Toward one nanometer X-ray focusing: a complex refractive lens design

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We report a design for one nanometer X-ray focusing by a complex refractive lens, which is capable of focusing 20 keV X-rays down to a lateral size of 0.92 nm (full-width at half-maximum (FWHM)) and an axial size of 98 nm (FWHM) with intensity gain of 49050. This complex refractive lens is comprised of a series of kinoform lenses, whose aperture is gradually matched to the converging trace of the X-ray beam so as to increase the numerical aperture (NA). The theoretical principle of the proposed complex refractive lens is presented. The NAs of these lenses are calculated. The numerical simulation results demonstrate that the proposed design can focus the X-ray beam into sub-nanometer while remaining high gain.

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X-ray nanoprobe can provide local information at nanoscale, which will benefit investigation on the nanoscale inhomogeneous samples. Nowadays, X-ray nanoprobe plays an important role in many research fields, ranging from materials science, to geophysics and environmental science, to biophysics and protein crystallography[1–5]. Toward focusing X-ray beam to nanoscale efficiently, great efforts have been paid. During the past decade, mirrors[6], zone plates[7], Laue lenses[8,9], and refractive lenses[10] have all been employed to focus the X-ray beam into a lateral size well below 50 nm. However, all these designs encounter problem in focusing the X-ray beam into less than 1 nm. It is of particular interest that whether focusing X-rays to atomic dimensions is possible.

Although X-ray focusing devices were formerly limited mainly by the perfection of the fabricated focusing structures, recent focusing optics has become more fundamentally constrained by the diffraction limit[1]. While X-rays have very short wavelengths, the long-standing difficulty in decreasing Airy disc size has been achieving a larger numerical aperture (NA)[11]. Owing to the tiny deviation of the refractive index from unity, $\delta = 1 - n$, in the X-ray regime, a NA limit with the critical angle $\theta_c = \sqrt{2\delta}$ for all single X-ray optics has been known for a long time[12,13], which limits the lateral focus size to 10 nm. Today, the 10 nm barrier in X-ray focusing has been broken by the combination of discrete devices[6,7]. Furthermore, a few designs toward sub-5 nm focusing of X-ray beam have been demonstrated[14–16]. However, till now, no experiment or design result shows that focusing X-ray beam to 1 nm is possible.

At present, X-ray refractive lens is one of the most popular X-ray focusing devices for synchrotron beamlines[17–21] due to its easy alignment and high efficiency. Previously, it was thought that refractive lens for X-rays are not feasible[22–24] until 1996 when Snigirev *et al.* invented the first X-ray focusing lens[25]. Nowadays, these so-called compound refractive lenses (CRLs) have been significantly improved by several ingenious designs[26–29], which is usually comprised of many refractive lenses integrated on one substrate by lithography and etching[30]. However, due to the strong absorption effect at the edge of the lens, it is hard to increase the NA of such a lens to focus X-ray beam into 1 nm.

To reduce the absorption effect and increase the NA, two approaches are practical. One of the design is to match the lens aperture gradually to the size of the beam, following the trace of the X-ray focusing[15]. The other is to use the kinoform-lens design by removing all the materials causing redundant 2π phase shifts in an individual lens without changing its overall focusing properties[16,28,31]. However, these two techniques independently cannot improve the NA large enough to meet the requirements for focusing the X-ray beam into 1 nm. In this letter, we consider a design by combining the complex kinoform lens with scaled reduction of curvature radii of individual lenses, which has the potential to increase the NA of the lens system and focus X-ray beam into atomic dimensions.

The proposed design for the CRL is shown in Fig. 1, which comprises a series of kinoform lenses. To avoid spherical aberration, each individual lens has a spherical shape. According to paraxial approximation, the

parabolic curvature of the lens is employed, in which the parameters are noted as length l_i, minimum thickness d_i, semi-aperture R_{0i}, and radius of curvature at the apex R_i. In order to achieve the optimal refractive power for each individual lens in the stack, the semi-aperture R_{0i} is set to the local beam size at the outline of the converging wave field. For the sake of convenience, this complex refractive lens is called gradually matched focusing lens (GMFL). In the following, the parameter optimization of a GMFL for one nanometer focusing will be carried out in two steps. At the first step, parameters of the GMFL composed of conventional refractive lenses are optimized to get the highest NA, with ratio of the radius of curvature to its semi-aperture keeping constant for all the lens elements. Then, kinoform lenses are employed to substitute the conventional refractive lenses to reduce the absorption effect and consequently further increase the effective aperture of the GMFL.

