

A proposal for maskless, zone-plate-array nanolithography

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We propose a novel form of x-ray projection lithography that: (1) requires no mask, and hence can be considered an x-ray pattern generator; (2) is, in principle, capable of reaching the limits of the lithographic process. The new scheme utilizes an array of Fresnel zone plates, and matrix-addressed micromechanical shutters to turn individual x-ray beamlets on or off in response to commands from a control computer. Zone plate resolution is approximately equal to the minimum zone width, which can approach 10 nm. Zone plates are narrow-band lensing elements: For a diffraction limited focus, the source bandwidth $\Delta\lambda/\lambda$ should be less than or equal to the reciprocal of the number of zones N . An undulator having N_u magnetic sections emits collimated radiation in a bandwidth $\Delta\lambda/\lambda = 1/N_u$. N_u is usually in the range 35–100. We present a system design based on 25 nm lithographic resolution using $\lambda=4.5$ nm. For $N=100$ the zone-plate diameter is 10 μm . Each zone plate of the array would be responsible only for exposure within its ‘unit cell.’ To fill in a full pattern, the stage holding the sample would be scanned in X and Y while the beamlets are multiplexed on and off. A microundulator designed for installation on a commercial compact synchrotron can provide 87 mW within a 2% bandwidth around 4.5 nm in a divergence cone of 0.28 mrad. The calculated efficiency of first-order focus for a zone plate operating at 4.5 nm is 31%, using 130 nm of spent U as the absorber/phase shifter. An exposure rate of ~ 1 cm^2/s at 25 nm resolution appears feasible. © 1996 American Vacuum Society.

I. INTRODUCTION

The relationship between mask–substrate gap G and minimum feature size W in conventional x-ray lithography is given by

$$G = \alpha W^2/\lambda, \quad (1)$$

where λ is the x-ray wavelength (~ 1 nm) and α is in the range 1–1.5.^{1–3} For feature sizes below 50 nm, the gap must be below 4 μm . Although such small gaps, and even mask–substrate contact, are feasible in research, it is questionable whether this would be acceptable in future manufacturing. Thus, one is persuaded to consider x-ray projection, especially for feature sizes below 50 nm. At x-ray and extreme UV (EUV) wavelengths there appear to be only three possible approaches to projection lithography: imaging with multilayer mirrors, in-line holography, and imaging with zone plates. The latter is the simplest and probably the most practical. In this article a novel maskless version of zone-plate-based lithography is proposed which appears to be highly attractive from the points-of-view of efficiency, throughput, and flexibility. It relies upon recent advances in micromechanics and spatial-phase-locked e-beam lithography, both of which are undergoing rapid development.

II. WHY ZONE PLATES FOR NANOLITHOGRAPHY

The imaging properties of Fresnel zone plates have been understood since the late 19th century.⁴ Zone plates have been used for many years in x-ray microscopy at the ‘‘water window’’ around 2.4 nm.^{5–9} In this application they work

extremely well, revealing details, for example, in biological specimens, that are not observable with either electron microscopy or conventional optical microscopy. Burge, Browne, and Charalambous were the first to propose the use of a zone plate in x-ray lithography.¹⁰ To circumvent the problem of the very limited field-of-view of zone plates, Hector and Smith¹¹ and Feldman¹² proposed two different lithography schemes using arrays of zone plates. Both schemes require a mask and two zone-plate arrays in tandem. Because the focusing efficiency of zone plates in the x-ray regime is in the range 10%–33%, the need for two tandem arrays implies a focusing efficiency of only 1%–9%, at best.

The appropriate wavelength to use for sub-100 nm lithography is either 4.5 nm, at the carbon K absorption edge, or around 1 nm.¹ At the C_K edge, resists such as PMMA, which are composed primarily of C and H, attenuate only about 2 dB/ μm , and hence can be quite thick. Early, Schattensburg, and Smith¹³ and Ocola *et al.*² showed that at a wavelength around 1 nm the ranges of photoelectrons and Auger electrons do not prevent one from achieving resolutions below 30 nm. The intrinsic resolution at the 4.5 nm wavelength is ~ 5 nm, which is probably at or just beyond the practical limit of the lithographic process itself. For the zone-plate-array scheme described here, 4.5 nm is the optimal wavelength from the points-of-view of resolution, source characteristics, and zone plate fabrication, as described below.

