MONOCAPILLARY OPTICS DEVELOPMENTS AND APPLICATIONS

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ABSTRACT

Hollow glass tubes are in use at Cornell High Energy Synchrotron Source (CHESS) as X-ray optics in the 4 to 25 keV range to take large diameter, fairly parallel X-ray beams and compress them into smaller diameter, more divergent beams for various kinds of X-ray applications. The optics come in two forms: 1) multibounce condensing capillaries for making submicron diameter beams and 2) one-bounce imaging capillaries with working distances of 20 to 50 mm from the tip of optic to the focal spot. The preliminary results of the applications, from X-ray fluorescence imaging to protein crystallography, are briefly described in this paper.

INTRODUCTION

It has been demonstrated by a number of research groups and companies over the last decade that hollow glass capillaries (operating at glancing angles by total reflection from their inner walls) are very useful focusing optics in synchrotron X-ray instrumentation [1-4]. Generally speaking, the polycapillary forms [5-7] are most useful for beam sizes greater than 40 microns and the monocapillary forms are especially good for micron sized beam applications [8-15]. There is strong overlap of the two types of optics for sizes between 1 and 40 microns.

By using glass capillaries, one can create a smaller beam size as well as increase the X-ray intensity per unit sample area. There are basically two different kinds of monocapillaries: condensing and imaging. In this paper we will first describe the state of condensing capillary developments and applications at CHESS and then discuss the imaging capillary progress.

CAPILLARIES DEVELOPED AT CHESS

For a condensing monocapillary, the X-rays passing through may be reflected many times by total reflection from the hollow inner wall of the capillary before they pass out the tip, as shown in figure 1. The X-ray beam size produced by a condensing capillary is mainly dependent on the opening of the capillary tip and not strongly related to the capillary figure. Therefore it is relatively easy to make sub-micron scale X-ray beams for experiments. One condensing capillary developed at CHESS and currently available for users has a 0.8 micron opening at the tip, a 50 micron opening at the capillary base entrance, and a length of 223 mm.

For an imaging capillary, all rays will be focused into a single focal point at some distance beyond the tip as shown in figure 2. The focal spot size of a one-bounce capillary is basically a demagnified image of the synchrotron source blurred out slightly from small residual slope errors from capillary manufacture. This size is not directly related to the capillary tip opening. The elliptical shape chosen depends on the capillary manufacture accuracy, required focal spot size, maximum focal divergence allowed, working distance and X-ray source parameters [16].
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have developed various sizes of one bounce capillaries, with spot sizes from under 10 µm to about 20 µm, focal distance from 30 mm to 55 mm, divergence angles from 2 mrad to 12 mrad, and the optimal elliptical figures appropriate for CHESS A2, B1, B2, D, and F2 stations using our in-house drawing machine [4]. The major parameters of the capillaries now available at CHESS are listed in table 1.

Figure 1. Schematic description of the X-ray reflections inside a condensing capillary. The X-ray beam size after a condensing capillary is dependent on the tip opening of the capillary. The X-ray spot increases as the sample to capillary distance is increased due to the beam divergence produced by the capillary. Typically, the sample needs to be placed within 10 to 100 diameters of the tip, i.e. to within 10 to 100 microns for a 1-micron opening capillary.

Figure 2. Schematic description of X-rays reflecting only once inside an imaging capillary; all rays are focused to one point. Here the focal spot size is dependent on the X-ray source parameters and capillary slope errors, but not on the capillary tip opening size.

Table 1. A brief list of capillaries in use at CHESS for fluorescence, spectroscopy, and diffraction applications in the energy range of 4 to 25 keV.

<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>Size (µm)**</th>
<th>G max ***</th>
<th>Div (mr)#</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>
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<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>
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<tr>
<td>BSG625</td>
<td>focusing</td>
<td>10.5</td>
<td>407/233</td>
<td>55</td>
<td>21</td>
<td>80</td>
<td>&lt;4.0</td>
</tr>
<tr>
<td>BSG631</td>
<td>focusing</td>
<td>10.5</td>
<td>292/162</td>
<td>40</td>
<td>20</td>
<td>10</td>
<td>&lt;4.0</td>
</tr>
<tr>
<td>BSGF413</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>
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<td>BSG7</td>
<td>focusing</td>
<td>28</td>
<td>800/300</td>
<td>25</td>
<td>20</td>
<td>150</td>
<td>&lt;12.0</td>
</tr>
<tr>
<td>BSG301</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>W102</td>
<td>focusing</td>
<td>28</td>
<td>800/300</td>
<td>25</td>
<td>20</td>
<td>150</td>
<td>&lt;12.0</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>
</tbody>
</table>

* F is the working distance from the capillary tip to the focal point (in mm).
** Size is the measured FWHM at D1 station convoluted with 10 µm pinhole, except BSG413 that was measured at the F2 station and A10 at the A2 station. (In effect, the real beam sizes produced by BSG7 and BSG301 should be approximately 10 µm smaller). The size measurement for CH015 was measured using a knife-edge scan.
*** G max is the maximum intensity gain at the image center measured using a 10 µm pinhole at D-line, with and without a capillary, except BSG413 which is measured at F2 station, A10 at A2 station and CH015 where the intensity has been scaled from knife-edge measurement.
# Div is the designed full divergence (Divergence of CH015 is measured value).

