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X-ray Microprobe: The Next Step in Microcharacterization

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ABSTRACT

The combination of high brilliance of third generation synchrotrons and advanced x-ray microfocusing optics will revolutionize microcharacterization. Kirkpatrick-Baez elliptical mirrors, zone plates, and condensing capillaries have all achieved intense submicron focused beams. Other focusing options are also under study including Bragg-Fresnel optics and compound refractive lenses. The intense micron-scale beams from advanced x-ray optics on third generation sources will provide unique information about the elemental and crystallographic distribution in samples and will enable a variety experiments previously unimaginable. X-ray microbeams can be used to map elemental distributions in two and three dimensions and can be used to study the phase, texture, and strain distributions of inhomogenous samples in two and three dimensions.

MICROBEAM FORMING

It has long been recognized that x-rays have unique advantages for the characterization of materials.[1] X-rays are inherently nondestructive with orders of magnitude lower power deposited for the same detectible limit as charged particles (Fig. 1). X-rays can penetrate below the surface and require little sample preparation. X-rays are also two orders of magnitude more sensitive to crystallographic strain than electron probes.[2] These unique advantages have been limited by relatively weak conventional x-ray sources.

The development of intense synchrotron x-ray sources, with at least 11 orders of magnitude greater brilliance than conventional x-ray sources (Fig. 2), has revived interest in x-ray optics. At least three microbeam forming options have emerged with various strengths and weaknesses for experiments.[3]

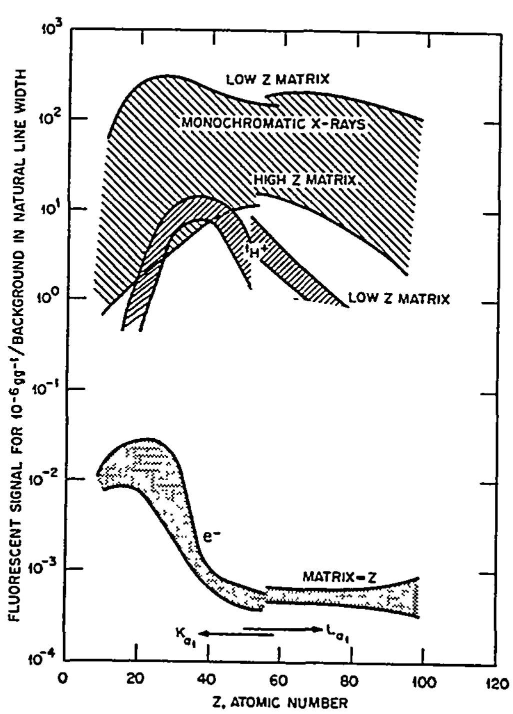

Fig. 1 Signal to noise of x-rays compared to electron and proton excitation (after ref. 1).

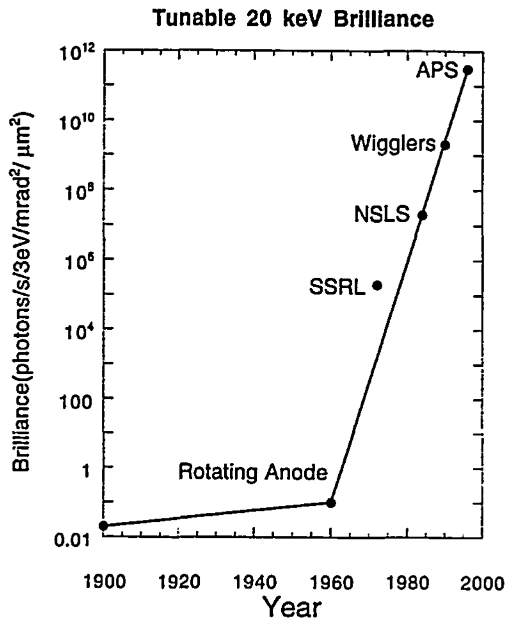

Fig. 2 X-ray brilliance over the last 100 years.

Tapered capillary optics (Fig. 3) have produced the smallest x-ray beams.[4] Bilderbach, et al.[5] have reported beams as small as 500 nm FWHM. This option appears to be the best for condensing beams below 0.1μm. One concern with capillary optics, however, is their effect on beam brilliance. Ray tracing and experimental measurements have found that the angular divergence following a capillary has a complex annular distribution. This distribution arises from roughness inside the capillary and from the non-equal number of reflections of different rays as they are propagated along the capillary.

