J. Synchrotron Rad. (1994). 1, 37–42

X-ray Applications with Glass-Capillary Optics

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(Received 23 May 1994; accepted 23 June 1994)

Glass capillaries have unique properties for guiding X-rays in experiments with micrometer precision. Design considerations of such optics are presented for X-ray applications involving macromolecular crystallography, tomography and high-pressure experiments at the Cornell High Energy Synchrotron Source. The authors propose that crystallography with protein crystals is feasible on a 50 μm or smaller scale using capillary optics along with a cold gas stream and precision rotation stages. For computed tomography experiments, capillary optics can produce X-ray beams on a submicrometer scale. The distribution of X-rays passing through the sample can then be blown up in size with a secondary capillary optic to match the ~10 μm pixel size of CCD detectors. For high-pressure experiments in diamond-anvil cells, mono- and polycapillary optics may provide 1–50 μm diameter beams for diffraction or X-ray absorption fine-structure applications.

Keywords: capillary optics; microbeams; total-reflection optics; microdiffraction; tomography; diamond-anvil cells; X-ray optics; high pressure.

Introduction

Capillary optics are rapidly evolving from novel X-ray optical components into standard beamline devices at synchrotron radiation facilities. As the capillary techniques are so new, we present a short review of the properties of capillaries. We anticipate that capillary optics will have a wide impact on synchrotron radiation research including the fields of X-ray crystallography (monochromatic and Laue), spectroscopy, fluorescence, imaging, tomography, high-pressure diffraction, *etc.* After the brief review, we discuss important design considerations in capillary applications to the specific areas of macromolecular crystallography, computed tomography and high-pressure diffraction, all areas of active research at CHESS.

Single-stranded tapered glass capillaries have efficiently concentrated monochromatic synchrotron radiation as well as broad-band 'white' synchrotron radiation (Thiel, Stern, Bilderback & Lewis, 1989; Engström *et al.*, 1991; Thiel, Bilderback, Lewis & Stern, 1992*a*). Capillaries are but one promising X-ray optical component that should soon allow many X-ray experiments traditionally conducted with large samples or with millimeter size beams to be undertaken on a much smaller scale. Other optical components such as multilayer mirrors (Underwood, Thompson, Wu & Giauque, 1988; Thompson *et al.*, 1988), zone plates (Lai *et al.*, 1992; Yun *et al.*, 1992), and Bragg–Fresnel lenses (Bonse, Riekel & Snigirev, 1992; Riekel, Bosecke & Sanchez del Rio, 1992; Kuznetsov, Snigireva, Snigirev, Engström & Riekel, 1994) are also under development for microbeam applications. In a related technology employing curved bundles of glass capillaries, Kumakhov & Komaorov (1990) have made X-ray beams on a millimeter-size scale.

Properties of capillary optics

A tapered capillary concentrator produces a small diameter X-ray beam from a larger beam as it passes through the long tapered needle-like structure. The X-rays are compressed in cross-sectional area as they successively reflect from the smooth inner wall of the device, Fig. 1. Like any other optical component, a capillary concentrator produces a smaller more-intense beam at the expense of divergence.

These optics are interesting because they are so simple and have potential to be replicated inexpensively. Beam sizes down to 0.05 μm (500 Å) (Bilderback, Hoffman & Thiel, 1994) have been made and gains in flux/area have been measured as high as 960 (Hoffman, Thiel & Bilderback, 1994*a*) with monocapillaries of leaded glass. In terms of focusing power, the performance of monocapillary optics *versus* that of optics made from classical X-ray mirrors and monochromator crystals is quite comparable for 100 μm diameter beams. The traditional optical methods, however, do not fare well for diminishing beam sizes.

