Electron Beam Lithography of Fresnel Zone Plates Using A Rectilinear Machine And Trilayer Resists

D. Tennant[1], S. Spector[2,3], A. Stein[2], C. Jacobsen[2]

[1] *Lucent Technologies Bell Laboratories*
[2] *Department of Physics and Astronomy, SUNY Stony Brook*
[3] *Present address: MIT Lincoln Laboratory*

Abstract. We describe the use of a commercial e-beam lithography system (JEOL JBX-6000FS) to fabricate Fresnel zone plates for x-ray microscopy. The machine is capable of controlling the pitch of optical gratings with sub-nanometer precision, so its beam placement properties are more than adequate for zone plate fabrication. The zone plate pattern is written into a thin top layer (PMMA or Calixarene) of a trilayer resist, and transferred into thick nickel zones using reactive ion etching (RIE) followed by electroplating. Zone plates with outermost zone widths of 30 nm have exhibited efficiencies up to 10.0% at a 390 eV photon energy and with diameters in the range 80 to 120 μm. Zone plates with outer zones of 18 to 20 nm were also fabricated in thinner Ni with correspondingly lower efficiencies of 2.6%. Zone plates with outermost zone widths of 45 nm have been fabricated with larger diameters up to 160 μm. All results reported were obtained with a 50 kV system with 80 μm field deflection size; future efforts will make use of a 100 kV, 500 μm field size system.

INTRODUCTION

Fresnel zone plates are the key optic in most x-ray microscopes, allowing high-resolution investigation of a wide range of samples. The X1A beamline at the National Synchrotron Light Source (NSLS) at Brookhaven National Laboratory uses zone plates as the focussing elements in scanning transmission x-ray microscopes (STXM) (see *e.g.*, [1, 2]). The zone plate is illuminated by x-rays from an undulator source and focussed to a diffraction-limited spot through which the sample is scanned. The transmitted x-rays are used to form an image of the object. By tuning the monochromator, the microscope can take absorption contrast images, elemental and chemical maps and spectroscopic data. The STXM plays a role in biology, polymer science, colloidal science, environmental science, and geochemistry research.

To achieve the high resolution patterning required for fabrication, the zone plates are exposed with a JEOL JBX-6000FS electron beam lithography (EBL) system at Lucent Technologies, Bell Laboratories. The system uses a ZrO/W field emission gun with a current density of about 2,000 A/cm^2. This machine delivers 1 nA of beam current at 50 keV into a 7 nm spot with 16 bit control of the beam position within a 80 μm writing field. A $\lambda/1024$ (~0.6 nm) interferometer is incorporated for deflector calibration and stage positioning to enable field stitching over larger areas. The precision of such an instrument for production of diffractive optical elements has been demonstrated in other applications such as optical gratings for wavelength selectable sources [3] needed for future lightwave communication systems. In that work, arrays of gratings with periods centered near 240 nm were produced in which adjacent grating were designed and measured to have consecutive period changes of 0.13 nm.

Fresnel zone plates are diffractive optics that give the highest resolution of any optic presently available in the soft x-ray region. The transverse spatial resolution, $\delta_{t,}$ is equal to $1.22\delta_{rN}$, where δ_{rN} is the outermost zone width. As with all imaging techniques, the highest resolution is the ultimate goal. Thus, the first fabrication challenge is to produce the smallest δ_{rN} while maintaining proper zone placement.

The focal length f of a zone plate with diameter d is given by $f=d\delta_{rN}/\lambda$. Thus, a decrease in δ_{rN} leads directly to a decrease in the focal length if the diameter d is not increased proportionally. Since the zone plates are diffractive, they have multiple orders, which are eliminated from 1st order imaging by an order-sorting aperture (OSA) - a pinhole placed between the zone plate and the sample (see Figure 1). Certain applications of STXM require large focal lengths for working with thick sample holders. The focal length is also inversely proportional to the wavelength of the x-rays. This means that certain spectral regions (e.g. the carbon edge) imply a smaller, i.e. more difficult, focal length. The second fabrication challenge, therefore, is to produce large diameter zone plates while maintaining proper zone placement and small δ_{rN}.

