第 15 卷　第 12 期
2007 年 12 月

光学 精密工程
Optics and Precision Engineering

Vol. 15　No. 12
Dec. 2007

Article ID　1004-924X(2007)12-1894-06

X 射线成像波带片及制作

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摘要：研究了 X 射线成像波带片的工作原理和制作工艺。从理论上分析了波带片的空间分辨率与最外环宽度的关系，以及波带片衍射效率与厚度和折射率的关系。利用国家同步辐射实验室发展的加工工艺，即电子束光刻技术和 X 射线光刻技术结合制作波带片。实验结果表明：波带片最外环宽度为 150 nm，高宽比为 4，基本满足高分辨 X 射线成像波带片的高空间分辨率、大高宽比、高精度等要求。

关　键　词：菲涅耳波带片；X 射线成像；空间分辨率；衍射效率
中图分类号：O434.1；O436.1　　**文献标识码：**A

X-ray imaging Fresnel zone plates and fabrication

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Abstract: In order to investigate the principle and fabrication technique of Fresnel zone plates (FZPs), a diffractive optical element used in X-ray imaging with the best resolution currently is presented. A theoretical analysis of the spatial resolution depending on the outermost zone width is given. An analytical expression of the diffraction efficiency in terms of thickness and X-ray refractive index of the zone plate is also provided. Electron beam lithography combined with X-ray lithography is employed to fabricate FZPs. Experimental results indicate that the outermost zone width is 150 nm and the aspect ratio is 4, which satisfies the X-ray imaging system requirements of high resolution, high aspect ratio as well as high precision.

Key words: Fresnel zone plate; X-ray imaging; spatial resolution; diffraction efficiency

1　Introduction

X-ray microscopy techniques can provide unique information for the environmental, materials and life sciences[1-4]. When Roentgen discovered X-rays in 1895, various optics were developed to focus them. However, the fabrication of such optics was delayed almost a century, due to the difficulty of realizing refraction and reflection at X-ray wavelengths. The physical origin of this technical difficulty is closely linked to the

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Received date: 2007-08-20; **Revised date**: 2007-10-10.
Foundation item: Supported by the National Natural Science Foundation of China (No. 10675113)

real, phase-shifting part of the refractive index that, for all materials at X-ray wavelengths, is very close to unity, *i. e.*, the contrast with respect to vacuum is very small[5].

The development of high-brilliance X-ray sources coupled with advances in X-ray optics has led to significant improvements in high-resolution X-ray microscopy recently. Now Fresnel zone plates are the key optics in most X-ray microscopes, allowing high-resolution investigation of a wide range of samples. Fresnel zone plates are known to be the most powerful focusing devices in the soft X-ray region, in which two-dimensional images resolved down to 15 nm have been achieved[6]. The soft X-ray microscopy can provide a unique insight into the cellular universe. The biological samples are imaged in the so called ′water window′, where X-ray photons have energies with a range between the K-shell absorption edges of carbon (284 eV) and oxygen (543 eV). In this range, organic materials absorb approximately an order of magnitude more strongly than water, which allows imaging of the ′living′ and untreated biological samples. More recently, X-ray microscopy has been extended to higher X-ray photon energies (hard X-ray). This extension enables the imaging of much thicker samples using phase contrast, encouraging the investigation of thicker biological samples in their natural environment and in material sciences[7].

The progress achieved in nanofabrication technologies has stimulated interest in the fabrication of diffractive optics and, in particular, Fresnel zone plates. For hard X-ray focusing applications, zone plates need a sufficiently large enough aspect ratio. Here we report the process for the fabrication of high-efficiency, high-resolution gold and nickel Fresnel zone plates for hard X-ray (6～12 keV) using electron beam lithography and X-ray lithography, with the outermost zone width as small as 150 nm. This research was undertaken at the National Synchrotron Radiation Laboratory.