Monocapillaries create a beam whose size is determined by the inside diameter of the glass tubing and not by the X-ray source size or the aberrations in the optical device

Figure 1
An X-ray beam can efficiently be transmitted through a capillary concentrator if reflection from the smooth walls takes place at angles less than the critical angle of reflection. For hard X-rays and typical borosilicate glasses, the critical angle for 10 keV X-rays is about 3 mrad or 0.2°.

itself, as would be true for the non-capillary type of optics mentioned earlier. In principle, capillaries should come close to preserving the brilliance of the X-ray source if the optics are designed to pass rays so that the critical angle for the X-rays is not exceeded, the number of bounces are not too many, and the shape of the reflecting wall is close to the ideal parabolic shape. Even if the component fabrication is not perfect, the exit beam size is still equal to the inside diameter of the tip, and only the throughput is possibly reduced (*i.e.* resulting in a loss of brilliance).

Since the critical angle for X-rays scales inversely with photon energy, there can be an energy-dependence term associated with the gain in X-ray flux/unit area. Engström *et al.* (1991) have shown this not to be an unreasonable problem for a capillary that creates a 0.1 μm beam size. Compared to a 20 × 20 μm aperture, their group calculated a gain in flux density of 100 at 60 keV and of 10 000 at 10 keV. These values are large enough to make optical designs at these high energies very interesting.

Experimental gain measurements have been made at Cornell University on a 1.6 m long hollow glass tube which accepted a 470 μm diameter beam and compressed it to 110 μm (Thiel, Bilderback, Lewis & Stern, 1992*b*). The borosilicate glass capillary was fabricated with an outer plastic jacket and was stretched taut between two supports to straighten it before performing the experiments. Irregularities in the taper manufacture limited its performance somewhat; nonetheless, it had a measured efficiency of 49% for 8 keV, 54% for 13 keV and 34% for 20 keV X-rays, and an overall gain of 6–10 in flux density/area.

One other important consideration with capillary optics is the 'depth of field' of the concentrated X-ray beam. For the monocapillaries that we have currently fabricated, the beam is smallest at the tip and the sample should be located no further away than 20–100 times the tip diameter (Bilderback *et al.*, 1994). For a 1 μm beam size, this working length amounts to 20–100 μm.

Alternatively, a bundle of channels can be pulled to create condensing optics (see Fig. 2) of a slightly different nature than that of monocapillaries. Such a device is significantly more compact in length than the comparable monocapillary optic. In addition, it allows the focal spot to be displaced by as much as a few centimeters from the tip (Hoffman, Thiel & Bilderback, 1994*b*). One drawback, however, is that these devices are not capable of producing the ultra-small beam sizes attainable with monocapillary devices.

In theory, tapered capillaries can be quite efficient in condensing down the radiation to a small size. Intensity gains (flux/area) for ideal optics can be exceedingly high, of up to 100 000 for an undulator source of radiation (Thiel *et al.*, 1989). In reality, capillaries of suitable figure (taper and straightness) are not easily produced and different groups are trying to perfect their fabrication. Furthermore, in the initial manufacturing of polycapillary tubing, great care must be taken to assure that all the parallel channels are without major defects. X-ray transmission is reduced by channels, holes, bumps and microscopic surface roughness in the thin-wall structure of the glass.

Macromolecular crystallography

While X-ray protein crystallography (Helliwell, 1992) continues to grow as the most powerful structural technique in molecular biology, one experimental aspect has failed to receive much attention – that of reducing the crystal sample size. Presently, crystals much smaller than 100 μm on an edge are generally considered unsuitable for X-ray diffraction measurements. This perceived limit is primarily based on earlier technological shortcomings, many of which have now been overcome by utilizing advanced X-ray sources and better optics.

At the heart of a macromolecular microdiffraction camera is the optics. Although not the only choice, the most promising optical components to date are capillary concentrators. These optics can be used with both monochromatic X-rays for standard oscillation photography and broad-band polychromatic radiation for Laue diffraction (see Fig. 3).

For the simplest capillary design, a small tip size is chosen to match the smallest dimension of the crystal sample. Other parameters to be chosen are the capillary divergence (determined by the slope at its tip) and the amount of beam compression desired. For example, a tapered monocapillary for use in protein crystallography with 15 keV X-rays might have a slope at the tip of 1 mrad, a 500 μm entrance diameter, a 50 μm exit diameter, and an overall length of 1.2 m. Here the sample would need to be placed 1–5 mm away from the tip of the capillary.