As shown in Fig. 1, the gradually matched aperture R_{0i} of the ith lens along the optical axis of the GMFL can be obtained based on geometric optics. Due to the fact that the refraction of each lens is tiny and the number of lenses N is large, the ray path inside the lenses can be determined according to the transfer matrix formalism in the continuum limit[32]. The vertical size $R_{0(z)}$ from outermost ray to the optical axis, defines the aperture of each lens as a function of distance along the optical axis and is given by

$$R'_{0(z)} = 2\sqrt{\delta'}\sqrt{\log\left(R_{01}\Big/R_{0(z)}\right)}, \tag{1}$$

where R_{01} is the semi-aperture of the first lens, $R_{0(z)}$ is the vertical size from outermost ray to the optical axis as shown in Fig. 1, and $\delta' = \left(1 - d_i/l_i\right)\delta$. The NA of the GMFL for a distant source is given by[11,15]

$$\mathrm{NA} = \sqrt{\delta'}\sqrt{\left(4a\Big/R_{01}\right)\left[1-\exp\left(-R_{01}\Big/a\right)\right]\left[\log\left(R_{01}\Big/R_{0e}\right)\right]}, \tag{2}$$

where R_{0e} is the radius of the last lens of the GMFL, $a = 4\sqrt{\delta'}\Big/\left(\sqrt{\pi}\mu'\right)$ is defined as a material specified characteristic aperture, where $\mu' = \left(1 - d_j/l_j\right)\mu$, μ is the linear adsorption coefficient of the lens material. According to Eq. (2), NA is a function of R_{01}, R_{0e}, a, and δ', where a depends on d_j/l_j, δ, and μ, and δ' relies on δ and d_j/l_j. Hence, in our design, R_{01}, R_{0e}, and d_j/l_j have to be optimized to achieve the largest NA, after material for the lens is selected. For example, to achieve the focus spot size of 1 nm (full-width at half-maximum (FWHM)), NA has to be larger than a threshold value, such as 0.02325 at 20 keV according to the formula[11] $dt = 0.75\lambda\Big/\left(2\mathrm{NA}\right)$. In the following, the parameters of the GMFL are optimized to achieve the critical value of NA for the one nanometer focusing.

First of all, the kind of material for the lens should be selected to get the relatively large NA, meanwhile remaining lowest absorption effect. Setting the initial values of the lens, including $R_{01} = 0.5$ mm, $d_i/l_i = 1/10$, and $R_{0e} = 100$ nm, which are commonly used for CRLs design[15,26], NA is calculated according to Eq. (2) for different lens materials, which are commonly used materials for CRLs including Ni, Si, Al, graphite, and diamond. According to simple calculation results, it is easy to know that diamond is the best choice for the materials of the GMFL with the largest NA, among the known lens materials, due to its relatively lower atomic number and higher mass density. Therefore, diamond is selected for the further design.

However, the NA of the common GMFL made of diamond is 0.002234 at 20 keV, only about one-tenth of the threshold value, which means that further efforts have to be paid to achieve larger NA. According to Eq. (2), we still have room to increase the NA by optimizing the three parameters R_{01}, R_{0e}, and d_i/l_i. By changing one of the three parameters with the other two fixed, the optimized NA can be obtained. For example, R_{01} is changed in the range of 500 nm $\le R_{01} \le$ 0.8 mm, setting $d_i/l_i = 1/10$ and $R_{0e} = 100$ nm, and then the maximum value of NA = 0.005291 can be found at $R_{01} = 0.016$ mm. Further optimization results show that NA grows with decreasing R_{0e} and d_i/l_i. Therefore, both R_{0e} and d_i/l_i should be chosen as small as possible. Considering the state of the art of nanofabrication techniques, $R_{0e} = 50$ nm and $d_i/l_i = 1/20$ are selected in this design. With these optimized parameters, the maximum NA of the GMFL can be improved to 0.005777, which is almost the upper limit of the conventional design, but still about a quarter of the threshold value required by the one nanometer X-ray focusing. This improvement of the NA value is constitutionally constrained by X-ray absorption at the

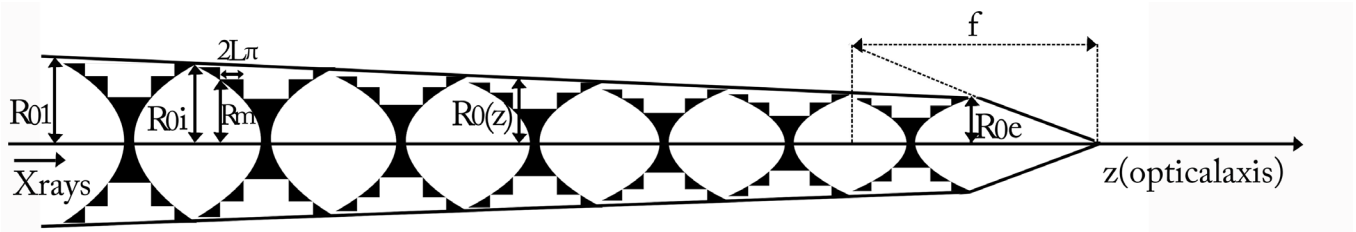

Fig. 1. Schematics for the gradually matched X-ray focusing lens.

outer range of the lens. Therefore, the critical point for further optimization is to reduce the absorption effect.