2 Zone plates based X-ray imaging

A Fresnel zone plate consists of concentric zones with alternate optical refractive indexes. The distribution of the zones in the zone plate alters an incident wavefront in such a way that the beam emerging from the zones interferes constructively at the focus of the zone plate. The wavefront alteration is obtained by either attenuation or phase change of the incident wave by neighbouring zones. According to the Rayleigh criterion for incoherent object illumination, the spatial resolution of a zone plate based microscope is given by

$$\Delta = 1.22 dr_n, \tag{1}$$

where dr_n is the outermost zone width of the objective zone plate[8-10].

A transmission X-ray microscope is installed at Beamline U7A of NSRL with a superconducting wiggler source, which provides intense X-rays in the 7～12 keV energy range. The X-rays are monochromatized by a crystal monochromator system. A high-resolution X-ray imaging end station consisting of a condenser system, a sample stage, mechanical and electrical systems is installed. There are four objective zone plate lenses covering X-ray energies in the range 7～12 keV. A schematic of the microscope is shown in Fig. 1. The outermost zone width of the objective zone plate is a 45 nm, so, according to formula (1), it is possible to achieve better than 60 nm diffraction-limited spatial resolution with well engineered mechanical and thermal stability[11].

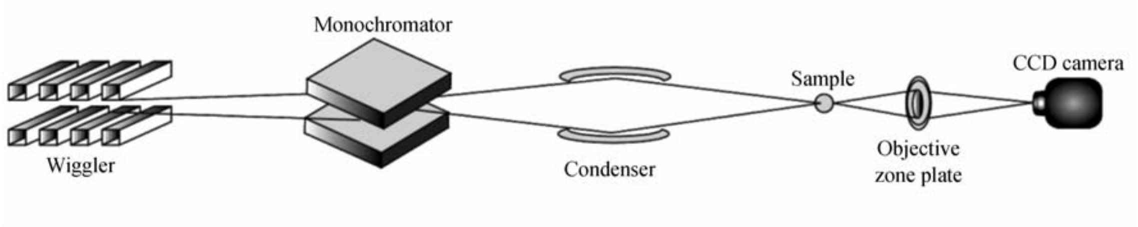


Fig. 1 Schematic of the transmission X-ray microscope installed at NSRL

3 Focusing efficiency of a zone plate

Together with lateral resolution, high efficiency and wide-energy operating range represent the other most important properties of a ZP. Fresnel zone plates currently give the best spatial resolution for X-ray microscopy, but they are usually limited to low photon energies. At high energies, their use is still limited by their efficiency. Note that an amplitude zone plate′s efficiency is no better than 10%, while the maximum focusing efficiency obtained from a phase zone plate is 4 times than that from an amplitude zone plate[10]. It is necessary that a zone plate has adequate thickness to modify the incident wave front for focusing. The thickness of a phase zone plate is determined by refraction of the material. To provide a π phase shift and form a hard X-ray phase ZP, a thickness of a few microns with a high aspect ratio is required.

High-Z materials are generally preferred for hard X-ray applications. The focusing efficiency of a ZP is generally expressed as a function of the zone plate thickness d and the refractive indexes n_1 and n_2, which are dependent on the materials from which the ZPs are made and the X-ray energy used. The diffraction efficiency of a ZP with a square zone profile and equal adjacent zones areas can be calculated[12-13]. The calculation of the diffraction efficiencies for first-order light were performed for a Au and a Ni zone plate at a photon energy of 7.5 keV. Fig. 2 shows the diffraction efficiencies without the influence of roughness and interdiffusion between zones. We can see that Au is a well-suited material for highly efficient, thick zone plates.

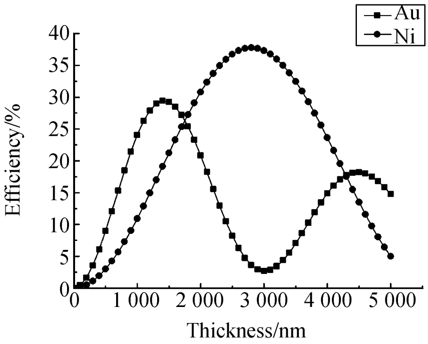


Fig. 2 Variation for first diffraction efficiency of FZPs together with the thickness of Au and Ni. The energy of hard X-ray is 7.5 keV at a wavelength of 0.1 653 nm. Density of Au is 19.3 g/cm^3 and Ni is 8.9g/cm^3. Diamonds plot the first diffraction efficiency of Au, and dots the first diffraction efficiency of Ni.