III. ARRAY WRITING STRATEGIES

The proposed lithography scheme is shown schematically in Fig. 1. It does not require a mask, and employs a single zone-plate array. At a wavelength of 4.5 nm, a focusing ef-

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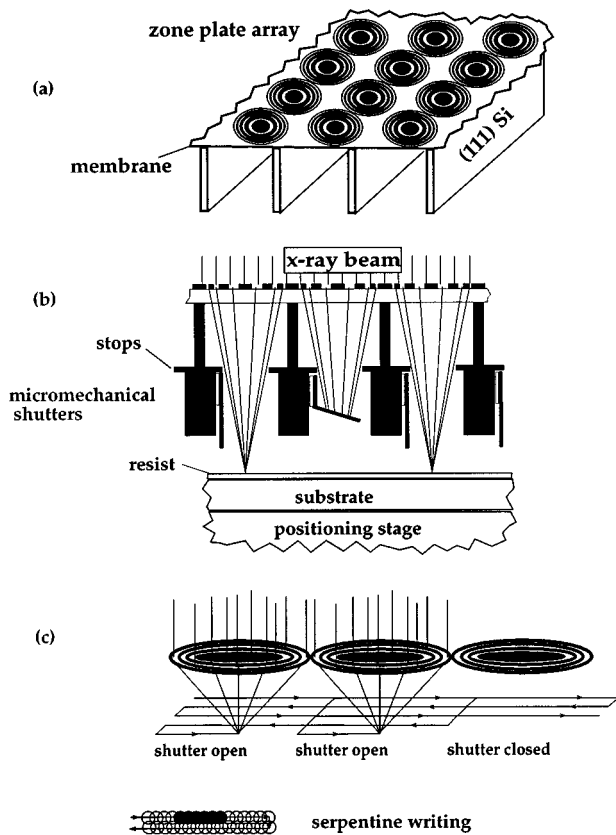


FIG. 1. Schematic view of the proposed maskless x-ray projection system. (a) Perspective view of an array of Fresnel zone plates on a (110) Si substrate. Each zone plate, which defines a "unit cell," is supported on a thin carbonaceous membrane, with vertical, anisotropically etched Si (111) "joists" for rigid mechanical support. Each zone plate is responsible for exposure only within its unit cell ($<16 \times 10^4$ pixels). (b) Cross section illustrating the focusing onto a resist-coated substrate, and "downstream" micromechanical shutters which turn beams on and off in response to commands from a control computer. (c) Illustration of one possible writing scheme, with the micromechanical shutters located "upstream" and the substrate scanned in X and Y by a fast piezoelectric system, thereby filling in the full pattern.

efficiency of 31% is feasible. The system is, in effect, a maskless zone-plate-array pattern generator. Each zone plate of the array is able to focus a collimated beam of x rays to a fine spot on a resist coated substrate. To write a pattern, the substrate is scanned under the array, while the individual beamlets are turned on and off as needed by means of micromechanical shutters, one associated with each zone plate. These shutters can either be "downstream," as depicted in Fig. 1(b), or "upstream," as in Fig. 1(c).

There are various ways in which the scanning and writing can be done. One is to employ a square array of zone plates, as depicted, with each zone plate "responsible" for patterning only within its "unit cell." The scanning in this case would be serpentine, with the stage moving only a distance equal to the zone-plate diameter ($\leq 10 \mu\text{m}$) in X and Y until all pixels within all the unit cell are addressed, and either written (i.e., shutter open) or not (i.e., shutter closed). The entire array would then be stepped a distance equal to its diameter and scanning repeated. An alternative to this "step-

and-scan" strategy is a linear-scan strategy described by Feldman.¹² It employs a close-packed array of zone plates, rotated in such a way that all pixels can be addressed when the substrate is scanned along one direction only.

In order that lithographic features which cross boundaries between unit cells are free of stitching errors it is necessary that the zone plates be arranged in the array with a placement precision much finer than a pixel diameter. Spatial-phase-locked e-beam lithography¹⁴ could be used to accomplish this.

