Hard X-ray Microimaging Techniques Based on Phase Zone Plates

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ABSTRACT

Phase zone plates of high focusing efficiency and submicron resolution have been demonstrated in the hard x-ray region. A scanning microscope based on these focusing optics will create many new applications. Preliminary results in the applications of the microscope are reported here. In the area of imaging, we have utilized absorption contrast to clearly identify the locations of Au and Ni constituents in a sample of two interleaved grids. Micro-EXAFS spectra has also been obtained on a Ni foil. Fluorescence from a nuclear fuel sample, as an example of microanalysis, has revealed the elemental distribution at the interfaces. Lastly, microdiffraction from AgBr crystallites has been studied.

Introduction

Many conventional x-ray techniques can be applied in conjunction with microfocusing optics, such as Fresnel zone plates, to provide information with high spatial resolution. This is an important development because most samples today have sizes, features, or inhomogeneities that are small compared to an unfocused x-ray beam. Recent advances with phase zone plate (PZP) optics in the hard x-ray regime¹⁻³ have facilitated this development. It is expected that with the brilliant third generation synchrotron sources,⁴ these techniques (such as microscopy, microspectroscopy, microanalysis, and microdiffraction) will evolve into powerful analytical tools. Some preliminary results in these areas are presented here. Another important application of PZP is in the area of coherence-based techniques, including holography, interferometry, speckle, and signal enhancement, in which the PZP is used as a coherence selection optic. Some of these coherence-based applications have been discussed before.³

Experimental

The experiments were performed at beamline X-18B of the National Synchrotron Light Source (Fig. 1). Bending magnet radiation was passed through a Si double crystal monochromator before reaching the zone plate located at ≈ 22 m from the source. Two types

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Fig. 1 Schematic of the scanning hard x-ray microscope using a Fresnel phase zone plate as the focusing element.

of zone plate were used during the experiments, an Al/Cu PZP and a Ni PZP. X-rays from unwanted orders were blocked by an order selection aperture (OSA) before reaching the sample. The sample transmission was measured by a scintillation counter (SC0) for microscopy and microspectroscopy applications. For microanalysis, the sample surface was rotated such that efficient collection of fluorescence radiation was obtained by a Si(Li) detector. The sample was also rotated for microdiffraction in order to diffract the beam into a scintillation counter (SC1). For the experiments, the Si monochromator used has a resolving power of about 10^4 , which is more than that required for zone plate focusing. If a wider bandpass was accepted, for instance by using a pair of multilayers, the flux available for some applications could be increased by 1-2 orders of magnitude.

The stringent requirements for hard x-ray PZP fabrication were met by two techniques: sputtering/slicing and x-ray lithography. The two techniques are discussed in detail elsewhere.⁵ In summary, both types of PZP have been tested at 8 keV; a focusing efficiency of 20% was measured from a 3π -phase-shifting Al/Cu sputtered PZP and that of 33% was measured from a π -phase-shifting Ni lithographic PZP. The Al/Cu PZP has a focal length of 13.3 cm (third order focus), and the Ni PZP has a focal length of 20 cm (first order focus). At 22 m from the source, the focal spot size is dominated by the demagnified image of the source, and spatial resolution of a few microns was easily obtained. Recently, by aperturing the source size, we were able to obtain a diffraction-limited resolution of $0.7 \mu\text{m}$ from the Ni PZP ($\Delta r_n = 0.5 \mu\text{m}$). These results show that the fabrication errors are well within tolerance at this level.

Microscopy

The hard x-ray regime includes the K-edge of transition metals and the L-edges of many heavier elements. This makes microscopy based on the high absorption contrast an attractive technique. Fig. 2 shows the transmission images of a Au grid and a Ni grid interleaved on one another. Each grid has 1000 mesh/inch, which is equivalent to a

Fig. 2 Transmission images of interleaved Au and Ni grid taken at (a) above the Ni K-edge (8.35 keV), and (b) below the Ni K-edge (8.30 keV). The Ni grid image was recovered in (c), as a subtracted image (a-b).

periodicity of 25 μm . Above the Ni edge, both grids are absorbent, and it is difficult to separate them based on the Moiré-pattern-like image. Below the Ni edge, the absorption coefficient of Ni decreases by about a factor of 7, and the transmission image shows only the Au grid. The position of the Ni grid is then easily identified by subtraction of these two images. The absorption contrast is particularly useful when several absorbent elements are distributed within a low-Z matrix. In that case, a spatial map of the elemental quantity for each element can be obtained.