4 Zone plate fabrication

It is well known that high-resolution resist features made with a high aspect ratio can collapse due to the lack of mechanical strength. This is a fabrication challenge to the energy operational range of the high resolution ZP. Here we report the fabrication of high-efficiency, high-resolution Au and Ni Fresnel zone plates for hard X-ray (6～12 keV), using electron beam lithography and X-ray lithography.

Electron beam lithography can produce high-resolution structures, but is limited in its aspect ratios to around unity. To overcome the aspect ratio limitation of e-beam lithography, the X-ray lithography is employed. First, a ZP X-ray mask with a low aspect ratio is fabricated using electron beam lithography. Then the required high aspect ratio zone plates are fabricated through X-ray lithography[14-15].

Fig. 3 shows the process flow chart for FZPs mask fabrication by e-beam lithography. The zone plate carrier, a Si_3N_4 membrane of 5 mm×5 mm formed on a Si wafer, is coated with a Cr/Au plating base and PMMA resist, and is then exposed by e-beam lithography. The structures are developed in a methyl-isobutyl ketone (MIBK): isopropanol (IPA) mixture. Patterns are transferred into Au by electroplating. The mask for FZPs is completed after removing the resist.

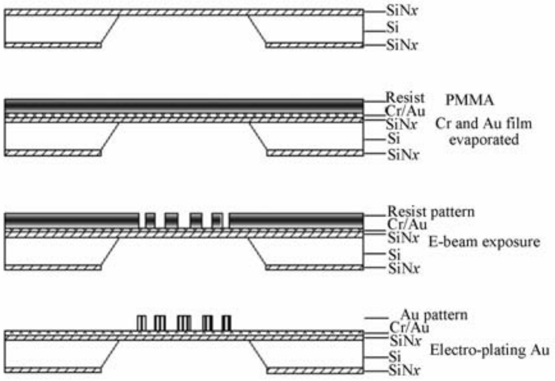


Fig. 3　Process flow chart for FZPs mask fabrication by e-beam lithography

The wavelength of the U1 (X-ray lithography) beamline at NSRL is between 0.5 nm and 2 nm. The exposure area is 30 mm×30 mm, with an asymmetry of ±5%. Fig. 4 shows the process flow chart for the fabrication of FZPs with high aspect ratio by X-ray lithography. The zone-plate carrier, a Si_3N_4 membrane of 5 mm×5 mm formed on a Si wafer, is coated with a Cr/Au plating base and 600 nm PMMA resist. The ZP patterns on the mask are transferred into PMMA through X-ray lithography. The structures are also developed in a methyl-isobutyl ketone (MIBK): isopropanol (IPA) mixture. The ZP's outermost pattern with a high aspect ratio is formed by an electroforming process. The FZPs with high aspect ratio for X-ray imaging is completed after removing the resist and etching the Cr/Au plating base.

