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Material Optimization for Hard X-Ray Fresnel Zone Plates

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Abstract

Fresnel zone plates have recently been used as the focusing optic for hard x-ray (5-11 keV) microscopy techniques. Fresnel zone plates used in the hard x-ray regime focus by constructive interference effects based on the phase modulation of the incident x-ray beam and have experimentally been shown to focus 20-30% of the incident photons to less than a one-micron focal spot. The materials of choice for these zone plates have been Al, Cu, Ni, and Au. The focus of this work is the theoretical optimization of the focusing efficiency of phase-modulating Fresnel zone plates in the hard x-ray regime by appropriate material selection. The optimal materials for three different energy ranges will be examined (1-5 keV, 5-20 keV, and > 20 keV) and a discussion of the selection criteria involved will be presented. The current zone plate fabrication techniques will be discussed as they pertain to the physical aspects of the zone plates such as thickness, finest zone width, and aspect ratio.

1. INTRODUCTION

Fresnel zone plates focus by constructive interference effects due to the modulation of the amplitude, phase, or a combination of both of an incident wave. Amplitude zone plates are constructed with alternating opaque and transparent zones that introduce a periodic modulation to the amplitude component of an incident wave but do not change the phase of the transmitted wave. A phase zone plate varies both the amplitude and the phase of an incident wave and can be constructed by two different methods - alternating material and transparent zones or by alternating zones of two different materials. The amplitude modulation from a phase zone plate reduces the focusing efficiency of the optic due to attenuation effects within the constituent material(s) and should be minimized.

The maximum theoretical focusing efficiency for an amplitude-modulating zone plate is approximately 10% due to the fact that 50% of the incident wave is lost to absorption within the opaque zones. The maximum theoretical focusing efficiency for the described phase-modulating zone plate profile is approximately 40% as the previously opaque zones now contribute to the image. Therefore, as this study is concerned with optimizing the focusing efficiency only phase-modulating zone plates will be considered and will be referred to as a ZP for brevity.

2. FABRICATION

A ZP must introduce an $N\pi$ phase modulation, where N is a nonzero odd integer in order to satisfy the constructive interference criterion. The phase modulation is controlled by the thickness of the ZP material(s), and an equation that defines the phase

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difference (ϕ) introduced between the same thickness of two different materials is given by

$$\phi = k t |\delta_1 - \delta_2|, \quad (1)$$

where k is the incident photon wave number ($2\pi/\lambda$) and δ is the real component of the materials complex index of refraction. The required zone plate thickness as given by equation 1 is an important fabrication parameter and can determine the feasibility of a ZP design.

There are two techniques currently available for the fabrication of hard x-ray Fresnel zone plates: sputter/slice [1] and x-ray lithography [2,3]. The sputter/slice technique uses two different materials, which are deposited onto a wire substrate by magnetron sputtering in alternating zones. The wire is then sliced and polished to the required thickness to achieve a π phase shift between the two different materials. The x-ray lithography technique requires the deposition of a photo sensitive material that is exposed to x-rays through a negative mask of the ZP. The material is then applied by an electroplating technique to the required thickness.

The three fabrication parameters that have the greatest effect on the performance (spatial resolution and focusing efficiency) of a zone plate are thickness, uniformity of the thickness, and finest zone width. The first two parameters affect the focusing efficiency while the finest zone width determines the intrinsic spatial resolution of the optic. The ratio of the thickness to the finest zone width is called the aspect ratio and will also be used as a fabrication parameter.

The sputter/slice technique has the advantage of theoretically providing finest zone widths on the order of tens of angstroms but has a problem with the thickness and the uniformity of the optic. Current Al/Cu sputter/slice zone plates have been reproducibly polished to a thickness of approximately 18 microns, which will provide a 3π phase shift for 8 keV photons. The polishing technique used has not been able to achieve the required 4 microns for a π phase shift. A larger phase shifting optic will result in a lower focusing efficiency due to increased attenuation effects within the materials. The current polishing technique also introduces a wedge shaped figure to the optic, which varies the thickness by approximately 1.0 micron. A nonuniform thickness results in a variation in the phase shifting power of the optic, which will lower the efficiency of the device. The registry of the zones has also been a problem with this technique. If the radial position of the zones is not accurate, then the higher number zones will not contribute to the image.

