Subgroup report on hard x-ray microprobes

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The increasing availability of synchrotron x-ray sources has stimulated the development of advanced hard x-ray (E≥5 keV) microprobes. New x-ray optics have been demonstrated which show promise for achieving intense submicron hard x-ray probes. These probes will be used for extraordinary elemental detection by x-ray fluorescence/absorption and for microdiffraction to identify phase and strain. The inherent elemental and crystallographic sensitivity of an x-ray microprobe and its inherently nondestructive and penetrating nature makes the development of an advanced hard x-ray microprobe an important national goal. Much of the work toward an advanced x-ray microprobe has been carried out by the national laboratories. In this workshop state-of-the-art hard x-ray microprobe optics were described and future directions were discussed. Gene Ice, Oak Ridge National Laboratory (ORNL), presented an overview of the current status of hard x-ray microprobe optics and described the use of crystal spectrometers to improve minimum detectable limits in fluorescent microprobe experiments. Al Thompson, Lawrence Berkeley Laboratory (LBL), described work at the Center for X-ray Optics to develop a hard x-ray microprobe based on Kirkpatrick-Baez (KB) optics. Al Thompson also showed the results of some experimental measurements with their KB optics. Malcolm Howells presented a method for bending elliptical mirrors and Troy Barbee commented on the use of graded d spacings to achieve highest efficiency in KB multilayer microfocusing. Richard Bionta, Lawrence Livermore National Laboratory (LLNL), described the development of the first hard x-ray zone plates and future promise of so called "jelly roll" or sputter slice zone plates. Wenbing Yun, Argonne National Laboratory (ANL), described characterization of jelly roll and lithographically produced zone plates and described the application of zone plates to focus extremely narrow bandwidths by nuclear resonance. This report summarizes the presentations of the workshop subgroup on hard x-ray microprobes.

INTRODUCTION

The increasing availability of intense hard x-ray second and third generation synchrotron sources has created new opportunities to develop diffraction and fluorescence x-ray microprobes. The achievable x-ray microprobe intensity is ultimately set by the source brilliance which for synchrotron sources is 8-12 orders of magnitude greater than for tunable laboratory sources. This source brilliance is sufficient to allow the development of x-ray microprobes with orders of magnitude better minimum-detectable-limits than electron or proton probes. It will also allow for microdiffraction with two orders of magnitude better sensitivity to strain and with the ability to nondestructively probe beneath surface layers. Techniques for developing x-ray microprobe beams from synchrotron radiation sources have been reviewed,[1-5] but the field is being revolutionized by recent developments.[5-8] The technology for microfocusing x rays is advancing rapidly due to new capabilities in figuring of mirror surfaces, in construction of layered structures and zone plates, and in drawing of tapered capillaries. Similarly the hardware and software for data acquisition, control and interpretation is rapidly changing. Scientists from the national laboratories are leading the development of new technologies required to achieve advanced x-ray microprobes. The state-of-the-art for hard x-ray microprobes is described and future directions are discussed. Hard x rays are qualitatively defined as x rays which can be efficiently transmitted through Be windows ($E \geq 5$ keV).

HARD X-RAY MICROPROBE OPTICS

Apertures and Pinholes

There are two generic ways to produce an x-ray microprobe beam. One approach is to directly image the source with high demagnification. The alternative is to achieve modest demagnification of the source and then to use an aperture to obtain a submicron beam. The aperture can be located either near the sample or at some intermediate focal plane. If located at an intermediate focal position, then the x-ray beam must be reimaged. Apertures have been successfully used to produce 2-10 μm x-ray beams.[9-11] For hard x rays the apertures are typically laser drilled pinholes, adjustable slits, or capillary condensers. Because of the added complexity of reimaging, almost all examples to date have placed the aperture near the sample. Apertures are efficient only if the beam divergence through the aperture exceeds the diffraction limited divergence. For hard x rays it is difficult to obtain apertures with the needed-thickness-to-hole-diameter aspect ratio.[9-10]