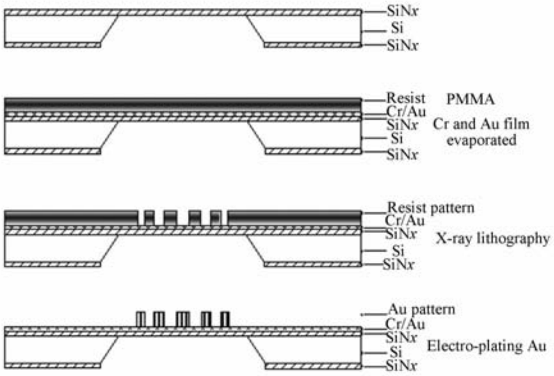
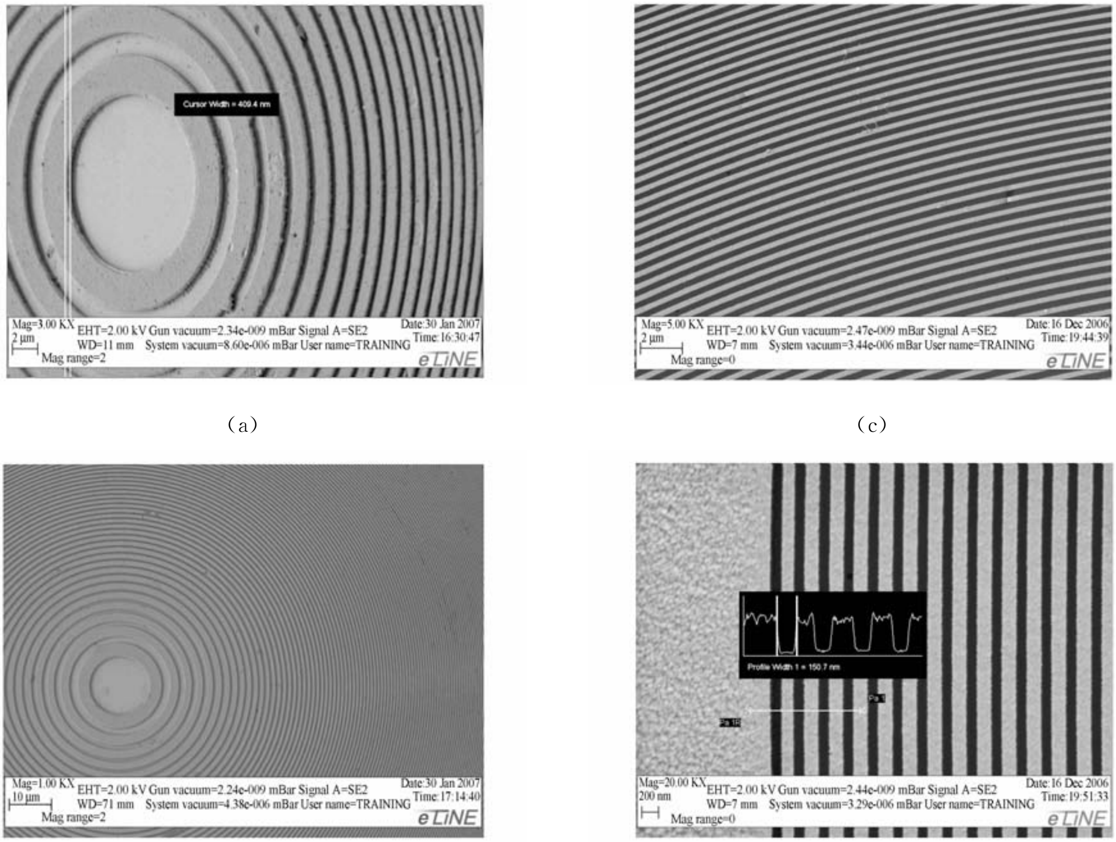


Fig. 4　Process flow chart for FZP fabrication with high aspect ratio by X-ray lithography

A ZP with an outermost zone width of 150 nm and thickness of 600 nm is fabricated by this techinique at NSRL. The material is Au and the aspect ratio is 4. Fig. 5 shows the scanning electron micrographs (SEMs) of the fabricated ZPs. Fig. 5(a) is the SEM of the central zone. The thickness of the FZP is 580 nm. Fig. 5(b) is the SEM of the central zone region of the fabricated ZP. Fig. 5(c) is the micrograph of the outer zone region of ZP. Also shown in Fig. 5(d) is a detailed view of the outermost zones of the FZPs. The outermost zone width, as indicated by the curve, is about 150 nm.



(a) (c)

(b) (d)

Fig. 5 SEMs of FZP made from Au. The outermost zone width of FZPs is 150 nm, with the thickness of 580 nm and the aspect ratio of 4.

5 Conclusions

The development of high-brilliance X-ray sources coupled with advances in nanofabrication technologies has led to significant improvements in high-resolution X-ray microscopy. The high aspect ratio nanodevices fabrication method has been developed at NSRL. Zone plates with an outermost zone width between 150 and 100 nm are fabricated using this technique.

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