Since the X-ray dose increases as the crystal volume shrinks, the camera must also accommodate a nitrogen cold stream directed at the sample to reduce radiation damage

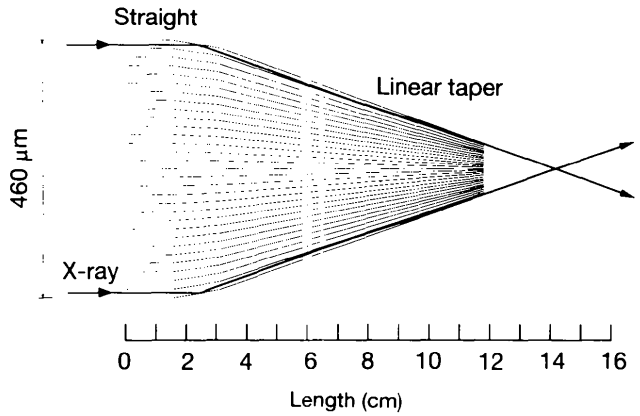

Figure 2
Schematic diagram of X-rays passing through a perfect polycapillary concentrator. A device has been used to condense 6 keV X-rays to a diameter of 68 μm while enhancing the intensity (flux/area) by a factor of 5 (Hoffman *et al.*, 1994*b*). Before pulling, the concentrator consisted of 330 parallel tubes of 18 μm inner diameter and 2 μm wall thickness (Gibson, 1994).

effects (Hope, 1988; Hedman, Hodgson, Helliwell, Liddington & Papiz, 1985). The particular radiation sensitivity of a crystal may turn out to be the factor that limits the smallest beam size that is practical to use for a given microbeam experiment.

In one preliminary Laue diffraction experiment (Bilderback *et al.*, 1994), a 5.6 μm diameter beam was used for Laue diffraction from a lysozyme single crystal on a CHESS bending-magnet beamline. Diffraction was observed over a 5–25 keV range of energies and the sample diffracted to a resolution of 2.2 Å. The sample was uncooled and had to be translated during the exposure to periodically expose fresh crystal volume to the X-ray beam.

For monochromatic oscillation experiments, it may be desirable to have a larger working distance than can be provided with monocapillaries. This can be achieved by a somewhat more sophisticated capillary optic – the tapered polycapillary, an optic that has a working distance of many centimeters, but typically is more difficult to fabricate.

A capillary-based microbeam oscillation camera requires that the crystal is easily centered in the small beam and that it does not wander out of the beam when rotated. Precision translation and rotation stages are needed for this purpose and are available commercially. An alternative idea is to work with existing rotary stages that have only a small wobble in the rotation axis and to compensate by translating the sample back into the beam after the crystal has been rotated.

For Laue diffraction experiments, the positioning requirements of the camera become somewhat less stringent. In some cases, radiation damage is controlled simply by reducing the exposure time with a fast shutter. Such fast diffraction techniques also open the possibility of time-resolved studies. For example, the small volume of the microcrystal along with the short duration of a rapidly shuttered X-ray beam bodes well for a laser-initiated structural measurement where the laser beam needs to adequately penetrate the sample in order to excite the sample fully.

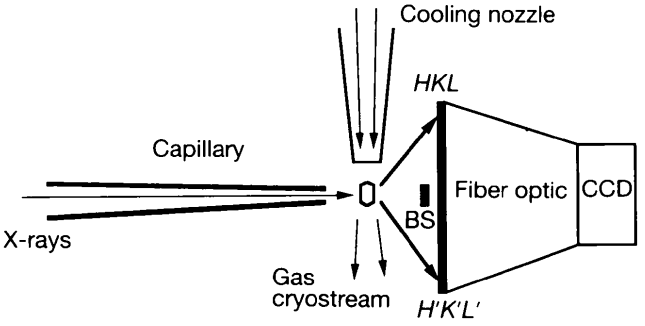

Figure 3
X-rays are directed by a capillary onto a very small single crystal protein sample of order 20–50 μm. The crystal is bathed by a low-temperature cryogenic gas stream to minimize radiation damage. Two reflections, *HKL*, and *H'K'L'* are shown intercepting the X-ray phosphor of a CCD area detector at wide angles. The beam stop, BS, prevents the transmitted beam from damaging the detector.