Glass Capillaries

The use of glass capillaries[12-14] appears to be a promising hybrid between direct imaging and aperturing methods. Research is being conducted in this area at the Cornell High Energy Synchrotron Source (CHESS) and at the Pacific Northwest Laboratory (PNL). Similar to an x-ray lens, capillaries provide some demagnification of the x-ray beam. Unlike a true lens, capillaries do not image the source; structure in the object is lost with multiple bounces through the capillary. Glass capillaries can, however, conserve emittance.[13] The efficiency of a condensing capillary is "in principle" proportional to the ratio of the critical angle squared to the incident beam divergences divided by the square of the effective magnification ($Eff=(M\theta_c)^2/\sigma'_h\sigma'_v$). This approximation ignores losses during multiple reflections along the capillary. In practice the efficiency of a capillary depends on the taper of the capillary, its mounting, its index of refraction, internal micro-roughness, and its orientation with respect to the incident beam. Condensing capillaries provide a beam with weak tails and operate over a wide bandpass (pass x rays with energies below the critical energy). The use of condensing capillaries is an area of active research.

Current state-of-the-art capillaries have achieved 2 μm diameter hard x-ray probes and have theoretical gains approaching 1000 in intensity. Glass capillaries have been reported which compressed a 22-μm-diam beam into a 2-μm beam with 50% transmission. Glass capillaries seem especially well suited as a second stage to reduce an already small beam.

A disadvantage of small aperture methods such as pinholes and glass capillaries is the need to have the aperture close to the sample. The pinhole or capillary may cover a substantial solid angle off the sample (if for example the capillary walls are thick). The minimum beam spread is limited by diffraction from the aperture. For example at 10 keV the diffraction limit requires that an aperture be within 7 mm of a sample to preserve a 1-μ beam. Even more stringent conditions are likely to result in real applications. The diffraction limited distance between an aperture and the sample decreases as the square of the aperture diameter. Therefore a 0.1-μm-diam aperture or capillary must be within 70 μm of the sample to preserve spot size.

A glass capillary with a critical angle of 5 mrad will have an exit beam of ~10 mrad. To preserve a submicron beam, the end of the capillary must be less than 100 μm from the sample. Glass capillaries appear to be better suited than pinholes as a final demagnifying

Here, F_1 is the object(source) distance and F_2 is the image(sample) distance and θ is the scattering angle. For F_1 of 70 m, and $\sin\theta \sim 0.008$(specular mirror), and $R_s \sim 1$ cm, we find a maximum demagnification of 110:1. Even if such a highly asymmetric mirror could be manufactured, the magnification will yield a focal spot size of 6 x 1.8 μm at the APS. To achieve 600:1 demagnification, the sagittal radius would be ~1.8 mm. Both the surface roughness and figure tolerance of such a mirror exceeds existing capabilities for such an asymmetric mirror. Ellipsoidal mirrors are probably a poor choice for direct microfocusing, although they might be combined with apertures or capillaries as at the ESRF.

Kirkpatrick-Baez Total Reflection Mirrors

A possible microfocusing option using total external reflection mirrors combines crossed elliptical mirrors in the KB geometry.[17] This geometry is illustrated in Fig. 1. Like the ellipsoidal mirror described above, a total-external-reflection KB mirror system has the advantage of being broad bandpass (separate function). The bandpass and bandwidth can, therefore, be controlled by an upstream monochromator of traditional design.

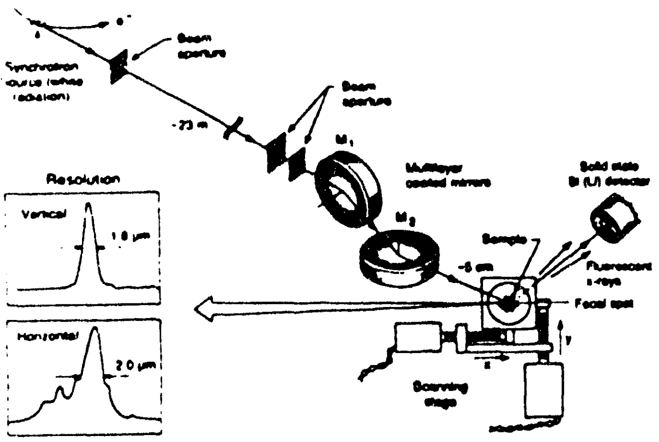

Fig. 1. Kirkpatrick-Baez mirror system used by the LBL group for microfluorescence analysis.