For one-bounce capillaries, the average slope errors are an important parameter influencing the overall optical quality. At CHESS, we have measured the one-bounce capillary inside surface
slope errors and confirmed that the measured value agrees with our capillary image simulation code[17]. Our current capillary overall slope errors are about 70 to 145 microradians, comparable to the CHESS X-ray source divergence before focusing. Further improvements to reduce the slope errors even further are under investigation and could lead to higher gains and smaller spot sizes.

**SOME APPLICATIONS WITH A CONDENSING CAPILLARY AT CHESS**

Two microbeam experiments using condensing capillaries illustrate some of the many possible applications. The first experiment measured the zinc distribution in a single layer of plant leaf cells and the second involved measuring the gallium to arsenic compositional profile ratio in a thin semiconductor film.

During a single layer plant cell experiment, one layer of "Thlaspi Caerulescens" leaf skin was peeled off and placed in front of the "CH015" condensing capillary. The zinc fluorescence was detected with a Si(Li) detector. Figure 3 shows an example data image [18]. Images like these raised interesting biological questions, such as “why are the strands of concentrated zinc several times larger than the size of individual cell?” These data were taken in collaboration with research group of Leon Kochian at Cornell University. The spatial resolution on this image was limited by the minimum motorized stage step size of about 3 µm.

![Figure 3. Zinc fluorescence from a single layer of plants cells (Thlaspi Caerulescens).](image)

The second example from solid-state physics illustrates materials characterization of InGaAs and InGaAsP semiconductor layers grown on oxide-patterned substrates in the narrow gaps between SiO₂ stripes. These areas, several microns in width, are important building blocks for modern optoelectronic devices. Information on the semiconductor layer composition (In, Ga, As, and P concentrations) is needed with a submicron in-plane resolution for understanding the growth kinetics in the regime of selective area growth (SAG). This knowledge is of fundamental importance and useful to help characterize the behavior of industrial metal-organic vapor phase epitaxy (MOVPE) reactors.

We investigated properties of the SAG layers of InGaAsP and InGaAs (0.3 µm thick) grown by MOVPE on InP substrates utilizing microbeam X-ray fluorescence at CHESS. The arsenic and
phosphorus composition stayed constant across the whole structure. In contrast, Ga and In concentrations varied near the oxide mask, where the diffusion process for In and Ga was influenced by the mask during the growth. In this example, the change in Ga composition was determined to be approximately 5% with a measurement error below 0.5%.

Figure 4. (a) Map of the Ga composition in InGaAs layer determined from the Ga fluorescence intensity taken with about 1µm spatial resolution using capillary CH015 at the D1 station. The dark regions in the map correspond to the oxide mask. (b) Map of the InGaAs layer thickness determined by the arsenic fluorescence intensity [19].

EXAMPLES OF IMAGING CAPILLARY APPLICATIONS AT CHESS

In addition to X-ray fluorescence imaging, capillaries can be quite useful for micro-crystal X-ray crystallography. For crystallography applications, we made an imaging capillary BSGF413 for the F2 station at CHESS that further focused an existing double focused X-ray beamline into a 17µm by 20µm spot size. An additional X-ray intensity gain by a factor of 10 was achieved.

Samples used for test were Lysozyme and Concanavalin A with crystal dimensions of 80-200 µm, exposure times of 5-15 seconds, an X-ray energy of 12.6 keV and with a total flux about 1.0x10^10 photons/sec (see raw CCD image in Figure 5). The crystallographic analysis yielded R-
factors of about 3-6% when processed with standard MOSFLM and DENZO data reduction software. Thus the imaging capillaries look very promising as microbeam optics for small crystal diffraction experiments.

Figure 5. X-ray diffraction image from a Lysozyme crystal taken with a Quantum 4 CCD X-ray area detector at a 100 mm film-to-specimen distance. The spots are well resolved in the image for successful integration of reflections. Low mosaic spread and adequate peak to background ratio yielded good statistics with R factor below 6%.

A second experiment used an imaging capillary to excite X-ray fluorescence from biological specimens, namely fish ear stones, to determine the concentrations of a number of elemental impurities. Fish ear stones (otoliths) are interesting to study because, as the fish grows, the stones record the life history of a fish in much the same way that tree rings record the life history of a tree. Figure 6 (below) shows an early example of a strontium concentration map that was collected by scanning the sample through an X-ray microbeam. Many other elemental fluorescence signals of interest can be detected using this technique, including (but not limited to) iron, calcium, zinc, and manganese.

Figure 6. Capillary BSG7 was used to make a 10 µm beam spot at D1 station with flux about 1.1x10^10 ph/s at 9keV. Using the second harmonic of the incident X-rays, a map of the Sr distribution inside a thin slice of ear stone was made. Sample capillary distance was 30 mm. A 1.5 year old blueback herring of 18" length was used for this image showing it swam in low Sr concentration water in its early life (blue center region) and then it went out to the Atlantic Ocean (middle red ring). Since the trace element ratios in water often vary along the length of a river, we can possibly trace out the fish life history if we can sufficiently simultaneously image the Ca, Sr, Fe, Mn, etc. content of the ear stone and compare with the known concentrations of river water. This is an exciting prospect for environmental scientists. The images of other elements have recently been made and will appear in a future publication.
CONCLUSION

In summary, there are many exciting projects that need microbeams and glass capillaries are proving useful for many applications.

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REFERENCES