Usually, NA grows with increasing geometric aperture of the lens system. However, due to the strong adsorption effect of X-ray in the outer range of the focusing lens, the effective aperture is limited intrinsically by the optical path length at the outer ring of the lens. Aiming at this problem, similar to optics in visible light range, kinoform lens design should be the right solution as shown in Fig. 1. The radius of the segment of the kinoform lens can be expressed as[33]

$$R_m = \sqrt{2m\lambda f_i},\ m = 1,2,3..., \tag{3}$$

where m is the segment number, λ is the X-ray wavelength, and f_i is the focal length.

By lessening the path length of X-ray in the kinoform lens design, absorption effect can be reduced especially at the outer range of the lens, which means that improvement of NA is practical. According to Eq. (3), by manifolding number of segments of the kinoform lens, R_{01} can be effectively increased. For example, R_{01} of kinoform-GMFL with four segments is 0.047 mm and that for eight segments is 0.067 mm. When 12 segments are employed, R_{01} of the GMFL is 0.082 mm and correspondingly the NA of 0.0241 is achieved larger than the threshold value mentioned above. This means that one nanometer X-ray focusing is possible by means of combining the GMFL with kinoform lens.

To confirm the proposed design, the wave propagation through the GMFL was simulated numerically based on the wave optics with the Fresnel–Kirchhoff approximation[34]. The incident X-rays are assumed to be monochromatic wave with unit complex amplitude from a distant point source. This wave field then propagates through each individual lens of the GMFL step by step, with the focused beam from the former lens acting as the source of the next one. The process above is repeated until the last individual lens is reached and finally the wave field at the focus plane is obtained.

As shown in Fig. 2(a), the lateral focused beam sizes for the common GMFL, GMFL-4segs, GMFL-8segs, and GMFL-12segs are 4.233, 1.568, 1.115, and 0.92 nm (FWHM), respectively. Figure 2(a) also shows that the peak intensity gains are 5101, 23100, 37520, and 49050, correspondently. Due to absorption effect, the transmission efficiency of GMFL-12segs is 30.97%. Nevertheless, the peak gain at the focal plane can still reach 49050, which is much higher than that of the pure CRLs design[21]. Axial beam profiles at the focal position for the GMFL are also given in Fig. 2(b), which is the so-called focus depth, and the value is 0.002 mm, 300, 140, and 98 nm (FWHM) for common, 4segs, 8segs, and 12segs, respectively. As shown in Fig. 2(b), the axial beam size decreases quickly with increasing number of segments. However, about 100 nm focus depth can still be achieved for the 1 nm nanoprobe, which should be large enough for all kinds of scanning investigations on samples in broad research fields.

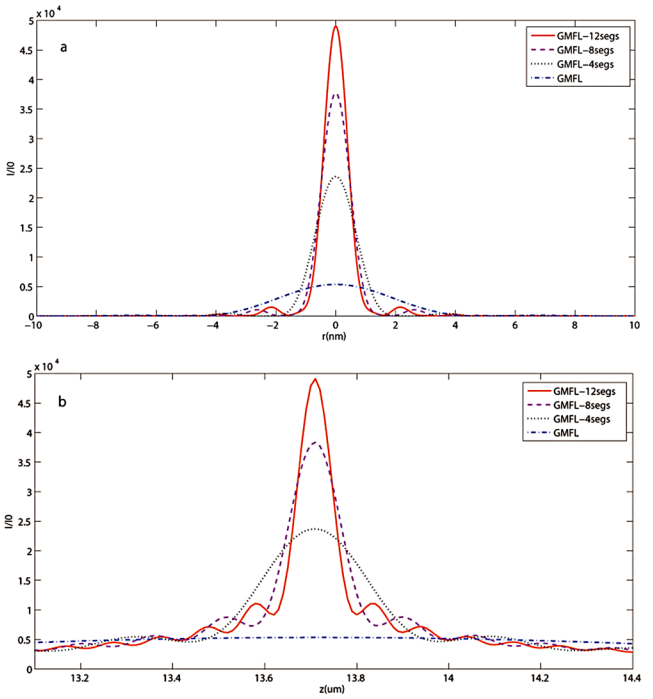

Fig. 2. Focusing properties for the kinoform combined GMFL, where (a) lateral beam profiles and (b) axial beam profiles at the focal position for a GMFL, GMFL-4segs, GMFL-8segs, and GMFL-12segs, respectively.

In conclusion, we propose a new design toward 1 nm X-ray focusing by combining GMFL with kinoform lens. Firstly, GMFL is employed to achieve larger NA than the conventional CRLs. Then, the GMFL is combined with kinoform lenses, and the NA can be significantly improved by lessening the absorption effect at the outer range of the individual lens. As a result, large enough NA can be achieved to focus the X-ray beam down to 1 nm with a relative high focus depth while remaining high peak intensity gain, as demonstrated by the simulation results. In addition, for conventional X-ray refractive lens at the high energy range of the spectrum, Compton scattering usually contributes a lot to the background. However, the kinoform lens is almost transparent, Compton scattered disturbance in the focusing plane shall be far less than that of CRLs. The proposed design can be a practical approach for focusing X-ray beam to atomic dimensions. State-of-the-art electron beam lithography and ion etching techniques should be able to fabricate this kind of lens system.

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