The lithographic technique has the advantage of a stronger development program as an application in different fields (e.g. semiconductor design). Therefore, the thickness and uniformity of a ZP can be controlled to high tolerances ($<1\%$). A problem with the lithographic technique is that the finest zone widths available are on the order of 0.2- 0.4 micron due to difficulty in manufacturing the masks. Currently, the lithographic technique is also limited to an aspect ratio of approximately 14 for a finest feature size of 0.4 micron. Therefore, the aspect ratio will limit the maximum ZP radius achievable, which determines the active area of the zone plate that will contribute to the image. The aspect ratio can determine the viability of a given ZP design based on the number of zones that can be fabricated, which can severely limit the incident flux and therefore the flux in the focal spot regardless of the optics focusing efficiency.

The x-ray microscopy project at the Advanced Photon Source (APS) has concentrated on lithographically fabricated zone plates because of their availability and

improved structural characteristics as compared to the sputter/slice optics. Therefore, this study will only consider lithographically fabricated zone plates.

The selection of materials for these zone plates will be examined for three different energy ranges: 1 - 5 keV, 5 - 20 keV, and > 20 keV. These energy ranges were selected for a number of reasons. The low energy range was selected to determine the feasibility of performing experiments (fluorescence, XAFS, and diffraction) on the SRI Cat sector II beamline of the APS, which is designed to provide high brilliance photons between 0.5 - 3.0 keV. The classical range of 5 - 20 keV was selected as it is the regime in which most spectroscopy and diffraction studies are performed. The high energy range was chosen to determine the feasibility of performing energy spectroscopy (fluorescence, XAFS, and DAFS) studies on the K absorption edges of high Z materials ($Z > 42$).

3. DISCUSSION

The theoretical focusing efficiency of a ZP can be calculated by^[4]

$$F_m = \left(\frac{1}{m\pi} \right)^2 (1 + \gamma^2 - 2\gamma \cos(kt\delta)) , \quad (2)$$

where m is the focal order and t is the thickness of the optic along the transmission direction. A material absorption term γ is given by

$$\gamma = \exp(-kt\beta) , \quad (3)$$

where β is the imaginary component of the complex index of refraction. The ZP parameters that affect the focusing efficiency are the thickness and the materials complex index of refraction (δ and β). A zone plate's thickness is determined by the energy at which the optic was designed to have a maximum focusing efficiency, that is, provide a π phase shift. The ratio of the complex to the real component of the index refraction (η) has been shown^[5] to indicate the relative theoretical focusing efficiency of a ZP; the smaller the η , the larger the theoretical focusing efficiency.

The energy dependence of η for Be, Al, Ni, and Au between 1-30 keV is shown in figure 1. In general, η for a lower Z material will be smaller than that of a high Z material except in the energy region near a material's absorption edges. The value for η will have a substantial increase at a material's absorption edge and then will decrease gradually due to the nonlinearity of β near an absorption edge. This indicates that materials should not be used for zone plates optimized for energies greater than their absorption edges, as a high Z material can provide a higher focusing efficiency.

The plots shown in figure 1 indicate that lower Z materials such as Be and Al should be preferred over higher Z materials such as Ni and Au, but the energy dependence of the required ZP thickness has not yet been discussed. The energy dependence of the optimum theoretical focusing efficiency for these materials over the same energy range are shown in figure 2. The focusing efficiencies are calculated using equation 2 but allowing the thickness to vary to maintain a π phase shift over the entire energy range. The required material thicknesses to maintain a π phase shift are also shown in figure 2.

In general, a material's phase shifting capability is proportional to its density. Therefore, as shown in figure 2, while Be and Al have higher theoretical focusing efficiencies than Ni and Au, the required thickness for a π phase shifting Be or Al zone plate can be very large for energies greater than 5-10 keV. The current aspect ratio of 14 for the lithographic technique will severely limit the radius of such a ZP thereby reducing the flux in the focal spot; eliminating any advantage of a marginally (3-5%) higher focusing efficiency. This would indicate that due to fabrication limitations higher Z materials should be used for zone plates optimized for higher energies.

The plots shown in figure 2 are for theoretically optimized focusing efficiencies, but real optics must have a fixed thickness. Using the criteria introduced in the previous sections, low Z materials for low energies, high Z materials for high energies, and avoiding material absorption edges below the zone plate's optimized energy, let us examine the three energy ranges of interest.