IV. ZONE-PLATE ARRAY DESIGN

The principle of operation of Fresnel zone plates has been described in detail elsewhere.^{4-12,15} A zone plate can be thought of as a structure of circular symmetry in which the local spatial period depends on radius in such a way that first-order diffracted radiation from any radius value crosses the axis at the same point, the focal length. For a plane wave incident the equation that describes the relationship among the first-order focal length f , the zone number n , and the zone radius R_n follows from the Pythagorean theorem and the condition for constructive interference,

$$(R_n)^2 + f^2 = [f + n\lambda/2]^2, \quad (2)$$

where λ is the wavelength. Letting p represent the "pitch" or period of the outermost zones, the angle of convergence to focus θ for a plane wave incident is given by

$$\sin \theta = \lambda/p. \quad (3)$$

Just as transmission diffraction gratings can be based on periodic obstruction or periodic phase shifting, so also zone plates can be based on obstruction ("amplitude" zone plates) or phase shifting ("phase" zone plates), and all intermediate types as well. Pure phase zone plates have a focusing efficiency of 40% whereas amplitude zone plates focus only 10% of the incident radiation into the positive first-order focus. Because zone plates are based on diffraction they are subject to chromatic aberration. That is, different wavelengths are focused at different axial distances. A zone plate will produce a diffraction-limited focal spot only for radiation in a bandwidth (BW) given by

$$\text{BW} = 1/N, \quad (4)$$

where N is the total number of zones.

The starting point of the design of a zone-plate-array pattern generator is the x-ray source since its bandwidth dictates other system parameters. The bandwidth of line radiation from inner-shell atomic transitions is sufficiently narrow in many cases for our purposes, however, such sources generally do not have sufficient brightness (i.e., photons emitted per unit area per unit solid angle). Expressed another way, such sources have sufficient temporal coherence but inadequate spatial coherence for high throughput. Synchrotron radiation has good collimation (i.e., spatial coherence) but inadequate spectral brightness (i.e., power in a narrow bandwidth). The optimal source for the zone-plate-array pattern generator is an undulator attached to a synchrotron. Such

sources, which consist of a linear array of alternating magnetic fields inserted into a straight section of a synchrotron orbit, have bandwidths given by an equation identical to Eq. (4), except that N in this case is the number of periods of the alternating magnetic field N_u . In modern undulators, permanent magnets are used with magnetic-field strengths ~ 0.35 T, and N_u between 35 and 100. The number of zones in the zone plates is therefore tied directly to the number of periods in the undulator. Synchrotrons, both superconducting¹⁶ and nonsuperconducting,¹⁷ designed specifically for lithography, are available commercially and can be provided with undulators. The following design is based on the specifications of one such undulator.^{17,18}

For $p = 50$ nm (i.e., 25 nm zone widths) and $\lambda = 4.5$ nm, $\sin \theta = 0.09$. Making the approximation $\sin \theta = \tan \theta$, we have $\lambda/p = R_N/f$. Substituting for f in Eq. (2) and solving for R_N we obtain

$$R_N = Np. \quad (5)$$

Thus, for $N = 35$ –100 and $p = 50$ nm, the zone plate diameter D is in the range 3.5–10 μm . Substituting Eq. (5) in Eq. (2), the focal length is given by

$$f = Np^2/\lambda, \quad (6)$$

to within 0.2% accuracy. Thus, f is in the range 19–56 μm . The minimum focal spot size is approximately equal to the width of the outermost zone, i.e., $p/2$. We take this to be the pixel diameter. If, for simplicity, we ignore the space taken up by the joists in Fig. 1 (in a final design they may not be needed) the number of pixels per unit cell is given by

$$\text{pixels per unit cell} = D^2/(p/2)^2 = 16N^2. \quad (7)$$

Thus, there are $< 1.6 \times 10^5$ pixels per unit cell.

The focal spot of a zone plate will be smeared out beyond its diffraction-limited value of $p/2$ because of the angular divergence of the source (i.e., nonperfect collimation). By straightforward geometry one can show that the focal spot is enlarged by the factor G , given by

$$G = 1 + (2Np\Delta\phi/\lambda), \quad (8)$$

where $\Delta\phi$ is the source divergence. Taking the value provided by a commercial undulator, $\Delta\phi = 2.8 \times 10^{-4}$ rad,¹⁷ $N = 50$, $p = 50$ nm, and $\lambda = 4.5$ nm we find that the focal spot is smeared out by the factor 1.3, i.e., from about 25 to 33 nm.