Finally, because the diffraction signal from a small crystal volume is likely to be weak, it may be advantageous to use a charge-coupled device (CCD)-based area detector. In particular, at CHESS an X-ray detector (Tate *et al.*, 1994) having a 50 × 50 mm active area directly coupled by a 2.6:1 reducing fiber-optic bundle to a 1k × 1k format CCD chip has proven to be extremely effective in a number of macromolecular crystallographic experiments involving 100 μm and larger crystals (Walter *et al.*, 1994), particularly for the recording of relatively weak, high-resolution reflections. The effective spatial resolution of this detector (80 μm) is fairly well matched to the spot size from microcrystals; plus, its large dynamic range aids in accurately recording the weak and the strong reflections simultaneously. Hence, this particular detector appears ideal for the recording of weak diffraction data from microcrystals.

CCD-based detectors may not be the only detector of choice. Snell *et al.* (1995) have also found image plates to be effective detectors of Laue patterns from a small crystal of Ni containing aluminophosphate of 20 × 25 × 250 μm dimension. Other detectors such as X-ray film and gas-filled proportional counters should find a useful role for detecting X-rays in various microbeam crystallography experiments.

Computed tomography

Computed tomography (CT) is a very powerful nondestructive tool with applications in biology, medicine, materials science, geology, *etc.* that can be used to determine the three-dimensional density map of an object. Of particular interest is the X-ray tomographic microscopy (XTM) technique of imaging in three dimensions at high spatial resolution. Instruments have been constructed at various synchrotron radiation laboratories [DORIS (Bonse *et al.*, 1991), SSRL (Kinney & Nichols, 1992) and NSLS (Spanne & Rivers, 1987)].

A variety of methods are used to obtain images having micrometer resolution. One approach utilizes microbeams with a size of less than 10 μm to map the elemental distributions based on the detection of transmitted or fluorescent X-rays. Briefly, an X-ray beam is collimated with tantalum slits. The object is then repeatedly scanned across the beam and rotated by a small angle between each scan to acquire the complete data set of projections necessary for the computed reconstruction. The most frequently chosen X-ray detectors for this application are NaI detectors or ionization chambers. The main obstacle for higher resolution has been the low photon flux as the beam size is reduced. Capillaries can potentially overcome this limitation since they intensify the X-ray beam onto a very small (micrometer size) focal area. For maximum compression of the beam size, a two-stage condenser may be useful, Fig. 4. The use of a dual-stage capillary optic has some technical advantages. It is much easier to control the optical parameters of capillaries of shorter length during manufacture and the capillary setup is also simplified since each part can be aligned separately.

The divergence of the capillary-focused beam offers the opportunity to improve the performance of the tomographic microbeam experiment even further. The magnification of the X-ray image caused by the fan-beam geometry in this instance could be used to enhance the spatial resolution, especially if it is employed with an area detector made from a CCD. At present, CCD X-ray detectors are the ultimate area detectors for microtomography applications, possessing a high spatial resolution (of order 10 μm) and reasonably high detection quantum efficiency (DQE) of greater than 40% (Eikenberry, Tate, Bilderback & Gruner, 1991).

An alternative approach for achieving high-resolution XTM has been developed by Bonse *et al.* (1991). This group uses an asymmetric channel-cut crystal to increase the X-ray beam profile behind the object. As this magnification is connected with a decrease in photon flux/area, it is usually applied only in one direction resulting in a distorted image of the specimen.

