DESIGN OF SINGLE-BOUNCE MONOCAPILLARY X-RAY OPTICS

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ABSTRACT

Elliptically-shaped hollow glass capillaries are the customary optic for micro X-ray beam experiments at the Cornell High Energy Synchrotron Source (CHESS). We have been able to manufacture optics that have produced x-ray spot sizes from 5 to 50 μm, gains in intensity of 10 to 500, divergences from 2 to 9 milliradians, and working distances between the tip of the capillary to the focus ranging from 20 to 150 mm. We discuss the basics in capillary design and explore the question of how well a single-bounce monocapillary can best match synchrotron sources to particular microbeam experiments, such as confocal X-ray fluorescence, microbeam powder X-ray diffraction, microbeam protein crystallography, and microbeam small angle X-ray scattering.

INTRODUCTION

X-ray focusing optics constructed of hollow glass have a large number of uses with both X-ray tube and synchrotron sources. The major reasons to employ capillary optics are to create a smaller x-ray beam size and to increase the X-ray flux density in that small spot.

There are two major branches of hollow glass optics; polycapillaries and monocapillaries. Both of these optics function by total external reflection of the X-rays from the inner walls of the glass at glancing angles. Polycapillaries have multiple small channels that guide X-rays by multiple internal reflections along the channel’s inner wall [1, 2]. Polycapillaries have focal spots ranging from 10 to 50 microns and have the major advantage of being able to collect large solid angles of X-rays [3]. Monocapillaries have two sub-categories, condensing capillaries and the single-bounce, ellipsoidal-shaped capillaries. The ellipsoidal shaped mono capillaries focus the X-ray beam by only one reflection of the X-ray beam from the inner surface of the channel. At CHESS we have demonstrated spot sizes ranging from 5 to 50 microns and controlled divergences of 2 to 9 mrad. In this paper we will describe in more detail the single-bounce monocapillary optics and their applications.

SINGLE-BOUNCE MONOCAPILLARY OPTICS

The single-bounce capillary optics are ellipsoidal shaped. A single reflection from the inner wall guides the rays to a focus. When inner glass surface is at or below grazing incidence. For glass, the critical angle for total internal reflection is roughly \( \theta_c = 32 \text{ keV} \times \text{mrad} / E_c(\text{keV}) \); i.e. if you want to reflect up to an energy of \( E_c = 16 \text{ keV} \), then the critical angle for the glass is about 2 milliradians (mrad) or 0.11°. For the ellipsoidal optic, the x-ray source is at one foci of the ellipsoid and the x-ray beam focus is at the other foci, as shown in Figure 1.
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Figure 1. The ratio of the source to reflection (F1) and the reflection to focus distance (F2) is the magnification (Magnification=F2/F1). In this figure, the image is ½ the size of the source. In practice, magnifications can be 1/100 to 1/1000. (monochromator not shown).

Because there is just one reflection from the inner surface of the glass, the inner ellipsoidal shape of the glass controls the focal spot location and the divergence of the beam. Also, because there is just one bounce, the optic is almost 100% efficient. Since the inner capillary shape is designed; a range of focal spot locations and beam divergences can be made (Table 1).

Divergences of fabricated optics range from 2 mrad to 9 mrad, which can range up to 4 times the critical angle for glass. Focal distances vary from 20 mm to 150 mm past the tip of the capillary. We have produced focal spots ranging from 5 μm to 50 μm FWHM depending on the capillary design, quality, source size and experimental setup. The focal spot size is independent of the x-ray energy, in contrast to the polycapillary, whose beam size grows larger as the x-ray energy is lowered. With low divergence designs, the optics can focus up to energies of 64 keV. We have found that an optimal length for an ellipsoidal shaped optic is about twice the length of the focal length [5].

