Nanometer focusing of hard x rays by phase zone plates

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Focusing of 8 keV x rays to a spot size of 150 and 90 nm full width at half maximum have been demonstrated at the first- and third-order foci, respectively, of a phase zone plate (PZP). The PZP has a numerical aperture of 1.5 mrad and focusing efficiency of 13% for 8 keV x rays. A flux density gain of 121 000 was obtained at the first-order focus. In this article, the fabrication of the PZP and its experimental characterization are presented and some special applications are discussed.

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I. INTRODUCTION

Zone plates (ZPs) are among the most promising microfocusing optics being developed for x-ray applications. In the soft x-ray spectral region, ZPs with a focal spot approaching 20 nm are being developed and will be used for various imaging techniques. In fact, the focal size is the smallest obtained in the electromagnetic spectrum. Correspondingly, the images produced using these ZPs are of the highest spatial resolution obtained in an imaging microscope in the electromagnetic spectral region. Although a sputtering/slicing technique, which in principle is capable of producing ZPs with spatial resolution approaching 10 nm, was proposed long ago and is being continuously developed, nearly all the ZPs in use today in the soft x-ray region are fabricated using various forms of an electron beam lithographic technique. These techniques, however, may not be directly used for producing ZPs for hard x-ray applications because the thickness required is beyond the fabrication capabilities. In order to fabricate ZPs with adequate thickness for hard x-ray applications, we have developed a technique to transfer a ZP pattern produced using electron beam lithography into one with adequate thickness. In this article, we report the first experimental demonstration of focusing 8 keV x rays to a 150 nm full width at half maximum (FWHM) focal spot size in the first-order focus and a FWHM focal spot size of 90 nm in the third-order focus. A flux density gain of 121 000 was obtained at the first-order focus. The fabrication process used for producing this PZP is briefly described and some of its applications to the study of biological systems are presented and discussed.

II. PZP PARAMETERS

Among many parameters, the spatial resolution and focusing efficiency of a microfocusing device are the most important ones that characterize its performance. Because the flux density at a focus is proportional to the square of the numerical aperture of the focusing optic, which is inversely proportional to its spatial resolution, a focusing optic with a high spatial resolution may be very desirable for experiments in which the high spatial resolution is not required but a high flux density is necessary.

The spatial resolution of a ZP is given by

$$\delta r_m = 1.22 \frac{\Delta r_k}{m},$$

where $$\delta r_m$$ is approximately the FWHM of the Airy disk of the zone plate for the $$m$$th diffraction order, and $$\Delta r_k$$ is the outer-most zone width. It should be noted that the spatial resolution of the third-order focus ($$m = 3$$) is 3 times smaller than that of the first-order focus ($$m = 1$$).

The focusing efficiency of a ZP depends on the materials used for construction of the zones and its profile. ZPs with a square wave zone profile are called Soret amplitude zone plates if the focusing originates mainly from the relatively different absorption of two neighboring zones, and are called PZPs if the focusing originates mainly from relative phase change between two neighboring zones. The zone plate presented in this paper is a PZP.

The thickness required for obtaining a $$\pi$$ phase shift for x rays of wavelength $$\lambda$$ is given by $$t = \lambda/2\delta$$, where $$n = 1 - \delta$$ is the real part of the refractive index of the material from which the zones are constructed, and $$\lambda$$ is the wavelength of the incident radiation. For Au and Ni, the required thickness $$t$$ is 1.5 and 3.3 $$\mu$$m, respectively, for 8 keV x rays. Adopting the definition of aspect ratio as the ratio of the thickness to the smallest line width in a pattern, the required aspect ratio for producing a PZP with the outer most zone width equal to 100 nm is 15 and 33 for Au and Ni, respectively. These requirements are beyond the capabilities of even state-of-the-art microfabrication facilities with a single processing step. Several approaches have been developed by us to effectively approach the required aspect ratio. One approach involves duplicating the PZP pattern on another side of a thin membrane, on which the original PZP pattern was made. In this
III. EXPERIMENT

The experiments for characterization of the focal spot size of the PZP were performed at the 2-ID beamline of the Advanced Photon Source (APS) at Argonne National Laboratory. X rays of 8 keV were selected using a water-cooled double crystal [Si(111)] monochromator from a beam of synchrotron radiation generated from APS undulator A and were reflected from a silicon mirror with an incident angle of 0.15°. The management of the thermal power of the undulator radiation using a mirror with a cut-off energy (12 keV) ensured the high performance of the crystal monochromator, which was located downstream of the x-ray mirror. The PZP was placed at 71.5 m from the x-ray source. Because the spatial resolution of the measurement would be limited by the vertical size of APS undulator A source (150 μm in FWHM) given a demagnification factor of 715 obtained from the ratio of source-to-zone-plate distance over the ZP focal length, a virtual source was made by placing a pair of water-cool slits with openings of $35 \times 35 \mu m$ at 43.5 m upstream of the ZP. As such, the FWHM size of the source image at the ZP focal plane was reduced from 210 to 80 nm. The monochromatic beam was apertured to a size of $200 \times 200 \mu m$ before it illuminated the ZP. A platinum order-sorting aperture of size $20 \mu m$ was placed 10 mm upstream from the image plane to increase the contrast of the source image.