Microspectroscopy

Additional information about a sample can be obtained when the absorption spectrum near an edge is recorded, that is by using micro-XANES and micro-EXAFS. The near edge structure can provide chemical information, while the extended structure reveals geometric arrangement around the atom. Fig. 3 shows an EXAFS spectrum taken with the focused beam on a Ni foil. It demonstrates that extended energy scanning can be accomplished with PZP optics. For an actual sample, the PZP will be translated relative to the sample in order to compensate for the focal length change. The motion accuracy does not need to be precise because of the large depth of focus ($\approx \text{mm}$). However, the translation axis must be critically aligned with the optical axis in order to track the same sample spot.

Fig. 3 X-ray absorption structure taken with a microfocused beam on a Ni foil.

Microanalysis

When x-rays impinge on a sample, it creates secondary products such as photoelectrons, fluorescence, and Auger electrons. Although these secondary particles all carry elemental information, the electron products are very surface sensitive while the fluorescence probes deeper into the sample. Because most elements have a fairly high fluorescence yield in the hard x-ray region, x-ray fluorescence is particularly attractive for trace element analysis of bulk samples.⁶ It is expected that femtogram sensitivity can be obtained by using microfocusing optics at the third generation synchrotron sources.

Fig. 4 Ni and Cr K_{α} fluorescence signal across the Ni-Cr/U-Zr interface. The line scans in (a) and (b) are separated by 20 μm laterally.

Fig. 4 shows an application of microanalysis using x-ray fluorescence. The object is a depleted nuclear fuel sample with an interface between Ni-(16%)Cr and U-(23%)Zr. The diffusion profile of Ni and Cr into the U region was studied with fluorescence. An interesting correlation between the Ni and the Cr distribution at the interface was found: a Cr enrichment is associated with depletion of Ni, which is also confirmed by the concentration profile measured with SEM. In addition, the interface is not uniform in the lateral direction; a different diffusion profile was obtained when the line scan was repeated at 20 μm away. These line scans could be assembled to create a complete 2D map of the interface. The excitation energy (10 keV) was chosen to maximize the fluorescence yield of Ni and Cr. By tuning to higher photon energy (≈ 20 keV), fluorescence from U and Zr can also be excited and detected. This is due to the fact that most elements contain some fluorescence lines in the hard x-ray region.

Also, if a high resolving power monochromator or detector is used, it is possible to extract the chemical information imbedded in the fluorescence signals. This has been demonstrated in the soft x-ray region with soft x-ray emission spectroscopy (SXES),⁷ which is a valuable tool for studying electronic structure of bulks and surfaces.

Microdiffraction

Another promising analytical tool under development is microdiffraction. Fig. 5 shows the x-ray diffraction signals measured from a sample with randomly oriented AgBr crystallites. The linear dimension of the crystals is about one micron, and the diffracted rays form a cone around the sample similar to powder diffraction. By positioning the

Fig. 5 Spatial map of the diffraction intensity from a AgBr sample. There might be other crystallites in this region, but they were not recorded because their orientations were different from that of the two crystallites in this figure.

detector at a fixed point along the diffraction cone, only crystallites with a particular orientation are selected and imaged. In other applications, one may want to obtain more detailed information by measuring the rocking curve locally. This can be achieved by rocking the sample or scanning the photon energy while keeping the illuminated spot on the sample fixed. Another possibility is to utilize a PZP with a sufficiently large numerical aperture so that the diffracted beam contains the entire rocking curve. The diffraction can then be recorded with a simple scanning detector or a position sensitive detector. This capability to obtain a rocking curve at any sample point is essential for characterization of imperfections such as grain boundary, dislocation, strain, and stress. This technique can be used for studies of mosaic crystals, thin films, or overlayers.

Summary

Recent success with hard x-ray phase zone plates has made possible the realization of many microfocusing-based x-ray techniques. In particular, experiments with microscopy, microspectroscopy, microanalysis, and microdiffraction have been performed. In the near future, we also plan to develop microtomography with increased spatial resolution. Together with the high brilliance offered by the third generation synchrotron sources, these techniques promise to become invaluable analytical methods in the hard x-ray regime.

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