<table>
<thead>
<tr>
<th>Capillary name</th>
<th>Type</th>
<th>Length (cm)</th>
<th>Base/Tip IDs (µm)</th>
<th>F (mm)</th>
<th>Spot Size (µm)</th>
<th>Gain (max)</th>
<th>Div (mrad)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CH015</td>
<td>concentrating</td>
<td>22.3</td>
<td>25 / 0.8</td>
<td>&lt;0.1</td>
<td>0.8</td>
<td>100</td>
<td>5.8</td>
</tr>
<tr>
<td>BSG2, BSG3</td>
<td>focusing</td>
<td>30.0</td>
<td>400/130</td>
<td>30</td>
<td>18</td>
<td>125</td>
<td>&lt;4.0</td>
</tr>
<tr>
<td>BSG7, BSG301</td>
<td>focusing</td>
<td>5.0</td>
<td>198/125</td>
<td>30</td>
<td>12</td>
<td>75</td>
<td>&lt;4.0</td>
</tr>
<tr>
<td>A10</td>
<td>focusing</td>
<td>10.5</td>
<td>211/123</td>
<td>55</td>
<td>14-18</td>
<td>70</td>
<td>2.0</td>
</tr>
<tr>
<td>Peb605</td>
<td>focusing</td>
<td>11.5</td>
<td>827/469</td>
<td>55</td>
<td>17-23</td>
<td>455</td>
<td>8.0</td>
</tr>
<tr>
<td>SF202</td>
<td>focusing</td>
<td>5.0</td>
<td>81/44</td>
<td>22</td>
<td>16-18</td>
<td>11</td>
<td>2.0</td>
</tr>
<tr>
<td>f1b_mr9f20_01</td>
<td>focusing</td>
<td>40</td>
<td>305/190</td>
<td>23</td>
<td>8-10</td>
<td>250</td>
<td>8.0</td>
</tr>
<tr>
<td>PEB_mr8f55_02</td>
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<td>11.5</td>
<td>815/470</td>
<td>55</td>
<td>13-18</td>
<td>550</td>
<td>8.0</td>
</tr>
<tr>
<td>F3_mr6f150_03</td>
<td>focusing</td>
<td>15</td>
<td>1270/905</td>
<td>150</td>
<td>30-50</td>
<td>70</td>
<td>6.0</td>
</tr>
</tbody>
</table>

Table 1. A table of capillaries used at CHESS outlining their dimensions, focal length, spot-size, gain and divergence. The spot size is the FWHM, measured through either a 5 µm or 10 µm pinhole. The maximum gain is measured comparing the flux through the pinhole at the focal spot with and without the capillary optic. The last three optics have been made with the new capillary puller.

The present capillary optics spot sizes are limited by the average inner surface slope errors and the straightness of the optic. These parameters are an important measure of the quality of the optic. In an effort to improve both the slope errors and the straightness of the optic, a new capillary puller has been built at CHESS which has been able to fabricate some high quality optics in the past year. For the first time with the new puller we were able to pull a capillary which was able to produce a 5 µm spot (Figure 3). With this new puller, we have been able to make capillaries with slope errors ranging from 40 µrad to 100 µrad and figure errors of order 1 to 5 µm rms. This level of quality is a good match for the source sizes that are available at CHESS. For other sources, such as the European Synchrotron Radiation Facility (ESRF) and the
proposed Energy Recover Linac (ERL) at Cornell, the slope errors will need to be improved by several orders of magnitude to have the spot size be limited by the source size (Figure 2). We are continuing to tune the capillary puller in an effort to produce optics with smaller slope errors.

Newly fabricated optics are evaluated in quality by both optical and x-ray methods. The first way that they are evaluated is directly on the new puller immediately after an optic is pulled. There are two Keyence LS-7010MR optical micrometers on the new puller which are used to scan the outside shape of the capillary and to give immediate feedback on the straightness and slope error. Later the capillary optics are evaluated on an x-ray beam line to both measure the spot size and to view a far field image. The spot size is measured by scanning a 5 μm pinhole across the focus. Additionally far field image is viewed on a high quality x-ray fluorescent screen about 25 cm to 75 cm down stream from the focus. This image contains information regarding the straightness and the slope errors of the optic (Figure 3).