It may be possible to overcome the disadvantages of this system with tapered polycapillary optics. A spatial resolution of 0.5 μm may be achieved using a CCD detector with a 10 μm pixel size by employing a polycapillary expander which will magnify the image 20 times, Fig. 5. The transmitted intensity is collected at the tip next to the object and guided by total reflection towards the other end. A phosphor coupled to this enlarged end is used to convert the X-ray image into visible light for registration with a CCD array. If perfectly tapered, this polycapillary optic performs a two-dimensional magnification without distortion.

In addition, one could also use another tapered polycapillary to intensify the incident flux onto the object. By constructing parallel sections at the tip of each capillary around the sample, one could discriminate against scattered radiation and might further improve the image contrast (see inset of Fig. 5).

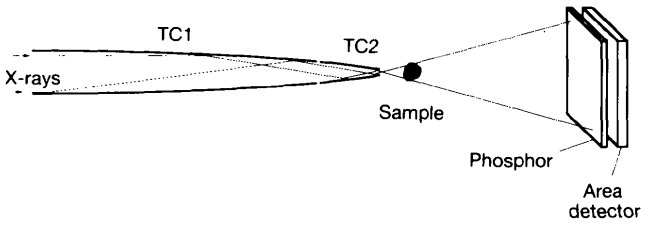

Figure 4
Projection tomography with a divergent microbeam made from capillary optics. A two-stage capillary condenser consisting of two tapered capillaries, TC1 and TC2, may be desirable since it is very difficult to pull a single glass tube precisely with an inside diameter that varies from millimeters to micrometers along its length. TC1 could be many meters long and fabricated from flexible hollow glass tubing that is stretched out straight (Thiel *et al.*, 1992*b*). TC2 would provide the final stage of compression.

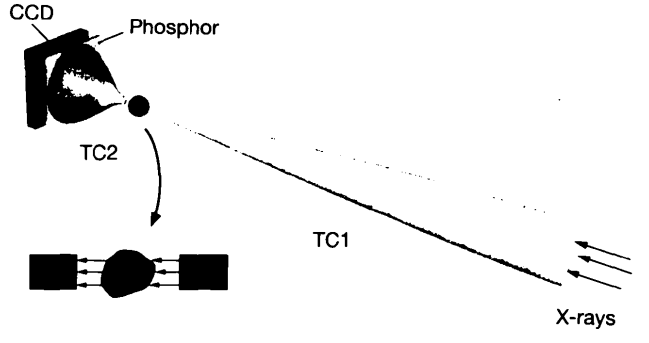

Figure 5
X-rays from a tapered polycapillary, TC1, are incident on a sample. After passing through the sample, the X-ray beam size is magnified by a second capillary, TC2, and then imaged with a CCD detector. The straight glass channels just before and after the sample form a collimating system to discriminate against Compton scattering, fluorescence, *etc.*

High-pressure diamond-anvil cell experiments

A promising application for tapered glass capillaries is for high-pressure diamond-anvil cell (DAC) experiments where a sample (single crystal or powder) is placed in a metal gasket and pressed between two diamonds (Jayaraman, 1986), Fig. 6. Because of the X-ray absorption in the several millimeter thickness of diamonds, X-rays of greater than 15 keV energy are generally employed.

Pressures greater than that at the center of the Earth (364 GPa; Dziewonski & Anderson, 1981) can be obtained in the laboratory (Xu, Mao & Bell, 1986; Ruoff, Xia, Luo & Vohra, 1990). For experiments at these pressures, there are large pressure gradients (Brister, Vohra & Ruoff, 1988), which means that the most precise measurements of the conditions in the DAC require the use of exceptionally