A disadvantage of total-external-reflection KB optics is the small aperture of the optics for modest length mirrors. This problem can be mitigated by using multilayer coating to increase the reflection angle.

Kirkpatrick-Baez Multilayer Mirrors

A practical approach for microfocusing is to use two crossed multilayer mirrors in the KB geometry. Several of these multilayer systems have been built and tested on synchrotron radiation facilities.[9-10] These devices were pioneered by Al Thompson, Jim Underwood and their colleagues at LBL. Richard Ryon and associates at LLNL are also collaborating on the development of these devices. The mirrors must be elliptical to achieve 1 μm probe size and should have graded d spacing to preserve bandpass.[1,3] Multilayer focusing combines some monochromatization with focusing so that for some applications an upstream monochromator will not be needed.

In comparison to a specular reflecting system, a multilayer system is improved by virtue of an approximately 5x larger reflecting angle. This means that these devices can collect larger divergences especially at high energies (larger aperture). A disadvantage of multilayer focusing is the difficulty of tuning energy beyond the bandpass of the multilayer and a need to grade the d spacing to compensate for different scattering angles along the multilayers. Troy Barbee (LLNL) discussed the technical feasibility of fabricating graded d spacing multilayer and concluded that this is not a barrier to continued development of KB multilayer optics.

The main technical challenge to achieving an advanced KB x-ray microprobe system is the difficulty of obtaining elliptical mirrors. Elliptical mirrors are difficult to polish to x-ray optical quality because the radius of curvature changes over the mirror surface. Although an elliptical mirror system has been reported[7] the surface figure and roughness were not adequate to achieve micron focusing.

stage; they have inherently high aspect ratio of diameter to thickness. Work is needed to demonstrate the potential of glass capillaries.

Ellipsoidal Total Reflection Mirrors

The alternative to aperturing the beam is to directly image the beam to a submicron spot. With a FWHM source size of 0.2 x 0.6 mm^2 at the Advance Proton Source (APS) the demagnification required to produce a 1μm spot is 200:1 in the vertical and 600:1 in the horizontal. At the Advanced Light Source (ALS), demagnifications of 60:1 and 200:1 will achieve 1 μm resolution. For specular reflection from a single mirror, ellipsoidal surfaces of revolution produce the smallest aberrations of the image. An ellipsoid combines sagittal (out-of-plane) and meridian (in-plane) focusing in one doubly curved optical element. Current plans at the European Synchrotron Radiation Facility (ESRF) call for the use of an ellipsoidal mirror on their microprobe beamline. This microprobe beamline has an early operation goal of a 100-μm^2 spot size.[15]

A current limit to demagnification with ellipsoidal mirrors arises from the difficult fabrication of highly asymmetric optics with acceptable surface roughness and figure errors. A discussion of this limit must consider the surface and figure accuracy requirements of x-ray mirrors.

The accuracy of a mirror finish can be divided into approximately two regimes. At short frequencies, surface roughness controls the scatter of the beam. At long spatial frequencies, slope errors (figure errors) cause the image to be blurred. The tolerance required for a mirror depends primarily on the source size and the distance of the mirror to the source. Consider, for example, the figure precision required for an APS mirror. The APS vertical source size, σ_y, is of the order of 85 μm and the distance from the source to a first mirror, F_1, is on the order of 30 m. If the slope deviates from the ideal figure with an root-mean-square (RMS) deviation, σ'_S, the image will be blurred by $2\sigma'_S MF_1$. The slope error must, therefore, have a standard deviation of less than $\sigma_y/2F_1$. At the APS this represents a standard deviation of 1.4 μrad or 0.25 arcsec.

The grating equation, $\lambda = d\theta_i\theta_s$, can be used to estimate the spatial frequency, d, below which surface roughness scatter lies outside the image region. With an incident beam angle, θ_i, of 3 mrad and a scatter angle, θ_s, due to roughness less than 2.8 μrad, spatial frequencies d(mm) below 12λ(Å) will scatter outside the geometrical image. Therefore at 10 keV, spatial frequencies below 1 cm can scatter out of the geometrical image. For spatial frequencies greater than ~3 mm, figure errors dominate beam blur. The total intensity into the geometrically demagnified beam is given by,

$$I = I_o \exp\left(\frac{-4\pi\sigma_S\theta}{\lambda}\right)^2 \quad . \qquad (1)$$

Here, σ_S is the RMS surface roughness below spatial frequencies of 1 cm. For 90% power in the geometrical image and with θ~3 mrad the surface roughness below 1 cm must satisfy σ_S(Å)~8.6λ(Å).