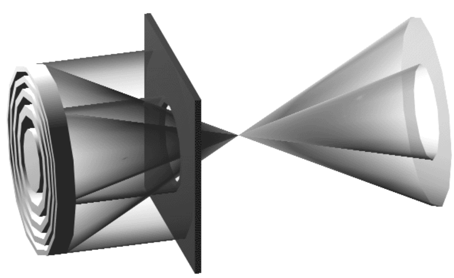

Figure 1: Schematic view of a Fresnel zone plate used in a scanning microscope. The first order focus is isolated from the non-diverging 0th order (and the diverging –1st order, not shown here) by the use of a central stop on the zone plate, and a pinhole called the Order Sorting Aperture (OSA) located at a distance of about (2/3)f from the zone plate of focal length f.

Lastly, the diffraction efficiency must be as high as possible, so that the sample is well illuminated in as short a time necessary for high-resolution imaging. This is achieved by fabricating zones which are thick enough to attenuate (for absorption zone plates) or phase-shift (for phase shifting zone plates) the incoming x-rays. Also, the zones should be made from a material that gives the desirable phase-shifting/absorption properties; a good choice for soft x rays is nickel [4].

To meet these challenges with a rectilinear scanned commercial electron beam lithography tool exploratory materials and software modifications were employed. Zone plates with minimum zone widths of δ_{rN}=18 nm, d=80 μm were fabricated in nickel [5].

Zone plates with δ_{rN}=45 nm, d=160 μm were also fabricated in nickel to give larger focal lengths while maintaining high resolution.

FABRICATION

Trilayer Resist and Electroplating

The zone plates are fabricated using a trilayer (Figure 2) scheme [6] on a Si_3N_4 membrane. The trilayer allows high-resolution e-beam lithography in a thin resist while still allowing higher aspect ratios to be achieved. Presently, the trilayer consists of a plating form of AZ 4110 resist deposited on a plating base of chrome and gold. A thin layer of germanium is evaporated for use as a hard etching mask upon which the resist is deposited. After exposure and development, the pattern is transferred by RIE and then electroplated with nickel.

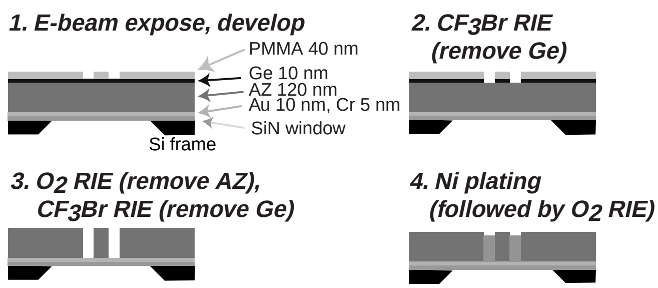

Figure 2: The use of trilevel resist schemes allows one to fabricate high aspect ratio structures at the resolution limits of electron beam lithography. The electron beam exposure is done in a thin resist to minimize linewidth degradation by electron forward scattering. This pattern is then transferred into a thicker resist by reactive ion etching, and that resist is then used as a plating mask. The particular scheme shown here is that used by Spector *et al.*, *J. Vac. Sci. Tech.* **B 15**, 2872 (1997).

Resists

Initially, poly(methyl methacrylate) (PMMA) was used for the zone plate patterning. This has proven to be adequate for δ_{rN} > 30 nm [7]. As the need for higher resolution (and thus smaller δ_{rN}) has grown, the performance of PMMA was not acceptable. The resist Calixarene, with its increased resolution [8], was used to fabricate test zone plates with δ_{rN} down to 18 nm in 60 nm thick nickel [5]. A δ_{rN}=20 nm test zone plate is shown in Figure 3 along with its diffraction efficiency measurements. The figure shows that the efficiency falls off only rather slowly as the zone width is decreased to 20 nm.