In the low energy range between 1-5 keV, Be and Ti appear to be good choices because their K edges are at 0.1 and 5.0 keV, respectively. The calculated focusing efficiencies for Be, Ti, and Ni zone plates optimized for approximately 2.0 keV are shown in figure 3; the Ni ZP is shown to illustrate the focusing efficiency of a material with a much higher K edge (8.3 keV). The focusing efficiencies for the higher Z materials are lower as expected, but the maximum focusing efficiency of the Ni ZP is still approximately 27%. An interesting feature illustrated in figure 3 is that, in energy regions not near a material's absorption edges, the energy dependent focusing efficiency is a smoothly varying function.

The classical energy range from 5-20 keV provides a wider choice of materials but utilizes essentially the same criteria over the entire regime. We will examine a ZP optimized for 10 keV to illustrate the selection criterion in this energy range. The four materials chosen were Ti, Cu, Zn, and Au, as each represents a different material property: a K edge a few keV below, a K edge less than 1 keV below, a K edge 1 keV above, and a K edge many keV above (but with L edges a few keV above) the optimized energy of 10.0 keV. The calculated focusing efficiencies for zone plates fabricated with these materials are shown in figure 4. The two interesting features shown in figure 4 are the effects of the absorption edges and the dynamic range over which a substantial focusing efficiency can be maintained.

The K edges of Cu and Zn both reduce the dynamic range of the ZP. The energies greater than the K edges have their focusing efficiencies reduced due to attenuation effects within the materials. The same effect can be seen due to the L edges of the Au ZP. The closer an absorption edge is to the optimized energy, the smaller a zone plate's dynamic range.

The two materials (Ti and Au) with K edges over 4 keV from the optimized energy provide the largest dynamic ranges. The focusing efficiencies of the Ti and Au zone plates remain over 30% from approximately 7.5-12 keV and 8-11 keV. A problem with the Ti ZP is the required thickness of 6.4 microns, which will limit the radius of the optic due to the current aspect ratio of the lithographic technique.

The material selection criteria for the high energy range will be identical to that of the classical energy range with one exception: there will be fewer materials available that can be used by the lithographic technique. We will illustrate this energy region by calculating the focusing efficiency for Mo, Ag, and W zone plates optimized for 30 keV. The calculated focusing efficiencies for these zone plates are shown in figure 5. The interesting feature of figure 5 is that each ZP can maintain a focusing efficiency above 30% over a 10 keV dynamic range. The problem with the Mo and Ag zone plates is once again the required thickness of 10 microns.

4. CONCLUSION

The theoretical focusing efficiencies of a number of materials for the energy range between 1-30 keV have been examined. The material selection for an x-ray lithographically fabricated ZP has been examined based on the criteria of maximum focusing efficiency and the current lithographic aspect ratio. The results of this study are that while lower Z materials have theoretically higher focusing efficiencies, the required optic thicknesses preclude the use of the current lithographic fabrication techniques. Material selection should be based on the energy range within which the optic is designed to operate with the criterion that the material should not have an absorption edge directly below the optimal energy. Lower Z materials such as Ti and Be should be used for optics designed to be used between 1-5 keV to minimize the reduction of the focusing efficiency due to attenuation effects within the material. Higher Z materials could also be used if one is willing to sacrifice focusing efficiency for ease of fabrication. The material selection for higher energies should be based on the location of absorption edges. Figure 4 illustrates that, for higher energies, the material chosen should have its K edge well removed from the optimized energy. However, the current lithographic fabrication technique precludes the use of lower Z materials due to the aspect ratio problem.

5. ACKNOWLEDGMENTS

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Figure Captions

- Figure 1. Calculated values of the energy dependence of η for Be, Al, Ni, and Au.
- Figure 2. Calculated values for the optimal theoretical focusing efficiency for zone plates fabricated with Be, Al, Ni, and Au.
- Figure 3. Calculated values for the focusing efficiency of Be, Ti, and Ni zone plates optimized for 2.0 keV.
- Figure 4. Calculated values for the focusing efficiency of Ti, Cu, Zn, and Au zone plates optimized for 10.0 keV.
- Figure 5. Calculated values for the focusing efficiency of Mo, Ag, and Au zone plates optimized for 30.0 keV.

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