There are four major parameters that need to be addressed for an experiment using the single-bounce capillary optic. They are the focal spot size, the gain, the divergence of the beam and the focal length of the optic [6]. The typical tradeoffs in design are that a smaller spot requires a smaller focal length and that a smaller divergence results in a smaller gain. For example, an
experiment may need additional room between the tip and the focus for a small ion chamber or a silicon channel-cut optic. In these cases we can design an optic with a longer focal length at the sacrifice of a smaller spot size. For micro x-ray fluorescent experiments, the spot size, and gain are most important; the working length and the divergence are usually not as critical. For x-ray diffraction experiments, the divergence of the beam is most important and has to be set to the experimental requirements. For small-angle x-ray diffraction experiments, the lower divergence needed will result in a lower gain. We can design the optic to optimize the experimental requirements. It is the designable aspect of the capillary’s manufacturing that has allowed it to be valuable in a large variety of experiments at CHESS.

EXAMPLES OF EXPERIMENTAL APPLICATIONS AT CHESS

A wide range of experiments use the single-bounce monocabillaries at CHESS. Some recent examples are x-ray high pressure powder diffraction, high resolution micro-diffraction, time-resolved powder diffraction on reactive foils, protein crystallography, SAXS, x-ray fluorescent imaging, and confocal x-ray fluorescent imaging [7-15]. We will briefly cover two of them, high pressure powder diffraction and the SAXS time-resolved folding of proteins and RNA.

For high pressure powder diffraction, the single-bounce capillary optic provides an increase in signal, a controlled divergence which is needed to resolve closely spaced lines and a small spot for the limited high pressure sample volumes. Small sample volumes are the major motivation for using the optic. We checked the feasibility of using the capillary optic for high pressure powder diffraction on two known samples: a NIST standard Lanthanum Hexaboride (LaB$_6$) at atmosphere and Structure I (cubic) Xenon Clathrate Hydrate (Xe$_8$(H$_2$O)$_{46}$) at 1.4 GPa [8]. With the capillary, the diffraction intensity increased by a factor of two and the FWHM of the diffraction peaks were only increased by 10% to 20%. The line widths allow for calculating lattice spacings on the order of one thousandth of an Angstrom.

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a 2 mrad divergence at a beam energy of 25 keV. The diffraction rings of Xenon Hydrate have an exceptionally narrow line width, which made it a good candidate for evaluating the effect the capillary’s divergence on line-width broadening as observed on a MAR345 area X-ray detector. Additionally only a smaller portion of the inside surface of the capillary was used to further narrow the divergence (Figure 4). The narrow diffraction rings expanded into closely spaced double rings when the entire inner surface of the optic was used, which was undesirable. The closely spaced double rings resulted from the optic having a different divergence coming from the lower and the upper part of the optic (Figure 4). To eliminate the double rings, a 2 mm thick tungsten blade was positioned down stream of the capillary and blocked the lower portion of the X-ray beam from being focused by the capillary. This eliminated the double ring and improved the angular resolution. This feasibility study showed that the capillaries provide many advantages for high pressure work. The advantages of using the optic are the signal intensity was roughly doubled, the flux per square micron in the sample was increased by a factor of 17 and the spot size in the sample was decreased from 50 µm with the collimator to about 16 µm with the optic. The disadvantages of using the optic are the line-widths of the peaks where broadened by 10 to 20%. This test showed that the A10 optic offers clear advantages in a majority of applications and the use of the collimator is necessary only when the resolution is of the greatest importance [8].

Capillaries have also enhanced small-angle X-ray scattering (SAXS) for protein folding experiments [9-10]. The ultimate motivation for these SAXS studies is to determine the size of either a protein or RNA as it folds over time, by measuring the radius of gyration from the SAXS profile [10]. Flow cells are one method for observing time resolved reactions (figure 5). A reaction is continuously initiated a point in the flow cell (point A in the figure). The sample continuously flows down the cell and is measured with an X-ray beam further down the stream (point B in the figure). The difference in the size of the structure between the two points (A and B) shows the folding response of the protein since the time of first mixing. By changing the distance between these two points, the timing for the folding process can be resolved. The smaller X-ray beam provided by the capillary is beneficial because the time resolution is in part affected by the width of the X-ray probe. Also, many of the sample solutions are very limited in volume. With an x-ray spot on the order of 15 to 20 µm provided by the optic, a much smaller flow cell can be accommodated to limit sample consumption.