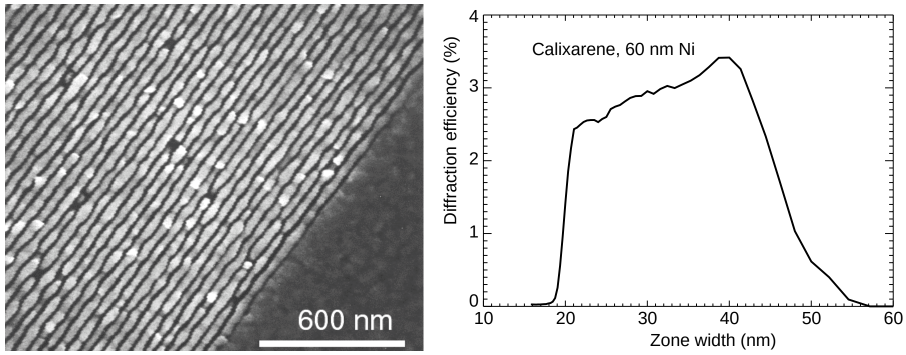

Figure 3: At left is shown 20 nm outermost zones in a test zone plate fabricated in 60 nm nickel using the photoresist Calixarene. The diffraction efficiency of this zone plates at 400 eV is shown at right; it is encouraging to note that the diffraction efficiency falls off only slowly as the zone width is decreased from 50 to 20 nm with this process.

Larger Diameter Zone Plates

Certain applications of the STXM and the Cryo-STXM [9], such as tomographic imaging [10] and wet specimen holder work [11, 12], require a working distance between order sorting aperture and specimen to be in the 300-1000 μm range. At the same time, the diameter of the central stop used for order sorting should be no more than about half the zone plate diameter to maintain good optical performance. As a result, it is important for many applications to have the focal length of the zone plate be significantly larger than 600-2000 μm.

This requirement for long focal lengths means that the zone plate diameter must be increased. For example, a δ_{rN}=30 nm, d=80 μm diameter zone plate has a focal length of only 560 μm at the carbon absorption edge (290 eV). Figure 4 shows how the diameter must increase as zone width is decreased to maintain a 1 mm focal length.

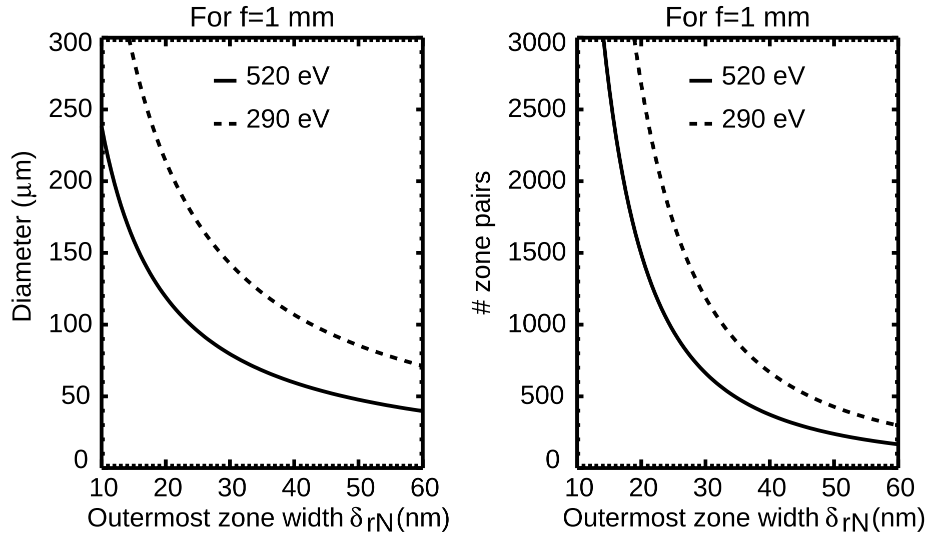

Figure 4 : Requirements on zone plate diameter and number of zone pairs to maintain a 1 mm focal length as outermost zone width is decreased, for 520 eV (oxygen edge) and 290 eV (carbon edge). At present, the largest zone plates we have fabricated have d=160 µm and δ_{rN} = 45nm. Future efforts will be directed towards increasing d for finer outermost zone widths δ_{rN}.

Because the JEOL's deflectors have an 80 µm range, the larger zone plates require movement of the stage and the stitching of multiple fields. Figure 5 shows examples of good and bad stitching as well as a 160 µm zone plate with 45 nm outer zone width. Good field stitching requires accurate alignment, thermal stability and good electrical conductivity on the sample during pattern writing.