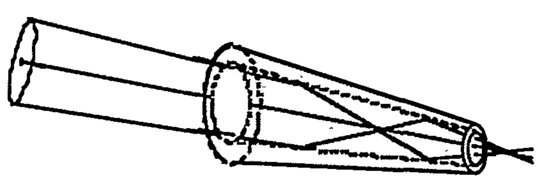

Fig. 3 Schematic of a tapered capillary condensing element.

Hard x-ray zone plates (Fig. 4) are a rapidly emerging option for focusing synchrotron radiation to mm dimensions.[6,7] This option appears especially promising for focusing monochromatic radiation ($dE/E \sim 10^{-4}$).

These devices are simple to align, allow good working distance between the optics and the sample, and have already achieved sub micron spots. Although zone plates are inherently chromatic, they can in principle be used with tunable radiation by a careful translation along the beam direction. State-of-the-art zone plates provide the most convenient optics for monochromatic experiments even though their focusing efficiency has not reached the 40-60% efficiency promised by more advanced designs.

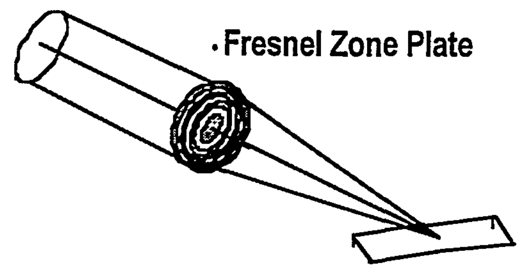

Fig. 4 Hard x-ray zone plate for focusing synchrotron radiation.

Kirkpatrick-Baez (KB) mirrors provide a third highly promising option for focusing synchrotron radiation.[8,9] A Kirkpatrick-Baez mirror pair consists of crossed total-external-reflecting mirrors which condense the beam in orthogonal directions (Fig. 5). Both multilayer and total-external reflection mirrors have been used for focusing synchrotron radiation. Multilayer mirrors appear most suitable for fluorescence measurements with a fixed wide-bandpass beam and where large divergences can be accepted. Total-external-reflection mirrors appear to offer the best option for focusing white beams to μm dimensions. The key challenge with KB mirrors is achieving low figure and surface roughness with elliptical surfaces. There are numerous parallel efforts currently underway to advance mirror figuring for advanced KB focusing schemes.

Kirkpatrick-Baez Mirrors

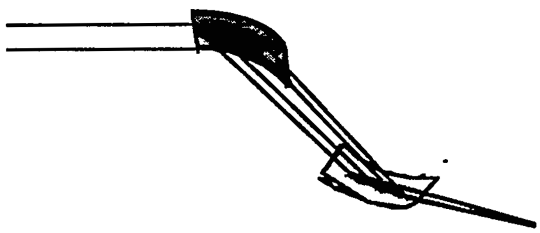

Fig. 5 Schematic of a two-mirror Kirkpatrick-Baez pair.

In addition to the focusing schemes mentioned above, there are two new options which have recently emerged from experiments at the European Synchrotron Radiation Facililty (ESRF). These are compound refractive lenses (Fig. 6) and Bragg Fresnel optics (Fig. 7). Compound refractive lenses are very interesting because they are relatively easy to manufacture.[10] Estimates of their theoretical efficiency, however, indicate that they cannot compete with the theoretical efficiency of KB mirrors or zone plates.

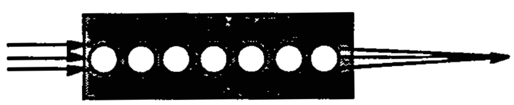

Fig. 6. X-ray compound refractive lense.

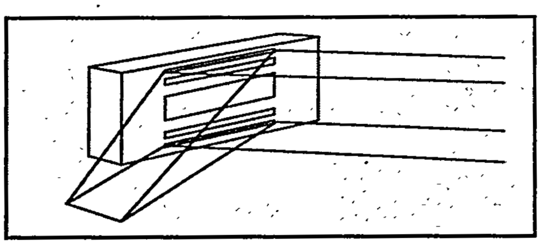

Fig. 7. Bragg Fresnel lense.

Bragg-Fresnel optics are also an interesting option.[11] With Bragg-Fresnel optics, the phase contrast steps are lithographically etched into a monolithic Si substrate. This offers a rugged substrate and serves to simultaneously focus and monochromate the beam. Some of the simplicity of operation with zone plates is lost with these devices because the beam is deflected.

MICROBEAM APPLICATIONS

The availability of intense tunable x-ray beams with micron scale dimensions will revolutionize our ability to characterize materials. There will be at least three immediate applications of x-ray microbeams: Microfluorescence analysis, microdiffraction, and three-dimensional x-ray microtomography.