V. THROUGHPUT

We first calculate the limit on throughput imposed by the incident x-ray flux, and then consider the problem of multiplexed parallel addressing. Although undulators installed at existing synchrotron facilities can provide adequate flux¹⁹ we consider instead a type of undulator that could be installed on a compact synchrotron, suitable for manufacturing. For example, a ‘‘microundulator’’ with a period of 14 mm, installed on the Aurora 2 synchrotron,¹⁷ would provide a first-order peak at 4.5 nm, and a flux of 1.97×10^{15} photons/s, or 87 mW, in a 2% bandwidth, suitable for diffraction-limited focusing by zone plates of ~ 50 zones.¹⁸

The power incident on a resist-coated substrate P' is given by

$$P' = P\epsilon(\pi/4)(F), \quad (9)$$

where ϵ is the efficiency of first-order focusing of the zone plate, and F accounts for loss due to various factors, including the fraction of area taken up by the joists and the attenuation of the membrane supporting the zone plate array. This membrane can also serve as the vacuum window. It would be made of diamond or other strong carbonaceous material. In our laboratory we use SiN_x membranes, 1.5 μm thick and spanning a diameter of 20 mm, as vacuum windows.²⁰ We show below that the overall size of the zone-plate array is $< 1.3 \times 1.3$ mm. Hence, it should be possible to make the membrane significantly thinner than 1 μm . In order to allow for future ingenuity in the design of either the array of zone plates or the source, we assume a generous factor, $F = 0.9$. As discussed below, we can take $\epsilon = 0.31$, in which case $P' = 19$ mW, which is spread out to fill the zone-plate array. For a resist with a sensitivity of 19 mJ/cm², a maximum throughput of 1 cm²/s is predicted.

Actually, at sub-50 nm feature sizes, one should estimate throughput taking into account the stochastic nature of the resist exposure process.²¹ A resist sensitivity of 19 mJ/cm² corresponds to an incident flux of 4.3 photons/nm², or ~ 2700 photons per pixel (25 \times 25 nm²). This is a very large number by the usual lithographic criterion.²¹ For example, if 50% of the incident radiation is absorbed in a 100-nm-thick resist film, this corresponds to 950 J/cm³, which is approximately the sensitivity of PMMA. It is generally understood that low-sensitivity resists such as PMMA will be required at sub-50 nm resolution.

One can expect further improvements in undulators and perhaps alternative sources (some of which are already in the conceptual stage). Hence, the calculated throughput, which is already attractive considering the fineness of the features projected, could be further enhanced. At 1 cm²/s the equivalent ‘‘data rate’’ is 1.6×10^{11} Hz.

Up to the point where the undulator flux is the limiter, the throughput T is given by

$$T = d^2RM, \quad (10)$$

where d is the pixel diameter, R is the rate at which micro-mechanical shutters can be switched, and M is the number of shutters that can be addressed in parallel. Because the shutters would have very small masses, it should be possible to switch them at rates of several megahertz. If we assume 10 MHz switching and $d = 25$ nm, $T = 6.3 \times 10^{-5}$ M cm²/s. Thus, the throughput would take on the maximum value of 1 cm²/s, set by the undulator flux, if 16 000 zone-plate shutters are addressed and multiplexed in parallel (i.e., an array of 126 \times 126). This appears to be feasible. The area occupied by 16 000 unit cells (ignoring joist area) is $< 1.3 \times 1.3$ mm². Thus, it may be possible to avoid use of joists except on the perimeter of the zone plate array.

