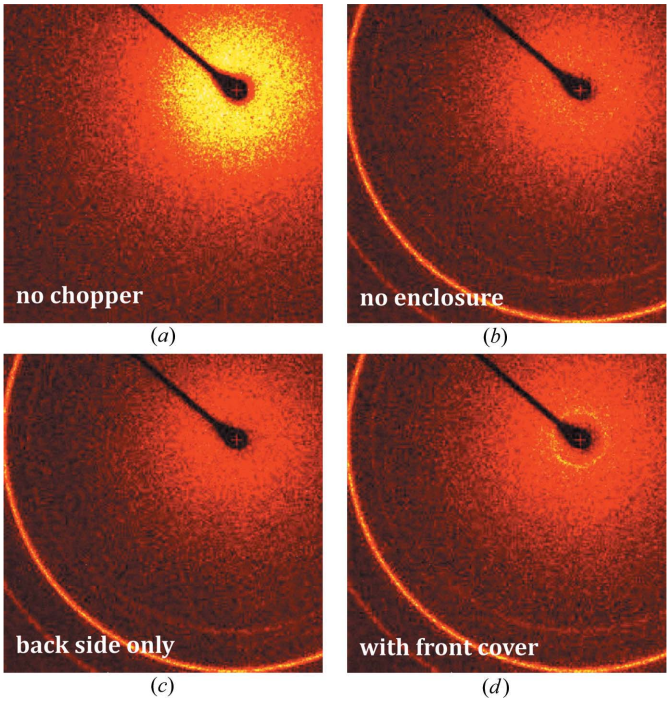

Figure 6
Single doubly correlated frames (30 s of exposure time) recorded with the CCD detector for the following cases: (*a*) no chopper, (*b*) rotating chopper (50 kHz) with no enclosure, (*c*) rotating chopper (50 kHz) without front cover, (*d*) rotating chopper (50 kHz) with front cover.

5. Summary

A chopper and enclosure design for in-house time-resolved diffraction is presented. The design eliminates the cryostream disturbance by airflow generated by the fast rotating chopper disc. Background and powder lines originating from the opening edges of the chopper need to be corrected during the integration procedure. The design is to facilitate more widespread application of crystallographic structural dynamics studies with a time resolution down to about 5 µs.

It may be noted that the availability of pixel-array detectors with their gating abilities allows elimination of a chopper (Ejdrup *et al.*, 2009). In that case, the selected time slices of the continuous X-ray beam can be used to accumulate both light-ON and light-OFF signals. Nevertheless, as the majority of diffraction studies nowadays are performed with integrating (CCD or CMOS) detectors, the presented solution allows for time-resolved photocrystallographic experiments with only minor instrument modifications.

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