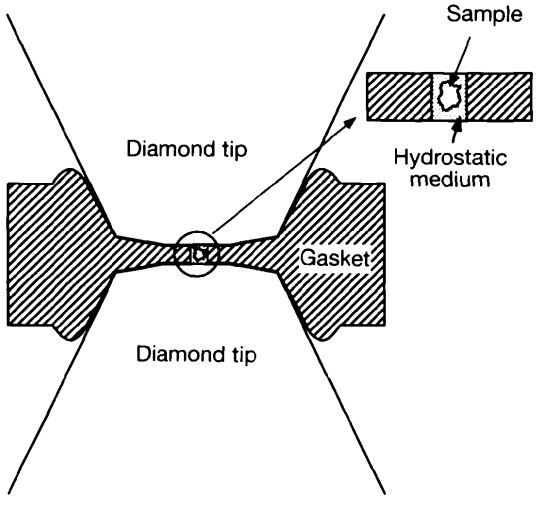

Figure 6
Schematic of a typical diamond-anvil cell. The diamonds are specially selected for low intrinsic stress and are typically 1/8 to 1/3 caret in size (several mm in diameter). The sample is loaded in a small hole (20–200 μm in diameter) which has been drilled in a metal gasket. A hydrostatic medium is often used to transmit the pressure to the sample.

small beams of X-rays: of the order of 1 μm or less is desirable although only beams of about 5 μm diameter have been used to date (Ruoff *et al.*, 1993). Unfortunately, the thickness of the diamonds (about 2 mm) prevents the capillaries from getting close enough to be effective, as the beam diverges rapidly from the tip of the capillary. There are several schemes that could be tried, such as bringing the X-rays in at a slight angle to the gasket (Fig. 7*a*), using a beryllium gasket and bringing the X-ray beam in perpendicular to the stress (Fig. 7*b*), or using a sapphire tube with a miniature diamond anvil mounted on its end (Fig. 7*c*).

Fortunately, the number of experiments that require pressures exceeding 100 GPa is quite small. Most experiments are carried out in the several GPa range where beam diameters of 20–50 μm are most useful. These experiments require no modification of the diamonds or the diffraction geometry to take advantage of tapered glass capillaries. The only requirement is to provide enough space upstream from the sample to position the capillary: as much as 1.2 m are needed to condense a beam from 500–50 μm.

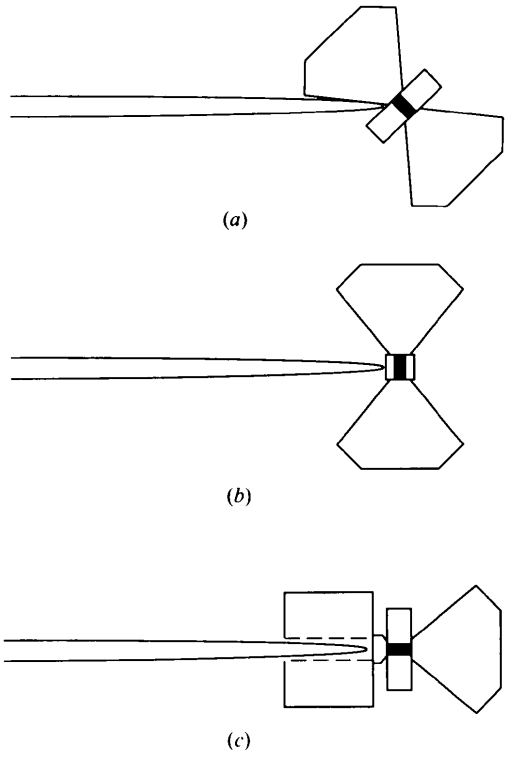

Figure 7
Possible configurations to illuminate a 0.1–1 μm diameter spot in a diamond-anvil cell. Since the capillary tip must be approximately within 100 times the exit diameter to make use of the X-ray concentrating effects, the geometry of the diamond-anvil cell must be altered to make use of the capillaries in ultra-high-pressure experiments. (*a*) The beam is brought in at a slight angle to the gasket. (*b*) The beam is brought in perpendicular to the stress axes of the diamond-anvil cell. The gasket would be made of beryllium. (*c*) One diamond tip is replaced by a miniature diamond while the bulk part of the diamond is replaced by a sapphire or tungsten-carbide tube.

This work is based upon research conducted at the Cornell High Energy Synchrotron Source (CHESS) which is supported by the National Science Foundation (DMR-9311772) and the National Institutes of Health (RR 01646). One of the authors (RP) acknowledges a fellowship from the Alexander von Humboldt Foundation, Germany.

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