To test the capillary used in conjunction with SAXS, a feasibility study was conducted using the single-bounce 4 mrad BSG644 capillary. This study measured optical performance, time resolve information was not collected for this experiment. The study compared the capillary to slits set at 100 µm by 100 µm at 9 keV with a 1.5% energy bandwidth on both a powdered silver stearate calibrant and a solution on the heme protein cytochrome c, used at a rate of 0.1 ml per minute in a flow cell (Figure 5). The flow cell channel was 2 mm and the size of the sample flowing within the center of the cell was 0.6 mm. In this experiment, again, only a smaller portion of inner surface of the capillary was illuminated to help eliminate parasitic scatter from the optic and to have a beam with a smaller divergence. The advantages of using the optic was the signal intensity was about 30% higher, in a much smaller 13 µm spot, which corresponds to a 60x increase in the flux density. There was only a small decrease in angular resolution [9]. The capillary provides a moderate increase in gain, a smaller X-ray probe that could enhance the time resolution. The smaller beam profile allows for using smaller flow cells, for example, a 1 mm
flow channel with the sample stream about 300 µm wide is presently being used. With the small spot provided by the optic, even smaller scale micro-flow cells could be used in the future [10]. Experiments using the capillary optic with the flow cell to obtain folding reaction times for both protein and RNA are presently on-going and have not yet been published.

Figure 5. The top left graph is a comparison of the resolution and intensity from a 100 µm by 100 µm slits vs. a mono-capillary with a 13 µm spot on a 10 sec exposures of silver stearate, a calibrant. The FWHM for the silver stearate peak was 0.0046 Å⁻¹ for the slits and 0.0053 Å⁻¹ with the capillary; this angular resolution does not limit the measurements [8]. The bottom left graph is a comparison of the resolution and intensity from the same 100 µm by 100 µm slits vs. a mono-capillary with a 20 µm spot on 60 sec exposures on the heme protein cytochrome c [8]. The unfocused beam has been rescaled upward by 30% to match the intensity from the capillary, the inset diagram shows the same graph before scaling. This verified that the information collected from the capillary and the slits were not different, therefore the capillary could be successfully used to collect SAXS information. Below is shown a diagram of the sample cell in which the protein or RNA sample is mixed. As it is mixed, it flows down the tube. At point A the sample comes in contact with the buffer, and starts to folded, or change shape. At point B the sample has been in contact with the buffer for a longer period of time, therefore, the amount of folding, or reaction time is determined by the distance between point A and B.

CONCLUSION

The new capillary puller has advantages over the old capillary puller. It has been able to pull optics which have smaller focal spots, and the new puller has design room for future incremental improvements. Another immense advantage the new puller has over the old puller is the onboard optical metrology which allows rapid 30 minute feedback on the freshly fabricated optic. This feed back of optic qualities such as the profile errors, slope errors, and straightness are much more detailed than the metrology done on a microscope previously.

There are many reasons that elliptically shaped capillaries have proven useful at CHESS. The optics can produce small spots at a wide range of energies (5-60 keV). They also have no chromatic aberration; therefore the spot size does not change over this wide range of energies. They have a designed divergence ranging from 2 to 9 mrad, which is independent of energy, as long as the critical angle is not exceeded. The capillary’s divergence can be further tuned with other components, such as up stream slits, to further lower the divergence of the resulting beam. The divergence for elliptical capillaries is at, or below the critical angle for total external reflection. This is the not the case for polycapillaries, where the divergence of the beam is at the critical angle for collimating optics and greater that the critical angle for focusing optics, due to
multiple reflections down the curved guiding channels. Because the divergence is a designed parameter, you can have an optic match the needs of an experiment’s requirements for angular resolution. For both experimental examples outlined, we were able to increase the signal, and allow for much smaller sample sizes, with only a slight decrease in the angular resolution.

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