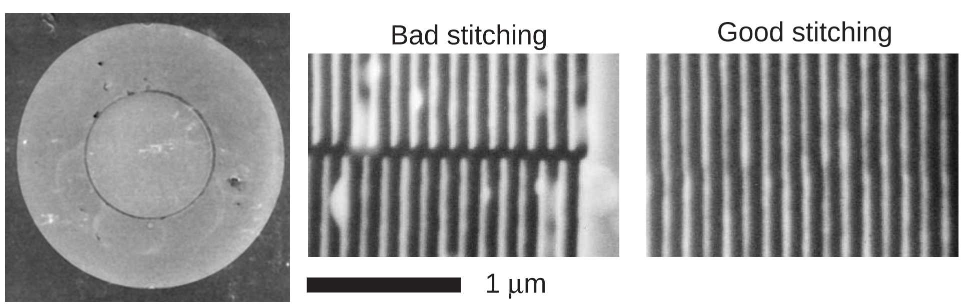

Figure 5: At left is shown a d=160 µm diameter, δ_{rN}=45 nm zone plate fabricated in 180 nm of nickel (no zones can be seen at this magnification scale, but the central stop can be distinguished). With the JEOL JBX-6000FS, zone plates of this type require the stitching of four 80 µm fields. Initial efforts in field stitching were less than fully successful (middle), but in subsequent work (right) we were able to obtain excellent field stitching by improving thermal equilibration and electrical conduction of the zone plate substrate so as to prevent electrostatic charging.

Future developments

A new EBL tool, the JEOL JBX-9300FS, is due to be delivered and installed at Bell Labs during 1Q 2000. This machine will provide several improvements over the present system that will facilitate an improvement in zone plate fabrication. The operating acceleration voltage is increased from 50 to 100 keV, thereby further reducing the forward scattering of electrons in thin resists [13] and increasing exposure resolution due to improved absorbed energy contours. The beam scanning resolution is increased to 19 bits (or 1.0 nm within a 500 μm field) and the spot size is expected to drop from 7 to 4 nm. The deflection range increases from 80 to 500 μm at the highest resolution setting so fabrication of large diameter zone plates will be possible without the movement of the stage, and therefore without field stitching.

REFERENCES

1. Jacobsen, C., *et al*. *New developments in scanning microscopy at Stony Brook*. These proceedings.

2. Feser, M., *et al*. *Instrumentation advances and detector development with the Stony Brook scanning transmission x-ray microscope*. These proceedings.

3. Tennant, D., *et al*., *Multiwavelength distributed Bragg reflector laser array fabricated using near field holographic printing with an electron-beam generated phase grating mask*. Journal of Vacuum Science and Technology, 1993. **B 11**(6): p. 2509-2513.

4. Anderson, E. and D. Kern. *Nanofabrication of zone plate lenses for high resolution x-ray microscopy*. In A.G. Michette, G.R. Morrison, and C.J. Buckley, ed.,*X-ray Microscopy III*. (Springer-Verlag, 1992), pp. 75--78.

5. Spector, S., C. Jacobsen, and D. Tennant, *Process optimization for production of sub-20 nm soft x-ray zone plates*. Journal of Vacuum Science and Technology, 1997. **B 15**(6): p. 2872--2876.

6. Tennant, D., *et al*., *25 nm features patterned with trilevel e-beam resist*. Journal of Vacuum Science and Technology, 1981. **19**: p. 1304-1307.

7. Spector, S.J., *Diffractive optics for soft x rays*. PhD dissertation, Department of Physics, State University of New York at Stony Brook: Stony Brook, NY, 1997.

8. Fujita, J., *et al*., *Ultrahigh resolution of calixarene negative resist in electron beam lithography*. Applied Physics Letters, 1996. **68**: p. 1297--1299.

9. Maser, J., *et al*., *Soft x-ray microscopy with a cryo STXM: I. Instrumentation, imaging, and spectroscopy*. Journal of Microscopy (in press).

10. Wang, Y., *et al*., *Soft x-ray microscopy with a cryo STXM: II. Tomography*. Journal of Microscopy (in press).

11. Neuhäusler, U., *et al*., *Soft x-ray spectromicroscopy on solid stabilized emulsions*. Colloid & Polymer Science, 1999. **277**: p. 719-726.

12. Neuhäusler, U., *et al*., *A specimen chamber for soft x-ray spectromicroscopy on aqueous and liquid samples*. Journal of Synchrotron Radiation (in press).

13. Kyser, D.F., *Spatial-resolution limits in electron beam nanolithography*. Journal of Vacuum Science and Technology B, 1983. **1**: p. 1391--1397.