The focal spot size was determined using a knife-edge scanning technique. The knife edge used was a 200 Å thick Cr deposited on a Si wafer. It was fabricated by first depositing the Cr film on a Si wafer and then cracking it at liquid nitrogen temperature. Using this method, we have successfully made sharp and straight knife edges with different metals. By scanning the knife edge across the focal spot in the focal plane while monitoring the Cr $K\alpha$ fluorescence, the intensity distribution of the focused beam is then determined. The scanning stage was a piezo-driven stage (Queensgate 100) capable of 10-nm resolution, and an energy-dispersive detector (Ge, Canberra) was used to measure the fluorescence signal. The focal plane was determined by making knife-edge scans at different distances between the ZP and the knife edge. Figure 1 shows the measured Cr ($K\alpha$) fluorescence intensity profile at the focal plane scanned in the vertical direction. For data analysis, the experimental data were fitted with an error function, and a uniform background was assumed. The vertical intensity profile of the source image was then obtained by taking the derivative over the fitted fluorescence intensity profile. The beam intensity profile gives a FWHM vertical spot size of 150 nm. Using the same procedure, we have determined that the focal spot size at the third-order focus to be about 90 nm.

The measured focal spot size $\delta_m'$ of the $mth$ order focus may be calculated by

$$\delta_m' = \left(\delta_{r,m}^2 + \delta_{i,m}^2 + \delta_s^2\right)^{1/2},$$

where $\delta_{r,m}$ is the intrinsic resolution of the ZP for the $mth$ order, $\delta_{i,m} = \delta f_m / L$, is the demagnified source size with $\delta_s = 35 \mu m$ being the effective source size, $L$ and $f_m$ the source PZP and PZP-image distances, and $\delta_r = D \Delta E / E$
14.5 nm is the chromatic aberration with $D = 145 \, \mu m$ being the diameter of the PZP and $E/\Delta E = 10^{-4}$ the resolving power of the Si(111) monochromator. Using expression (1), we have $\delta_{r,1} = 126$ and 42 nm for $m = 1$ and 3, respectively. Using $L = 43.5 \, m$, $f_1 = 10 \, cm$, and $f_3 = 3.3 \, cm$, we have $\delta_{r,1} = 80 \, nm$, and $\delta_{r,3} = 27 \, nm$. Using expression (2) we obtain $\delta'_{r,1} = 150 \, nm$, $\delta'_{r,3} = 52 \, nm$. While the calculated result for the first-order focus agrees well with that measured, the difference between the calculated and measured results is large for the third-order focus. There are several possible reasons why the measured focal spot size is larger than the calculated one, including mechanical vibration between the PZP and the knife edge, error in the placement accuracy of the zones, and error in the determination of the exact focal plane. The latter one may be the most likely reason because it was hard to determine the exact focal plane as the fluorescence signal from the knife edge was weak and a knife-edge scan took about several hours to complete.

The focusing efficiency has been defined as the percentage of incident x-ray beam delivered to the focus. In our experiment, it is determined by the ratio of the intensity of the focused beam and the total integrated intensity illuminating the ZP. The incident beam intensity was sampled using a 20-\mu m-diameter pinhole at several positions of the illuminating beam. The intensity variation found to be less than 15%. The average flux density over 20 sample points was then used to calculate the integrated incident beam intensity, which was obtained as the product of the average flux density and the area of the ZP. The detector used was an ionization chamber. To measure the focused beam intensity, a 20-\mu m pinhole was scanned in the focal plane, and the focus spot intensity distribution was obtained. The focused beam intensity was then obtained by subtracting from the maximum intensity at the focus the intensity near but outside the focus, which accounts for all contributions other than the positive first-order focused beam, including undiffracted zero order and all diffraction orders except the first positive order. Using this procedure, we obtained a focusing efficiency of 13%. This value is smaller than the calculated value of 19%, based on a previously published formula assuming a Au-zone/open-zone ratio of unity. The actual Au-zone/open-zone ratio in the ZP varies along the radial direction and this may explain the difference between the measured and calculated values. Furthermore, the actual thickness of the ZP may be smaller than 0.9 \mu m, which could also contribute to the difference. Nevertheless, even with 13% focusing efficiency, the flux density gain at the primary focus of the zone plate was about $10^5$.

IV. X-RAY MICROPROBE

We have developed an x-ray microprobe using a PZP as the main focusing optic at the APS. This microprobe combines the high performance of a PZP with the high brilliance of the APS undulator radiation. A photon flux density exceeding $5 \times 10^{10}/(s \mu m^2 \times 0.01\%BW)$ has been obtained. The scanning microprobe has many unprecedented capabilities by adding submicron spatial resolving power to the wide range applications of x rays. The microprobe is very flexible and versatile and can be used simultaneously to perform x-ray diffraction microscopy, fluorescence microscopy, spectromicroscopy, and three-dimensional (3D) tomography with submicron spatial resolution. It has been used for a broad range of applications since its commissioning in December, 1996. For example, it has been used in the study of the symbiotic relation between plant root and fungi, platinum-based anticancer agents, integrated laser/modulator microdevices, electromigration in Al wires in semiconductor devices,
and submicron spatial resolution 3D tomography of a single cell. As an example, Fig. 2 shows images of the spatial distribution of P, Ca, Fe, and Cu in a single biological cell. These images were obtained by recording fluorescence signals of those elements as a function of the relative position of the cell with respect to the focus of the PZP. While the significance of the results obtained from these images is outside the context of this article, the ability of imaging many elements in a single biological cell, including minor trace elements such as Fe and Cu, with a spatial resolution of 150 nm is a first of its kind.

In conclusion, we have developed a high performance PZP and demonstrated the focusing of 8 keV x-rays to a spot size less than 100 nm (FWHM) for the first time using the third-order focus. The first-order focal spot size (FWHM) is 150 nm, and the flux density gain is about 121,000. The PZP has been used to construct a state-of-the-art x-ray microprobe that has been used for a variety of important microscopic studies.

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16 B. Lai et al. (to be published).