1. Abstract

Polycapillary optics, shaped arrays consisting of hundreds of thousands of hollow glass capillary tubes, can be used to redirect, collimate, or focus x-ray beams. X rays emitted over a large angular range from conventional, laboratory-based sources can be transformed into a beam with a small angular divergence or focused onto a small sample or sample area. Convergent beams of x-rays, with convergence angles as high as six degrees, have been produced using polycapillary x-ray optics. Focused spot sizes as small as 20 μm have been achieved, with flux densities more than two orders of magnitude larger than that produced by pinhole collimation. For microdiffraction studies, this results in a comparable decrease in data collection times due to the increase in direct beam intensity and reciprocal space coverage.

In addition, the optics can be employed to reduce background and provide more convenient alignment geometries. The inverse dependence of the critical angle for total external reflection on photon energy results in suppression of high energy photons. This effect can be employed to allow the use of higher tube potentials to increase characteristic line emission and also has been employed to significantly increase the Kα/Kβ ratio in Cu radiation.

Measurements of x-ray diffraction data and crystallographic analysis have been performed for systems ranging from elemental crystals to proteins. Data from a lysozyme protein “standard” with a slightly convergent beam, taken in 3 minutes per frame with 2° oscillation with a 2.8 kW source, refined to intensity variance of 5%. High quality data was also obtained with a 0.03 kW fixed anode source and a 2° convergent lens in 5 minutes per frame.

2. Introduction to Polycapillary Optics

2.1. Description

Polycapillary optics are arrays of hollow glass tubes used to collect, focus, and redirect x-ray and neutron beams. These optics guide x rays by total external reflection from the capillary surfaces at very small angles in the manner of grazing incidence mirrors commonly used in synchrotron beam lines and x-ray telescopes. The critical angle for total external reflection from glass surfaces, θc, is 3 mrad for 8 keV photons and is inversely proportional to photon energy. X rays can be transmitted down hollow glass tubes with high efficiency so long as the incidence angles are kept smaller than the critical angle. As shown in Figure 1, this limits the maximum permissible channel diameter. This consideration leads to the use of polycapillary fibers with hundreds or thousands of hollow channels with diameters ranging from 4-50 μm. A micrograph of a typical fiber cross section is shown in Figure 2.
Thousands of such fibers are strung through lithographically produced metal grids to produce a multifiber lens, such as shown Figure 3. The fibers are parallel at the output (visible) end and point toward a common focus at the input end. Alternatively, a larger diameter polycapillary fiber can be shaped into a monolithic optic, as shown in Figure 4. Monolithic refers to the one piece nature of the optic, which typically has hundreds of thousands of channels.

2.2. Collimation

2.2.1. Theory

The output from a multifiber polycapillary collimating optic has both global divergence, $\alpha$, and local divergence, $\beta$. Even if the fibers are parallel ($\alpha=0$), the output divergence, $\beta$, is not zero, but is determined by the critical angle. The exit divergence from capillary optics is typically measured by rotating a high quality crystal in the beam and measuring the angular width of a Bragg peak. Since the Darwin width of the crystal is much smaller than the exit divergence from the optics, the measurement yields the divergence directly. The result at 8 keV for a 5 mm diameter monolithic collimating optic, is a full width at half maximum of 3 mrad. A large multifiber optic similar to that in Figure 3 has a divergence width of 3.8 mrad at 8 keV over the full 3 cm diameter field. The small difference between the output divergence from a small optic and the full lens is a measure of the alignment of the individual fibers. The full field divergence for the lens at 18 keV is 2.6 mrad.

2.2.2. Gain Measurements

The transmission of a 3 cm square multifiber collimating lens is shown as a function of photon energy in Figure 5. An x-ray micrograph of the output of the lens is shown in Figure 6.

A smaller lens, designed for 8 keV operation, had an output area of 2 cm square, a capture angle of 7°, focal distance of 132 mm, and transmission at Cu Ka of 25%. An out of plane $\psi$ diffraction scan of a Si (100) test specimen gave a measured count rate a factor of 20 higher with the optic than with a 1.35 mm pinhole at 155 mm from the source. The resolution of the scan with the pinhole was 0.5°. The resolution with the optic was 0.34°. The detector used for this measurement was only one quarter the size of the output beam of the collimating optic, resulting in a substantial reduction of gain. A measurement with a 2 cm diameter detector would have yielded a gain of 60. A measurement with a 3 cm diameter lens, detector and sample would yield a gain of ~100.
2.2.3. Collimation for Protein Crystallography

For the small beam sizes required for protein crystallography, a different polycapillary optics technology, monolithic lens construction, is employed. These are constructed by taking a large diameter fiber, in this case 5.1 mm, and shaping it in one piece, so that each individual channel tapers and points toward a common focus, as shown in left half of Figure 6. This results in a smaller optic, with smaller focal distance and acceptance spot. This spot, 170 μm across, with a depth of field of 3.7 mm, is well matched to a fine focus rotating anode x-ray generator. The 0.19° divergence is less than the omega crystal oscillations typically employed to increase the density of reflections captured in a single image in protein crystallography.

