Fabrication of x-ray zone plates by surface-plasma chemical vapor deposition

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A new cost-efficient sputter-slice technology for hard x-ray (10–30 keV) Fresnel zone plates fabrication, imposing no limitation to aspect ratio, is proposed. By means of a plasma chemical process, SiO2/SiGeO2 glassy film multilayer structures are deposited on a lateral surface of a silica rod, outermost layers being as thin as 100 nm. It has been shown by numerical simulation that for \( x = 0.2 \) germanium fraction, 100–300 \( \mu \text{m} \) zone plate thickness and the number of zones of about 1000, first order diffraction efficiency as high as 20%–30% at the energy of approximately 20 keV can be achieved. © 2007 Optical Society of America


Fresnel x-ray zone plates (FXZP) are x-ray optical elements, which are widely used for x-ray imaging and sharp focusing of x-rays in the wavelength range from soft (\( \lambda = 46.9 \text{ nm} \)) [1,2] to hard (energy > 10 keV) [3] radiation. These optical elements have been extensively studied for the past 25 years, but still remain rather expensive and difficult to manufacture.

A typical FXZP is a set of concentric rings (zones) with alternating refractive indices or absorption coefficients. Formation of an image occurs due to coherent summation of the radiation passing through the transparent zones [4]. For a given wavelength \( \lambda \), zone radii \( r_n \) must obey the relation

\[
r_n = \sqrt{r_0^2 + \lambda F n}, \quad n = 1, 2, \ldots, N,
\]

where \( r_0 \) is the radius of the central zone, \( F \) is the focal distance, and \( n \) is the zone number. Relation (1) provides constructive interference at the focus due to equality of the optical paths and independence of the zone areas on the zone number. Spatial resolution of the FXZP (or the size of its focal spot) is determined by the width of the outermost zone [4]. The best spatial resolution obtained by soft x-ray FXZP used for scanning and imaging microscopy in the so-called “water window” (2.4 < \( \lambda < 4.4 \text{ nm} \)) is about 20–50 nm [5].

The main technology currently used for the production of zone plates is e-beam lithography [4]. This method enables the fabrication of FXZP with external zone thicknesses down to 10 nm and numbers of zones \( N \) up to 10^4. The main disadvantages intrinsic to this technology are limited aspect ratio (FXZP thickness/zone width), typically not exceeding 10–20, and a rather high fabrication cost. So-called sputter-slice technique [6] provides a partial solution to these problems. Under this technology, FXZP are made as transverse slices of a filament laterally covered by properly structured layers of alternate composition. This multilayer coating is deposited by magnetron sputtering from two targets of corresponding materi-
als. Although this technique imposes no limitation on the aspect ratio, a rather accurate filament rotation and a stable sputtered flow are necessary to deposit uniform and axially symmetric layers of predefined thicknesses.

We modified the sputter-slice technology by substituting magnetron sputtering with the surface-plasma chemical vapor deposition (SPCVD). Such a process was first applied to glass deposition on the inner surface of a substrate tube in the technology for optical fiber preforms fabrication [7]. Later, a similar process was also applied to the films deposition [8] and multilayer structure formation for antiresonance reflecting optical waveguides [9].

The SPCVD setup used in our experiments is shown in Fig. 1. A 40 cm long plasma column is sustained in a silica tube (inner diameter being 16 mm, and wall thickness = 2 mm) by the excitation of microwave induced surface plasma waves by means of a launcher. A 2.45 GHz, up to 5 kW, magnetron is used as a microwave oscillator. In such a configuration, stationary glow discharge is located in the vicinity of the inner surface of the tube, thus forming an axially symmetric source of active agents’ species created in plasma from the gas molecules passing through the tube.

In the course of the deposition process, oxygen, together with vapors of silicon and germanium tetrachloride, is fed toward the plasma column through mass flow controllers (Fig. 1), total pressure of the gas mixture in the tube being several torr. Entering the plasma region, chlorides are transformed into oxides, forming a uniform omnidirectional flow of silicon and germanium oxide molecules as well as oxygen and chlorine atoms. A section of the tube approximately several cm in length at the top of the plasma column is the deposition zone, where the oxide molecules created in plasma are being adsorbed by the surfaces of the tube and the substrate inserted into it. In our experiments, a silica rod of approximately 0.5–5 mm in diameter mounted on the axis of the tube in the deposition zone is used as a substrate for subsequent FXZP fabrication. When passing through the tube with the surface plasma, silicon and germanium containing species disappear, first being transformed to oxides, and then adsorbed by the surfaces in the deposition zone. As a consequence, the released chlorine and the oxygen excess are the only plasma forming gases, which pass through the rest of the tube toward the pump inlet (Fig. 1).

The SPCVD process we outlined is characterized by a film deposition rate as high as several microns per minute. The necessary variation of glass composition in the neighboring Fresnel zones occurs due to alternately switching germanium doping of the deposited silicon dioxide on and off. This process is computer controlled by temporally varying the germanium tetrachloride portion in the input gas mixture. As the time constant for the corresponding flow controller used in our experiments is as small as 1 s, a rather thick net glassy coating, compositionally structured along the radius at a scale of hundreds of nanometers, can be synthesized at high deposition rates. This is illustrated by Fig. 2, where scanning electron microscopy (SEM) pictures of a transverse slice of the deposited structure are shown at two various magnifications.

In the 10–30 keV range, the first order diffraction efficiency of thus made FXZP calculated by the analytical theory [10] lies in the 10%–20% range, if
The optimal zone plate thicknesses for wave field amplitude (arbitrary units).

SiO$_2$/Si$_{0.8}$Ge$_{0.2}$O$_2$ glass composition is used for zones structuring. The optimal zone plate thicknesses for the 10–30 keV x-rays are about 100–300 µm.

To evaluate focusing performance of this type of FXZP, numerical simulation of the wave field pattern, based on the parabolic wave equation [11], was carried out. To reduce computational time required for the simulation, the number of layers $N$ and the central rod diameter of the FXZP were scaled down to 100 and 0.8 mm, respectively. The results of this simulation are depicted in Fig. 3. One can see an elongated first order focus at a distance of about 1100 mm.

In summary, a novel type of technology for Fresnel x-ray zone plates fabrication is sketched in this paper. The technology is based on the surface-plasma chemical vapor deposition process initially developed for optical fiber preforms synthesis. The first samples of zone plates fabricated by this method show quite regular layered structures, down to 100 nm layer thickness, obtained at a very high deposition rate. This enables low-cost manufacturing of high aspect ratio FXZP with the number of zones greater than 1000.

Numerical simulation of the FXZP fabricated by SPCVD has been carried out. It demonstrates the ability of thus made FXZP to focus 10–30 keV x-ray radiation. The next immediate step is to check the predicted characteristics at a synchrotron radiation facility. The suggested technology promises a dramatic cost reduction of the imaging optical elements in the hard x-ray spectral region, as well as improvement of their diffraction efficiency while maintaining spatial resolution high enough for application in x-ray microanalysis, x-ray microscopy, and focusing of x-ray radiation.

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References