Both the ALS and the APS have source size to object distance ratios, σ/F_1, of about 1×10^{-5}. The surface roughness and figure requirements for ALS optics are, therefore, similar to those of the APS when used to focus the same x-ray energy.

To illustrate the technical challenge of a microfocusing ellipsoidal mirror we calculate the asymmetric radii of an ellipsoidal condensing mirror. The sagittal, R_s, and meridional, R_m, radii of a ellipsoidal mirror are given by,

$$R_s = 2F_1 F_2 \sin\theta/(F_1 + F_2) \qquad (2a)$$

$$R_m = R_s/\sin^2\theta \qquad (2b)$$

Several schemes have been proposed to bend mirrors to achieve a good approximation to an ellipse. These are currently under investigation at the Center for X-ray Optics (LBL). Al Thompson and Jim Underwood from the Center for X-ray Optics propose to instrument a bending magnet microprobe beamline on the ALS using x-ray mirrors bent with unequal couples. These mirrors will approximate an ellipse and reduce spherical aberration for a fixed aperture.

Malcolm Howells (LLNL) discussed an even more advanced bender concept based on nearly cube-root-thickness varying mirrors.[20] Both methods have the advantage that the mirror surfaces are polished as flats.

A practical compromise to the ideal elliptical mirror surface is the use of spherical optics which can be obtained with good surface roughness and figure. Although these optics have an aperture limited by spherical aberration, they can achieve near-micron focusing with a carefully collimated incident beam. The best performance reported to date has achieved image sizes with FWHM of ~ 2-μm but with small angular acceptance.[10]

Zone Plates

Another promising technology is the use of zone plates for hard x-ray imaging. Zone plates have been used to focus soft x rays with great success as reported in this conference and elsewhere.[17] Their application to hard x rays has been limited by the difficulty of obtaining the required high-aspect ratio. This problem is similar to the difficulty of procuring high aspect apertures for x rays. The construction of high-aspect-ratio zone plates has now been achieved by Richard Bionta and associates at LLNL using a sputtered-slice technique.[18] With this technique, a high Z wire has concentric layers of alternating Z deposited. The resulting "jelly roll" structure is sliced to create the x-ray lens (Fig. 2). A state-of-the-art phase zone plate can currently be constructed using this method with a ~0.3-mm aperture and a 30-cm focal length and with roughly 30% efficiency (Fig 3). The ultimate resolution of a zone plate depends on the thickness of the last zone and on the depth of the zone plate.[19] It is possible that zone plates with resolutions down to 0.1 μm or less can be fabricated. The aperture of the zone plate can be reduced to decrease the focal length. With current jelly roll techniques it should be possible to fabricate a zone plate with a 10 cm focal length and with a 100-μm-diam. At 60 m from the source, such a zone plate would have a 600:1 demagnification. Bionta (LLNL) also described a prototype 3-step blazed zone plate with improved efficiency and plans for an even more efficient zone plates.

Compared to the other optics discussed, zone plates are much easier to align although their focused intensity distribution is complicated and requires secondary apertures to obtain a clean beam. Even greater demagnifications (at reduced efficiency) are possible with the zone plates used in higher orders.

Recent advances in lithographic techniques have also been used to develop x-ray zone plates. Wenbing Yun (ANL) described a lithographic zone plate with 0.25 μm outer line width and a 60-μm-diam. This zone plate is capable of achieving very small spot size, although the efficiency is not expected to match an advanced jelly roll zone plate.

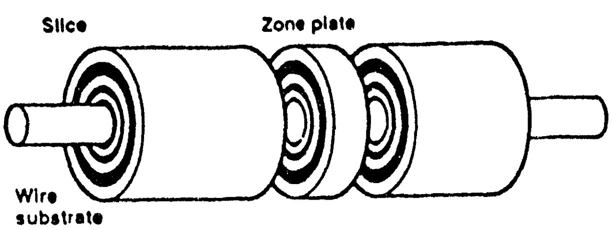

Fig. 2. "Jelly roll" zone plate fabrication method for hard x-ray zone plates.