Microfluorescence analysis has already been applied to many problems with second generation x-ray beams.[12,13] Microfluoresence exploits many of the unique properties of x-rays: high signal-to-noise, depth probing, minimum sample preparation, and the ability to measure local bond distances with NEXAFS/EXAFS methods.

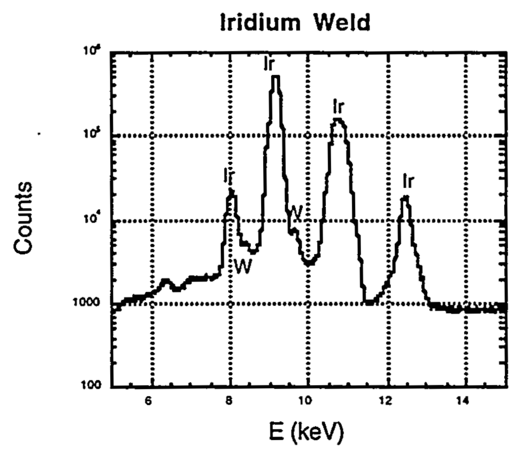

Fig. 8 Fluorescence spectra from a iridium sample with trace elements of W and Th. The W fluoresence peaks are swamped by the intense Ir lines. Because the Ir edges are above W, the Ir fluoresence can be removed by tuning the x-ray energy below the Ir edges.

As shown in Fig. 1, the signal-to-noise of microfluorescence is far higher than for other non-destructive probes. The availability of even more intense and tunable x-ray microbeams from third generation sources will allow the use of analyzing crystal optics with far higher signal-to-noise but relatively poor efficiency compared to solid state detectors. This will revolutionize the search for trace element distributions in heavy Z materials where the fluorescence background from the matrix can paralyze solid state detectors (Fig. 8).

X-ray microdiffraction[14] also have a major impact on the characterization of materials. Fig. 9 illustrates many of the powerful attributes of microdiffraction. The figure shows the x-ray microdiffraction pattern from a ferroelectric thin film. The single-crystal thin film Bragg peak appears as a lenticular intense pattern in the CCD image even though it is covered with a 200 nm cap of aluminum. The strain in the thin film can be measured to $\Delta d/d \sim 5x10^{-4}$. Texture, particle size, and strain of the aluminum overlayer can be inferred directly from the 111 and 200 Debye rings.

X-ray microdiffraction can be widely applied to the study of texture and strain in inhomogeneous materials.[15] X-ray microdiffraction has already achieved great interest from the semi-conductor community for understanding the strain distribution in advanced materials and integrated-circuit interconnects.[16,17]

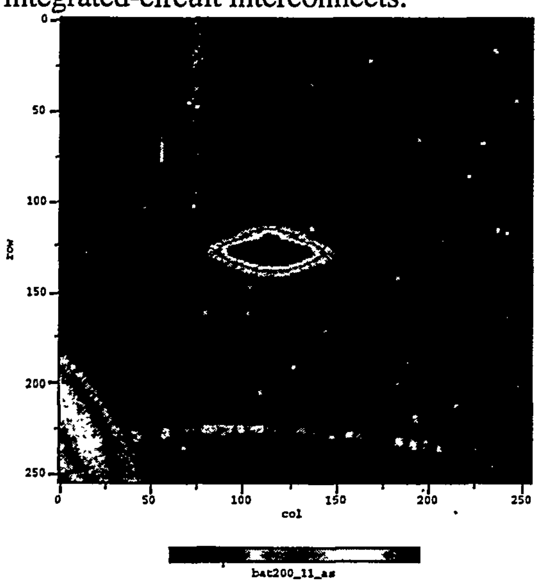

Fig. 8. X-ray microdiffraction image from an advanced thin film. The CCD image shows the single crystal reflection and the powder-like image from the Al overlayer.

X-ray microtomography will also benefit from the ability to produce small x-ray beams. As shown in Fig. 9, transmission tomography can already probe the density and defect structure of very small particles. X-ray microbeams, however, offer the promise of both fluorescence microtomography for advanced characterization of 3-D distributions of trace elements in materials, and x-ray diffraction tomography.[18] Efforts to demonstrate these methods are already underway, and methods for automating the indexing of Laue patterns from one or more crystallites will greatly accelerate the study of materials by microdiffraction and microdiffraction-tomography methods.[17,19]

Fig. 9 Iso-intensity contours from an x-ray transmission tomography measurement of an advanced fuel sphere taken on X-2B at the NSLS.

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