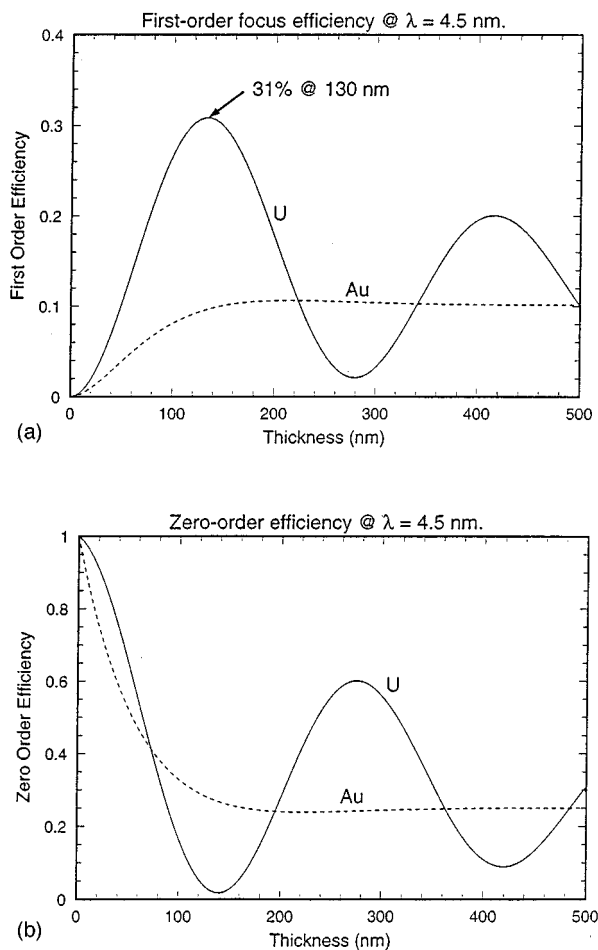


FIG. 2. (a) Plots of the fraction of incident 4.5 nm x radiation that is focused in the first-order focal spot, for zone plates made of gold and uranium, as a function of the thickness of the absorber. Note the 31% efficiency for a 130 nm thickness of U. (b) Plots of the zeroth-order efficiency for gold and U at 4.5 nm wavelength. At 130 nm thickness, uranium is nearly an ideal phase shifter.

VI. FABRICATION OF ZONE PLATES

Electron-beam lithography provides the optimal path to fabricating zone plates.²² As first demonstrated by Shaver *et al.*,²³ it is necessary to eliminate various sources of distortion. This can be done most effectively by comparing the electron-beam scan raster to a distortion free reference grid made using interferometric lithography.²⁴

Figure 2(a) is a plot of the first-order efficiency of zone plates made of uranium and gold as a function of the thickness of the absorber. Note that the efficiency of a gold zone plate never exceeds 10% whereas the efficiency of zone plates made of uranium reaches a maximum of 31% at a uranium thickness of 130 nm. This is because uranium is a nearly ideal phase shifter at $\lambda = 4.5$ nm. This is further illustrated in Fig. 2(b) in which the zeroth-order (i.e., the radiation that is propagated straight ahead) is attenuated about 98% at 130 nm thickness. Spent uranium is available in large quantities. Its fluoride UF_6 is a gas, hence the material can be reactively etched in fluorocarbon plasmas, presumably at high resolution and aspect ratios compatible with the 130 nm

thickness. Uranium does have the problem that it is pyrophoric, i.e., it will ignite in air; however, techniques for working with uranium have been developed at various laboratories around the world and this proposed peaceful use should be entirely welcome.

VII. SUMMARY

We have proposed a system for performing nanolithography (i.e., lithography below 100 nm feature sizes) that employs an array of Fresnel zone plates in conjunction with an undulator source and micromechanical shutters, all relatively new technologies, but separately proven. The size of the array of zone plates depends on how many shutters can be addressed in parallel. For the design example presented, i.e., 25 nm resolution, this number need not exceed 1.6×10^4 since the throughput at that point is limited by the undulator flux. Such an array would occupy an area of about 1.3×1.3 mm². Of course, it can be larger than this if necessary to match the area of an expanded undulator beam, in which case the shutters would be operated at a rate slower than 10 MHz.

From Eq. (7), and assuming a square array of zone plates, measuring 1.3×1.3 mm², a pixel diameter of 25 nm, and $N = 100$, an exposure rate of 1 cm²/s corresponds to a substrate scanning rate of 24 cm/s. Laser interferometer control of the stage should be sufficient at least in the case of a linear-scan strategy.

The maskless nanolithography system proposed here can be operated in a He gas environment for temperature homogeneity and control, which is a significant advantage over EUV and charged-particle projection systems which must be operated in vacuum.

The 4.5 nm photon is probably the ideal particle for nanolithography. The difference in energy between the C_K emission line and the binding energy of the K shell electron in carbon, the predominant species in most resists, is only 7 eV. Thus, there are no proximity effects with 4.5 nm photons. In the late 1970s, Flanders demonstrated the replication of 18 nm lines and spaces in PMMA using x rays of 4.5 nm, with the mask in contact with the resist.²⁵ The absorption in resist of 4.5 nm photons can be easily increased above that of PMMA by adding elements other than H or C, i.e., absorption can be tailored as lithographic considerations require (e.g., to absorb 50% in 100 nm).

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