Performance of zone plates fabricated by a lithographic method and a sputtering/slicing method have been characterized using synchrotron radiation. The zone plates have been used successfully to focus synchrotron x rays to a small focal spot with focusing efficiency close to that predicted theoretically. For a Ni zone plate fabricated using the lithographic technique, a focusing efficiency of 33% was experimentally measured and a diffraction-limited focal spot was obtained. The smallest zone width obtained by the lithography technique is 0.25 μ and that obtained by the sputtering/slicing technique is slightly less than 0.2 μ.

While the capabilities of the two fabrication methods are quite complimentary, the sputtering/slicing method is, in principle, capable of producing zone plates with large thickness and zone width as small as a few tens of angstroms. However, many technical problems need to be overcome to realize the full potential of this method. On the other hand, the lithography method is better developed for producing zone plates with submicron spatial resolutions, but it will be difficult to produce zone plates with spatial resolutions less than 0.1 μ for hard x-ray applications (Fig. 5).

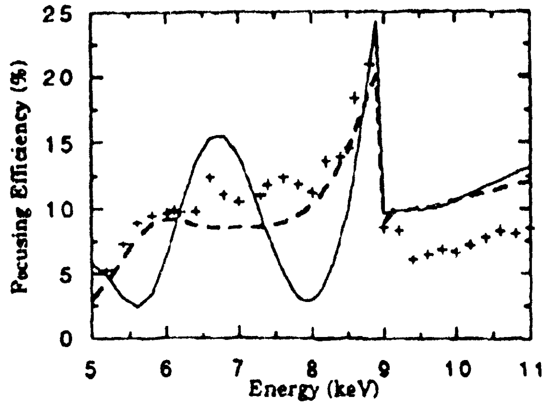

Fig. 3. Focusing efficiency for a "jelly roll" zone plate measured by Yun et al. Experimental measurements (crosses), are compared to theoretical calculations assuming a uniform thickness of 19 μm (solid line) and a non uniform (wedged) thickness profile (dashed line).

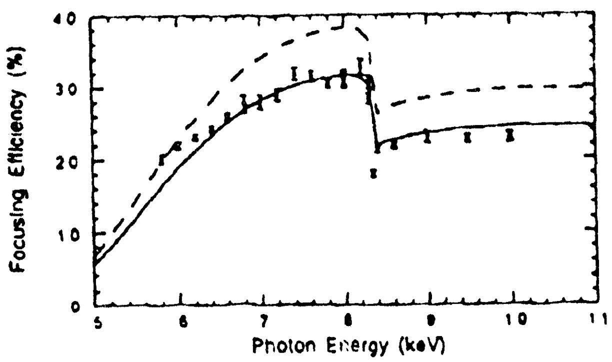

Fig. 4. Focusing efficiency for a lithographically produced hard x-ray zone plate as a function of x-ray energy. Experimental measurements (dots) are compared with calculations assuming an area ratio between the Ni and open zones equal to 1:1 (dashed line), and 1:1.75 (solid line). In the calculation, a square wave Ni zone profile of 3.1 μm thick was assumed.