2.2.4. Slightly Convergent Protein Crystallography

Protein crystallography does not require highly parallel beams. A lens with a focal length of 10 cm, shown in Figure 7, apered to give a convergence angle of 0.5°, produced an intensity gain of 60 on the sample.

A 2 degree oscillation image of lysozyme, shown in Figure 8 was produced in 20 seconds. Analysis of the data show that it is equivalent to the collimated beam results.

An additional benefit of the polycapillary optics is their performance as a low pass filter, due to dependence of the critical angle for reflection on the reciprocal of photon energy. For protein crystals this obviates the need for a monochromator to suppress the high energy Brehmsstrahlung, as shown in Figure 9. Molybdenum filtered Mo anode radiation could similarly be used without a monochromator with an optic designed for 17 keV. The removal of the monochromator significantly reduces alignment complexity.
2.3. Focused Beam Diffraction

2.3.1. Intensity Gain

Focusing the beam yields a number of important results. Firstly, the intensity on the sample is increased by more than two orders of magnitude compared to pinhole collimation. The intensity gain depends on the spot size produced by the optic. The measured FWHM for Mo Kα is 21 μm.

Using a lens input diameter of d_{lens}=3 mm, an input focal length of f_{in}=22 mm, and an output focal distance of D=9 mm gives a \( \text{LSouTCB-sample}=D+f_{in}=31 \text{ mm} \). The transmission of the lens will depend on the photon energy and the capillary channel size. Using a very conservative estimate of T=1% gives a gain of 396. A one watt source with this focusing lens will give about as much intensity on a 21 μm spot as a 400 W source without the lens. The ability to downsize the source is real and dramatic.

Measurements of a 300 μm lysozyme crystal are normally performed with a 3 kilowatt rotating anode system at 20 minutes per exposure. The data shown in Figure 10 was obtained with a 30 Watt sealed tube source in 5 minutes. The diffraction signal from high Z inorganics would be even higher. The lens employed had an output focal spot size of 100 μm, a convergence angle of 2.1°, and was 29 mm from the source.

2.3.2. Parallel Beam Focusing

Focusing beam measurements can also be performed with parallel input beams, as from synchrotrons. Focused beam intensity gain measurements were performed with a monolithic optic with a focal length of f=17 mm. The minimum focal spot size is roughly \( \sqrt{(1.50c_f)^2 + c^2} = 77 \mu m \), close to the measured value of 80 μm. This is also in good agreement with the input focal spot size of this lens measured with a laboratory source in collimating geometry.

Figure 8. Lysozyme, 0.5° convergence, 8 μm Ni, 20 seconds exposure, 2° oscillation, 70 mA, 40 kV, RAXIS II detector.

Figure 9. Spectrum of a copper tube source with and without a slightly focusing optic. The optic suppresses the high energy Bremsstrahlung. The nickel filter further suppresses the Cu Kβ peak.

Figure 10. Lysozyme image taken in 5 minutes with a 30 W sealed tube source, a 2.1 degree convergence lens and a Large area multiwire detector.
The transmission of the monolithic optic is as high as 49% at 8 keV, which was the design energy for this lens. This gives a measured gain of 96 at 8 keV for a 350 μm pinhole. Because the lens increases the photon density in the focal spot area relative to the case without the lens, the gain will be large for pinholes that are as small as, or smaller than, the spot size. The expected gain for a 10 μm pinhole is approximately 1000. The good agreement between calculated and measured gain for the larger pinhole yields some confidence in this high calculated value for a small pinhole.

3. Discussion and Conclusions

Using polycapillary optics to collimate the output from a point source provides in most cases much higher diffraction intensity than pinhole collimation, particularly if two dimensional collimation is required. Focusing the beam yields even higher intensity gains. Measured gains agree with computations. Diffraction analysis remains possible for fairly high values of the convergence angle. Calculations have been made of the resulting spot streaking, which agree well with measured results. A detailed report of measurements and calculations for quasiparallel and convergent beam microdiffraction have been reported elsewhere.

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