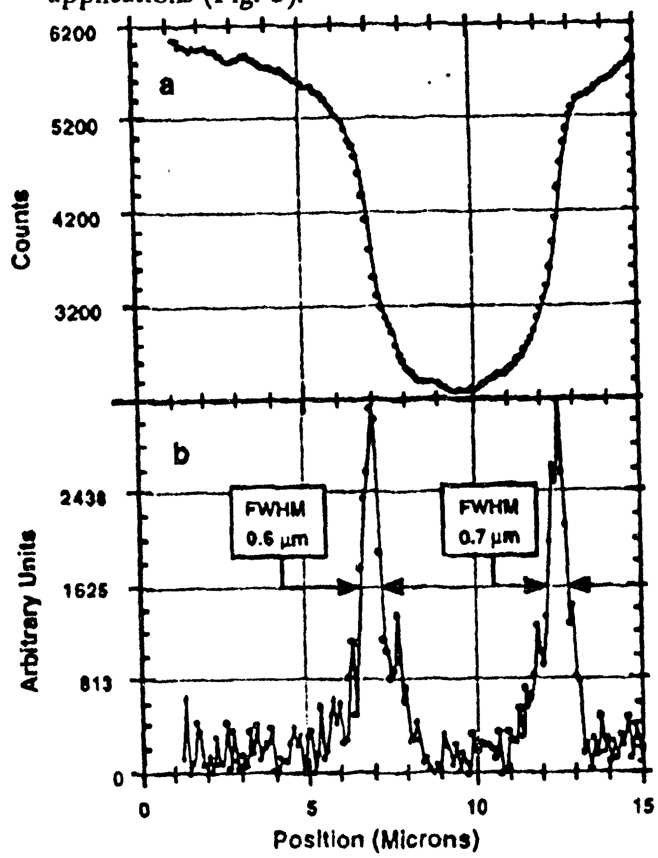

Fig. 5 The transmitted intensity measured when a 6-μm bar of a Au grid was scanned across the primary focus of a lithographically produced hard x-ray zone plate (a), and the derivative of the intensity with respect to the bar position (b).

Two practical problems with zone plates help to put their performance in perspective with the previous optics. Highest zone plate performance occurs for phase-type zone plates. Such devices have been demonstrated and more advanced phase-zone plates are being developed. These devices work best over a restricted range of about 500 eV to 1 keV. This means that many zone plates will be required to cover the range from 4-40 keV. Because they are easily aligned, it will fortunately be possible to have an assortment of zone plates optimized for selected energies. These can be inserted and aligned rapidly.

Another limitation of zone plates is their inherent chromatic aberration. The focal length of a zone plate is inversely proportional to the wavelength. Therefore as x-ray energy increases, the zone plate focal length also increases. This chromatic aberration limits the useful aperture of a zone plate for a given bandpass and a given focal spot size. For small bandpass ΔE/E and an aperture diameter A, the chromatic blur of a zone plate is given by,

$$\sigma_{chromatic} \sim \frac{A}{2}\,\frac{\Delta E}{E} \quad . \qquad (3)$$

Hence, with a 1% bandpass the aperture of a zone plate must be less than 100 μm to achieve a 1-μm-diam focal spot. This limits the phase space acceptance of a zone plate with 1% ΔE/E below the acceptance for mirrors considered previously. Where small divergences or small bandwidths are acceptable however the zone plate technology may be the first to achieve resolutions to 0.1 μm. The focal length must decrease with decreasing aperture to reduce the effect of the diffraction limit on spatial resolution.

In addition to the work at LLNL, zone plate technology is being pursued for lower energies (2-4 keV) by Anderson and Attwood at LBL. At these energies they hope to achieve 0.1 μm resolution.

Summary

The properties of the various optical devices which can be used to microfocus hard x rays are given in Table 1. Several devices show the potential to achieve submicron resolution. In addition to choosing the kind of optics to be used, it is critical to decide on the object and image distances. In general, aberrations due to source size are smaller for large object distances. The heat load/unit area is also smaller for large object distances. However, aberrations due to surface roughness and figure error are worse for large object distances.

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Table 1. The relative demagnifications for various x-ray optical systems and their efficiencies for an APS undulator A. Demagnifications are for a single stage.				
Element	Mag	Efficiency	Divergence (mrad)	Comments
K-B external reflection mirrors	1/600	2%[a]	1x2	Wide bandpass
K-B multilayers	1/1000	4%[b]	3x5	1-10% fixed bandpass
Ellipsoidal mirrors	1/100	15%[c]	1x2	Difficult to produce;wide bandpass
Ellipsoidal multilayer	1/500	10%[d]	5x10	Difficult to produce;1-10% fixed bandpass
Tapered capillary	1/30	15%[e]	10x10	Wide bandpass;requires prefocused beam.
Zone plate	1/600	2%[f]	2x2	Best efficiency in narrow bandpass; easily aligned.

[a]Efficiency estimated for 20 keV x rays and 3 mrad mirrors.
[b]Efficiency estimated for 20 keV x rays and 5% bandpass multilayers.
[c]Performance limited by lower limit to sagittal radius.
[d]Multilayer must be graded to achieve 10% efficiency.
[e]Assumes prefocused beam to 30 μm spot.
[f]Efficiency limited by technical limit to zone fabrication.

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